A new method to analyze and understand molecular linear and nonlinear optical responses via Field-Induced Functions. A straightforward alternative to Sum-Over-States (SOS) analysis

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Abstract

Sum-Over-States (SOS) method allows the computation of polarizabilities and hyperpolarizabilities, additively, from the contributions of different electronic excited states in a given molecule or cluster. Subsequent analysis of the main excited configurations contributing to the relevant excited states allows characterizing the orbitals involved in the linear and nonlinear optical response. Unfortunately, the chemically relevant information that can be obtained by SOS is hindered by a series of methodological and computational drawbacks. Among these drawbacks, we can remark the large computational cost, problems arising from nonconvergent series and errors caused by inaccurate description of excitation energies and/or higher excited state matrix elements. For this reason, coupled-perturbed schemes are currently widely used to determine the NLO potential of molecules and materials. However, such a choice limits the amount of intuitive chemical information that, on the other hand, can be retrieved by a successful SOS computation. In this work, we present and discuss a novel computational strategy that offers the means to extract the useful chemical insights from a coupled-perturbed calculation at almost negligible extra computational cost providing

a transparent picture about orbital contributions on the properties of interest. The proposed method is based on the generation and further analysis of field-induced orbitals, FIOs, from the analytic or numerical derivatives of the dipole moment. Orbital symmetry rules are derived using group theory and the method is tested for a series of small and medium size systems.

I. Introduction

The nonlinear optical susceptibilities of molecules are routinely studied within a robust semiclassical theoretical framework which describes the nonlinear dependence of the molecular dipole moment (or the molecular energy) on an external electric perturbation.[1-4] In this context, higher order properties referred as electric dipole (hyper)polarizabilities[5] play the pivotal role and they are routinely employed as qualitative indices for the establishment of comprehensive structure-property relations to be exploited in the development of new nonlinear optical (NLO) materials.[6-11] In the realm of NLO materials, built from molecules of high nonlinear optical responses, current theoretical investigations mostly focus on the quantitative determination of the relevant (non)linear optical coefficients related to the electronic part of the response. Owing to the vast development of *ab initio* and density functional methods that treat, in principle, electron-electron coulomb interactions, a vast majority of these studies rely on coupled-perturbed[9-14] Hartree-Fock or Kohn-Sham approaches which have become available in most of the commercial quantum chemical codes. Such methods provide analytic (hyper)polarizability values of negligible numerical errors as compared, for instance, to finite field self-consistent field (FF-SCF) techniques[15] (see also ref [16] and refs therein), the application of which is currently limited to computations performed in molecules of small sizes at post-HF levels (MPn, CC) due to considerable computational cost. Provided that the suitable method/basis set is used, such approaches are able to deliver reliable static and/or dynamic (hyper)polarizability results as compared to the experiment, but they offer no information about crucial intramolecular physical processes from which the molecular (non)linear optical responses stem. To overcome this limitation, (hyper)polarizability investigations are frequently complemented by qualitative or semiquantitative studies within the sum-over-states (SOS)[17-19] perturbative

approximation. SOS relies on the description of the perturbed electronic wave function in terms of all eigenfunctions of the unperturbed Hamiltonian. In such a manner, any (hyper)polarizability tensorial component can be computed as a sum of infinite terms comprising excitation energies and dipole transitions. Few decades ago, this treatment, coupled with Configuration Interaction (CI) schemes or semiempirical wave functions, was the method of choice also in the quantitative prediction of the NLO properties of molecules.[4] Nowadays, the application of SOS is mostly restricted to qualitative explorations[20,21] of predominantly contributing electronic excitations on the (non)linear optical properties of interest. Such a choice stems from the success of few-state variations of the SOS approximation in the prediction of NLO responses of molecules featuring low-lying quantum transitions associated to "long-distance" charge transfer across their molecular structure (as for instance in donor-acceptor chromophores).[4] In such cases, the SOS series converge rather fast to the correct value of the first hyperpolarizability returning useful structure property relations which can be generalized through carefully adapted semiempirical few-state SOS models.[4,22-24] On the other hand, for systems of large sizes and complex structures in which more than few transitions contribute in their NLO responses, few-state SOS models do not guarantee the complete resolution of the underlying NLO intramolecular mechanism. Therefore, one has to perform computations across a large number of excited states, by means of CI or time-dependent methods, to determine a sufficient number of higher matrix elements, paying a high price in computational resources with no guarantee that useful intuitive data can be retrieved especially in cases of nonconvergent series. A significant lowering of the computational cost could be achieved through the so-called uncoupled HF approach (UCHF),[25] in which the excited state wave-functions are approximated by Slater determinants formed from Hartree-Fock (or Kohn-Sham) orbitals. In this case, the energies of each "state" are taken

as the sum of the orbital energies of each determinant. However, in UCHF/SOS the field-induced effects stemming from electron-electron interaction are not taken into account. As a result, significant overestimations of the differences between the excited states are delivered which, in turn, lead to underestimated (non)linear optical properties and, in some cases, artifactual orbital-pair contributions.

Bearing in mind the shortcomings of the conventional application of SOS methods in the qualitative understanding of the (non)linear optical properties of molecules, in this work we propose an alternative approach based on field-induced orbitals (FIOs), taken from analytic or numerical derivatives of the molecular dipole moment with respect to a homogeneous external electric field. We show how this approach, contrary to other previously proposed orbital partitionings of the (non)linear optical properties,[26-28] decomposes the response into orthogonal parts (FIOs) built from the combination of occupied and virtual unperturbed orbitals. This leads to an arbitrariness-free assignment of the different orbital contributions, allows understanding the response properties using orbital symmetry rules and, as will be shown in the following, provides a visualization tool similar to that based on natural transition orbitals (NTOs) and employed in SOS methods.[29,20]

II. Theoretical Background

The total dipole moment of a molecule subjected to a uniform electric field is given by the following Taylor expansion,

$$\mu_\sigma = \mu_\sigma^0 + \alpha_{\sigma\sigma'} E_{\sigma'} + \frac{1}{2}\beta_{\sigma\sigma'\sigma''} E_{\sigma'} E_{\sigma''} + \ldots \quad (1)$$

where μ_σ^0 represents the permanent dipole moment and α and β the dipole polarizability and first-order hyperpolarizability, respectively. In this work we will focus only on β for the analysis of the nonlinear response, as the generalization of the present formulation to higher-order polarizabilities is trivial.

In the Taylor expansion given above, the electric polarizability, α, corresponds to the derivative of the dipole moment, μ, with respect to the electric field, E. Since this derivative depends on both the dipole moment and field directions, α is represented by a 3x3 tensor, where each component is given by the following expression,

$$\alpha_{\sigma\sigma'} = \left(\frac{d\mu_\sigma}{dE_{\sigma'}} \right)_{E=0} \tag{2}$$

Eqn (2) can be rewritten as the integral to the whole space of the product of the electron density derivative and the corresponding Cartesian component.

$$\alpha_{\sigma\sigma'} = \int \sigma \left(\frac{d\rho(\mathbf{r})}{dE_{\sigma'}} \right)_{E=0} d\mathbf{r} \tag{3}$$

Introducing a basis set to represent the electron density derivative, in our case the set of unperturbed orbitals $\{\phi_i\}$, the polarizability can be expressed in a matrix fashion,

$$\alpha_{\sigma\sigma'} = \sum_{ij} D_{ij}^{\sigma'} \int \phi_i(\mathbf{r}) \sigma \phi_j(\mathbf{r}) d\mathbf{r} \tag{4}$$

where $D_{ij}^{\sigma'}$ are the components of the first derivative of the electron density matrix with respect to an electric field applied on the σ' direction. Diagonalization of this matrix leads to a set of orthogonal eigenfunctions, $\{\zeta_k^{\sigma'}\}$, and their corresponding eigenvalues, n_k. Then, the components of the polarizability tensor can be reduced to a single summation using these eigenfunctions.

$$\alpha_{\sigma\sigma'} = \sum_{k} n_k \int \zeta_k^{\sigma'}(\mathbf{r}) \sigma \zeta_k^{\sigma'}(\mathbf{r}) d\mathbf{r} \tag{5}$$

Since these eigenfunctions are linear combinations of the set of unperturbed orbitals, we refer to them as Field-Induced Orbitals (FIOs), by analogy with the Electron Deformation Orbitals (EDOs) previously introduced to analyze electric conductivities.[30-32] As in EDOs, FIOs are grouped in pairs of functions with the same absolute eigenvalue but opposite sign. Then, the polarizability tensor of eqn (5) can be reduced to a summation over the number of different pairs,

$$\alpha_{\sigma\sigma'} = \sum_k \int \mathrm{Tr}\left(\left(\mathbf{\Theta}_k^{\sigma'}\right)^t \hat{\boldsymbol{\sigma}} \mathbf{\Theta}_k^{\sigma'}\right) d\mathbf{r} \tag{6}$$

where each pair in eqn (6) is represented by the following complex 2x2 matrix,

$$\mathbf{\Theta}_k = n_k^{1/2} \begin{pmatrix} \zeta_k^+ & 0 \\ 0 & i\zeta_k^- \end{pmatrix} \tag{7}$$

By analogy with the analysis of electric conductivity, where $\mathbf{\Theta}_k$ forms an electron transport channel,[31] herein they represent polarizability channels, whose formation in one-determinant wave functions is ruled by the following symmetry constrain,

$$\Gamma_{occ} \otimes \Gamma_{unocc} = \Gamma_{\sigma'} \tag{8}$$

Eqn (8) is rooted in group theory and restricts the combinations of occupied and unoccupied orbitals to those where the symmetry product corresponds or contains the symmetry irreducible representation (*irrep*) of the field direction. It must be remarked that, in a multideterminant wave function, eqns (5) to (8) can be applied using the natural orbital representation of the one-electron density.

Similar steps may be followed to extend this formalism to the analysis of hyperpolarizabilities. In the case of the first hyperpolarizability, β, the second derivative of the electron density with respect to electric fields applied on the σ' and σ'' directions is required,

$$\beta_{\sigma\sigma'\sigma''} = \int \sigma \left(\frac{\partial^2 \rho(\mathbf{r})}{\partial E_{\sigma'} \partial E_{\sigma''}} \right)_{E=0} d\mathbf{r} \tag{9}$$

Analogous equations to eqns (4) and (5) lead to an expression in terms of FIOs, obtained in this case by diagonalization of the second derivative of the electron density matrix.

$$\beta_{\sigma\sigma'\sigma''} = \sum_k n_k \int \zeta_k^{\sigma'\sigma''}(\mathbf{r})\sigma\zeta_k^{\sigma'\sigma''}(\mathbf{r})d\mathbf{r} \quad (10)$$

However, for β the FIOs can be grouped in pairs only when $\sigma' \neq \sigma''$, as the trace of the second derivative of the electron density matrix is not zero when $\sigma' = \sigma''$.[30] Thus, in the case of $\sigma' \neq \sigma''$, differences in the absolute eigenvalues between the FIOs of a given pair may occur. However, they can be also grouped in pairs as they still share the same orbital mixing.

FIOs allow decomposing the linear and nonlinear optical responses in individual contributions. The fact that these functions are constructed as linear combinations of unperturbed orbitals allows also quantifying the weight of each occupied and virtual orbital through the square of its coefficient in the FIOs expansions. In other words, the square of the coefficient of a given unperturbed orbital in an FIO measures its relative weight. Then, for a given FIO, k, the contribution of orbital i to the value of α may be measured as,

$$\alpha_{\sigma\sigma'}^k(i) = c_{i,k}^2 \alpha_{\sigma\sigma'}^k \quad (11)$$

By adding the whole set of FIOs we get a measure of the contribution of i to the total value of α.

$$\alpha_{\sigma\sigma'}(i) = \sum_k c_{i,k}^2 \alpha_{\sigma\sigma'}^k \quad (12)$$

An equivalent expression to eqn (12) can be written for β.

$$\beta_{\sigma\sigma'\sigma''}(i) = \sum_k c_{i,k}^2 \beta_{\sigma\sigma'\sigma''}^k \quad (13)$$

In symmetric molecules, the orbital symmetry rule given in eqn (8) allows understanding qualitatively the contributions of different occupied and virtual orbitals to the values of α and β. In the case of β, the rule is written as,

$$\Gamma_{occ} \otimes \Gamma_{unocc} = \Gamma_{\sigma'\sigma''} \quad (14)$$

where the rhs term refers to the *irrep* of the product $\sigma'\sigma''$.

III. Computational Details

To test the performance of the method presented in the previous section, the following set of molecules has been analyzed: hydrohelium cation (HHe^+), nitrogen molecule (N_2), lithium fluoride (LiF), hydrogen fluoride (HF), hydrogen chloride (HCl), water (H_2O), hydrogen sulphide (H_2S), ammonia (NH_3), fluoroethyne (C_2HF), (trifluoromethyl)acetylene (C_3HF_3), cyanoacetylene (C_3HN), 1-cyano-1,3-butadiyne (C_5HN), 1-cyano-1,3,5-hexatriyne (C_7HN), *p*-cyanoaniline ($C_7N_2H_6$), *p*-nitroaniline ($C_6O_2N_2H_6$) and *p*-methoxy-nitrobenzene ($C_7O_2NH_7$). This set covers small and medium size molecules for which optical properties were previously investigated (see for instance refs 33-37). Our intention is to test the FIOs method in model molecules of different characteristics. Therefore, we chose systems of different polarities, comprising hard/soft central atoms bonded with single/double/triple bonds and bearing aromatic rings containing activating/deactivating groups.

Calculations of α and β tensors using FIOs were performed with a FORTRAN code developed in our lab, which interfaces with the Gaussian09 suite of programs[38] to read the density matrix derivatives obtained from a coupled or uncoupled perturbed HF calculation (CPHF-FIOs or UPHF-FIOs). In order to account for dynamic correlation effects, calculations were also performed using the coupled perturbed Kohn-Sham

method (CPKS-FIOs), with the CAM-B3LYP functional.[39] The results were confronted with those obtained using the SOS method, with the excited states calculated using the configuration interaction singles approach (CIS-SOS) and the time-dependent density functional theory (TDDFT-SOS). For these calculations, the information of excited states was obtained with Gaussian09, whereas the α and β tensors were computed with the Multiwfn program.[40] All the SOS calculations were performed including a sufficient number of excited states until convergence was reached. Triple-zeta basis sets including polarization functions (6-311G(d,p)) and polarization and diffuse functions (6-311G++(d,p)) were employed for all the calculations. Even though diffuse *s*, *p* gaussian-type functions may be particularly important for the calculation of hyperpolarizabilities in molecules containing third-row and heavier elements, they were found to show a very slow convergence in the SOS calculations and, in some particular cases, caused instabilities to the ground state wave functions. Moreover, the effect of basis sets including larger polarization such as 6-311G(2d,2p) or 6-311G(3df,3pd), which are expected to improve the results for hyperpolarizabilities, was also assessed. Significant differences in β were only detected for HCl and H_2S, which are molecules built from third-row elements. Therefore, taking into account that our approach is general and not limited by the electronic structure method or the basis set size, we will discuss only results obtained with smallest basis set (6-311G(d,p)). At this point it must be noted that highly accurate hyperpolarizabilities obtained with heavily polarized basis sets is out of the scope of this work which aims at demonstrating the validity and capabilities of the FIOs analysis in comparison with SOS approximation. For the sake of completeness, results obtained including diffuse functions are given as Supplementary Material.

In this work, only static properties will be analyzed. Implementation of the CP-FIOs method for frequency-dependent polarizabilities and first-order hyperpolarizabilities,

which requires the diagonalization of nonsymmetric matrices, is currently in progress. First derivatives are obtained analytically, whereas second derivatives are computed numerically with a symmetric derivative formula, using an electric field strength of 10^{-3} au. Total values of α and β obtained from CP-FIOs (eqns (6) and (10)) were compared with the values obtained directly with the CP method using Gaussian09. Due to the use of analytical first derivatives of the density matrix, both CP and CP-FIOs values of α are the same, whereas negligible differences in β arise from the use of numerical second derivatives. These differences are indicated for each molecule in the Supplementary Information.

IV. Results and Discussion

Due to the large amount of data arising from the six and nine nonsymmetric components of α and β tensors, in this section we will discuss only the dominant components in each case that define the nonlinear response of each system considered. For all the molecules studied here, these components correspond to the orientation of the principal symmetry axis, α_{zz} and β_{zzz}. Plots representing the accumulative value of α_{zz} and β_{zzz} with respect to the occupied and virtual molecular orbitals (obtained by eqns (12) and (13)) are confronted, in this section, with those obtained with SOS in terms of excited states. These plots are included as Supporting Information for the sake of simplicity, and a general comparison is presented in the following pages. Nevertheless, some of these data are also discussed in detail in this section.

Confronting FIOs orbital plots and SOS excited state plots reveals a series of advantages of the former. The first one and, for obvious reasons, the most important one, is that the FIOs analysis is performed on the base of coupled perturbed theories, so

that the total values of α and β are expected to be more accurate than those obtained from a truncated SOS series.

The second advantage is that convergence to the final property value with the number of orbitals is much faster than with the number of excited states. This is obvious since the contributions of a given MO in different excited states using SOS are gathered in just one using FIOs. It must be noticed that, even though convergence in SOS is reached for all the molecules investigated here, this is not always the case for larger molecules, the second nonlinear optical responses of which are defined more than few dominant charge transfer excited states. On the contrary, the fast convergence in the CP-FIOs calculations allows truncating the analysis to a reduced set of occupied and virtual orbitals in the case of large molecules. Such a choice reduces the dimension of the density matrices, lightening the diagonalization process, speeding up in such a manner the calculations. In this work, we have checked that reducing the number of MOs from 240 to 115 in the largest molecule (*p*-methoxy-nitrobenzene) changes the total value of α and β less than 1.1%.

Besides the advantages mentioned above, it could be argued that an orbital-based CP-FIOs analysis loses somehow the solid bridge between SOS and spectroscopic information obtained experimentally. However, we will show in the following discussion that this bridge is recovered when a deeper analysis of the different pairs of FIOs is performed. Therefore, similar information given by excited states in SOS is provided by the set of occupied and virtual orbitals involved in the FIOs. This is also not surprising since the orbital symmetry rule represented by eqns (8) and (14) also rules the magnitude of the components of the transition dipole and quadrupole moments, respectively, between different excited states and the ground state. If this rule is not satisfied for any of the excited configurations that contribute significantly to a

given excited state, the transition dipole/quadrupole moment will be essentially null. Moreover, as will be shown in the following, FIOs are in most cases constructed from the combination of just one occupied and one virtual orbital, so linking with the orbital picture extensively employed to interpret spectroscopic data.

Analysis of Model Molecules

In this subsection, six molecules will be analyzed, HHe^+, HF, H_2O, H_2S, *p*-nitroaniline and *p*-methoxy-nitrobenzene. Except for the last two molecules, the number of MOs that contribute significantly to the linear and nonlinear response is relatively small. Thus, the number of relevant FIOs is manageable and the symmetry rules can be exploited to understand the couplings between occupied and virtual MOs.

Starting with the simplest molecule, HHe^+, the SOS analysis (see Fig. 1(b)) indicates the α_{zz} value arises mainly from the first excited state. The main contribution to this excited state comes from the HOMO(H)→LUMO(L) excited configuration (see Supporting Information). Two additional steps up can be clearly observed in the plot, corresponding to excited states 2 and 6. The main contributions to states 2 and 6 come from the H→L+1 and H→L+5 excited configurations, respectively. Similar steps up are observed in the orbital representation of α_{zz} at orbitals H, L, L+1 and L+5 (Fig. 1(a)). The increase of the α_{zz} value at the L orbital using FIOs is similar to that observed at the first excited state using SOS. The same can be said for the L+1 and L+5 orbitals and excited states 2 and 6. Differences between SOS and FIOs in the polarizability of this molecule are found in the total values calculated with the different approaches. Whereas at DFT level FIOs and SOS values are identical, at the HF level small differences arise. The effect of increasing the value of α when introducing dynamic correlation is only appreciated using FIOs. In fact, SOS predicts a slightly smaller value at DFT level. On

the other hand, as can be seen in Fig. 1(a), removal of the orbital relaxation using UPHF as expected reduces the polarizability.

The analysis of β_{zzz} reflects significant differences between total values obtained with FIOs and SOS. In the excited state analysis provided by SOS (Fig. 1(d)) we can observe a large leap in β_{zzz} at the first excited state. As mentioned before this state corresponds mainly to the H→L orbital excitation. Afterwards, two drops of the hyperpolarizability occur at the excited states 2 and 6, which, as also discussed previously, involve excitations to L+1 and L+5 orbitals. The same information is extracted from the orbital analysis using FIOs (Fig. 1(c)), but with some nuances. Thus, the largest contributions arise from H and L orbitals, but mainly from the second one. Afterwards, two steps down are observed at the L+1 and L+5 orbitals. However, the fluctuations in the β_{zzz} value observed in the orbital representation are significantly smaller than those observed in the excited state representation. As remarked before, a much faster convergence of the optical properties is observed with the orbital representation obtained from FIOs.

Additionally, visualization of the main FIOs involved in the optical response may give useful information similar to visualization of hole-electron natural transition orbitals of the crucial excited states[19,20] that can be retrieved when SOS approximation is applied. Thus, the representation of the hole and electron distributions provided by the main pair of FIOs is confronted with those obtained using the principal NTOs of the first excited state in HHe^+. A step-by-step description of the process to construct the net charge transfer representation is given in the plot. We can see first the FIO and NTO hole and electron functions. These functions are squared to get the corresponding distributions and finally they are scaled using their eigenvalues and added to obtain the net charge transfer plot. In the case of FIOs, we have two different plots, one for α and another one for β. This is an advantage of FIOs over NTOs, since the formers are directly obtained

from the perturbed density, which differs for α and β, whereas the later stem from a unique transition density employed to calculate both α and β. Then, if the same electronic excitation dominates the charge transfer representations of α and β, only the FIOs' are able to detect subtle differences between both. In the case of HHe^+, we can observe in Fig. 2 how the FIOs functions are quite similar for α and β, but not identical. On the other hand, comparing with the NTOs important differences are observed in the hole and electron functions. However, the net charge transfer is almost the same as that one obtained with β FIOs.

In Fig. 2, the FIOs values for α_{zz} and β_{zzz} are also shown, together with the main MOs involved in its formation and their corresponding relative weight. Thus, the main pair of FIOs, both in α_{zz} and β_{zzz}, is formed by the combination of H and L orbitals in more than 90%. Comparing with the total values shown in Fig. 1, one can say that α_{zz} is exclusively given by this pair, whereas for β_{zzz} a small difference (+0.26 au) is observed, indicating that other FIOs contribute slightly to β_{zzz}. Both the z and zz functions transform following the Σ^+ *irrep*. According to eqn (8), the symmetry product of occupied and virtual orbitals involved in the FIOs must contain Σ^+. This is the case of H (Σ^+) and L (Σ^+) orbitals.

Let us analyze now a more polar molecule, namely, hydrogen fluoride (HF). The substitution of helium by a more polarizable atom (fluorine) increases the value of α_{zz}. Due to the larger molecular dipole moment, β_{zzz} significantly increases with respect to HHe^+. As can be observed in Fig. 3(a), the FIOs analysis reflects that the most relevant contributions to α_{zz} come from the H-2, L and, to a lesser extent, from L+1. However, additional, but weaker, orbital contributions are also noticeable (H-1, H, L+4, +6 and L+7). SOS analysis of α_{zz} indicates that the most important contributions come from

states 3 and 6 (Fig. 3(b)), where the main excitations correspond to H-2→L and H-2→L+1, respectively (see Supporting Information). Other noticeable changes are observed at states 13 and 23. For state 13 the main orbital transition corresponds to H-2→L+4, whereas for state 23 three orbital excitations contribute almost equally, H-2→L+5, H-1→L+6 and H→L+7. Then, FIOs and SOS analysis agree quite well in the main orbitals involved in the linear response.

Looking at the plot obtained for β_{zzz} with FIOs (Fig. 3(c)), we can see that H-2, L and L+1 are again the orbitals of the most important contribution on the second-order nonlinear response in hydrogen fluoride. In this case, contribution stemming from the rest of the orbitals considered is negligible. Since excited states 3 and 6 contribute the most on the final SOS value for β_{zzz} (Fig. 3(d)), the main orbitals involved coincide with FIOs. However, the SOS plot shows again important fluctuations in comparison with the FIOs orbital representation, requiring in this case more than forty states to reach convergence in the β_{zzz} value.

The charge transfer stemmed from the main pairs of FIOs responsible of α_{zz} and β_{zzz} values in HF are depicted in Fig. 4. As can be observed, the electron charge flows from fluorine to hydrogen in all pairs. In the case of α_{zz} (Fig. 4(a)), three different pairs contribute, although only one displays a significant value. The participation of orbitals H-2 and L in this pair is 87.7%, and the L+1 has a contribution of 10%. The other two pairs involve the rest of orbitals with a nonnegligible weight in the plot of Fig. 4(a), as mentioned before.

In the case of β_{zzz}, the total value is almost the same as the value shown in Fig. 4(b) for the main pair of FIOs, with participation of orbitals H-2 and L in more than 95%. According to eqns (8) and (13), the symmetry product of occupied and virtual orbitals must contain, as in HHe^+, the *irrep* of α_{zz} and β_{zzz} (Σ^+). This is fulfilled by the orbitals

involved in each FIO shown in Fig. 5, H-2 (Σ^+) and L(Σ^+), H-2 (Σ^+) and L+1 (Σ^+), H-1(Π) and L+6 (Π) and H(Π) and L+7(Π).

The first nonlinear molecule studied is H_2O, with C_{2v} symmetry. Fig. 5 collects the excited state and orbital representations of α_{zz} and β_{zzz} obtained with SOS and FIOs, respectively. In Fig. 5(a), one can distinguish five orbitals that contribute clearly to the value of α_{zz}, H-2, H-1, L, L+1 and L+2, then α_{zz} continues growing slightly up to orbital L+10 where the convergence is reached. The SOS analysis is more complicated since many excited states significantly contribute and convergence requires the inclusion of more than eighty states (Fig. 5(b)). However, three states clearly stand out over the rest, state 3, 6 and 11, with the main contributions coming, respectively, from the excited configurations H-1→L, H-2→L+1 and H-2→L+2 (see Supporting Information). Once more, SOS and FIOs agree with respect to the orbitals involved. In terms of charge transfer, FIOs representations suggest that in the most important transition the electron charge should flow from oxygen to hydrogen.

In the FIOs plot of Fig. 5(c), we can clearly remark the contributions of orbitals H-1 and L. Convergence is rapidly reached with ten orbitals. On the other hand, state 3 stands out over the rest in the SOS plot (Fig. 5(d)), but many important fluctuations in the value of β_{zzz} are observed until the convergence is reached with more than fifty states. As mentioned in the previous paragraph for the discussion of α_{zz}, the main contribution to state 3 comes from the H-1→L excitation, the outstanding occupied and virtual orbitals in the FIOs plot of Fig. 5(c).

Fig. (6) shows the electron charge transfer associated to the main pairs of FIOs for α_{zz} and β_{zzz}, together with the occupied and virtual orbitals involved in their formation. These occupied-virtual orbital combinations, as in the previous molecules discussed, agree with the most important excitations in SOS. In the C_{2v} symmetry group, both *z*

and *zz* functions transform as the A_1 *irrep*. Then, the close-in-energy combinations of occupied and virtual orbitals in H_2O whose symmetry product contains the A_1 *irrep* are: H-1(A_1) with L(A_1), H-2(B_2) with L+1(B_2) and H-2(B_2) with L+2(B_2). The values of α_{zz} and β_{zzz} obtained from the FIOs pairs and shown in Fig. (6) indicate that, for α_{zz}, two pairs are required to recover more than 90% of the total value, whereas only one is necessary in the case of β_{zzz}. The main orbitals involved in each pair coincide, as expected, with the orbital combinations derived from the symmetry rules.

Another interesting example to analyze in detail is the H_2S molecule, which shows strong fluctuations in the orbital and excited state representation of β_{zzz}. Many other molecules show these fluctuations but neither of the changes are so important, as in the case of small molecules, as those discussed above, or they are larger systems of more electrons, a feature that makes the respective analysis less obvious. In Fig. (7), the excited state and orbital representations of α_{zz} and β_{zzz} obtained with SOS and FIOs, respectively, are presented. In the SOS plot of α_{zz}, three excited states clearly stand out over the rest: 3, 9 and 37. The dominant excited configurations are: in state 3 H-1→L and, to a lesser extent, H-2→L+1; in state 9 H-2→L+1 and, to a lesser extent, H→L+4; and in state 37 H-1→L+8 and H→L+10 (see Supporting Information). The MOs involved in the excited configurations of states 3 and 9 correspond to the MOs with the largest contributions to α_{zz} in the FIOs plot, H-2, H-1, L and L+1. Other nonnegligible contributions come from H, L+4 and L+10, MOs involved in state 37 and, to a lesser extent, in state 9. Thus, once again the orbital and excited state representations score a perfect match.

More challenging is the analysis of β_{zzz}, due to the strong fluctuations observed. In the SOS plot (Fig. 7(d)) important positive and negative contributions to the value of β_{zzz} come from states 3 and 9, respectively. Other states also contribute significantly, but for

the sake of simplicity we are going to focus our attention on these two states. As mentioned above, excited configuration H-1→L dominates state 3. These orbitals are the ones with the largest positive contributions to β_{zzz} in the FIOs plot (Fig. 7(c)). On the other hand, the transition H-2→L+1 dominates state 9, and certainly these orbitals contribute negatively to β_{zzz} in the FIOs plot. There is, however, an orbital whose negative contribution is even larger in Fig. 7(c), L+3, but whose participation in state 9 is irrelevant. The contribution of this orbital seems to be underestimated in the SOS calculation and can partially explain the large difference between the total values of β_{zzz} obtained with CPKS and SOS.

Fig. 8 represents the main FIOs obtained for α_{zz} and β_{zzz} in H_2S. In the case of α_{zz}, the electron charge transfer associated to these FIOs is identical to that obtained for H_2O and depicted in Fig. 6. On the contrary, they differ significantly for β_{zzz}. In Fig. 8(b), we have depicted the positive and negative FIOs separately instead of grouped in pairs, since there are some important details of this molecule that can be noticed only by looking at the hole and electron distributions separately. For instance, although H-1 contributes to two different FIOs, one with a positive value of β_{zzz} and another one with a negative value, for the latter its relative weight is much larger, explaining the net positive contribution of this MO. Additionally, L and H-2 almost form by themselves individual FIOs, with relative weights around 75%. These FIOs account for the value of these MOs in Fig. 7. However, the most interesting FIO is the one formed mostly by L+3, since the contribution of this MO to the β_{zzz} value is apparently overlooked by SOS. This FIO is the third one in Fig. 8(b) and shows the largest β_{zzz} value (in absolute terms) together with the FIO formed by L. As can be seen, it is localized far over the S atom, in agreement with the high energy of the L+3 MO.

To finish this section, we will analyze and compare two widely known and extensively studied prototype book-text donor/acceptor chromophores exhibiting large molecular second-order nonlinear optical responses. These are *p*-nitroaniline and *p*-methoxy-nitrobenzene.[4] In Fig. 9, we can observe a slow convergence of α_{zz} with the number of orbitals and excited states in the FIOs and SOS plots, respectively. The number of FIOs to take into account is too large, thus not appropriate, for testing the method. This, however, is not the case for β_{zzz}, the value of which is clearly dominated by the contribution of the frontier MOs in both molecules. SOS plots also reflect that state 2 stands out over the rest, which, in both molecules, involves mainly the H→L excited configuration (see Supporting Information).

Visualization of the main pairs of FIOs of β_{zzz} in *p*-nitroaniline and *p*-methoxy-nitrobenzene (Fig. 10 and 11, respectively) reveals the nonlinear response is almost identical within the nitrobenzene fragment. The main differences are spotted, as expected, on the distinct groups (-NH_2 and -OCH_3). So, in terms of FIOs, the larger hyperpolarizability of *p*-nitroaniline could be attributed to the smaller inductive effect of -NH_2 since the long-range resonance effects should be reflected throughout the benzene ring. In both molecules, frontier orbitals contribute more than 90% to these FIOs. The charge transfer implied by the FIO representations occurs from the donors (-NH_2, -OCH_3) to the acceptor (-NO_2). This picture is in line with the push/pull properties of these groups as they have been determined in earlier investigations.[4] What is more, the obtained FIOs representations can be compared in a straightforward manner to the dominant hole-electron NTOs shown in the same figures. As in the example of HHe^+ discussed at the beginning of this section, some differences between the NTOs and FIOs functions are appreciated, however, the net charge transfer obtained is practically identical. The observed similarities, obviously stemming in this case from the

predominance of the H-L transitions, clearly point out that the FIOs approach can be used as an alternative handy tool to analyze, in a chemist-friendly fashion, not only the linear and nonlinear optical responses of any molecule but also to retrieve information about crucial intramolecular charge transfer interactions avoiding costly excited state computations.

V. Concluding remarks

In this work we have presented a rigorous and efficient method to be used as a straightforward tool for the analysis and better understanding of the linear and nonlinear optical properties of molecules. The proposed method relies on the decomposition of the tensorial components of the electric dipole polarizability and the first dipole hyperpolarizability into contributions of field-induced orbitals (FIOs). These can be retrieved either analytically from coupled-perturbed (CP) Hartree-Fock, CP Kohn-Sham approximations or numerically from finite field (FF) field self-consistent computations. Its strongest feature lies on its ability to provide crucial information about the principal intramolecular physical processes related to the (non)linear optical responses of molecules by simply analyzing the ground state electronic state of a given molecule, exploiting in turn group-theory symmetry rules in an elegant and efficient manner in case of highly symmetric systems. This characteristic is of decisive importance since, as we have shown in this article, it distinguishes the proposed method from other methodologies that require excited state computations of higher computational cost, as, for instance, methods or models built within the realm of sum-over state perturbative approximation. What is more, the presented FIO-based analysis of the optical response of molecules can be used to obtain easy-to-interpret and quantitatively defined intuitive hole-electron representations of electron quantum transitions representing intramolecular charge transfer processes. Finally, it is important to stress that the

proposed method is easily implementable to any electronic-structure computational code as a low cost *a posteriori* analysis method of a successful CP (HF, KS) computation, provided that an efficient diagonalization process of the density matrix derivatives is available.

Acknowledgements

N.O. thanks "Xunta de Galicia" for a postdoctoral contract. N. O. and M. M. thank "Xunta de Galicia" for financial support through the project GRC2015/017. Part of this work was granted access to the HPC resources of [CCRT/CINES/IDRIS] under the allocation 2018-2019 [A0040807031] made by GENCI ("Grand Equipement National de Calcul Intensif"). We also acknowledge the "Direction du Numérique" of the "Université de Pau et des Pays de l'Adour" for the computing facilities provided.

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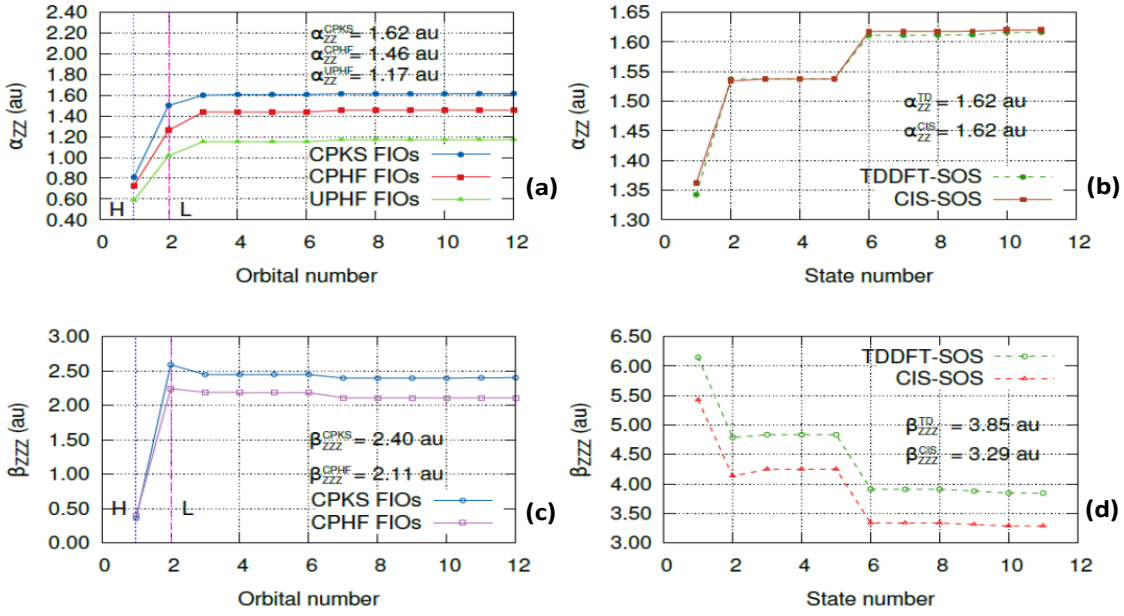

Fig. 1 For HHe^{+} molecule, Variation of α_{zz} (top) and β_{zzz} (bottom) with respect to the number of orbitals (plots **(a)** and **(c)** obtained using FIOs) and excited states (plots **(b)** and **(d)** obtained using SOS approach), respectively. For the FIOs plots, up to three approaches were applied: CPKS, CPHF and UPHF. For the SOS plots, TDDFT (CAM-B3LYP) and CIS methods were employed. Total values of α_{zz} and β_{zzz} are shown in the plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.

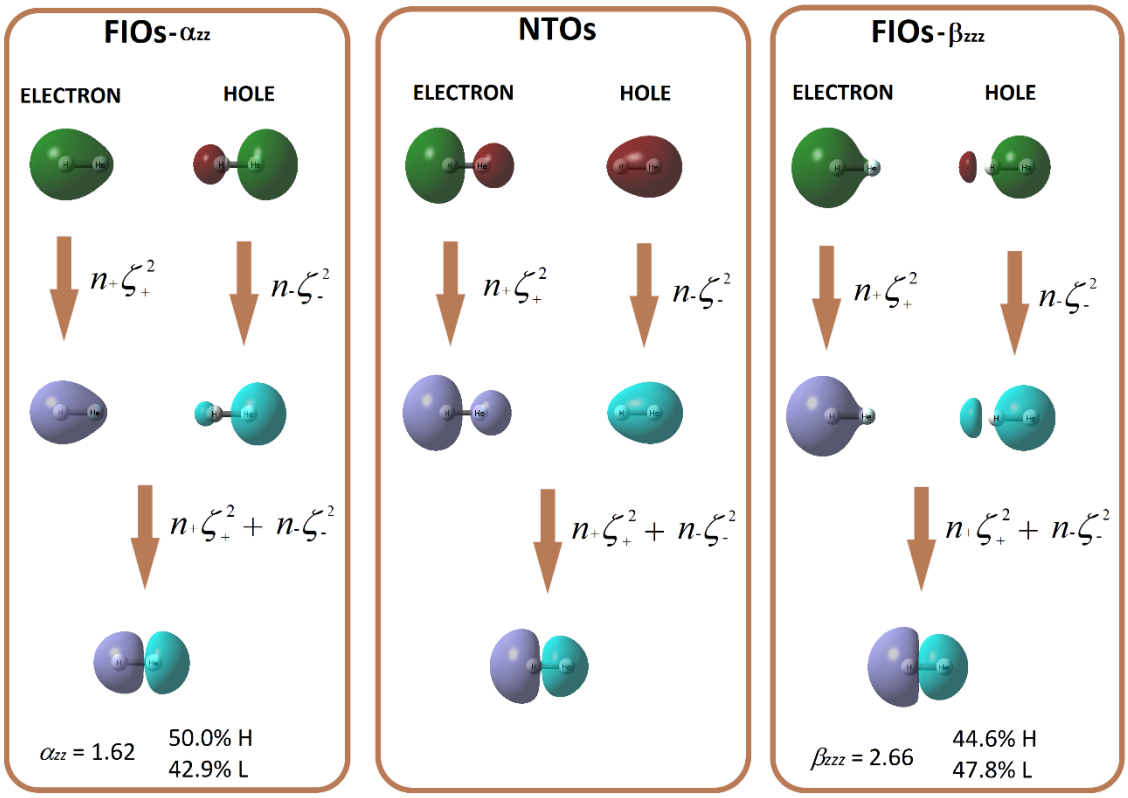

Fig. 2 Representation of the electron and hole functions obtained from the main pairs of FIOs and NTOs responsible of the α_{zz} and β_{zz} values in HHe^+. Below the functions are represented the corresponding distributions (obtained by scaling the squared functions with the eigenvalues) and below the electron and hole distributions is depicted the net electron charge transfer (obtained by adding the electron and hole distributions). Value of the (hyper)polarizability associated to the FIOs are also included together with the relative weight of the main occupied and virtual MOs involved.

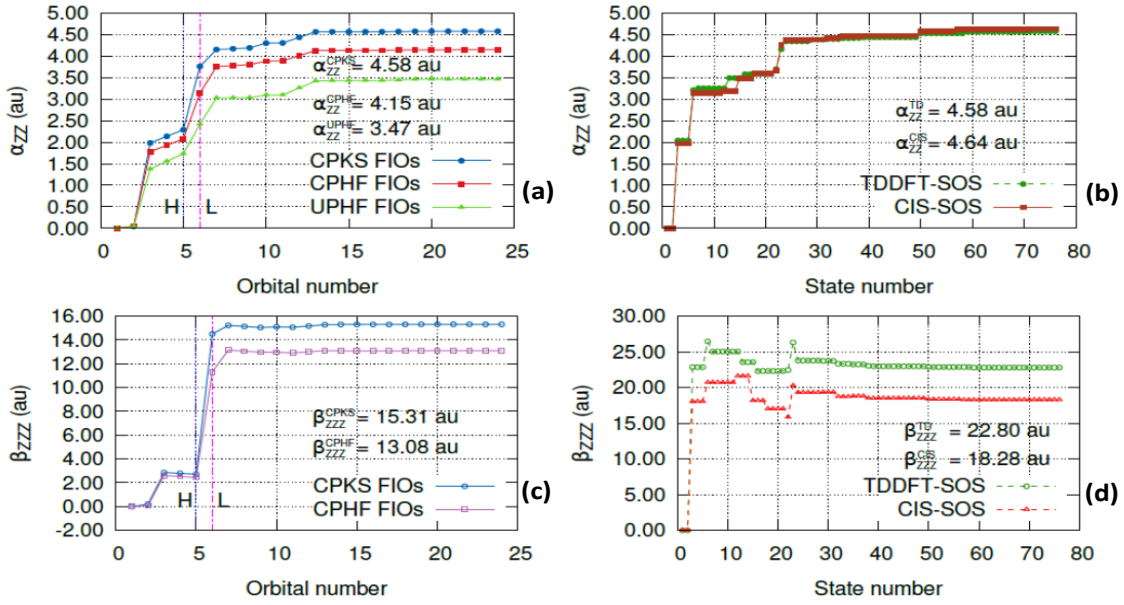

Fig. 3 For HF molecule, variation of α_{zz} (top) and β_{zzz} (bottom) with respect to the number of orbitals (plots **(a)** and **(c)** obtained using FIOs) and excited states (plots **(b)** and **(d)** obtained using SOS). For the FIOs plots, up to three approaches were applied: CPKS, CPHF and UPHF. For the SOS plots, TDDFT (CAM-B3LYP) and CIS methods were employed. Total values of α_{zz} and β_{zzz} are shown in the plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.

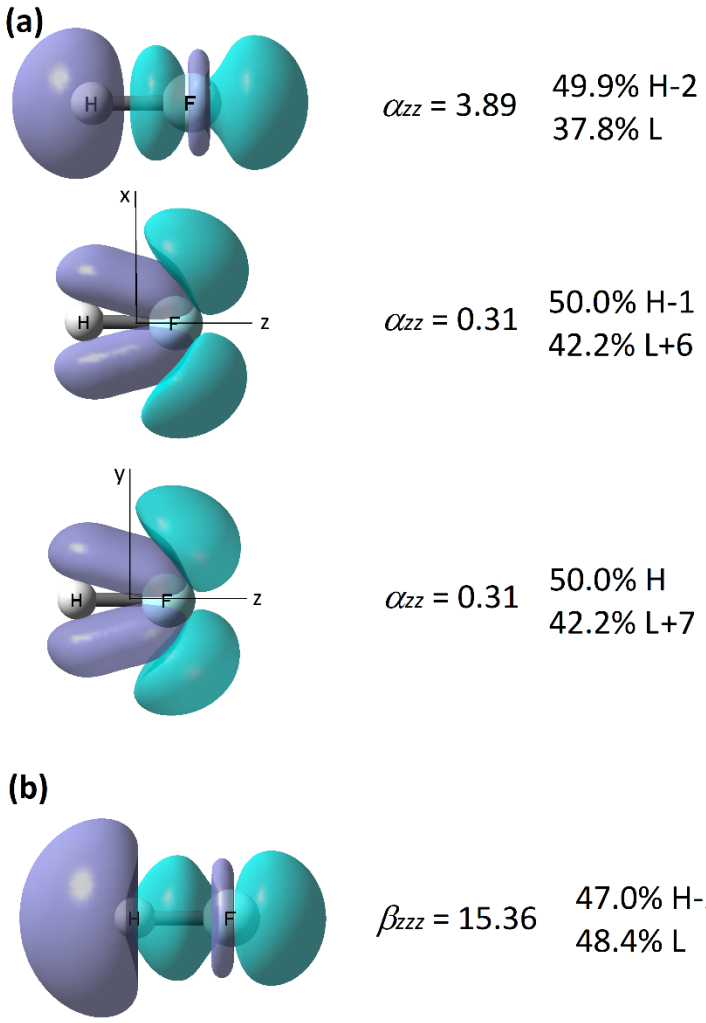

Fig. 4 Representation of the electron charge distribution in the main pairs of FIOs obtained for α_{zz} (plot **(a)**) and β_{zzz} (plot **(b)**) in HF molecule. Values of the (hyper)polarizability corresponding to these pairs are included in the Figure together with the relative weight of the main occupied and virtual MOs involved.

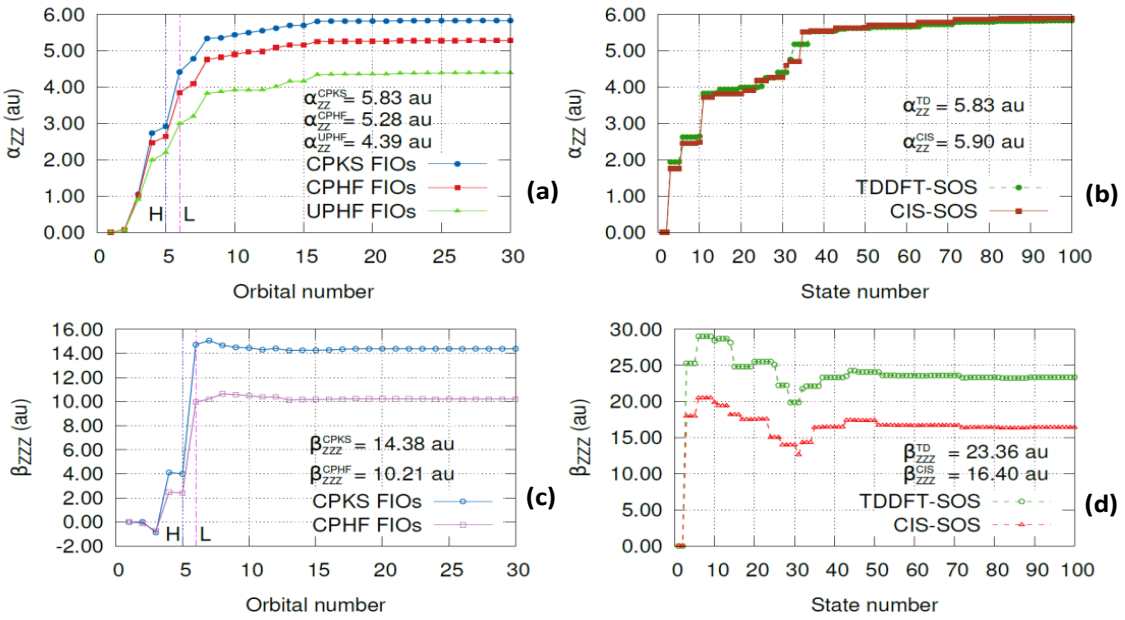

Fig. 5 For H_2O molecule, variation of α_{zz} (top) and β_{zzz} (bottom) with respect to the number of orbitals (plots **(a)** and **(c)** obtained using FIOs) and excited states (plots **(b)** and **(d)** obtained using SOS). For the FIOs plots, up to three approaches were applied: CPKS, CPHF and UPHF. For the SOS plots, TDDFT (CAM-B3LYP) and CIS methods were employed. Total values of α_{zz} and β_{zzz} are shown in the plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.

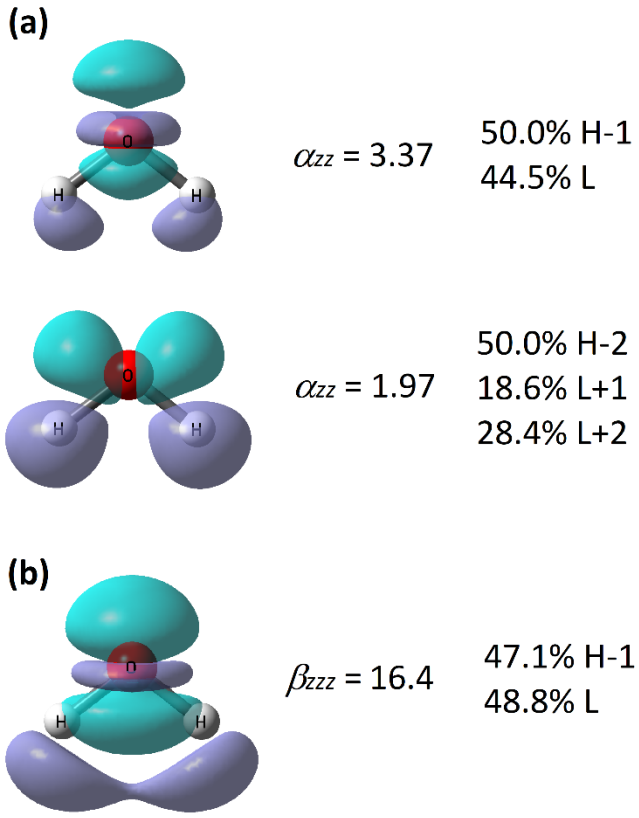

Fig. 6 Representation of the electron charge distribution in the main pairs of FIOs obtained for α_{zz} (plot **(a)**) and β_{zzz} (plot **(b)**) in H_2O molecule. Values of the (hyper)polarizability corresponding to these pairs are included in the Figure together with the relative weight of the main occupied and virtual MOs involved.

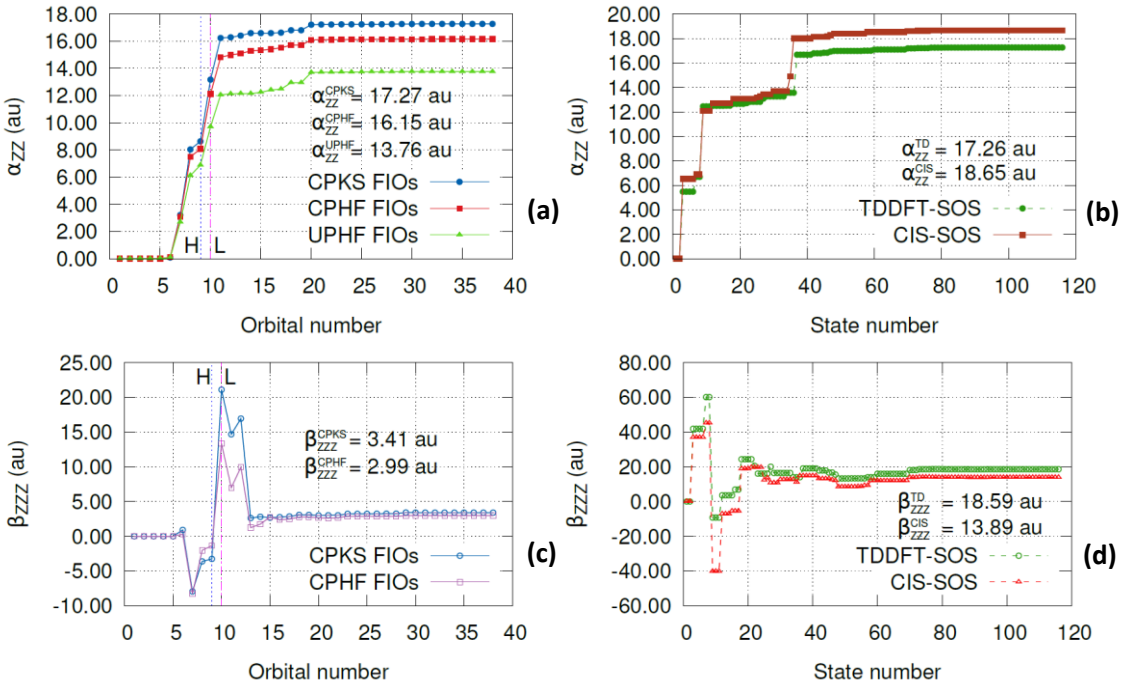

Fig. 7 For H_2S molecule, variation of α_{zz} (top) and β_{zzz} (bottom) with respect to the number of orbitals (plots **(a)** and **(c)** obtained using FIOs) and excited states (plots **(b)** and **(d)** obtained using SOS). For the FIOs plots, up to three approaches were applied: CPKS, CPHF and UPHF. For the SOS plots, TDDFT (CAM-B3LYP) and CIS methods were employed. Total values of α_{zz} and β_{zzz} are shown in the plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.

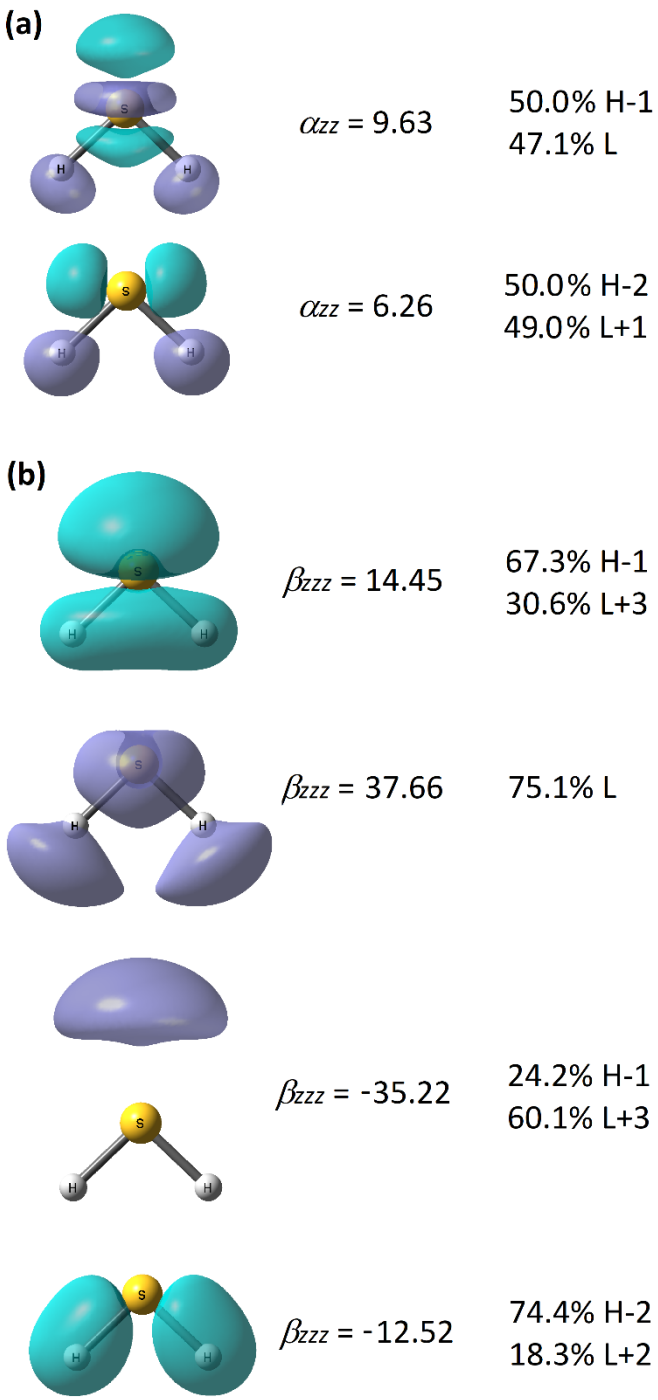

Fig. 8 Representation of the electron charge distribution in the main pairs of FIOs obtained for α_{zz} (plot **(a)**) and the main FIOs of β_{zzz} (plot **(b)**) in H_2S molecule. Values of the (hyper)polarizability corresponding to these pairs are included in the Figure together with the relative weight of the main occupied and virtual MOs involved.

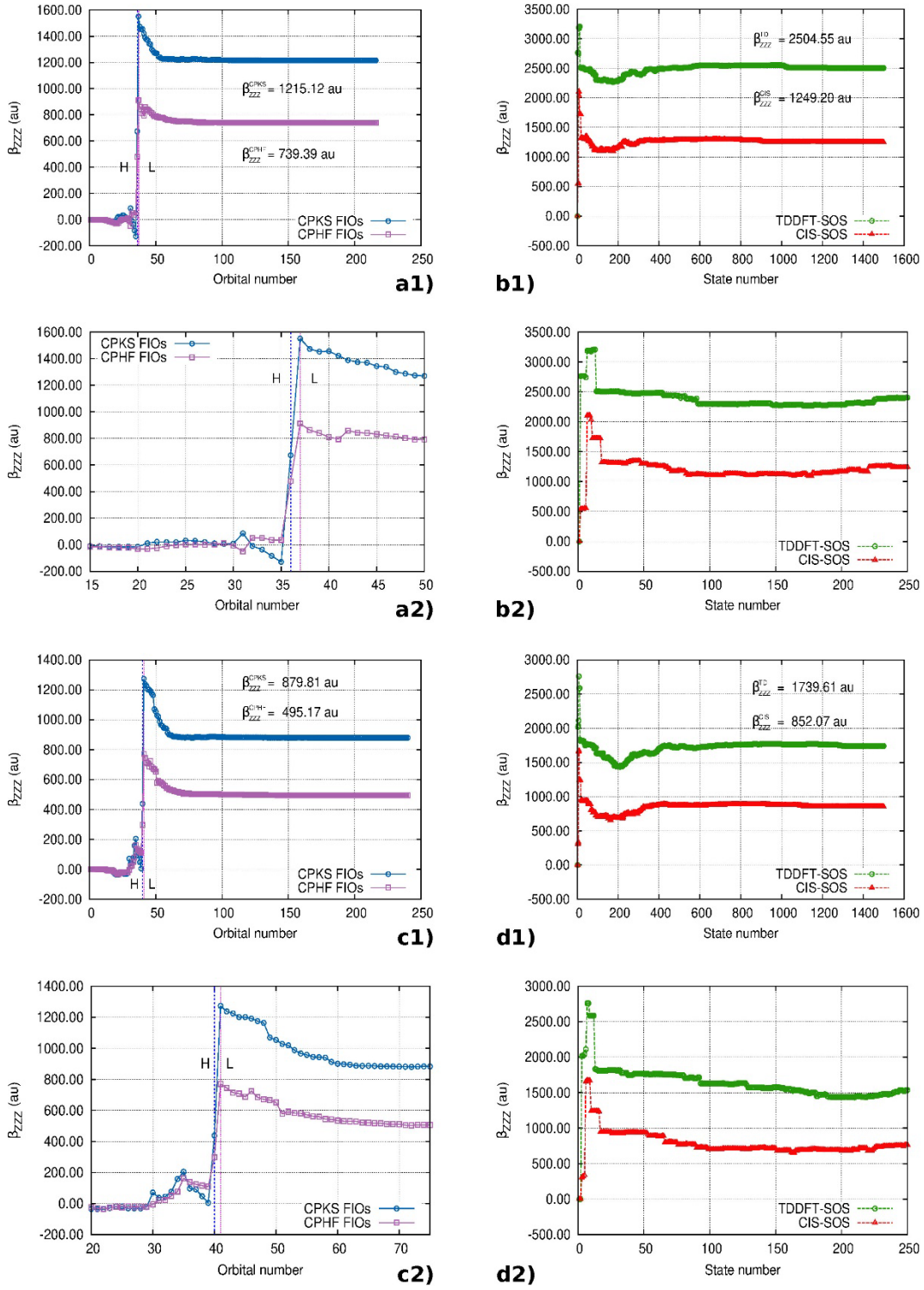

Fig. 9 Variation of β_{zzz} with respect to the number of orbitals obtained using FIOs for *p*-nitroaniline (plots (**a1)** and **(a2)**) and *p*-methoxy-nitrobenzene (plots (**c1)** and (**c2)**), and variation of β_{zzz} with respect to the number of excited states obtained using SOS for *p*-nitroaniline (plots **(b1)** and **(b2)**) and *p*-methoxy-nitrobenzene (plots **(d1)** and **(d2)**). For the FIOs plots, two approaches were applied: CPKS and CPHF. For the SOS plots, TDDFT (CAM-B3LYP) and CIS methods were employed. Total values of β_{zzz} are shown in the plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively. The region showing the most important changes is reproduced right below each plot by reducing the number of MOs or excited states.

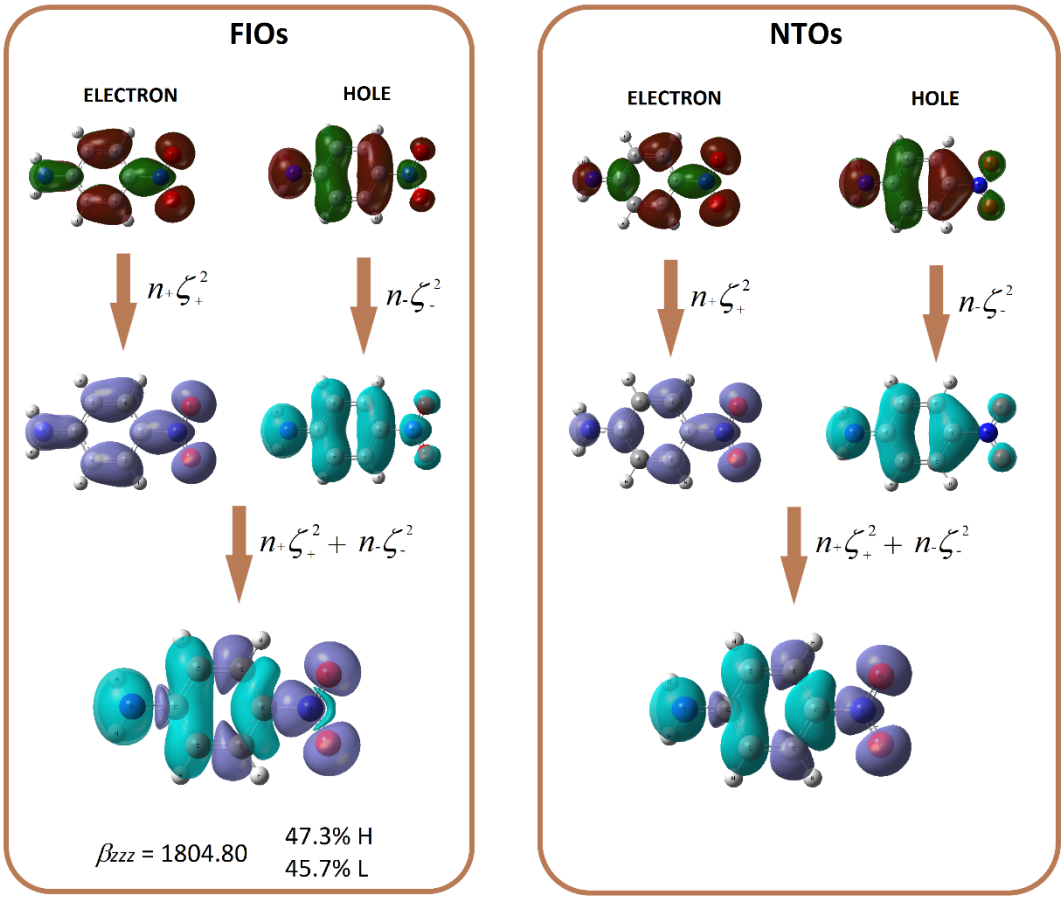

Fig. 10 Representation of the electron and hole functions obtained from the main pairs of FIOs and NTOs responsible of the β_{zz} value in p-nitroaniline. Below the functions are represented the corresponding distributions (obtained by scaling the squared functions with the eigenvalues) and below the electron and hole distributions is depicted the net electron charge transfer (obtained by adding the electron and hole distributions). The value of β_{zz} associated to the FIOs is also included together with the relative weight of the main occupied and virtual MOs involved.

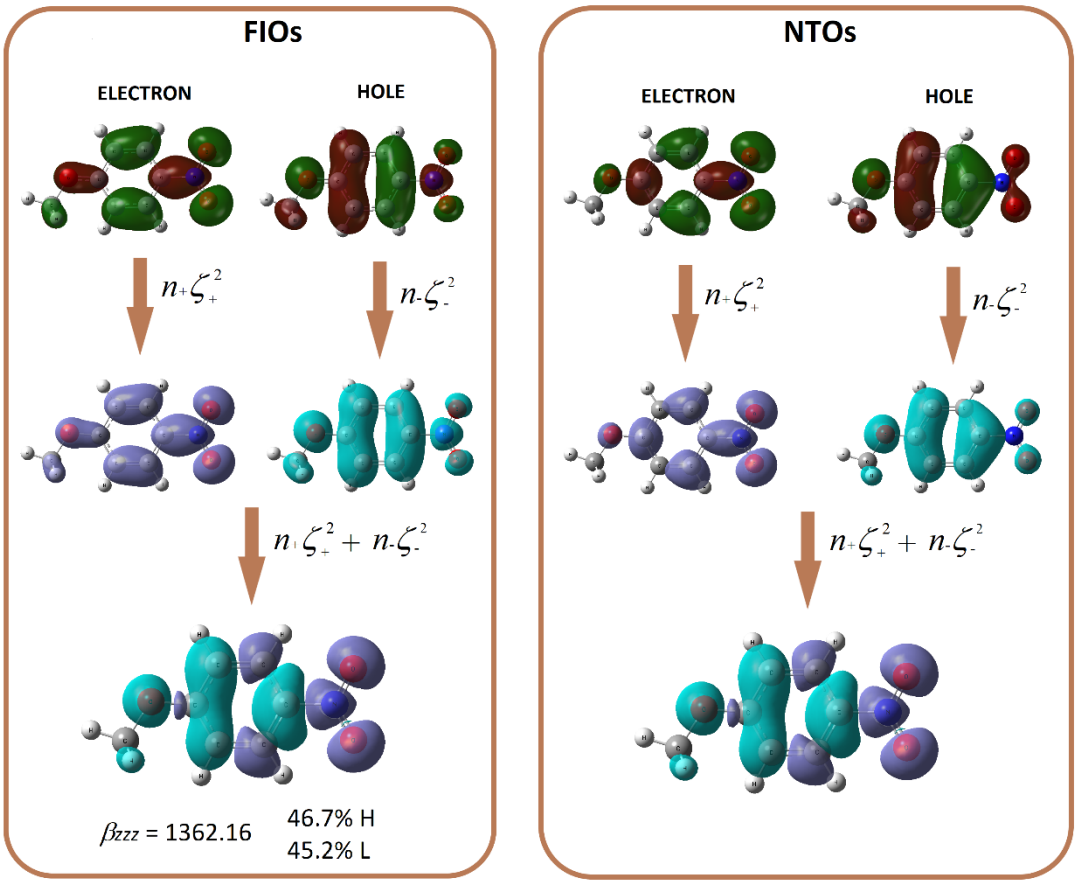

Fig. 11 Representation of the electron and hole functions obtained from the main pairs of FIOs and NTOs responsible of the β_{zz} value in p-methoxynitrobenzene. Below the functions are represented the corresponding distributions (obtained by scaling the squared functions with the eigenvalues) and below the electron and hole distributions is depicted the net electron charge transfer (obtained by adding the electron and hole distributions). The value of β_{zz} associated to the FIOs is also included together with the relative weight of the main occupied and virtual MOs involved.

Supporting Information: A New Method to Analyze and Understand Molecular Linear and Nonlinear Responses via Field-Induced Functions. A Straightforward Alternative to Sum-Over-States (SOS) Analysis

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Contents

1 Detailed description of the electronic supporting information

This electronic supporting information (ESI) contains two kinds of information:

- Plots with the representations of the variation of a component of the polarizability, α, or one of the 1st hyperpolarizability, β, with respect to the number of molecular orbitals (MOs) or excited states, depending on the approach considered. If it is not said otherwise, the principal axis is considered along z direction.

  Each figure is formed by four subfigures, except N_2 molecule, whose full β tensor is zero. Subfigures (a) and (c) were obtained by means of our field-induced orbital (FIO) decomposition and summed over the number of MOs. For α, three approaches were considered: coupled perturbed Kohn-Sham (CPKS), coupled perturbed Hartree-Fock (CPHF) and uncoupled perturbed Hartree-Fock (UPHF). In the case of β, only CPKS and CPHF approaches were taken into account.

  The values shown in subfigures (b) and (d) were computed through the TDDFT and CIS *sum over states* method (SOS) as implemented in Multiwfn.[S1] In the case of larger molecules, the number of excited states considered to compute the (hyper)polarizabilities were truncated to a value of 1500.

  In addition to the four subfigures and caused by the enormous number of states and MOs, subfigures (e) and (f) of benzene derivatives, the series of molecules based on CN$-$C$\equiv$C$-$H as well as H$-$C$\equiv$C$-$CF$_3$ are also included with a shorten range on the x axis to observe the most important variations.

  The plots were obtained at CAM-B3LYP and HF approaches combined in both cases with 6-311G(d,p) and 6-311++G(d,p) basis sets for the optimized structures. The FIOs and SOS results of these basis sets are included in the corresponding subsections, as can be seen in the table of contents. Since the FIOs and TDDFT-SOS approaches

are independent of the functional to be considered, CAM-B3LYP was employed due to the well-known results obtained in calculations of (hyper)polarizabilities.[S2–S6] The first derivative of the density matrices with respect to an uniform electric field was obtained analytically from the corresponding Gaussian calculations.[S7] On the contrary, the second derivative was computed by the symmetric numerical definition of the finite field (FF) approach with an electric field strength of 0.001 au.

In all cases, the recomputed total values of the corresponding component are included in plots (a)–(d).

- Outputs from our either TDDFT (CAM-B3LYP) or CIS calculations of the molecules commented in the main text of the work, *i.e.*, the following set of molecules are shown: **HHe$^+$** (Subsections 3.1.2 and 3.1.3), **HF** (Subsections 5.1.2 and 5.1.3), **H_2O** (Subsections 7.1.2 and 7.1.3), **H_2S** (Subsections 8.1.2 and 8.1.3), **p-nitroaniline** (Subsections 16.1.2 and 16.1.3), **m-nitroaniline** (Subsections **??** and **??**) and **p-methoxy-nitrobenzene** (Subsections 18.1.2 and 18.1.3). These outputs, considering the aforementioned two basis sets, are formed by a truncated set of excited states (except HHe$^+$, the full number of excited states was printed) adequate to represent the most important contributions to recover the final SOS value (see the corresponding figures of the same section and read the main text of the work). The symmetry determined by Gaussian 09 (`symm`), the energy (`Exc.E`), the oscillator strength (`Osc.Strength`), the transition dipole moment (`f`) and $\langle S^2 \rangle$ (`<S**2>`) are included together with the main excited configurations ordered by the five first largest and the five first lowest CI coefficients. In the last line of each excited state, the number of nonnegligible excited configurations with respect to the CI coefficient (`#CIs`), as well as the number of positive (`#CIs>0`) and negative (`#CIs<0`) values considered to obtain the SOS plots are shown.

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2 Molecular polarizability and 1st hyperpolarizability tensor components

Table S1: Tensor components of polarizability α of the molecules considered in this work. 1 CAM-B3LYP/6-311G**, 2 CAM-B3LYP/6-311++G*, 3 HF/6-311G**, 4 HF/6-311++G**

Molecule	Level	α_{XX}	α_{XY}	α_{YY}	α_{XZ}	α_{YZ}	α_{ZZ}
HHe^+	1	0.62	0.00	0.62	0.00	0.00	1.62
	2	0.62	0.00	0.62	0.00	0.00	1.63
	3	0.60	0.00	0.60	0.00	0.00	1.46
	4	0.60	0.00	0.60	0.00	0.00	1.46
N_2	1	6.05	0.00	6.05	0.00	0.00	13.66
	2	7.81	0.00	7.81	0.00	0.00	15.23
	3	5.73	0.00	5.73	0.00	0.00	13.68
	4	7.00	0.00	7.00	0.00	0.00	14.66
CO	1	8.61	0.00	8.61	0.00	0.00	13.04
	2	9.92	0.00	9.92	0.00	0.00	15.27
	3	8.23	0.00	8.23	0.00	0.00	12.45
	4	9.06	0.00	9.06	0.00	0.00	14.01
LiF	1	7.33	0.00	7.33	0.00	0.00	8.08
	2	7.31	0.00	7.31	0.00	0.00	9.31
	3	4.84	0.00	4.84	0.00	0.00	5.64
	4	5.17	0.00	5.17	0.00	0.00	6.58
HF	1	1.82	0.00	1.82	0.00	0.00	4.58
	2	2.82	0.00	2.82	0.00	0.00	5.05
	3	1.70	0.00	1.70	0.00	0.00	4.15
	4	2.32	0.00	2.32	0.00	0.00	4.42

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Table S1 – *Continued from previous page*

Molecule	Level	α_{XX}	α_{XY}	α_{YY}	α_{XZ}	α_{YZ}	α_{ZZ}
HCl	1	6.82	0.00	6.82	0.00	0.00	13.32
	2	9.43	0.00	9.43	0.00	0.00	13.48
	3	6.56	0.00	6.56	0.00	0.00	12.82
	4	8.70	0.00	8.70	0.00	0.00	12.95
H_2O	1	4.02	0.00	7.66	0.00	0.00	5.83
	2	6.81	0.00	8.21	0.00	0.00	6.76
	3	3.77	0.00	7.08	0.00	0.00	5.28
	4	5.50	0.00	7.42	0.00	0.00	5.85
H_2S	1	11.59	0.00	18.52	0.00	0.00	17.27
	2	19.27	0.00	18.64	0.00	0.00	17.31
	3	11.24	0.00	18.19	0.00	0.00	16.15
	4	17.99	0.00	18.31	0.00	0.00	16.17
NH_3	1	10.18	0.00	10.18	0.00	0.00	7.89
	2	10.95	0.00	10.95	0.00	0.00	12.89
	3	9.65	0.00	9.65	0.00	0.00	7.38
	4	10.19	0.00	10.19	0.00	0.00	10.70
F–CC–H	1	10.43	0.00	10.43	0.00	0.00	30.09
	2	13.09	0.00	13.09	0.00	0.00	32.28
	3	10.21	0.00	10.21	0.00	0.00	28.49
	4	12.51	0.00	12.51	0.00	0.00	29.96
$HCCCF_3$	1	20.33	0.00	20.33	0.00	0.00	43.04
	2	25.83	0.00	25.83	0.00	0.00	47.35
	3	18.88	0.00	18.88	0.00	0.00	39.89
	4	42.88	0.00	23.26	0.00	0.00	23.26

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Table S1 – *Continued from previous page*

Molecule	Level	α_{XX}	α_{XY}	α_{YY}	α_{XZ}	α_{YZ}	α_{ZZ}
CN−CC−H	1	15.74	0.00	15.74	0.00	0.00	63.21
	2	18.53	0.00	18.53	0.00	0.00	65.17
	3	15.50	0.00	15.50	0.00	0.00	59.87
	4	18.04	0.00	18.04	0.00	0.00	63.11
CN−CC−CC−H	1	22.20	0.00	22.20	0.00	0.00	135.18
	2	26.54	0.00	26.54	0.00	0.00	142.87
	3	22.03	0.00	22.03	0.00	0.00	121.44
	4	25.87	0.00	25.87	0.00	0.00	126.60
CN−CC−CC−CC−H	1	28.56	0.00	28.56	0.00	0.00	236.59
	2	34.28	0.00	34.28	0.00	0.00	247.52
	3	28.46	0.00	28.46	0.00	0.00	201.60
	4	33.62	0.00	33.62	0.00	0.00	208.47
p−cyanoaniline	1	37.39	−0.31	138.11	0.00	0.00	81.57
	2	51.43	0.17	147.59	0.00	0.00	86.80
	3	37.25	−0.51	126.22	0.00	0.00	78.37
	4	50.36	−0.09	133.04	0.00	0.00	83.05
p−cyanoaniline	1	81.57	0.00	37.39	0.00	−0.31	138.11
	2	86.80	0.00	51.43	0.00	0.17	147.59
	3	78.37	0.00	37.25	0.00	−0.51	126.22
	4	83.05	0.00	50.36	0.00	−0.09	133.04
p−nitroaniline	1	91.94	0.00	36.12	0.00	0.25	131.30
	2	98.17	0.00	50.40	0.00	−0.20	143.48
	3	88.50	0.00	35.99	0.00	0.44	115.25
	4	93.83	0.00	49.25	0.00	0.05	122.90

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Table S1 – *Continued from previous page*

Molecule	Level	α_{XX}	α_{XY}	α_{YY}	α_{XZ}	α_{YZ}	α_{ZZ}
$m-$nitroaniline	1	95.68	18.61	44.15	−1.38	−12.71	109.43
	2						
	3	78.01	27.89	57.77	2.74	−12.20	98.47
	4						
$p-$methoxy$-$nitrobenzene	1	44.02	0.00	96.90	0.00	2.02	137.08
	2	56.62	0.00	103.91	0.00	1.37	147.44
	3	43.18	0.00	93.02	0.00	2.07	122.03
	4	55.18	0.00	98.91	0.00	1.81	128.82

Table S2: Nonzero tensor components of the 1st hyperpolarizability β of the molecules considered in this work. 1 CAM-B3LYP/6-311G**, 2 CAM-B3LYP/6-311++G*, 3 HF/6-311G**, 4 HF/6-311++G**

Molecule	Level	β_{XXX}	β_{XXY}	β_{XYY}	β_{YYY}	β_{XXZ}	β_{XYZ}	β_{YYZ}	β_{XZZ}	β_{YZZ}
Molecule	Level	β_{XXX}	β_{XXY}	β_{XYY}	β_{YYY}	β_{XXZ}	β_{XYZ}	β_{YYZ}	β_{XZZ}	β_{YZZ}
HHe^+	1	0.00	0.00	0.00	0.00	0.12	0.00	0.12	0.00	0.00
	2	0.00	0.00	0.00	0.00	0.12	0.00	0.12	0.00	0.00
	3	0.00	0.00	0.00	0.00	0.11	0.00	0.11	0.00	0.00
	4	0.00	0.00	0.00	0.00	0.12	0.00	0.12	0.00	0.00
CO	1	0.00	0.00	0.00	0.00	−0.64	0.00	−0.64	0.00	0.00
	2	0.00	0.00	0.00	0.00	4.05	0.00	4.05	0.00	0.00
	3	0.00	0.00	0.00	0.00	−1.06	0.00	−1.06	0.00	0.00
	4	0.00	0.00	0.00	0.00	1.98	0.00	1.98	0.00	0.00
LiF	1	0.00	0.00	0.00	0.00	73.27	0.00	73.27	0.00	0.00
	2	0.00	0.00	0.00	0.00	50.88	0.00	50.88	0.00	0.00
	3	0.00	0.00	0.00	0.00	32.53	0.00	32.53	0.00	0.00
	4	0.00	0.00	0.00	0.00	22.48	0.00	22.48	0.00	0.00
HF	1	0.00	0.00	0.00	0.00	2.42	0.00	2.42	0.00	0.00
	2	0.00	0.00	0.00	0.00	0.28	0.00	0.28	0.00	0.00

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Table S2 – *Continued from previous page*

Molecule	Level	β_{XXX}	β_{XXY}	β_{XYY}	β_{YYY}	β_{XXZ}	β_{XYZ}	β_{YYZ}	β_{XZZ}	β_{YZZ}
	3	0.00	0.00	0.00	0.00	1.55	0.00	1.55	0.00	0.00
	4	0.00	0.00	0.00	0.00	−0.05	0.00	−0.05	0.00	0.00
HCl	1	0.00	0.00	0.00	0.00	7.57	0.00	7.57	0.00	0.00
	2	0.00	0.00	0.00	0.00	−0.75	0.00	−0.75	0.00	0.00
	3	0.00	0.00	0.00	0.00	5.27	0.00	5.27	0.00	0.00
	4	0.00	0.00	0.00	0.00	−1.48	0.00	−1.48	0.00	0.00
H_2O	1	0.00	0.00	0.00	0.00	4.41	0.00	18.81	0.00	0.00
	2	0.00	0.00	0.00	0.00	3.04	0.00	17.76	0.00	0.00
	3	0.00	0.00	0.00	0.00	2.63	0.00	15.71	0.00	0.00
	4	0.00	0.00	0.00	0.00	0.04	0.00	14.49	0.00	0.00
H_2S	1	0.00	0.00	0.00	0.00	6.76	0.00	17.74	0.00	0.00
	2	0.00	0.00	0.00	0.00	0.16	0.00	30.68	0.00	0.00
	3	0.00	0.00	0.00	0.00	3.91	0.00	19.92	0.00	0.00
	4	0.00	0.00	0.00	0.00	−2.40	0.00	29.41	0.00	0.00
NH_3	1	0.00	20.36	0.00	−20.36	15.50	0.00	15.50	0.00	0.00
	2	0.00	16.26	0.00	−16.24	0.90	0.00	0.90	0.00	0.00
	3	0.00	18.34	0.00	−18.34	12.44	0.00	12.44	0.00	0.00

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Table S2 – *Continued from previous page*

Molecule	Level	β_{XXX}	β_{XXY}	β_{XYY}	β_{YYY}	β_{XXZ}	β_{XYZ}	β_{YYZ}	β_{XZZ}	β_{YZZ}
	4	0.00	15.57	0.00	−15.57	2.99	0.00	2.99	0.00	0.00
F−CC−H	1	0.00	0.00	0.00	0.00	1.28	0.00	1.28	0.00	0.00
	2	0.00	0.00	0.00	0.00	13.29	0.00	13.29	0.00	0.00
	3	0.00	0.00	0.00	0.00	−0.05	0.00	−0.05	0.00	0.00
	4	0.00	0.00	0.00	0.00	7.95	0.00	7.95	0.00	0.00
$HCCCF_3$	1	0.00	−3.96	0.00	3.96	−16.42	0.00	−16.42	0.00	0.00
	2	0.00	−9.46	0.00	9.45	−11.65	0.00	−11.64	0.00	0.00
	3	0.00	−2.71	0.00	2.71	−11.78	0.00	−11.78	0.00	0.00
	4	0.00	−3.43	−7.02	3.43	−8.42	0.00	−8.42	0.00	7.02
CN−CC−H	1	0.00	0.00	0.00	0.00	9.45	0.00	9.45	0.00	0.00
	2	0.00	0.00	0.00	0.00	9.23	0.00	9.23	0.00	0.00
	3	0.00	0.00	0.00	0.00	10.16	0.00	10.16	0.00	0.00
	4	0.00	0.00	0.00	0.00	11.58	0.00	11.58	0.00	0.00
CN−CC−CC−H	1	0.00	0.00	0.00	0.00	12.16	0.00	12.16	0.00	0.00
	2	0.00	0.00	0.00	0.00	17.70	0.00	17.70	0.00	0.00
	3	0.00	0.00	0.00	0.00	12.58	0.00	12.58	0.00	0.00
	4	0.00	0.00	0.00	0.00	17.46	0.00	17.46	0.00	0.00

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Table S2 – *Continued from previous page*

Molecule	Level	β_{XXX}	β_{XXY}	β_{XYY}	β_{YYY}	β_{XXZ}	β_{XYZ}	β_{YYZ}	β_{XZZ}	β_{YZZ}
CN−CC−CC−CC−H	1	0.00	0.00	0.00	0.00	14.11	0.00	14.11	0.00	0.00
	2	0.00	0.00	0.00	0.00	24.09	0.00	24.09	0.00	0.00
	3	0.00	0.00	0.00	0.00	14.15	0.00	14.15	0.00	0.00
	4	0.00	0.00	0.00	0.00	21.85	0.00	21.85	0.00	0.00
p−cyanoaniline	1	0.00	4.22	0.00	3.63	−94.92	0.00	−14.54	0.00	28.60
	2	0.00	3.20	0.00	9.11	−84.28	0.00	−44.47	0.00	7.34
	3	0.00	4.44	0.00	3.42	−100.81	0.00	−14.12	0.00	23.41
	4	0.00	4.48	0.00	9.30	−96.13	0.00	−31.04	0.00	5.61
p−nitroaniline	1	0.00	−3.71	0.00	−3.22	−143.65	0.00	−15.50	0.00	−28.53
	2	0.00	−2.31	0.00	−7.68	−142.01	0.00	−60.12	0.00	−10.54
	3	0.00	−4.04	0.00	−3.18	−165.83	0.00	−10.48	0.00	−22.73
	4	0.00	−3.50	0.00	−8.39	−174.41	0.00	−37.66	0.00	−7.14
m−nitroaniline	1	−17.72	−31.14	−50.88	−87.01	13.48	33.60	74.67	−57.98	16.35
	2									
	3	−47.85	−44.53	−43.13	−49.85	20.89	21.95	16.41	−33.82	11.14
	4									
p−methoxy−nitrobenzene	1	0.00	−33.44	0.00	7.09	−37.53	0.00	−113.23	0.00	−82.49

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Table S2 – *Continued from previous page*

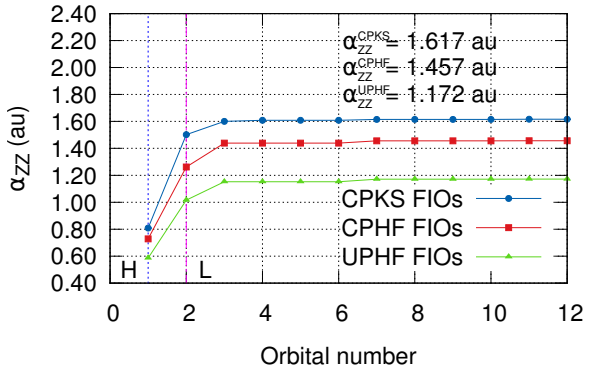
Molecule	Level	β_{XXX}	β_{XXY}	β_{XYY}	β_{YYY}	β_{XXZ}	β_{XYZ}	β_{YYZ}	β_{XZZ}	β_{YZZ}
	2	0.00	−30.04	0.00	2.45	−39.28	0.00	−110.52	0.00	−105.12
	3	0.00	−26.81	0.00	9.20	−26.03	0.00	−136.73	0.00	−61.17
	4	0.00	−21.62	0.00	9.85	−28.48	0.00	−140.21	0.00	−71.73

3 HHe^+

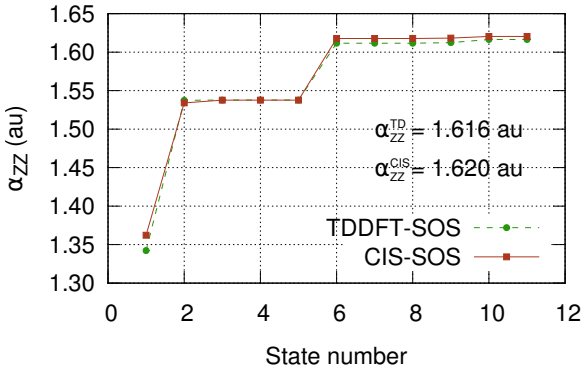
3.1 6-311G(d,p)

3.1.1 Plots

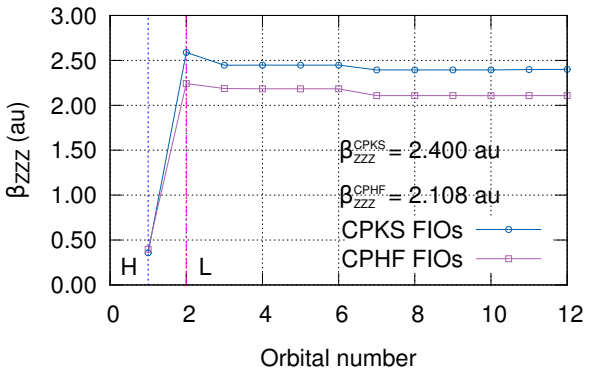
Figure S1: For HHe^+ molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S1a and S1c) or states (SOS approaches, in Plots S1b and S1d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.04 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



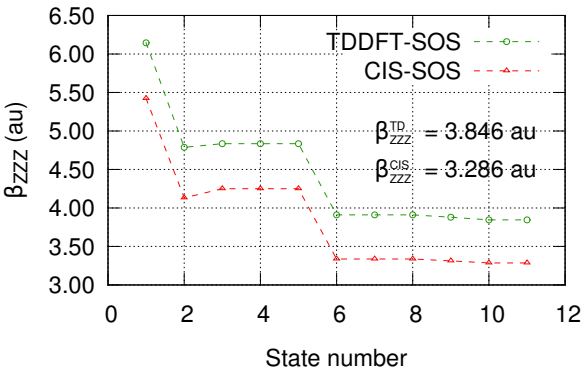
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

3.1.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_          min(4)=        1 ->  3         0.00130
                                                                        min(5)=        1 ->  7         0.00683
1        Singlet-SG   25.3777         48.86    0.3892    0.000          #CIs=7 #CIs>0=4 #CIs<0=3
               max(1)=        1 ->  2         0.70588
               max(2)=        1 ->  7         0.01962   4        Singlet-?Sym   49.6261         24.98    0.6477    0.000
               max(3)=        1 -> 11        -0.00521                 max(1)=        1 ->  5         0.68562
               max(4)=        1 -> 10        -0.00607                 max(2)=        1 ->  6         0.17112
               max(5)=        1 -> 12        -0.00815                 max(3)=        1 ->  8         0.03117
               min(1)=        1 ->  4        -0.05874                 max(4)=        1 ->  9         0.00778
               min(2)=        1 ->  3        -0.02041                 max(5)=        1 ->  4         0.70534
               min(3)=        1 -> 12        -0.00815                 min(1)=        1 ->  9         0.00778
               min(4)=        1 -> 10        -0.00607                 min(2)=        1 ->  8         0.03117
               min(5)=        1 -> 11        -0.00521                 min(3)=        1 ->  6         0.17112
               #CIs=7 #CIs>0=2 #CIs<0=5                               min(4)=        1 ->  5         0.68562
                                                                       min(5)=        1 -> 10        -0.01534
2        Singlet-SG   32.8423         37.75    0.0947    0.000          #CIs=4 #CIs>0=4 #CIs<0=0
               max(1)=        1 ->  3         0.70684
               max(2)=        1 ->  2         0.02014   5        Singlet-?Sym   49.6261         24.98    0.6477    0.000
               max(3)=        1 -> 10         0.00422                 max(1)=        1 ->  6         0.68562
               max(4)=        1 -> 12        -0.00125                 max(2)=        1 ->  9         0.03117
               max(5)=        1 ->  4        -0.00287                 max(3)=        1 ->  8        -0.00778
               min(1)=        1 ->  7        -0.01227                 max(4)=        1 ->  5        -0.17112
               min(2)=        1 -> 11        -0.00600                 max(5)=        1 ->  5         0.68562
               min(3)=        1 ->  4        -0.00287                 min(1)=        1 ->  5        -0.17112
               min(4)=        1 -> 12        -0.00125                 min(2)=        1 ->  8        -0.00778
               min(5)=        1 -> 10         0.00422                 min(3)=        1 ->  9         0.03117
               #CIs=7 #CIs>0=3 #CIs<0=4                               min(4)=        1 ->  6         0.68562
                                                                       min(5)=        1 ->  9         0.00778
3        Singlet-SG   46.8246         26.48    0.0004    0.000          #CIs=4 #CIs>0=2 #CIs<0=2
               max(1)=        1 ->  4         0.70534
               max(2)=        1 ->  2         0.06098   6        Singlet-SG   62.9858         19.68    0.1317    0.000
               max(3)=        1 ->  7         0.00683                 max(1)=        1 ->  7         0.70660
               max(4)=        1 ->  3         0.00130                 max(2)=        1 ->  3         0.01341
               max(5)=        1 -> 11        -0.01096                 max(3)=        1 -> 11         0.00424
               min(1)=        1 -> 10        -0.01534                 max(4)=        1 -> 12         0.00210
               min(2)=        1 -> 12        -0.01193                 max(5)=        1 ->  4        -0.00606
               min(3)=        1 -> 11        -0.01096                 min(1)=        1 -> 10        -0.02235
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                min(2)=        1 ->  2        -0.02114                max(3)=        1 ->  4         0.01452
                min(3)=        1 ->  4        -0.00606                max(4)=        1 ->  2         0.00667
                min(4)=        1 -> 12         0.00210                max(5)=        1 -> 12        -0.00146
                min(5)=        1 -> 11         0.00424                min(1)=        1 -> 11        -0.02345
                #CIs=7 #CIs>0=4 #CIs<0=3                              min(2)=        1 ->  3        -0.00411
                                                                      min(3)=        1 -> 12        -0.00146
7       Singlet-?Sym   67.8741         18.27    0.0646    0.000      min(4)=        1 ->  2         0.00667
                max(1)=        1 ->  8         0.70122                min(5)=        1 ->  4         0.01452
                max(2)=        1 ->  6         0.00392                #CIs=7 #CIs>0=4 #CIs<0=3
                max(3)=        1 ->  5        -0.03213
                max(4)=        1 ->  9        -0.08553   10       Singlet-SG   96.1022         12.90    0.0172    0.000
                max(5)=        1 ->  7         0.70660                max(1)=        1 -> 11         0.70657
                min(1)=        1 ->  9        -0.08553                max(2)=        1 -> 10         0.02337
                min(2)=        1 ->  5        -0.03213                max(3)=        1 ->  4         0.01134
                min(3)=        1 ->  6         0.00392                max(4)=        1 ->  2         0.00711
                min(4)=        1 ->  8         0.70122                max(5)=        1 ->  3         0.00569
                min(5)=        1 -> 10        -0.02235                min(1)=        1 -> 12        -0.01013
                #CIs=4 #CIs>0=2 #CIs<0=2                              min(2)=        1 ->  7        -0.00336
                                                                      min(3)=        1 ->  3         0.00569
8       Singlet-?Sym   67.8741         18.27    0.0646    0.000      min(4)=        1 ->  2         0.00711
                max(1)=        1 ->  9         0.70122                min(5)=        1 ->  4         0.01134
                max(2)=        1 ->  8         0.08553                #CIs=7 #CIs>0=5 #CIs<0=2
                max(3)=        1 ->  5        -0.00392
                max(4)=        1 ->  6        -0.03213   11       Singlet-SG  190.6269          6.50    0.0000    0.000
                max(5)=        1 ->  8         0.70122                max(1)=        1 -> 12         0.70701
                min(1)=        1 ->  6        -0.03213                max(2)=        1 ->  4         0.01214
                min(2)=        1 ->  5        -0.00392                max(3)=        1 ->  2         0.01028
                min(3)=        1 ->  8         0.08553                max(4)=        1 -> 11         0.00999
                min(4)=        1 ->  9         0.70122                max(5)=        1 -> 10         0.00163
                min(5)=        1 ->  9        -0.08553                min(1)=        1 ->  7        -0.00194
                #CIs=4 #CIs>0=2 #CIs<0=2                              min(2)=        1 ->  3         0.00107
                                                                      min(3)=        1 -> 10         0.00163
9       Singlet-SG   77.1426         16.07    0.0019    0.000      min(4)=        1 -> 11         0.00999
                max(1)=        1 -> 10         0.70618                min(5)=        1 ->  2         0.01028
                max(2)=        1 ->  7         0.02284                #CIs=7 #CIs>0=6 #CIs<0=1
```

3.1.3 Main contributions from different excited states at CIS approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_   Singlet-SG   27.3850         45.27    0.4599    0.000
                                                                      max(1)=        1 ->  2         0.69064
```

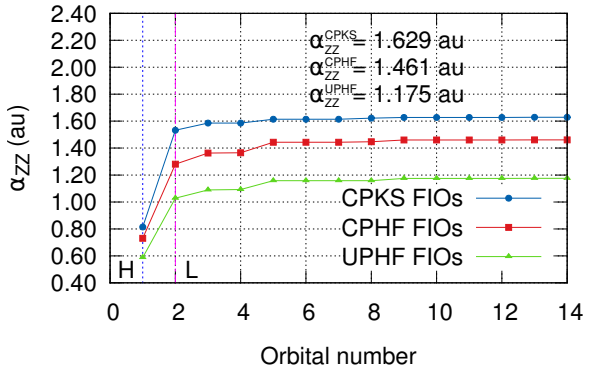
```
              max(2)=        1 ->  3         0.15138
              max(3)=        1 ->  4         0.00085
              max(4)=        1 -> 11        -0.00083
              max(5)=        1 -> 10        -0.00419
              min(1)=        1 ->  7        -0.00761
              min(2)=        1 -> 12        -0.00534
              min(3)=        1 -> 10        -0.00419
              min(4)=        1 -> 11        -0.00083
              min(5)=        1 ->  4         0.00085
              #CIs=7 #CIs>0=3 #CIs<0=4              5        Singlet-?Sym   50.2671         24.67    0.7000    0.000
                                                             max(1)=        1 ->  6         0.59801
2        Singlet-SG   35.3079         35.12    0.0965    0.000          max(2)=        1 ->  5         0.37670
              max(1)=        1 ->  3         0.68769                   max(3)=        1 ->  9        -0.01161
              max(2)=        1 -> 11         0.00673                   max(4)=        1 ->  8        -0.01844
              max(3)=        1 -> 12         0.00078                   max(5)=        1 ->  5         0.59801
              max(4)=        1 -> 10        -0.01229                   min(1)=        1 ->  8        -0.01844
              max(5)=        1 ->  7        -0.02391                   min(2)=        1 ->  9        -0.01161
              min(1)=        1 ->  2        -0.15098                   min(3)=        1 ->  5         0.37670
              min(2)=        1 ->  4        -0.05936                   min(4)=        1 ->  6         0.59801
              min(3)=        1 ->  7        -0.02391                   min(5)=        1 ->  6        -0.37670
              min(4)=        1 -> 10        -0.01229                   #CIs=4 #CIs>0=2 #CIs<0=2
              min(5)=        1 -> 12         0.00078
              #CIs=7 #CIs>0=3 #CIs<0=4              6        Singlet-SG   64.5538         19.21    0.1502    0.000
                                                             max(1)=        1 ->  7         0.70500
3        Singlet-SG   48.2215         25.71    0.0038    0.000          max(2)=        1 ->  3         0.02445
              max(1)=        1 ->  4         0.70452                   max(3)=        1 -> 11         0.02297
              max(2)=        1 ->  3         0.05741                   max(4)=        1 ->  4         0.00516
              max(3)=        1 -> 11         0.01036                   max(5)=        1 ->  2         0.00216
              max(4)=        1 -> 12        -0.00016                   min(1)=        1 -> 10        -0.04254
              max(5)=        1 -> 10        -0.00303                   min(2)=        1 -> 12        -0.00179
              min(1)=        1 ->  2        -0.01354                   min(3)=        1 ->  2         0.00216
              min(2)=        1 ->  7        -0.00763                   min(4)=        1 ->  4         0.00516
              min(3)=        1 -> 10        -0.00303                   min(5)=        1 -> 11         0.02297
              min(4)=        1 -> 12        -0.00016                   #CIs=7 #CIs>0=5 #CIs<0=2
              min(5)=        1 -> 11         0.01036
              #CIs=7 #CIs>0=3 #CIs<0=4              7        Singlet-?Sym   69.9228         17.73    0.0531    0.000
                                                             max(1)=        1 ->  9         0.53751
4        Singlet-?Sym   50.2671         24.67    0.7000    0.000          max(2)=        1 ->  5         0.01657
              max(1)=        1 ->  5         0.59801                   max(3)=        1 ->  6        -0.01415
              max(2)=        1 ->  8         0.01161                   max(4)=        1 ->  8        -0.45891
              max(3)=        1 ->  9        -0.01844                   max(5)=        1 ->  7         0.70500
```

```
                min(1)=        1 ->  8        -0.45891                 min(4)=        1 -> 12         0.00432
                min(2)=        1 ->  6        -0.01415                 min(5)=        1 ->  3         0.01496
                min(3)=        1 ->  5         0.01657                 #CIs=7 #CIs>0=6 #CIs<0=1
                min(4)=        1 ->  9         0.53751
                min(5)=        1 -> 10        -0.04254   10       Singlet-SG  100.0171         12.40    0.0099    0.000
                #CIs=4 #CIs>0=2 #CIs<0=2                                max(1)=        1 -> 11         0.70591
                                                                        max(2)=        1 -> 10         0.03320
8       Singlet-?Sym   69.9228         17.73    0.0531    0.000        max(3)=        1 ->  2         0.00245
                max(1)=        1 ->  8         0.53751                 max(4)=        1 -> 12         0.00040
                max(2)=        1 ->  9         0.45891                 max(5)=        1 ->  3        -0.00734
                max(3)=        1 ->  6         0.01657                 min(1)=        1 ->  7        -0.02067
                max(4)=        1 ->  5         0.01415                 min(2)=        1 ->  4        -0.00982
                max(5)=        1 ->  9         0.53751                 min(3)=        1 ->  3        -0.00734
                min(1)=        1 ->  5         0.01415                 min(4)=        1 -> 12         0.00040
                min(2)=        1 ->  6         0.01657                 min(5)=        1 ->  2         0.00245
                min(3)=        1 ->  9         0.45891                 #CIs=7 #CIs>0=4 #CIs<0=3
                min(4)=        1 ->  8         0.53751
                min(5)=        1 ->  8        -0.45891   11       Singlet-SG  195.2285          6.35    0.0002    0.000
                #CIs=4 #CIs>0=4 #CIs<0=0                                max(1)=        1 -> 12         0.70707
                                                                        max(2)=        1 ->  2         0.00538
9       Singlet-SG   79.7245         15.55    0.0013    0.000        max(3)=        1 ->  7         0.00150
                max(1)=        1 -> 10         0.70491                 max(4)=        1 ->  3         0.00038
                max(2)=        1 ->  7         0.04303                 max(5)=        1 ->  4         0.00024
                max(3)=        1 ->  3         0.01496                 min(1)=        1 -> 10        -0.00445
                max(4)=        1 -> 12         0.00432                 min(2)=        1 -> 11        -0.00016
                max(5)=        1 ->  4         0.00278                 min(3)=        1 ->  4         0.00024
                min(1)=        1 -> 11        -0.03170                 min(4)=        1 ->  3         0.00038
                min(2)=        1 ->  2         0.00147                 min(5)=        1 ->  7         0.00150
                min(3)=        1 ->  4         0.00278                 #CIs=7 #CIs>0=5 #CIs<0=2
```

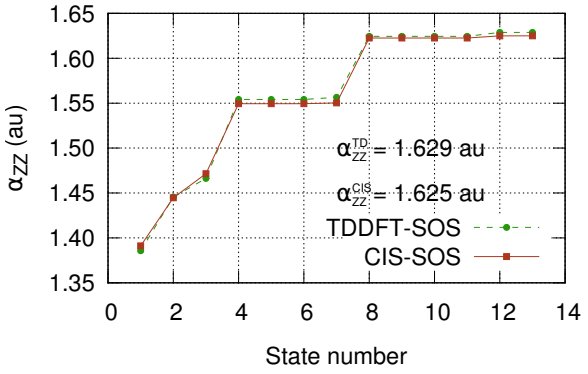
3.2 6-311G++(d,p)

3.2.1 Plots

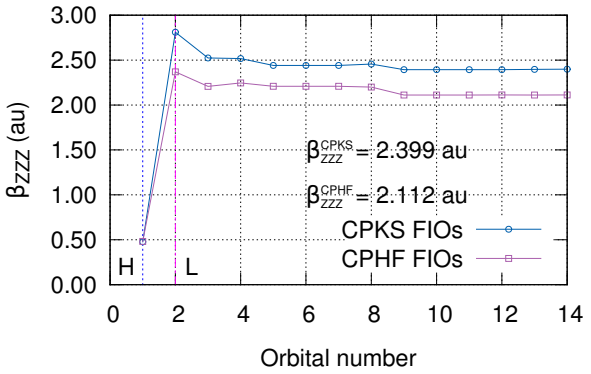
Figure S2: For HHe^+ molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S2a and S2c) or states (SOS approaches, in Plots S2b and S2d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.01 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



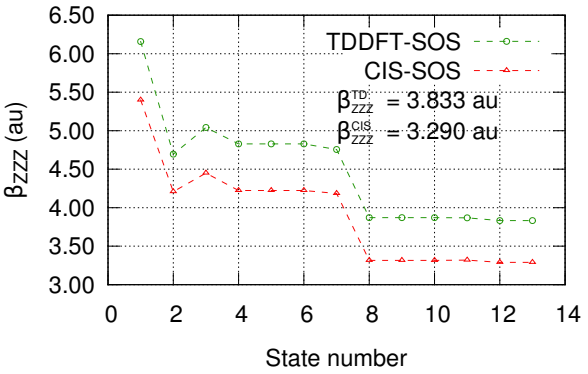
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

3.2.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_   Singlet-SG   38.2925         32.38    0.0580    0.000
                                                                        max(1)=        1 ->  5         0.70329
1        Singlet-SG   25.1296         49.34    0.3939    0.000         max(2)=        1 ->  4         0.05189
               max(1)=        1 ->  2         0.70332                max(3)=        1 ->  8         0.01362
               max(2)=        1 ->  9         0.02163                max(4)=        1 -> 12         0.00803
               max(3)=        1 ->  5         0.01447                max(5)=        1 -> 14         0.00137
               max(4)=        1 -> 12        -0.00201                min(1)=        1 ->  3        -0.04807
               max(5)=        1 -> 13        -0.00342                min(2)=        1 ->  2        -0.01391
               min(1)=        1 ->  4        -0.05371                min(3)=        1 ->  9        -0.01248
               min(2)=        1 ->  3        -0.05257                min(4)=        1 -> 13        -0.00457
               min(3)=        1 ->  8        -0.03704                min(5)=        1 -> 14         0.00137
               min(4)=        1 -> 14        -0.00709                #CIs=9 #CIs>0=5 #CIs<0=4
               min(5)=        1 -> 13        -0.00342
               #CIs=9 #CIs>0=3 #CIs<0=6              5        Singlet-?Sym   49.6273         24.98    0.6480    0.000
                                                                       max(1)=        1 ->  6         0.58275
2        Singlet-SG   30.6326         40.47    0.0252    0.000         max(2)=        1 -> 11         0.02657
               max(1)=        1 ->  3         0.70046                max(3)=        1 -> 10        -0.01822
               max(2)=        1 ->  4         0.06388                max(4)=        1 ->  7        -0.39970
               max(3)=        1 ->  2         0.05598                max(5)=        1 ->  5         0.70329
               max(4)=        1 ->  5         0.04451                min(1)=        1 ->  7        -0.39970
               max(5)=        1 -> 12        -0.00111                min(2)=        1 -> 10        -0.01822
               min(1)=        1 ->  8        -0.01699                min(3)=        1 -> 11         0.02657
               min(2)=        1 ->  9        -0.00620                min(4)=        1 ->  6         0.58275
               min(3)=        1 -> 13        -0.00407                min(5)=        1 ->  3        -0.04807
               min(4)=        1 -> 14        -0.00290                #CIs=4 #CIs>0=2 #CIs<0=2
               min(5)=        1 -> 12        -0.00111
               #CIs=9 #CIs>0=4 #CIs<0=5              6        Singlet-?Sym   49.6273         24.98    0.6480    0.000
                                                                       max(1)=        1 ->  7         0.58275
3        Singlet-SG   34.7510         35.68    0.0113    0.000         max(2)=        1 ->  6         0.39970
               max(1)=        1 ->  4         0.70028                max(3)=        1 -> 10         0.02657
               max(2)=        1 ->  2         0.05004                max(4)=        1 -> 11         0.01822
               max(3)=        1 ->  9         0.00912                max(5)=        1 ->  6         0.58275
               max(4)=        1 -> 12        -0.00367                min(1)=        1 -> 11         0.01822
               max(5)=        1 -> 13        -0.00392                min(2)=        1 -> 10         0.02657
               min(1)=        1 ->  3        -0.06453                min(3)=        1 ->  6         0.39970
               min(2)=        1 ->  5        -0.05487                min(4)=        1 ->  7         0.58275
               min(3)=        1 ->  8        -0.02113                min(5)=        1 ->  7        -0.39970
               min(4)=        1 -> 14        -0.00632                #CIs=4 #CIs>0=4 #CIs<0=0
               min(5)=        1 -> 13        -0.00392
               #CIs=9 #CIs>0=3 #CIs<0=6              7        Singlet-SG   54.0800         22.93    0.0032    0.000
                                                                       max(1)=        1 ->  8         0.70582
```

```
              max(2)=        1 ->  2         0.04125                 max(4)=        1 -> 10        -0.11508
              max(3)=        1 ->  4         0.01935                 max(5)=        1 -> 10         0.69698
              max(4)=        1 ->  9         0.01682                 min(1)=        1 -> 10        -0.11508
              max(5)=        1 ->  3         0.01352                 min(2)=        1 ->  6        -0.03202
              min(1)=        1 ->  5        -0.01333                 min(3)=        1 ->  7         0.00529
              min(2)=        1 -> 14        -0.01019                 min(4)=        1 -> 11         0.69698
              min(3)=        1 -> 13        -0.00972                 min(5)=        1 ->  7        -0.03202
              min(4)=        1 -> 12        -0.00861                 #CIs=4 #CIs>0=2 #CIs<0=2
              min(5)=        1 ->  3         0.01352
              #CIs=9 #CIs>0=5 #CIs<0=4              11       Singlet-SG   81.6646         15.18    0.0000    0.000
                                                                      max(1)=        1 -> 12         0.70620
8         Singlet-SG   64.0602         19.35    0.1254    0.000      max(2)=        1 ->  9         0.02804
              max(1)=        1 ->  9         0.70608                 max(3)=        1 ->  8         0.00742
              max(2)=        1 ->  5         0.01421                 max(4)=        1 ->  4         0.00289
              max(3)=        1 ->  3         0.00753                 max(5)=        1 ->  2         0.00201
              max(4)=        1 -> 13         0.00678                 min(1)=        1 -> 13        -0.01945
              max(5)=        1 -> 14         0.00307                 min(2)=        1 ->  5        -0.00788
              min(1)=        1 -> 12        -0.02742                 min(3)=        1 -> 14        -0.00016
              min(2)=        1 ->  2        -0.02419                 min(4)=        1 ->  3         0.00158
              min(3)=        1 ->  8        -0.01586                 min(5)=        1 ->  2         0.00201
              min(4)=        1 ->  4        -0.00683                 #CIs=9 #CIs>0=6 #CIs<0=3
              min(5)=        1 -> 14         0.00307
              #CIs=9 #CIs>0=5 #CIs<0=4              12       Singlet-SG   97.3985         12.73    0.0185    0.000
                                                                      max(1)=        1 -> 13         0.70669
9         Singlet-?Sym   67.8710         18.27    0.0651    0.000      max(2)=        1 -> 12         0.01962
              max(1)=        1 -> 10         0.69698                 max(3)=        1 ->  8         0.00999
              max(2)=        1 -> 11         0.11508                 max(4)=        1 ->  2         0.00511
              max(3)=        1 ->  6        -0.00529                 max(5)=        1 ->  4         0.00491
              max(4)=        1 ->  7        -0.03202                 min(1)=        1 -> 14        -0.00857
              max(5)=        1 ->  9         0.70608                 min(2)=        1 ->  9        -0.00577
              min(1)=        1 ->  7        -0.03202                 min(3)=        1 ->  3         0.00334
              min(2)=        1 ->  6        -0.00529                 min(4)=        1 ->  5         0.00400
              min(3)=        1 -> 11         0.11508                 min(5)=        1 ->  4         0.00491
              min(4)=        1 -> 10         0.69698                 #CIs=9 #CIs>0=7 #CIs<0=2
              min(5)=        1 -> 12        -0.02742
              #CIs=4 #CIs>0=2 #CIs<0=2              13       Singlet-SG  194.5931          6.37    0.0000    0.000
                                                                      max(1)=        1 -> 14         0.70703
10        Singlet-?Sym   67.8710         18.27    0.0651    0.000      max(2)=        1 ->  8         0.01027
              max(1)=        1 -> 11         0.69698                 max(3)=        1 ->  2         0.00937
              max(2)=        1 ->  7         0.00529                 max(4)=        1 -> 13         0.00846
              max(3)=        1 ->  6        -0.03202                 max(5)=        1 ->  4         0.00666
```

```
                min(1)=        1 ->  9        -0.00279                 min(4)=        1 ->  3         0.00218
                min(2)=        1 ->  5        -0.00189                 min(5)=        1 ->  4         0.00666
                min(3)=        1 -> 12         0.00036                 #CIs=9 #CIs>0=7 #CIs<0=2
```

3.2.3 Main contributions from different excited states at CIS approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_         min(1)=        1 ->  3        -0.18050
                                                                       min(2)=        1 ->  5        -0.16569
1        Singlet-SG   27.2151         45.56    0.4638    0.000        min(3)=        1 ->  2        -0.09407
               max(1)=        1 ->  2         0.68603                 min(4)=        1 ->  9        -0.00754
               max(2)=        1 ->  4         0.03846                 min(5)=        1 -> 12        -0.00595
               max(3)=        1 -> 13        -0.00033                 #CIs=9 #CIs>0=3 #CIs<0=6
               max(4)=        1 -> 12        -0.00190
               max(5)=        1 ->  9        -0.00389   4       Singlet-SG   41.4654         29.90    0.0604    0.000
               min(1)=        1 ->  3        -0.14286                 max(1)=        1 ->  5         0.66948
               min(2)=        1 ->  5        -0.08511                 max(2)=        1 ->  4         0.12336
               min(3)=        1 ->  8        -0.01384                 max(3)=        1 ->  2         0.03956
               min(4)=        1 -> 14        -0.00462                 max(4)=        1 ->  8         0.03758
               min(5)=        1 ->  9        -0.00389                 max(5)=        1 ->  9         0.02596
               #CIs=9 #CIs>0=2 #CIs<0=7                               min(1)=        1 ->  3        -0.18026
                                                                       min(2)=        1 -> 13        -0.00917
2        Singlet-SG   33.4779         37.03    0.0270    0.000        min(3)=        1 -> 14         0.00045
               max(1)=        1 ->  3         0.64369                 min(4)=        1 -> 12         0.01865
               max(2)=        1 ->  4         0.22561                 min(5)=        1 ->  9         0.02596
               max(3)=        1 ->  2         0.13717                 #CIs=9 #CIs>0=7 #CIs<0=2
               max(4)=        1 ->  5         0.12136
               max(5)=        1 ->  8         0.03399   5       Singlet-?Sym   50.2689         24.66    0.7008    0.000
               min(1)=        1 -> 13        -0.00281                 max(1)=        1 ->  6         0.56957
               min(2)=        1 -> 14        -0.00215                 max(2)=        1 -> 10         0.01283
               min(3)=        1 -> 12         0.00259                 max(3)=        1 -> 11        -0.01746
               min(4)=        1 ->  9         0.00699                 max(4)=        1 ->  7        -0.41848
               min(5)=        1 ->  8         0.03399                 max(5)=        1 ->  5         0.66948
               #CIs=9 #CIs>0=7 #CIs<0=2                               min(1)=        1 ->  7        -0.41848
                                                                       min(2)=        1 -> 11        -0.01746
3        Singlet-SG   38.1164         32.53    0.0175    0.000        min(3)=        1 -> 10         0.01283
               max(1)=        1 ->  4         0.65507                 min(4)=        1 ->  6         0.56957
               max(2)=        1 ->  8         0.04347                 min(5)=        1 ->  3        -0.18026
               max(3)=        1 -> 13         0.00254                 #CIs=4 #CIs>0=2 #CIs<0=2
               max(4)=        1 -> 14        -0.00334
               max(5)=        1 -> 12        -0.00595   6       Singlet-?Sym   50.2689         24.66    0.7008    0.000
```

```
              max(1)=        1 ->  7         0.56957                 max(3)=        1 ->  7         0.02081
              max(2)=        1 ->  6         0.41848                 max(4)=        1 ->  6         0.00603
              max(3)=        1 -> 11        -0.01283                 max(5)=        1 ->  9         0.70507
              max(4)=        1 -> 10        -0.01746                 min(1)=        1 ->  6         0.00603
              max(5)=        1 ->  6         0.56957                 min(2)=        1 ->  7         0.02081
              min(1)=        1 -> 10        -0.01746                 min(3)=        1 -> 11         0.19673
              min(2)=        1 -> 11        -0.01283                 min(4)=        1 -> 10         0.67884
              min(3)=        1 ->  6         0.41848                 min(5)=        1 -> 12        -0.03697
              min(4)=        1 ->  7         0.56957                 #CIs=4 #CIs>0=4 #CIs<0=0
              min(5)=        1 ->  7        -0.41848
              #CIs=4 #CIs>0=2 #CIs<0=2              10       Singlet-?Sym   69.9154         17.73    0.0538    0.000
                                                                     max(1)=        1 -> 11         0.67884
 7        Singlet-SG   55.4610         22.36    0.0011    0.000      max(2)=        1 ->  6         0.02081
              max(1)=        1 ->  8         0.70363                 max(3)=        1 ->  7        -0.00603
              max(2)=        1 -> 13         0.01144                 max(4)=        1 -> 10        -0.19673
              max(3)=        1 ->  2         0.01060                 max(5)=        1 -> 10         0.67884
              max(4)=        1 ->  9         0.00999                 min(1)=        1 -> 10        -0.19673
              max(5)=        1 -> 14         0.00250                 min(2)=        1 ->  7        -0.00603
              min(1)=        1 ->  4        -0.05715                 min(3)=        1 ->  6         0.02081
              min(2)=        1 ->  5        -0.03340                 min(4)=        1 -> 11         0.67884
              min(3)=        1 ->  3        -0.01315                 min(5)=        1 ->  6         0.00603
              min(4)=        1 -> 12         0.00112                 #CIs=4 #CIs>0=2 #CIs<0=2
              min(5)=        1 -> 14         0.00250
              #CIs=9 #CIs>0=6 #CIs<0=3              11       Singlet-SG   84.3348         14.70    0.0001    0.000
                                                                     max(1)=        1 -> 12         0.70522
 8        Singlet-SG   65.8644         18.82    0.1411    0.000      max(2)=        1 ->  9         0.03721
              max(1)=        1 ->  9         0.70507                 max(3)=        1 -> 14         0.00281
              max(2)=        1 -> 13         0.02621                 max(4)=        1 ->  4         0.00166
              max(3)=        1 ->  4         0.00118                 max(5)=        1 ->  3         0.00035
              max(4)=        1 ->  2        -0.00012                 min(1)=        1 -> 13        -0.02834
              max(5)=        1 ->  3        -0.00226                 min(2)=        1 ->  5        -0.02154
              min(1)=        1 -> 12        -0.03697                 min(3)=        1 ->  8        -0.00207
              min(2)=        1 ->  5        -0.02612                 min(4)=        1 ->  2        -0.00057
              min(3)=        1 ->  8        -0.01155                 min(5)=        1 ->  3         0.00035
              min(4)=        1 -> 14        -0.00266                 #CIs=9 #CIs>0=5 #CIs<0=4
              min(5)=        1 ->  3        -0.00226
              #CIs=9 #CIs>0=3 #CIs<0=6              12       Singlet-SG  101.3328         12.24    0.0110    0.000
                                                                     max(1)=        1 -> 13         0.70589
 9        Singlet-?Sym   69.9154         17.73    0.0538    0.000      max(2)=        1 -> 12         0.02994
              max(1)=        1 -> 10         0.67884                 max(3)=        1 ->  5         0.01038
              max(2)=        1 -> 11         0.19673                 max(4)=        1 ->  2         0.00152
```

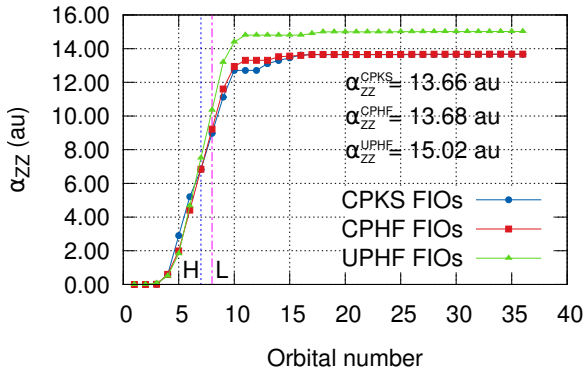
		max(5)=	1 -> 3	0.00111			max(2)=	1 -> 2	0.00439
		min(1)=	1 -> 9	-0.02446			max(3)=	1 -> 4	0.00416
		min(2)=	1 -> 8	-0.01060			max(4)=	1 -> 9	0.00241
		min(3)=	1 -> 3	0.00111			max(5)=	1 -> 3	0.00033
		min(4)=	1 -> 4	0.00111			min(1)=	1 -> 12	-0.00299
		min(5)=	1 -> 2	0.00152			min(2)=	1 -> 8	-0.00233
		#CIs=8 #CIs>0=6 #CIs<0=2					min(3)=	1 -> 5	-0.00129
							min(4)=	1 -> 13	0.00018
13	Singlet-SG	199.0088		6.23	0.0000	0.000	min(5)=	1 -> 3	0.00033
		max(1)=	1 -> 14	0.70707			#CIs=9 #CIs>0=6 #CIs<0=3		

4 N_2

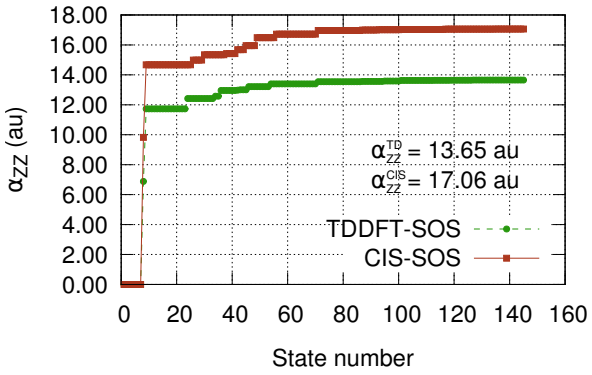
4.1 6-311G(d,p)

4.1.1 Plots

Figure S3: For N_2 molecule and 6-311G(d,p) basis set, plots of variation of α_{ZZ} with respect to the number of orbitals (FIO decomposition presented in this work, in Figure ZZZ) or states (SOS approach, in Figure S3a). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. Recomputed values of α_{ZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.

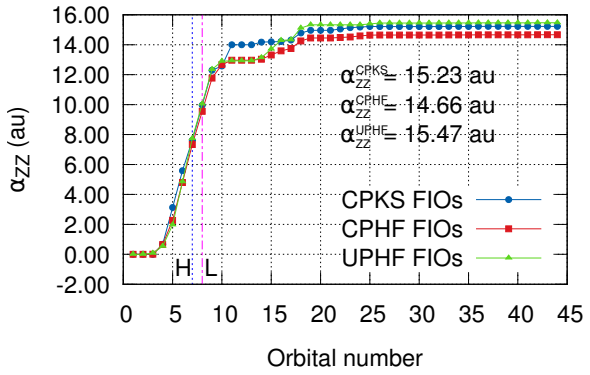


(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

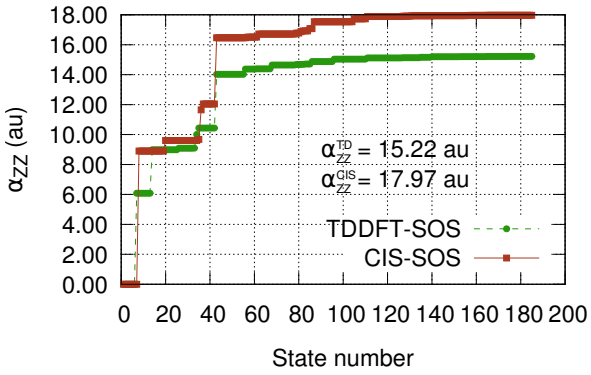
4.2 6-311++G(d,p)

4.2.1 Plots

Figure S4: For N_2 molecule and 6-311++G(d,p) basis set, plots of variation of α_{ZZ} with respect to the number of orbitals (FIO decomposition presented in this work, in Figure ZZZ) or states (SOS approach, in Figure S4a). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. Recomputed values of α_{ZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



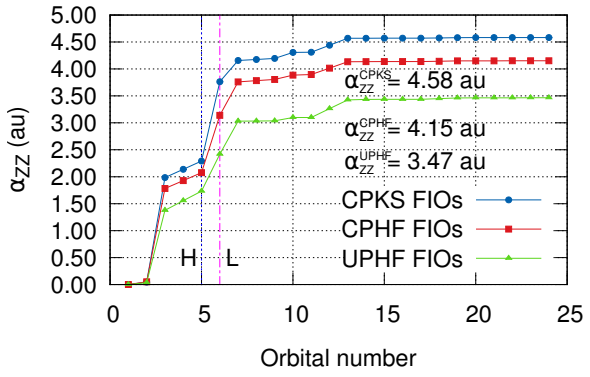
(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

5 HF

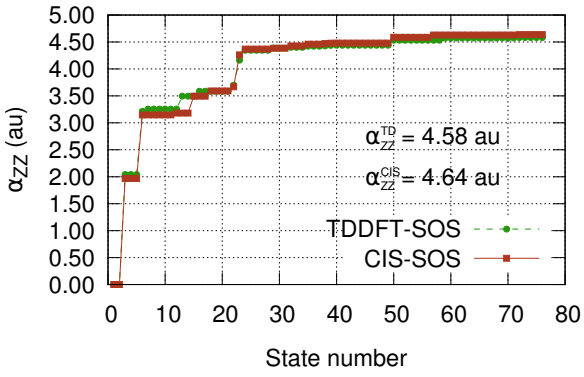
5.1 6-311G(d,p)

5.1.1 Plots

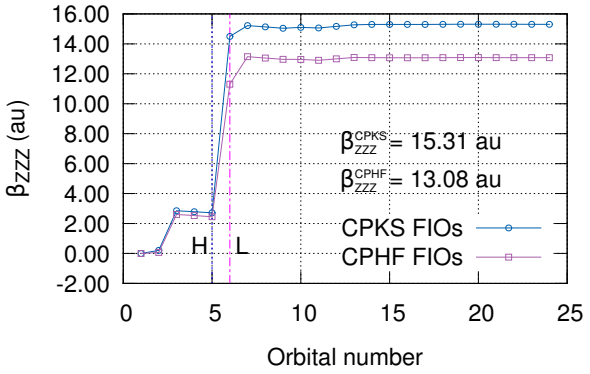
Figure S5: For HF molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S5a and S5c) or states (SOS approaches, in Plots S5b and S5d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.02 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



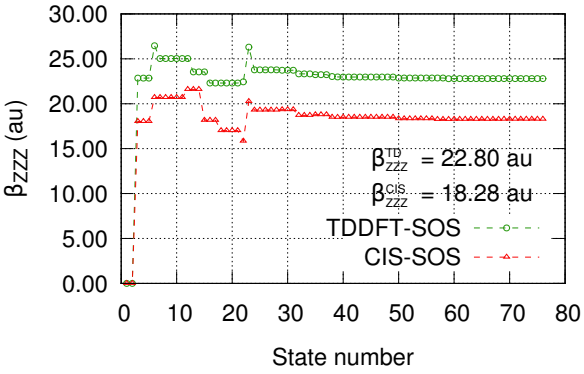
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

5.1.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_          min(4)=        2 -> 10        -0.00114
                                                                        min(5)=        3 -> 23        -0.00071
1        Singlet-?Sym    9.7484        127.18    0.0196    0.000        #CIs=24 #CIs>0=16 #CIs<0=8
               max(1)=        5 ->  6         0.70013
               max(2)=        5 ->  7         0.05593   4        Singlet-?Sym   19.1128         64.87    0.0380    0.000
               max(3)=        5 -> 11         0.00895                 max(1)=        4 ->  7         0.65596
               max(4)=        4 -> 17         0.00353                 max(2)=        5 ->  7         0.25691
               max(5)=        5 -> 16         0.00353                 max(3)=        5 -> 17         0.00522
               min(1)=        4 ->  6        -0.08111                 max(4)=        2 -> 12         0.00518
               min(2)=        5 -> 10        -0.01210                 max(5)=        3 -> 18         0.00240
               min(3)=        4 ->  7        -0.00648                 min(1)=        4 ->  6        -0.05264
               min(4)=        2 ->  9        -0.00395                 min(2)=        5 ->  6        -0.02062
               min(5)=        2 -> 19        -0.00188                 min(3)=        4 -> 10        -0.01632
               #CIs=36 #CIs>0=19 #CIs<0=17                             min(4)=        4 -> 14        -0.01092
                                                                       min(5)=        3 -> 12        -0.00821
2        Singlet-?Sym    9.7484        127.18    0.0196    0.000        #CIs=37 #CIs>0=17 #CIs<0=20
               max(1)=        4 ->  6         0.70013
               max(2)=        5 ->  6         0.08111   5        Singlet-?Sym   19.1128         64.87    0.0380    0.000
               max(3)=        4 ->  7         0.05593                 max(1)=        5 ->  7         0.65596
               max(4)=        4 -> 11         0.00895                 max(2)=        4 ->  6         0.02062
               max(5)=        5 ->  7         0.00648                 max(3)=        4 -> 10         0.00639
               min(1)=        4 -> 10        -0.01210                 max(4)=        4 -> 17         0.00522
               min(2)=        2 ->  8        -0.00395                 max(5)=        5 -> 16         0.00522
               min(3)=        4 -> 16        -0.00353                 min(1)=        4 ->  7        -0.25691
               min(4)=        2 -> 18        -0.00188                 min(2)=        5 ->  6        -0.05264
               min(5)=        4 -> 20        -0.00186                 min(3)=        5 -> 10        -0.01632
               #CIs=36 #CIs>0=17 #CIs<0=19                             min(4)=        5 -> 14        -0.01092
                                                                       min(5)=        3 -> 13        -0.00821
3        Singlet-SG   14.2483         87.02    0.1866    0.000        #CIs=37 #CIs>0=19 #CIs<0=18
               max(1)=        3 ->  6         0.70537
               max(2)=        4 ->  8         0.03354   6        Singlet-SG   24.8444         49.90    0.3252    0.000
               max(3)=        5 ->  9         0.03354                 max(1)=        3 ->  7         0.69678
               max(4)=        3 -> 11         0.01300                 max(2)=        4 ->  8         0.07887
               max(5)=        2 -> 11         0.01211                 max(3)=        5 ->  9         0.07887
               min(1)=        3 ->  7        -0.02865                 max(4)=        4 -> 12         0.03505
               min(2)=        2 -> 14        -0.00293                 max(5)=        5 -> 13         0.03505
               min(3)=        2 -> 20        -0.00183                 min(1)=        2 -> 14        -0.00837
```

```
              min(2)=        4 -> 18        -0.00807                min(4)=        5 -> 12        -0.00011
              min(3)=        5 -> 19        -0.00807                min(5)=        5 ->  9        -0.00001
              min(4)=        3 -> 20        -0.00445                #CIs=12 #CIs>0=7 #CIs<0=5
              min(5)=        3 -> 15        -0.00239
              #CIs=25 #CIs>0=16 #CIs<0=9             10       Singlet-?Sym   33.5752         36.93    0.0000    0.000
                                                                     max(1)=        4 ->  8         0.49999
7       Singlet-SG   31.7579         39.04    0.0200    0.000          max(2)=        4 -> 22         0.00348
              max(1)=        2 ->  6         0.69080                 max(3)=        5 -> 19         0.00012
              max(2)=        2 ->  7         0.02635                 max(4)=        5 -> 13         0.00011
              max(3)=        3 -> 11         0.02229                 max(5)=        4 ->  9        -0.00001
              max(4)=        4 -> 12         0.01501                 min(1)=        5 ->  9        -0.49999
              max(5)=        5 -> 13         0.01501                 min(2)=        5 -> 21        -0.00348
              min(1)=        4 ->  8        -0.10235                 min(3)=        2 -> 16        -0.00256
              min(2)=        5 ->  9        -0.10235                 min(4)=        3 -> 16        -0.00126
              min(3)=        3 -> 10        -0.01609                 min(5)=        4 -> 18        -0.00012
              min(4)=        4 -> 18        -0.00780                 #CIs=12 #CIs>0=4 #CIs<0=8
              min(5)=        5 -> 19        -0.00780
              #CIs=25 #CIs>0=13 #CIs<0=12            11       Singlet-?Sym   34.6083         35.82    0.0004    0.000
                                                                     max(1)=        5 -> 10         0.70260
8       Singlet-?Sym   33.2149         37.33    0.0000    0.000          max(2)=        5 ->  7         0.01815
              max(1)=        5 ->  8         0.50001                 max(3)=        5 ->  6         0.01069
              max(2)=        5 -> 22         0.00357                 max(4)=        5 -> 14         0.00742
              max(3)=        4 -> 19         0.00026                 max(5)=        4 -> 17         0.00561
              max(4)=        5 -> 12         0.00022                 min(1)=        3 ->  9        -0.05751
              max(5)=        4 -> 13        -0.00022                 min(2)=        4 -> 10        -0.04755
              min(1)=        4 ->  9        -0.50001                 min(3)=        3 -> 13        -0.01150
              min(2)=        4 -> 21        -0.00357                 min(4)=        2 ->  9        -0.00870
              min(3)=        5 -> 18        -0.00026                 min(5)=        5 -> 15        -0.00464
              min(4)=        4 -> 13        -0.00022                 #CIs=36 #CIs>0=18 #CIs<0=18
              min(5)=        5 -> 12         0.00022
              #CIs=8 #CIs>0=4 #CIs<0=4               12       Singlet-?Sym   34.6083         35.82    0.0004    0.000
                                                                     max(1)=        4 -> 10         0.70260
9       Singlet-?Sym   33.5752         36.93    0.0000    0.000          max(2)=        5 -> 10         0.04755
              max(1)=        4 ->  9         0.49999                 max(3)=        4 ->  7         0.01815
              max(2)=        5 ->  8         0.49999                 max(4)=        4 ->  6         0.01069
              max(3)=        4 -> 21         0.00348                 max(5)=        4 -> 14         0.00742
              max(4)=        5 -> 22         0.00348                 min(1)=        3 ->  8        -0.05751
              max(5)=        2 -> 17         0.00256                 min(2)=        3 -> 12        -0.01150
              min(1)=        4 -> 19        -0.00012                 min(3)=        2 ->  8        -0.00870
              min(2)=        5 -> 18        -0.00012                 min(4)=        4 -> 16        -0.00561
              min(3)=        4 -> 13        -0.00011                 min(5)=        4 -> 15        -0.00464
```

```
                #CIs=36 #CIs>0=21 #CIs<0=15             16       Singlet-SG   41.0629         30.19    0.0690    0.000

                                                                        max(1)=        2 ->  7         0.67483
13       Singlet-SG   37.0885         33.43    0.1478    0.000         max(2)=        3 -> 11         0.06680
                max(1)=        3 -> 10         0.52121                 max(3)=        2 -> 11         0.00524
                max(2)=        2 ->  7         0.10466                 max(4)=        3 ->  7         0.00470
                max(3)=        3 ->  7         0.06640                 max(5)=        4 -> 22         0.00198
                max(4)=        3 ->  6         0.02774                 min(1)=        3 -> 10        -0.18857
                max(5)=        2 -> 11         0.02669                 min(2)=        2 ->  6        -0.04316
                min(1)=        4 ->  8        -0.31905                 min(3)=        4 ->  8        -0.03670
                min(2)=        5 ->  9        -0.31905                 min(4)=        5 ->  9        -0.03670
                min(3)=        2 ->  6        -0.08795                 min(5)=        4 -> 18        -0.00650
                min(4)=        2 -> 14        -0.00782                 #CIs=26 #CIs>0=7 #CIs<0=19
                min(5)=        3 -> 15        -0.00628
                #CIs=26 #CIs>0=15 #CIs<0=11             17       Singlet-?Sym   43.6287         28.42    0.2384    0.000

                                                                        max(1)=        5 -> 11         0.70330
14       Singlet-?Sym   37.4368         33.12    0.0010    0.000         max(2)=        4 -> 11         0.04835
                max(1)=        3 ->  9         0.70452                 max(3)=        3 -> 13         0.04384
                max(2)=        5 -> 10         0.05715                 max(4)=        3 ->  9         0.00823
                max(3)=        4 -> 17         0.00543                 max(5)=        3 -> 19         0.00359
                max(4)=        5 -> 16         0.00543                 min(1)=        2 ->  9        -0.03136
                max(5)=        3 -> 19         0.00455                 min(2)=        5 ->  6        -0.00890
                min(1)=        3 -> 13        -0.01455                 min(3)=        2 -> 13        -0.00477
                min(2)=        5 -> 11        -0.00788                 min(4)=        5 -> 20        -0.00454
                min(3)=        2 ->  9        -0.00740                 min(5)=        2 ->  8        -0.00216
                min(4)=        2 -> 13        -0.00370                 #CIs=36 #CIs>0=17 #CIs<0=19
                min(5)=        5 -> 20        -0.00153
                #CIs=28 #CIs>0=17 #CIs<0=11             18       Singlet-?Sym   43.6287         28.42    0.2384    0.000

                                                                        max(1)=        4 -> 11         0.70330
15       Singlet-?Sym   37.4368         33.12    0.0010    0.000         max(2)=        3 -> 12         0.04384
                max(1)=        3 ->  8         0.70452                 max(3)=        3 ->  8         0.00823
                max(2)=        4 -> 10         0.05715                 max(4)=        3 -> 18         0.00359
                max(3)=        5 -> 17         0.00543                 max(5)=        5 -> 17         0.00326
                max(4)=        3 -> 18         0.00455                 min(1)=        5 -> 11        -0.04835
                max(5)=        3 -> 22         0.00446                 min(2)=        2 ->  8        -0.03136
                min(1)=        3 -> 12        -0.01455                 min(3)=        4 ->  6        -0.00890
                min(2)=        4 -> 11        -0.00788                 min(4)=        2 -> 12        -0.00477
                min(3)=        2 ->  8        -0.00740                 min(5)=        4 -> 20        -0.00454
                min(4)=        4 -> 16        -0.00543                 #CIs=36 #CIs>0=17 #CIs<0=19
                min(5)=        2 -> 12        -0.00370
                #CIs=28 #CIs>0=14 #CIs<0=14             19       Singlet-?Sym   45.5255         27.23    0.0000    0.000

                                                                        max(1)=        4 -> 13         0.49999
```

```
                max(2)=        5 -> 12         0.49999                 min(2)=        5 -> 18        -0.00391
                max(3)=        3 -> 17         0.00086                 min(3)=        5 -> 22        -0.00026
                max(4)=        2 -> 17         0.00085                 min(4)=        5 ->  8        -0.00022
                max(5)=        5 -> 13         0.00075                 min(5)=        4 ->  9         0.00022
                min(1)=        4 -> 19        -0.00318                 #CIs=8 #CIs>0=4 #CIs<0=4
                min(2)=        5 -> 18        -0.00318
                min(3)=        4 -> 12        -0.00075   22       Singlet-SG   46.1756         26.85    0.1086    0.000
                min(4)=        4 -> 21        -0.00001                 max(1)=        3 -> 11         0.35181
                min(5)=        5 -> 22        -0.00001                 max(2)=        2 -> 11         0.12650
                #CIs=12 #CIs>0=7 #CIs<0=5                               max(3)=        3 ->  7         0.08663
                                                                        max(4)=        3 -> 14         0.04823
20       Singlet-?Sym   45.5255         27.23    0.0000    0.000       max(5)=        3 ->  6         0.03473
                max(1)=        4 -> 12         0.49999                 min(1)=        3 -> 10        -0.31846
                max(2)=        5 -> 19         0.00318                 min(2)=        4 ->  8        -0.27020
                max(3)=        4 -> 13         0.00075                 min(3)=        5 ->  9        -0.27020
                max(4)=        5 -> 12         0.00075                 min(4)=        4 -> 12        -0.19263
                max(5)=        4 ->  8         0.00010                 min(5)=        5 -> 13        -0.19263
                min(1)=        5 -> 13        -0.49999                 #CIs=26 #CIs>0=13 #CIs<0=13
                min(2)=        4 -> 18        -0.00318
                min(3)=        3 -> 16        -0.00086   23       Singlet-SG   47.4801         26.11    0.4714    0.000
                min(4)=        2 -> 16        -0.00085                 max(1)=        3 -> 11         0.41253
                min(5)=        5 ->  9        -0.00010                 max(2)=        4 -> 12         0.39957
                #CIs=12 #CIs>0=6 #CIs<0=6                               max(3)=        5 -> 13         0.39957
                                                                        max(4)=        4 -> 18         0.01712
21       Singlet-?Sym   45.5781         27.20    0.0000    0.000       max(5)=        5 -> 19         0.01712
                max(1)=        5 -> 12         0.49999                 min(1)=        3 -> 14        -0.05195
                max(2)=        4 -> 19         0.00391                 min(2)=        2 -> 10        -0.04999
                max(3)=        4 -> 21         0.00026                 min(3)=        2 ->  7        -0.04505
                max(4)=        4 ->  9         0.00022                 min(4)=        3 ->  7        -0.03926
                max(5)=        5 ->  8        -0.00022                 min(5)=        2 ->  6        -0.02861
                min(1)=        4 -> 13        -0.49999                 #CIs=25 #CIs>0=15 #CIs<0=10
```

5.1.3 Main contributions from different excited states at CIS approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_          max(4)=        4 -> 10         0.02682
                                                                        max(5)=        5 -> 15         0.00971
1        Singlet-?Sym   12.0824        102.62    0.0238    0.000       min(1)=        4 ->  6        -0.42020
                max(1)=        5 ->  6         0.53959                 min(2)=        4 ->  7        -0.10263
                max(2)=        5 ->  7         0.13179                 min(3)=        5 -> 10        -0.03444
                max(3)=        5 -> 11         0.03502                 min(4)=        4 -> 11        -0.02727
```

```
                min(5)=        5 -> 14        -0.01179
                #states=34 #states>0=16 #states<0=18     5        Singlet-?Sym   21.6414         57.29    0.0565    0.000
                                                                        max(1)=        4 ->  7         0.56812
 2        Singlet-?Sym   12.0824        102.62    0.0238    0.000        max(2)=        5 ->  7         0.37572
                max(1)=        4 ->  6         0.53959                 max(3)=        4 -> 15         0.01263
                max(2)=        5 ->  6         0.42020                 max(4)=        5 -> 15         0.00835
                max(3)=        4 ->  7         0.13179                 max(5)=        2 -> 12         0.00763
                max(4)=        5 ->  7         0.10263                 min(1)=        4 ->  6        -0.14313
                max(5)=        4 -> 11         0.03502                 min(2)=        5 ->  6        -0.09466
                min(1)=        4 -> 10        -0.03444                 min(3)=        4 -> 10        -0.05179
                min(2)=        5 -> 10        -0.02682                 min(4)=        4 -> 14        -0.03867
                min(3)=        4 -> 14        -0.01179                 min(5)=        5 -> 10        -0.03425
                min(4)=        5 -> 14        -0.00918                 #states=36 #states>0=17 #states<0=19
                min(5)=        4 -> 23        -0.00311
                #states=34 #states>0=21 #states<0=13     6        Singlet-SG   27.0282         45.87    0.3872    0.000
                                                                        max(1)=        3 ->  7         0.68277
 3        Singlet-SG   16.4027         75.59    0.2385    0.000        max(2)=        4 ->  8         0.10281
                max(1)=        3 ->  6         0.69825                 max(3)=        5 ->  9         0.10281
                max(2)=        3 ->  7         0.07072                 max(4)=        4 -> 12         0.03791
                max(3)=        4 ->  8         0.05015                 max(5)=        5 -> 13         0.03791
                max(4)=        5 ->  9         0.05015                 min(1)=        3 ->  6        -0.08600
                max(5)=        3 -> 11         0.04267                 min(2)=        3 -> 10        -0.02836
                min(1)=        3 -> 10        -0.00654                 min(3)=        2 ->  7        -0.02226
                min(2)=        2 ->  6        -0.00604                 min(4)=        3 -> 14        -0.01898
                min(3)=        2 -> 14        -0.00566                 min(5)=        4 -> 19        -0.00846
                min(4)=        3 -> 23        -0.00179                 #states=24 #states>0=14 #states<0=10
                min(5)=        2 -> 23        -0.00155
                #states=24 #states>0=19 #states<0=5      7        Singlet-?Sym   34.1898         36.26    0.0000    0.000
                                                                        max(1)=        5 ->  8         0.49975
 4        Singlet-?Sym   21.6414         57.29    0.0565    0.000        max(2)=        5 -> 22         0.01482
                max(1)=        5 ->  7         0.56812                 max(3)=        5 -> 12         0.00500
                max(2)=        4 ->  6         0.09466                 max(4)=        4 -> 18         0.00063
                max(3)=        4 -> 10         0.03425                 max(5)=        5 -> 19        -0.00063
                max(4)=        4 -> 14         0.02557                 min(1)=        4 ->  9        -0.49975
                max(5)=        5 -> 15         0.01263                 min(2)=        4 -> 21        -0.01482
                min(1)=        4 ->  7        -0.37572                 min(3)=        4 -> 13        -0.00500
                min(2)=        5 ->  6        -0.14313                 min(4)=        5 -> 19        -0.00063
                min(3)=        5 -> 10        -0.05179                 min(5)=        4 -> 18         0.00063
                min(4)=        5 -> 14        -0.03867                 #states=8 #states>0=4 #states<0=4
                min(5)=        3 -> 13        -0.00879
                #states=36 #states>0=17 #states<0=19     8        Singlet-?Sym   34.5435         35.89    0.0000    0.000
```

```
                max(1)=        5 ->  9         0.49980                 max(3)=        4 -> 11         0.03841
                max(2)=        5 -> 21         0.01358                 max(4)=        4 ->  6         0.02249
                max(3)=        5 -> 13         0.00344                 max(5)=        3 ->  9         0.01699
                max(4)=        2 -> 17         0.00333                 min(1)=        5 -> 10        -0.31508
                max(5)=        3 -> 17         0.00067                 min(2)=        3 ->  8        -0.03369
                min(1)=        4 ->  8        -0.49980                 min(3)=        5 ->  7        -0.03223
                min(2)=        4 -> 22        -0.01358                 min(4)=        5 -> 11        -0.01936
                min(3)=        4 -> 12        -0.00344                 min(5)=        3 -> 12        -0.01257
                min(4)=        5 -> 18        -0.00066                 #states=35 #states>0=18 #states<0=17
                min(5)=        4 -> 19         0.00066
                #states=10 #states>0=6 #states<0=4      12       Singlet-SG   38.0466         32.59    0.0230    0.000
                                                                        max(1)=        2 ->  6         0.53808
9       Singlet-?Sym   34.5435         35.89    0.0000    0.000        max(2)=        3 -> 10         0.25213
                max(1)=        4 ->  9         0.49980                 max(3)=        2 ->  7         0.14849
                max(2)=        5 ->  8         0.49980                 max(4)=        3 ->  7         0.08412
                max(3)=        4 -> 21         0.01358                 max(5)=        2 -> 11         0.03692
                max(4)=        5 -> 22         0.01358                 min(1)=        4 ->  8        -0.23618
                max(5)=        4 -> 13         0.00344                 min(2)=        5 ->  9        -0.23618
                min(1)=        2 -> 16        -0.00333                 min(3)=        2 -> 14        -0.01129
                min(2)=        3 -> 16        -0.00067                 min(4)=        2 -> 10        -0.00979
                min(3)=        4 -> 18        -0.00066                 min(5)=        4 -> 19        -0.00574
                min(4)=        5 -> 19        -0.00066                 #states=25 #states>0=13 #states<0=12
                min(5)=        4 -> 13         0.00344
                #states=10 #states>0=6 #states<0=4      13       Singlet-?Sym   38.5159         32.19    0.0001    0.000
                                                                        max(1)=        3 ->  8         0.69780
10      Singlet-?Sym   36.1022         34.34    0.0014    0.000        max(2)=        3 ->  9         0.10225
                max(1)=        5 -> 10         0.62498                 max(3)=        4 -> 10         0.03564
                max(2)=        4 -> 10         0.31508                 max(4)=        4 -> 11         0.02616
                max(3)=        5 ->  7         0.06394                 max(5)=        3 -> 22         0.01715
                max(4)=        5 -> 11         0.03841                 min(1)=        2 -> 12        -0.01288
                max(5)=        4 ->  7         0.03223                 min(2)=        2 -> 19        -0.00511
                min(1)=        3 ->  9        -0.03369                 min(3)=        4 ->  6        -0.00402
                min(2)=        3 ->  8        -0.01699                 min(4)=        2 -> 13        -0.00189
                min(3)=        3 -> 13        -0.01257                 min(5)=        4 -> 20        -0.00109
                min(4)=        5 -> 15        -0.01026                 #states=33 #states>0=24 #states<0=9
                min(5)=        3 -> 12        -0.00634
                #states=35 #states>0=21 #states<0=14     14       Singlet-?Sym   38.5159         32.19    0.0001    0.000
                                                                        max(1)=        3 ->  9         0.69780
11      Singlet-?Sym   36.1022         34.34    0.0014    0.000        max(2)=        5 -> 10         0.03564
                max(1)=        4 -> 10         0.62498                 max(3)=        5 -> 11         0.02616
                max(2)=        4 ->  7         0.06394                 max(4)=        3 -> 21         0.01715
```

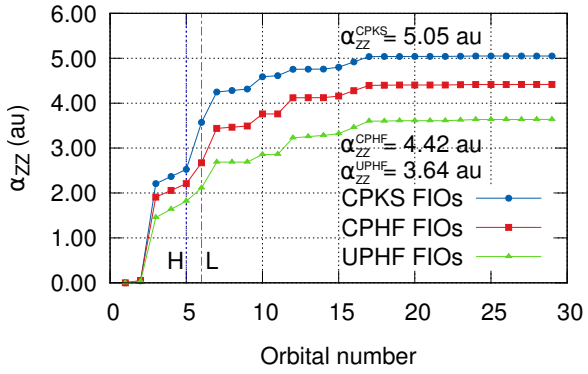
```
                max(5)=        2 ->  9         0.00592                 min(2)=        4 -> 10        -0.04034
                min(1)=        3 ->  8        -0.10225                 min(3)=        3 ->  8        -0.02493
                min(2)=        2 -> 13        -0.01288                 min(4)=        4 ->  7        -0.01206
                min(3)=        4 -> 10        -0.00522                 min(5)=        4 -> 20        -0.01188
                min(4)=        2 -> 18        -0.00511                 #states=31 #states>0=14 #states<0=17
                min(5)=        5 -> 17        -0.00421
                #states=33 #states>0=16 #states<0=17     18       Singlet-SG   47.2064         26.26    0.0982    0.000
                                                                        max(1)=        2 ->  7         0.62911
15       Singlet-SG   38.8694         31.90    0.2130    0.000          max(2)=        4 -> 12         0.08063
                max(1)=        3 -> 10         0.49289                 max(3)=        5 -> 13         0.08063
                max(2)=        3 ->  7         0.08178                 max(4)=        3 ->  7         0.03327
                max(3)=        2 -> 10         0.04860                 max(5)=        2 -> 11         0.01882
                max(4)=        3 ->  6         0.02090                 min(1)=        2 ->  6        -0.19425
                max(5)=        3 -> 14         0.02041                 min(2)=        3 -> 10        -0.17280
                min(1)=        2 ->  6        -0.40590                 min(3)=        4 ->  8        -0.09351
                min(2)=        4 ->  8        -0.19908                 min(4)=        5 ->  9        -0.09351
                min(3)=        5 ->  9        -0.19908                 min(5)=        2 -> 10        -0.04884
                min(4)=        2 ->  7        -0.05090                 #states=25 #states>0=13 #states<0=12
                min(5)=        3 -> 15        -0.01043
                #states=23 #states>0=14 #states<0=9      19       Singlet-?Sym   48.4145         25.61    0.0000    0.000
                                                                        max(1)=        5 -> 13         0.49976
16       Singlet-?Sym   45.1100         27.48    0.2896    0.000          max(2)=        4 -> 19         0.01496
                max(1)=        5 -> 11         0.70043                 max(3)=        4 ->  8         0.00346
                max(2)=        2 ->  9         0.02993                 max(4)=        4 -> 22         0.00015
                max(3)=        3 -> 13         0.02980                 max(5)=        2 -> 17        -0.00011
                max(4)=        5 -> 15         0.02857                 min(1)=        4 -> 12        -0.49976
                max(5)=        4 -> 16         0.00348                 min(2)=        5 -> 18        -0.01496
                min(1)=        5 ->  6        -0.04560                 min(3)=        5 ->  9        -0.00346
                min(2)=        4 -> 11        -0.04529                 min(4)=        3 -> 17        -0.00072
                min(3)=        5 -> 10        -0.04034                 min(5)=        5 -> 21        -0.00015
                min(4)=        3 ->  9        -0.02493                 #states=12 #states>0=4 #states<0=8
                min(5)=        5 ->  7        -0.01206
                #states=31 #states>0=13 #states<0=18     20       Singlet-?Sym   48.4145         25.61    0.0000    0.000
                                                                        max(1)=        4 -> 13         0.49976
17       Singlet-?Sym   45.1100         27.48    0.2896    0.000          max(2)=        5 -> 12         0.49976
                max(1)=        4 -> 11         0.70043                 max(3)=        3 -> 16         0.00072
                max(2)=        5 -> 11         0.04529                 max(4)=        5 -> 13         0.00012
                max(3)=        2 ->  8         0.02993                 max(5)=        2 -> 16         0.00011
                max(4)=        3 -> 12         0.02980                 min(1)=        4 -> 18        -0.01496
                max(5)=        4 -> 15         0.02857                 min(2)=        5 -> 19        -0.01496
                min(1)=        4 ->  6        -0.04560                 min(3)=        4 ->  9        -0.00346
```

```
                min(4)=        5 ->  8        -0.00346                max(4)=        2 -> 11         0.01561
                min(5)=        4 -> 21        -0.00015                max(5)=        3 ->  7         0.01530
                #states=12 #states>0=5 #states<0=7                     min(1)=        4 -> 12        -0.12835
                                                                       min(2)=        5 -> 13        -0.12835
21      Singlet-?Sym   48.5122         25.56    0.0000    0.000       min(3)=        3 -> 10        -0.08210
                max(1)=        5 -> 12         0.49972                min(4)=        4 ->  8        -0.05871
                max(2)=        4 -> 18         0.01595                min(5)=        5 ->  9        -0.05871
                max(3)=        4 ->  9         0.00501                #states=26 #states>0=13 #states<0=13
                max(4)=        4 -> 21         0.00045
                max(5)=        5 -> 22        -0.00045   23      Singlet-?Sym   50.5029         24.55    0.6813    0.000
                min(1)=        4 -> 13        -0.49972                max(1)=        4 -> 12         0.42459
                min(2)=        5 -> 19        -0.01595                max(2)=        5 -> 13         0.42459
                min(3)=        5 ->  8        -0.00501                max(3)=        3 -> 11         0.21226
                min(4)=        5 -> 22        -0.00045                max(4)=        4 ->  8         0.15236
                min(5)=        4 -> 21         0.00045                max(5)=        5 ->  9         0.15236
                #states=8 #states>0=4 #states<0=4                      min(1)=        3 ->  7        -0.09358
                                                                       min(2)=        3 -> 14        -0.08157
22      Singlet-SG   48.8362         25.39    0.0809    0.000         min(3)=        2 -> 11        -0.06761
                max(1)=        3 -> 11         0.66958                min(4)=        2 -> 10        -0.04123
                max(2)=        2 -> 10         0.03031                min(5)=        3 ->  6        -0.03072
                max(3)=        3 -> 15         0.02673                #states=26 #states>0=15 #states<0=11
```

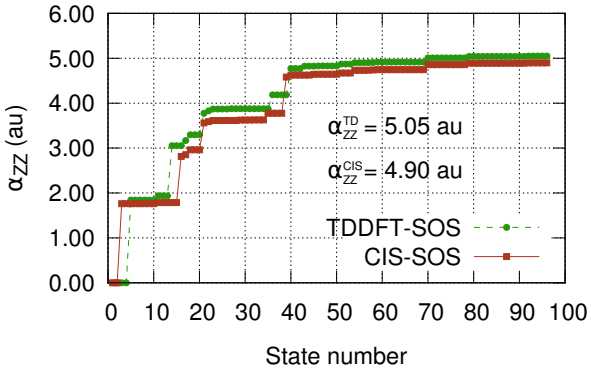
5.2 6-311++G(d,p)

5.2.1 Plots

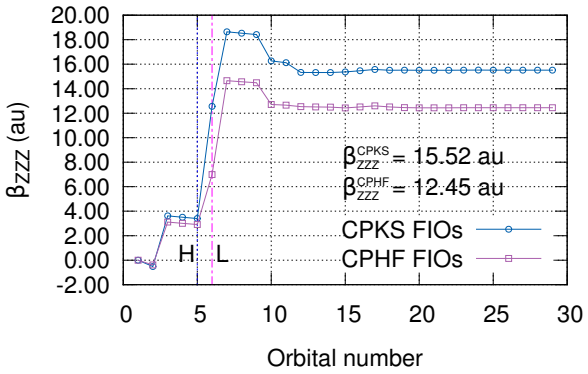
Figure S6: For HF molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S6a and S6c) or states (SOS approaches, in Plots S6b and S6d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.01 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



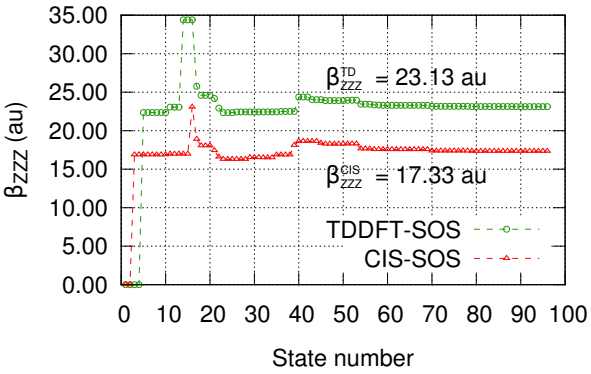
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

5.2.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_   Singlet-?Sym   13.2757         93.39    0.0161    0.000
                                                                        max(1)=        5 ->  7         0.59902
1        Singlet-?Sym    9.5784        129.44    0.0302    0.000        max(2)=        4 ->  6         0.07589
               max(1)=        5 ->  6         0.56118                 max(3)=        4 -> 10         0.04715
               max(2)=        4 ->  6         0.39106                 max(4)=        4 -> 12         0.03511
               max(3)=        5 ->  7         0.13820                 max(5)=        4 -> 15         0.00482
               max(4)=        4 ->  7         0.09630                 min(1)=        4 ->  7        -0.31328
               max(5)=        5 -> 11         0.03273                 min(2)=        5 ->  6        -0.14512
               min(1)=        5 -> 12        -0.02299                 min(3)=        5 -> 10        -0.09015
               min(2)=        4 -> 12        -0.01602                 min(4)=        5 -> 12        -0.06714
               min(3)=        5 -> 15        -0.00525                 min(5)=        5 -> 15        -0.00921
               min(4)=        4 -> 15        -0.00366                 #CIs=46 #CIs>0=22 #CIs<0=24
               min(5)=        2 -> 13        -0.00248
               #CIs=46 #CIs>0=23 #CIs<0=23            5        Singlet-SG   13.5686         91.38    0.1527    0.000
                                                                        max(1)=        3 ->  6         0.68540
2        Singlet-?Sym    9.5784        129.44    0.0302    0.000        max(2)=        4 ->  8         0.09770
               max(1)=        4 ->  6         0.56118                 max(3)=        5 ->  9         0.09770
               max(2)=        4 ->  7         0.13820                 max(4)=        3 ->  7         0.08276
               max(3)=        4 -> 11         0.03273                 max(5)=        3 -> 10         0.04809
               max(4)=        4 -> 10         0.03139                 min(1)=        2 -> 12        -0.00588
               max(5)=        5 -> 12         0.01602                 min(2)=        4 -> 17        -0.00316
               min(1)=        5 ->  6        -0.39106                 min(3)=        5 -> 16        -0.00316
               min(2)=        5 ->  7        -0.09630                 min(4)=        2 -> 15        -0.00314
               min(3)=        4 -> 12        -0.02299                 min(5)=        3 -> 15        -0.00280
               min(4)=        5 -> 11        -0.02281                 #CIs=32 #CIs>0=19 #CIs<0=13
               min(5)=        5 -> 10        -0.02187
               #CIs=46 #CIs>0=24 #CIs<0=22            6        Singlet-?Sym   15.0780         82.23    0.0000    0.000
                                                                        max(1)=        5 ->  8         0.49967
3        Singlet-?Sym   13.2757         93.39    0.0161    0.000        max(2)=        5 -> 14         0.01782
               max(1)=        4 ->  7         0.59902                 max(3)=        5 -> 17         0.00529
               max(2)=        5 ->  7         0.31328                 max(4)=        5 -> 26         0.00034
               max(3)=        3 -> 17         0.00278                 max(5)=        4 -> 24         0.00003
               max(4)=        4 -> 21         0.00174                 min(1)=        4 ->  9        -0.49967
               max(5)=        5 -> 22         0.00174                 min(2)=        4 -> 13        -0.01782
               min(1)=        4 ->  6        -0.14512                 min(3)=        4 -> 16        -0.00529
               min(2)=        4 -> 10        -0.09015                 min(4)=        4 -> 27        -0.00034
               min(3)=        5 ->  6        -0.07589                 min(5)=        5 -> 23        -0.00003
               min(4)=        4 -> 12        -0.06714                 #CIs=10 #CIs>0=5 #CIs<0=5
               min(5)=        5 -> 10        -0.04715
               #CIs=46 #CIs>0=23 #CIs<0=23            7        Singlet-?Sym   15.1837         81.66    0.0000    0.000
                                                                        max(1)=        4 ->  9         0.49978
```

```
              max(2)=        5 ->  8         0.49978                 max(4)=        4 -> 15         0.01138
              max(3)=        4 -> 13         0.01406                 max(5)=        5 -> 11         0.00746
              max(4)=        5 -> 14         0.01406                 min(1)=        5 -> 10        -0.24426
              max(5)=        4 -> 16         0.00458                 min(2)=        4 ->  6        -0.05631
              min(1)=        4 -> 24        -0.00002                 min(3)=        5 ->  7        -0.03112
              min(2)=        5 -> 23        -0.00002                 min(4)=        4 -> 11        -0.01997
              min(3)=        4 -> 27         0.00051                 min(5)=        4 -> 12        -0.01377
              min(4)=        5 -> 26         0.00051                 #CIs=47 #CIs>0=25 #CIs<0=22
              min(5)=        3 -> 22         0.00083
              #CIs=12 #CIs>0=10 #CIs<0=2              11       Singlet-?Sym   16.4171         75.52    0.0115    0.000
                                                                      max(1)=        4 ->  8         0.39821
8       Singlet-?Sym   15.1837         81.66    0.0000    0.000      max(2)=        5 ->  9         0.39821
              max(1)=        4 ->  8         0.49978                 max(3)=        3 ->  7         0.36890
              max(2)=        4 -> 14         0.01406                 max(4)=        3 -> 11         0.02117
              max(3)=        4 -> 17         0.00458                 max(5)=        2 -> 12         0.00980
              max(4)=        2 -> 21         0.00199                 min(1)=        3 ->  6        -0.14835
              max(5)=        3 -> 21         0.00083                 min(2)=        3 -> 10        -0.14758
              min(1)=        5 ->  9        -0.49978                 min(3)=        3 -> 12        -0.04469
              min(2)=        5 -> 13        -0.01406                 min(4)=        3 -> 15        -0.02312
              min(3)=        5 -> 16        -0.00458                 min(5)=        2 ->  7        -0.01835
              min(4)=        5 -> 27        -0.00051                 #CIs=34 #CIs>0=8 #CIs<0=26
              min(5)=        4 -> 23        -0.00002
              #CIs=12 #CIs>0=7 #CIs<0=5               12       Singlet-?Sym   18.2404         67.97    0.0924    0.000
                                                                      max(1)=        4 -> 11         0.69518
9       Singlet-?Sym   15.6619         79.16    0.0309    0.000      max(2)=        5 -> 11         0.10727
              max(1)=        5 -> 10         0.65417                 max(3)=        3 ->  8         0.05108
              max(2)=        4 -> 10         0.24426                 max(4)=        4 -> 10         0.01910
              max(3)=        5 ->  7         0.08336                 max(5)=        4 -> 12         0.01572
              max(4)=        4 ->  7         0.03112                 min(1)=        4 ->  6        -0.04058
              max(5)=        5 -> 15         0.01138                 min(2)=        4 -> 15        -0.01356
              min(1)=        5 ->  6        -0.05631                 min(3)=        5 ->  6        -0.00626
              min(2)=        4 ->  6        -0.02102                 min(4)=        3 -> 17        -0.00440
              min(3)=        5 -> 11        -0.01997                 min(5)=        2 -> 14        -0.00284
              min(4)=        5 -> 12        -0.01377                 #CIs=47 #CIs>0=22 #CIs<0=25
              min(5)=        4 -> 11        -0.00746
              #CIs=47 #CIs>0=16 #CIs<0=31             13       Singlet-?Sym   18.2404         67.97    0.0924    0.000
                                                                      max(1)=        5 -> 11         0.69518
10      Singlet-?Sym   15.6619         79.16    0.0309    0.000      max(2)=        3 ->  9         0.05108
              max(1)=        4 -> 10         0.65417                 max(3)=        5 -> 10         0.01910
              max(2)=        4 ->  7         0.08336                 max(4)=        5 -> 12         0.01572
              max(3)=        5 ->  6         0.02102                 max(5)=        4 ->  6         0.00626
```

```
                min(1)=        4 -> 11        -0.10727                min(3)=        5 -> 15        -0.00237
                min(2)=        5 ->  6        -0.04058                min(4)=        5 -> 21        -0.00234
                min(3)=        5 -> 15        -0.01356                min(5)=        5 -> 18        -0.00177
                min(4)=        3 ->  8        -0.00788                #CIs=43 #CIs>0=21 #CIs<0=22
                min(5)=        3 -> 16        -0.00440
                #CIs=47 #CIs>0=25 #CIs<0=22            17       Singlet-SG   19.9758         62.07    0.0203    0.000
                                                                       max(1)=        3 -> 10         0.64226
14       Singlet-SG   18.5639         66.79    0.1732    0.000       max(2)=        4 ->  8         0.15845
                max(1)=        3 ->  7         0.58095                 max(3)=        5 ->  9         0.15845
                max(2)=        3 -> 10         0.23673                 max(4)=        3 -> 11         0.09867
                max(3)=        3 -> 12         0.03949                 max(5)=        2 -> 12         0.00754
                max(4)=        4 -> 14         0.03630                 min(1)=        3 ->  7        -0.12997
                max(5)=        5 -> 13         0.03630                 min(2)=        3 ->  6        -0.07934
                min(1)=        4 ->  8        -0.22540                 min(3)=        3 -> 12        -0.05143
                min(2)=        5 ->  9        -0.22540                 min(4)=        4 -> 14        -0.03507
                min(3)=        3 -> 11        -0.03182                 min(5)=        5 -> 13        -0.03507
                min(4)=        3 ->  6        -0.02141                 #CIs=34 #CIs>0=13 #CIs<0=21
                min(5)=        2 -> 12        -0.01690
                #CIs=33 #CIs>0=19 #CIs<0=14            18       Singlet-SG   21.9019         56.61    0.0283    0.000
                                                                       max(1)=        3 -> 11         0.69699
15       Singlet-?Sym   18.8792         65.67    0.0014    0.000       max(2)=        3 ->  7         0.02726
                max(1)=        3 ->  8         0.70473                 max(3)=        3 -> 12         0.02201
                max(2)=        3 -> 14         0.01951                 max(4)=        2 ->  6         0.02033
                max(3)=        3 -> 17         0.00878                 max(5)=        2 ->  7         0.01850
                max(4)=        4 -> 10         0.00439                 min(1)=        3 -> 10        -0.07745
                max(5)=        3 -> 23         0.00235                 min(2)=        4 ->  8        -0.04874
                min(1)=        4 -> 11        -0.05198                 min(3)=        5 ->  9        -0.04874
                min(2)=        3 ->  9        -0.01326                 min(4)=        3 ->  6        -0.02803
                min(3)=        2 ->  8        -0.00249                 min(5)=        2 -> 12        -0.00687
                min(4)=        4 -> 15        -0.00237                 #CIs=33 #CIs>0=21 #CIs<0=12
                min(5)=        4 -> 18        -0.00177
                #CIs=43 #CIs>0=23 #CIs<0=20            19       Singlet-?Sym   22.5967         54.87    0.0250    0.000
                                                                       max(1)=        4 -> 12         0.59791
16       Singlet-?Sym   18.8792         65.67    0.0014    0.000       max(2)=        5 -> 12         0.36723
                max(1)=        3 ->  9         0.70473                 max(3)=        4 ->  7         0.06976
                max(2)=        3 -> 13         0.01951                 max(4)=        5 ->  7         0.04284
                max(3)=        3 ->  8         0.01326                 max(5)=        4 -> 15         0.01263
                max(4)=        3 -> 16         0.00878                 min(1)=        4 -> 11        -0.01271
                max(5)=        5 -> 10         0.00439                 min(2)=        4 -> 19        -0.01107
                min(1)=        5 -> 11        -0.05198                 min(3)=        5 -> 11        -0.00781
                min(2)=        2 ->  9        -0.00249                 min(4)=        3 -> 17        -0.00683
```

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                min(5)=        5 -> 19        -0.00680
                #CIs=46 #CIs>0=27 #CIs<0=19             23       Singlet-SG   35.2070         35.22    0.0229    0.000
                                                                        max(1)=        2 ->  7         0.68655
20       Singlet-?Sym   22.5967         54.87    0.0250    0.000        max(2)=        4 ->  8         0.01356
                max(1)=        5 -> 12         0.59791                 max(3)=        5 ->  9         0.01356
                max(2)=        5 ->  7         0.06976                 max(4)=        3 -> 18         0.01188
                max(3)=        5 -> 15         0.01263                 max(5)=        3 -> 12         0.00803
                max(4)=        2 ->  9         0.01024                 min(1)=        2 -> 10        -0.11122
                max(5)=        5 ->  6         0.00794                 min(2)=        2 ->  6        -0.09972
                min(1)=        4 -> 12        -0.36723                 min(3)=        2 -> 12        -0.04923
                min(2)=        4 ->  7        -0.04284                 min(4)=        2 -> 11        -0.03786
                min(3)=        5 -> 11        -0.01271                 min(5)=        4 -> 14        -0.01987
                min(4)=        5 -> 19        -0.01107                 #CIs=32 #CIs>0=8 #CIs<0=24
                min(5)=        4 -> 15        -0.00775
                #CIs=46 #CIs>0=24 #CIs<0=22             24       Singlet-?Sym   37.0121         33.50    0.0164    0.000
                                                                        max(1)=        2 ->  8         0.68553
21       Singlet-SG   28.0003         44.28    0.1677    0.000        max(2)=        3 -> 17         0.01537
                max(1)=        3 -> 12         0.69279                 max(3)=        2 -> 14         0.01383
                max(2)=        4 ->  8         0.04307                 max(4)=        4 -> 21         0.00817
                max(3)=        5 ->  9         0.04307                 max(5)=        5 -> 22         0.00817
                max(4)=        3 -> 10         0.02267                 min(1)=        2 ->  9        -0.16898
                max(5)=        4 -> 17         0.02231                 min(2)=        3 -> 14        -0.01642
                min(1)=        4 -> 14        -0.06640                 min(3)=        4 -> 15        -0.01635
                min(2)=        5 -> 13        -0.06640                 min(4)=        4 -> 18        -0.01217
                min(3)=        2 ->  6        -0.06520                 min(5)=        4 -> 12        -0.01178
                min(4)=        2 -> 10        -0.03705                 #CIs=47 #CIs>0=25 #CIs<0=22
                min(5)=        2 ->  7        -0.02746
                #CIs=33 #CIs>0=15 #CIs<0=18             25       Singlet-?Sym   37.0121         33.50    0.0164    0.000
                                                                        max(1)=        2 ->  9         0.68553
22       Singlet-SG   31.5816         39.26    0.0249    0.000        max(2)=        2 ->  8         0.16898
                max(1)=        2 ->  6         0.69172                 max(3)=        3 -> 16         0.01537
                max(2)=        2 ->  7         0.09990                 max(4)=        2 -> 13         0.01383
                max(3)=        3 -> 12         0.06033                 max(5)=        4 -> 22         0.00817
                max(4)=        2 -> 10         0.02592                 min(1)=        3 -> 13        -0.01642
                max(5)=        4 ->  8         0.02170                 min(2)=        5 -> 15        -0.01635
                min(1)=        4 -> 14        -0.04758                 min(3)=        5 -> 18        -0.01217
                min(2)=        5 -> 13        -0.04758                 min(4)=        5 -> 12        -0.01178
                min(3)=        3 -> 15        -0.02596                 min(5)=        5 -> 21        -0.00817
                min(4)=        3 -> 11        -0.01921                 #CIs=47 #CIs>0=26 #CIs<0=21
                min(5)=        4 -> 17        -0.01828
                #CIs=33 #CIs>0=13 #CIs<0=20             26       Singlet-SG   37.4162         33.14    0.0010    0.000
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              max(1)=        2 -> 10         0.69005                  max(3)=        4 -> 16         0.00601
              max(2)=        2 ->  7         0.09841                  max(4)=        5 -> 17         0.00601
              max(3)=        3 -> 12         0.02113                  max(5)=        4 -> 27         0.00356
              max(4)=        4 ->  8         0.01193                  min(1)=        4 ->  9        -0.01412
              max(5)=        5 ->  9         0.01193                  min(2)=        5 ->  8        -0.01412
              min(1)=        4 -> 14        -0.07067                  min(3)=        4 -> 24        -0.00099
              min(2)=        5 -> 13        -0.07067                  min(4)=        5 -> 23        -0.00099
              min(3)=        2 ->  6        -0.05244                  min(5)=        3 -> 22         0.00114
              min(4)=        3 -> 15        -0.01965                  #CIs=12 #CIs>0=8 #CIs<0=4
              min(5)=        3 ->  7        -0.01064
              #CIs=34 #CIs>0=17 #CIs<0=17             30       Singlet-?Sym   42.0714         29.47    0.0000    0.000
                                                                      max(1)=        5 -> 13         0.49975
27       Singlet-SG   39.8666         31.10    0.0044    0.000        max(2)=        4 ->  8         0.01412
              max(1)=        2 -> 11         0.69972                  max(3)=        5 -> 16         0.00601
              max(2)=        2 -> 12         0.03729                  max(4)=        5 -> 27         0.00356
              max(3)=        2 ->  7         0.02974                  max(5)=        4 -> 23         0.00099
              max(4)=        3 -> 10         0.00959                  min(1)=        4 -> 14        -0.49975
              max(5)=        2 -> 18         0.00497                  min(2)=        5 ->  9        -0.01412
              min(1)=        4 -> 14        -0.05116                  min(3)=        4 -> 17        -0.00601
              min(2)=        5 -> 13        -0.05116                  min(4)=        4 -> 26        -0.00356
              min(3)=        2 ->  6        -0.03258                  min(5)=        2 -> 21        -0.00201
              min(4)=        4 -> 17        -0.02218                  #CIs=12 #CIs>0=5 #CIs<0=7
              min(5)=        5 -> 16        -0.02218
              #CIs=33 #CIs>0=13 #CIs<0=20             31       Singlet-?Sym   43.4895         28.51    0.0018    0.000
                                                                      max(1)=        5 -> 15         0.58358
28       Singlet-?Sym   41.8623         29.62    0.0000    0.000        max(2)=        4 -> 15         0.39205
              max(1)=        4 -> 13         0.49947                  max(3)=        3 -> 16         0.01818
              max(2)=        5 ->  8         0.01793                  max(4)=        3 -> 17         0.01221
              max(3)=        4 -> 16         0.01420                  max(5)=        5 -> 11         0.01217
              max(4)=        4 -> 27         0.00364                  min(1)=        3 -> 13        -0.05338
              max(5)=        5 -> 23         0.00128                  min(2)=        3 -> 14        -0.03586
              min(1)=        5 -> 14        -0.49947                  min(3)=        5 -> 12        -0.01262
              min(2)=        4 ->  9        -0.01793                  min(4)=        5 -> 10        -0.01083
              min(3)=        5 -> 17        -0.01420                  min(5)=        4 -> 12        -0.00848
              min(4)=        5 -> 26        -0.00364                  #CIs=46 #CIs>0=29 #CIs<0=17
              min(5)=        4 -> 24        -0.00128
              #CIs=10 #CIs>0=5 #CIs<0=5               32       Singlet-?Sym   43.4895         28.51    0.0018    0.000
                                                                      max(1)=        4 -> 15         0.58358
29       Singlet-?Sym   42.0714         29.47    0.0000    0.000        max(2)=        3 -> 13         0.03586
              max(1)=        4 -> 13         0.49975                  max(3)=        3 -> 17         0.01818
              max(2)=        5 -> 14         0.49975                  max(4)=        4 -> 11         0.01217
```

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                max(5)=        2 ->  8         0.01202                 min(2)=        5 -> 21        -0.00797
                min(1)=        5 -> 15        -0.39205                 min(3)=        2 -> 13        -0.00703
                min(2)=        3 -> 14        -0.05338                 min(4)=        5 -> 19        -0.00687
                min(3)=        4 -> 12        -0.01262                 min(5)=        5 -> 12        -0.00451
                min(4)=        3 -> 16        -0.01221                 #CIs=41 #CIs>0=26 #CIs<0=15
                min(5)=        4 -> 10        -0.01083
                #CIs=46 #CIs>0=24 #CIs<0=22             36       Singlet-SG   46.0142         26.94    0.2948    0.000
                                                                        max(1)=        3 -> 15         0.49866
33       Singlet-SG   44.3180         27.98    0.0000    0.000         max(2)=        2 -> 12         0.25054
                max(1)=        2 -> 12         0.64836                 max(3)=        3 ->  7         0.03128
                max(2)=        4 -> 14         0.17401                 max(4)=        2 -> 18         0.02582
                max(3)=        5 -> 13         0.17401                 max(5)=        3 ->  6         0.01620
                max(4)=        2 ->  7         0.06048                 min(1)=        4 -> 14        -0.29831
                max(5)=        2 -> 10         0.03196                 min(2)=        5 -> 13        -0.29831
                min(1)=        3 -> 15        -0.11051                 min(3)=        3 -> 12        -0.05786
                min(2)=        3 -> 18        -0.02780                 min(4)=        2 -> 11        -0.04490
                min(3)=        2 -> 11        -0.01301                 min(5)=        2 -> 10        -0.04298
                min(4)=        2 -> 19        -0.01013                 #CIs=33 #CIs>0=14 #CIs<0=19
                min(5)=        2 -> 18        -0.00901
                #CIs=34 #CIs>0=23 #CIs<0=11             37       Singlet-?Sym   46.4005         26.72    0.0000    0.000
                                                                        max(1)=        4 -> 16         0.49993
34       Singlet-?Sym   46.0076         26.95    0.0141    0.000         max(2)=        5 -> 17         0.49993
                max(1)=        3 -> 14         0.70034                 max(3)=        4 -> 24         0.00303
                max(2)=        3 -> 17         0.06914                 max(4)=        5 -> 23         0.00303
                max(3)=        4 -> 15         0.06144                 max(5)=        4 -> 27         0.00115
                max(4)=        2 ->  8         0.01662                 min(1)=        4 -> 13        -0.00614
                max(5)=        4 -> 21         0.00797                 min(2)=        5 -> 14        -0.00614
                min(1)=        3 ->  8        -0.02005                 min(3)=        4 ->  9        -0.00441
                min(2)=        3 -> 13        -0.00839                 min(4)=        5 ->  8        -0.00441
                min(3)=        2 -> 14        -0.00703                 min(5)=        3 -> 22        -0.00054
                min(4)=        4 -> 19        -0.00687                 #CIs=12 #CIs>0=6 #CIs<0=6
                min(5)=        4 -> 12        -0.00451
                #CIs=41 #CIs>0=23 #CIs<0=18             38       Singlet-?Sym   46.4005         26.72    0.0000    0.000
                                                                        max(1)=        5 -> 16         0.49993
35       Singlet-?Sym   46.0076         26.95    0.0141    0.000         max(2)=        4 -> 14         0.00614
                max(1)=        3 -> 13         0.70034                 max(3)=        4 ->  8         0.00441
                max(2)=        3 -> 16         0.06914                 max(4)=        5 -> 24         0.00303
                max(3)=        5 -> 15         0.06144                 max(5)=        5 -> 27         0.00115
                max(4)=        2 ->  9         0.01662                 min(1)=        4 -> 17        -0.49993
                max(5)=        3 -> 14         0.00839                 min(2)=        5 -> 13        -0.00614
                min(1)=        3 ->  9        -0.02005                 min(3)=        5 ->  9        -0.00441
```

```
                min(4)=        4 -> 23        -0.00303                max(4)=        3 ->  7         0.02273
                min(5)=        4 -> 26        -0.00115                max(5)=        3 -> 18         0.02161
                #CIs=12 #CIs>0=7 #CIs<0=5                              min(1)=        3 -> 15        -0.18122
                                                                       min(2)=        4 -> 14        -0.14689
39      Singlet-?Sym   46.4344         26.70    0.0000    0.000       min(3)=        5 -> 13        -0.14689
                max(1)=        4 -> 16         0.49976                min(4)=        3 -> 12        -0.05964
                max(2)=        5 -> 14         0.01437                min(5)=        3 -> 19        -0.05207
                max(3)=        5 ->  8         0.00477                #CIs=34 #CIs>0=18 #CIs<0=16
                max(4)=        4 -> 24         0.00368
                max(5)=        4 -> 27         0.00135  41      Singlet-?Sym   50.8965         24.36    0.3830    0.000
                min(1)=        5 -> 17        -0.49976                max(1)=        3 -> 17         0.59677
                min(2)=        4 -> 13        -0.01437                max(2)=        3 -> 16         0.36549
                min(3)=        4 ->  9        -0.00477                max(3)=        4 -> 19         0.01624
                min(4)=        5 -> 23        -0.00368                max(4)=        5 -> 19         0.00995
                min(5)=        5 -> 26        -0.00135                max(5)=        4 -> 12         0.00823
                #CIs=10 #CIs>0=5 #CIs<0=5                              min(1)=        3 -> 14        -0.05625
                                                                       min(2)=        4 -> 18        -0.05329
40      Singlet-?Sym   47.7941         25.94    0.6017    0.000       min(3)=        3 -> 13        -0.03445
                max(1)=        4 -> 17         0.45504                min(4)=        5 -> 18        -0.03264
                max(2)=        5 -> 16         0.45504                min(5)=        4 -> 15        -0.02262
                max(3)=        2 -> 12         0.03458                #CIs=48 #CIs>0=23 #CIs<0=25
```

5.2.3 Main contributions from different excited states at CIS approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_         max(1)=        4 ->  6         0.52373
                                                                       max(2)=        4 ->  7         0.30641
1       Singlet-?Sym   11.9145        104.06    0.0300    0.000       max(3)=        5 ->  6         0.26468
                max(1)=        5 ->  6         0.52373                max(4)=        5 ->  7         0.15485
                max(2)=        5 ->  7         0.30641                max(5)=        4 -> 11         0.11227
                max(3)=        5 -> 11         0.11227                min(1)=        4 -> 12        -0.10838
                max(4)=        5 -> 10         0.06641                min(2)=        5 -> 12        -0.05477
                max(5)=        4 -> 12         0.05477                min(3)=        4 -> 15        -0.02697
                min(1)=        4 ->  6        -0.26468                min(4)=        5 -> 15        -0.01363
                min(2)=        4 ->  7        -0.15485                min(5)=        4 -> 19        -0.01297
                min(3)=        5 -> 12        -0.10838                #CIs=44 #CIs>0=29 #CIs<0=15
                min(4)=        4 -> 11        -0.05674
                min(5)=        4 -> 10        -0.03356  3       Singlet-SG   15.7111         78.91    0.1958    0.000
                #CIs=44 #CIs>0=21 #CIs<0=23                            max(1)=        3 ->  6         0.60306
                                                                       max(2)=        3 ->  7         0.26720
2       Singlet-?Sym   11.9145        104.06    0.0300    0.000       max(3)=        4 ->  9         0.13778
```

```
              max(4)=        5 ->  8         0.13778                  min(1)=        4 ->  8        -0.49683
              max(5)=        3 -> 11         0.11576                  min(2)=        4 -> 13        -0.05371
              min(1)=        3 -> 12        -0.05736                  min(3)=        4 -> 16        -0.01621
              min(2)=        3 -> 15        -0.01597                  min(4)=        4 -> 26        -0.00329
              min(3)=        2 ->  7        -0.01411                  min(5)=        5 -> 23        -0.00013
              min(4)=        2 ->  6        -0.00498                  #CIs=10 #CIs>0=5 #CIs<0=5
              min(5)=        2 -> 19        -0.00458
              #CIs=30 #CIs>0=21 #CIs<0=9             7        Singlet-?Sym   17.2989         71.67    0.0000    0.000
                                                                      max(1)=        4 ->  8         0.49736
4         Singlet-?Sym   15.7979         78.48    0.0261    0.000        max(2)=        5 ->  9         0.49736
              max(1)=        5 ->  7         0.55625                  max(3)=        4 -> 13         0.04884
              max(2)=        4 ->  7         0.04469                  max(4)=        5 -> 14         0.04884
              max(3)=        5 -> 20         0.00696                  max(5)=        4 -> 16         0.01529
              max(4)=        3 -> 16         0.00371                  min(1)=        2 -> 22        -0.00166
              max(5)=        4 -> 22         0.00185                  min(2)=        4 ->  9        -0.00070
              min(1)=        5 ->  6        -0.33730                  min(3)=        3 -> 22        -0.00027
              min(2)=        5 -> 10        -0.20533                  min(4)=        4 -> 24        -0.00019
              min(3)=        5 -> 12        -0.17067                  min(5)=        5 -> 23        -0.00019
              min(4)=        5 -> 15        -0.04318                  #CIs=14 #CIs>0=9 #CIs<0=5
              min(5)=        4 ->  6        -0.02709
              #CIs=41 #CIs>0=17 #CIs<0=24            8        Singlet-?Sym   17.2989         71.67    0.0000    0.000
                                                                      max(1)=        5 ->  8         0.49736
5         Singlet-?Sym   15.7979         78.48    0.0261    0.000        max(2)=        5 -> 13         0.04884
              max(1)=        4 ->  7         0.55625                  max(3)=        5 -> 16         0.01529
              max(2)=        5 ->  6         0.02709                  max(4)=        5 -> 26         0.00282
              max(3)=        5 -> 10         0.01650                  max(5)=        2 -> 21         0.00166
              max(4)=        5 -> 12         0.01371                  min(1)=        4 ->  9        -0.49736
              max(5)=        4 -> 20         0.00696                  min(2)=        4 -> 14        -0.04884
              min(1)=        4 ->  6        -0.33730                  min(3)=        4 -> 17        -0.01529
              min(2)=        4 -> 10        -0.20533                  min(4)=        4 -> 27        -0.00282
              min(3)=        4 -> 12        -0.17067                  min(5)=        4 ->  8        -0.00070
              min(4)=        5 ->  7        -0.04469                  #CIs=14 #CIs>0=7 #CIs<0=7
              min(5)=        4 -> 15        -0.04318
              #CIs=41 #CIs>0=23 #CIs<0=18            9        Singlet-?Sym   18.1234         68.41    0.0314    0.000
                                                                      max(1)=        5 -> 10         0.66525
6         Singlet-?Sym   17.2784         71.76    0.0000    0.000        max(2)=        5 ->  7         0.12381
              max(1)=        5 ->  9         0.49683                  max(3)=        5 -> 11         0.06687
              max(2)=        5 -> 14         0.05371                  max(4)=        5 -> 18         0.03270
              max(3)=        5 -> 17         0.01621                  max(5)=        5 -> 15         0.03232
              max(4)=        5 -> 27         0.00329                  min(1)=        5 ->  6        -0.18092
              max(5)=        4 -> 24         0.00013                  min(2)=        5 -> 12        -0.04824
```

```
              min(3)=        5 -> 21        -0.00408                 min(5)=        5 ->  7        -0.06087
              min(4)=        4 -> 10        -0.00379                 #CIs=43 #CIs>0=21 #CIs<0=22
              min(5)=        5 -> 19        -0.00280
              #CIs=31 #CIs>0=17 #CIs<0=14            13       Singlet-?Sym   20.7628         59.71    0.0525    0.000
                                                                     max(1)=        4 -> 11         0.53783
10       Singlet-?Sym   18.1234         68.41    0.0314    0.000   max(2)=        3 ->  9         0.41629
              max(1)=        4 -> 10         0.66525                 max(3)=        5 -> 11         0.10691
              max(2)=        4 ->  7         0.12381                 max(4)=        3 ->  8         0.08275
              max(3)=        4 -> 11         0.06687                 max(5)=        3 -> 14         0.04214
              max(4)=        4 -> 18         0.03270                 min(1)=        4 ->  6        -0.07307
              max(5)=        4 -> 15         0.03232                 min(2)=        4 -> 10        -0.07215
              min(1)=        4 ->  6        -0.18092                 min(3)=        4 ->  7        -0.06087
              min(2)=        4 -> 12        -0.04824                 min(4)=        4 -> 15        -0.03193
              min(3)=        4 -> 19        -0.00280                 min(5)=        5 ->  6        -0.01453
              min(4)=        4 -> 25        -0.00105                 #CIs=43 #CIs>0=28 #CIs<0=15
              min(5)=        5 ->  6        -0.00103
              #CIs=31 #CIs>0=22 #CIs<0=9             14       Singlet-?Sym   20.9412         59.21    0.0439    0.000
                                                                     max(1)=        3 ->  9         0.47181
11       Singlet-?Sym   18.6919         66.33    0.0039    0.000   max(2)=        5 -> 11         0.22691
              max(1)=        4 ->  9         0.36939                 max(3)=        4 ->  6         0.04261
              max(2)=        5 ->  8         0.36939                 max(4)=        3 -> 14         0.04137
              max(3)=        3 ->  7         0.30592                 max(5)=        4 ->  7         0.03615
              max(4)=        2 -> 12         0.01600                 min(1)=        4 -> 11        -0.35287
              max(5)=        4 -> 14         0.01134                 min(2)=        3 ->  8        -0.30340
              min(1)=        3 ->  6        -0.28933                 min(3)=        5 ->  6        -0.02740
              min(2)=        3 -> 10        -0.19081                 min(4)=        3 -> 13        -0.02660
              min(3)=        3 -> 12        -0.10075                 min(5)=        5 ->  7        -0.02326
              min(4)=        3 -> 15        -0.04466                 #CIs=44 #CIs>0=23 #CIs<0=21
              min(5)=        2 ->  7        -0.01668
              #CIs=31 #CIs>0=10 #CIs<0=21            15       Singlet-?Sym   20.9412         59.21    0.0439    0.000
                                                                     max(1)=        3 ->  8         0.47181
12       Singlet-?Sym   20.7628         59.71    0.0525    0.000   max(2)=        3 ->  9         0.30340
              max(1)=        5 -> 11         0.53783                 max(3)=        5 ->  6         0.04261
              max(2)=        3 ->  8         0.41629                 max(4)=        3 -> 13         0.04137
              max(3)=        3 -> 13         0.04214                 max(5)=        5 ->  7         0.03615
              max(4)=        5 -> 12         0.02634                 min(1)=        5 -> 11        -0.35287
              max(5)=        5 -> 18         0.02127                 min(2)=        4 -> 11        -0.22691
              min(1)=        4 -> 11        -0.10691                 min(3)=        5 -> 12        -0.01739
              min(2)=        3 ->  9        -0.08275                 min(4)=        5 -> 18        -0.01524
              min(3)=        5 ->  6        -0.07307                 min(5)=        4 -> 12        -0.01119
              min(4)=        5 -> 10        -0.07215                 #CIs=44 #CIs>0=27 #CIs<0=17
```

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                                                                max(1)=        5 -> 12         0.66461
16       Singlet-SG   21.1870         58.52    0.2080    0.000  max(2)=        5 ->  7         0.21985
       max(1)=        3 ->  7         0.52266                  max(3)=        4 -> 12         0.06741
       max(2)=        3 -> 10         0.29800                  max(4)=        5 -> 19         0.04840
       max(3)=        4 -> 14         0.03320                  max(5)=        5 -> 15         0.04231
       max(4)=        5 -> 13         0.03320                  min(1)=        3 -> 16        -0.00948
       max(5)=        3 -> 15         0.02683                  min(2)=        5 -> 20        -0.00686
       min(1)=        4 ->  9        -0.21817                  min(3)=        4 -> 22        -0.00629
       min(2)=        5 ->  8        -0.21817                  min(4)=        3 -> 24        -0.00431
       min(3)=        3 ->  6        -0.18874                  min(5)=        5 -> 11        -0.00397
       min(4)=        3 -> 12        -0.04468                  #CIs=43 #CIs>0=26 #CIs<0=17
       min(5)=        2 -> 12        -0.01017
       #CIs=32 #CIs>0=20 #CIs<0=12             20       Singlet-?Sym   25.2680         49.07    0.0272    0.000
                                                                max(1)=        4 -> 12         0.66461
17       Singlet-SG   22.1522         55.97    0.0082    0.000  max(2)=        4 ->  7         0.21985
       max(1)=        3 -> 10         0.58476                  max(3)=        4 -> 19         0.04840
       max(2)=        4 ->  9         0.19111                  max(4)=        4 -> 15         0.04231
       max(3)=        5 ->  8         0.19111                  max(5)=        4 ->  6         0.01222
       max(4)=        3 -> 11         0.13729                  min(1)=        5 -> 12        -0.06741
       max(5)=        2 -> 12         0.01959                  min(2)=        5 ->  7        -0.02230
       min(1)=        3 ->  7        -0.21739                  min(3)=        3 -> 17        -0.00948
       min(2)=        3 ->  6        -0.10687                  min(4)=        4 -> 20        -0.00686
       min(3)=        3 -> 12        -0.04714                  min(5)=        4 -> 21        -0.00629
       min(4)=        4 -> 14        -0.03978                  #CIs=43 #CIs>0=20 #CIs<0=23
       min(5)=        5 -> 13        -0.03978
       #CIs=30 #CIs>0=13 #CIs<0=17             21       Singlet-SG   30.4963         40.66    0.2501    0.000
                                                                max(1)=        3 -> 12         0.67977
18       Singlet-SG   24.3185         50.98    0.0292    0.000  max(2)=        3 ->  7         0.09924
       max(1)=        3 -> 11         0.68114                  max(3)=        4 ->  9         0.06274
       max(2)=        3 -> 18         0.03006                  max(4)=        5 ->  8         0.06274
       max(3)=        3 -> 12         0.02777                  max(5)=        3 -> 10         0.03869
       max(4)=        4 -> 14         0.01805                  min(1)=        4 -> 14        -0.08800
       max(5)=        5 -> 13         0.01805                  min(2)=        5 -> 13        -0.08800
       min(1)=        3 -> 10        -0.14011                  min(3)=        2 -> 10        -0.01804
       min(2)=        3 ->  6        -0.07064                  min(4)=        2 -> 18        -0.01759
       min(3)=        4 ->  9        -0.05794                  min(5)=        2 ->  6        -0.01605
       min(4)=        5 ->  8        -0.05794                  #CIs=32 #CIs>0=15 #CIs<0=17
       min(5)=        3 ->  7        -0.02786
       #CIs=31 #CIs>0=18 #CIs<0=13             22       Singlet-SG   38.2279         32.43    0.0194    0.000
                                                                max(1)=        2 ->  6         0.60203
19       Singlet-?Sym   25.2680         49.07    0.0272    0.000  max(2)=        2 ->  7         0.32218
```

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              max(3)=        2 -> 11         0.10171                  max(5)=        2 -> 13         0.03162
              max(4)=        2 -> 10         0.06264                  min(1)=        4 -> 15        -0.03279
              max(5)=        3 ->  7         0.01055                  min(2)=        3 -> 14        -0.01831
              min(1)=        2 -> 12        -0.09953                  min(3)=        5 -> 15        -0.01774
              min(2)=        4 -> 14        -0.05109                  min(4)=        4 -> 11        -0.01409
              min(3)=        5 -> 13        -0.05109                  min(5)=        4 -> 18        -0.00994
              min(4)=        4 -> 17        -0.03419                  #CIs=46 #CIs>0=21 #CIs<0=25
              min(5)=        5 -> 16        -0.03419
              #CIs=32 #CIs>0=14 #CIs<0=18             26       Singlet-?Sym   43.5482         28.47    0.0187    0.000
                                                                      max(1)=        2 ->  8         0.61668
23       Singlet-SG   42.0036         29.52    0.0192    0.000      max(2)=        2 -> 13         0.05847
              max(1)=        2 ->  7         0.57611                  max(3)=        3 -> 16         0.03289
              max(2)=        4 ->  9         0.01274                  max(4)=        4 -> 15         0.01774
              max(3)=        5 ->  8         0.01274                  max(5)=        2 -> 16         0.01267
              max(4)=        3 -> 10         0.01250                  min(1)=        2 ->  9        -0.33351
              max(5)=        3 ->  7         0.00911                  min(2)=        5 -> 15        -0.03279
              min(1)=        2 ->  6        -0.32206                  min(3)=        2 -> 14        -0.03162
              min(2)=        2 -> 10        -0.17300                  min(4)=        3 -> 13        -0.01831
              min(3)=        2 -> 12        -0.16783                  min(5)=        3 -> 17        -0.01779
              min(4)=        4 -> 17        -0.03956                  #CIs=46 #CIs>0=22 #CIs<0=24
              min(5)=        5 -> 16        -0.03956
              #CIs=32 #CIs>0=9 #CIs<0=23              27       Singlet-?Sym   43.7371         28.35    0.0000    0.000
                                                                      max(1)=        4 -> 13         0.49404
24       Singlet-?Sym   43.5292         28.48    0.0000    0.000      max(2)=        5 -> 14         0.49404
              max(1)=        5 -> 14         0.49317                  max(3)=        4 -> 16         0.05619
              max(2)=        5 -> 17         0.05902                  max(4)=        5 -> 17         0.05619
              max(3)=        4 ->  8         0.05534                  max(5)=        4 -> 26         0.01376
              max(4)=        5 -> 27         0.01491                  min(1)=        4 ->  8        -0.05033
              max(5)=        4 -> 24         0.00373                  min(2)=        5 ->  9        -0.05033
              min(1)=        4 -> 13        -0.49317                  min(3)=        4 -> 14        -0.00494
              min(2)=        4 -> 16        -0.05902                  min(4)=        4 -> 24        -0.00365
              min(3)=        5 ->  9        -0.05534                  min(5)=        5 -> 23        -0.00365
              min(4)=        4 -> 26        -0.01491                  #CIs=20 #CIs>0=10 #CIs<0=10
              min(5)=        5 -> 23        -0.00373
              #CIs=10 #CIs>0=5 #CIs<0=5               28       Singlet-?Sym   43.7371         28.35    0.0000    0.000
                                                                      max(1)=        5 -> 13         0.49404
25       Singlet-?Sym   43.5482         28.47    0.0187    0.000      max(2)=        5 -> 16         0.05619
              max(1)=        2 ->  9         0.61668                  max(3)=        4 ->  9         0.05033
              max(2)=        2 ->  8         0.33351                  max(4)=        5 -> 26         0.01376
              max(3)=        2 -> 14         0.05847                  max(5)=        4 -> 23         0.00365
              max(4)=        3 -> 17         0.03289                  min(1)=        4 -> 14        -0.49404
```

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                min(2)=        4 -> 17        -0.05619                min(4)=        4 -> 20        -0.01536
                min(3)=        5 ->  8        -0.05033                min(5)=        5 -> 12        -0.00196
                min(4)=        4 -> 27        -0.01376                #CIs=41 #CIs>0=30 #CIs<0=11
                min(5)=        4 -> 13        -0.00494
                #CIs=20 #CIs>0=9 #CIs<0=11             32       Singlet-SG   46.8521         26.46    0.0025    0.000
                                                                       max(1)=        2 -> 11         0.68763
29       Singlet-SG   44.3839         27.93    0.0107    0.000         max(2)=        2 -> 12         0.04870
                max(1)=        2 -> 10         0.65724                max(3)=        2 -> 18         0.02444
                max(2)=        2 ->  7         0.09934                max(4)=        3 -> 10         0.01646
                max(3)=        2 -> 11         0.06909                max(5)=        2 -> 20         0.00519
                max(4)=        3 -> 15         0.03702                min(1)=        2 -> 10        -0.09090
                max(5)=        2 -> 15         0.03142                min(2)=        2 ->  6        -0.08807
                min(1)=        2 ->  6        -0.15705                min(3)=        3 -> 15        -0.04482
                min(2)=        4 -> 14        -0.10467                min(4)=        2 ->  7        -0.03966
                min(3)=        5 -> 13        -0.10467                min(5)=        2 -> 15        -0.03861
                min(4)=        2 -> 12        -0.05324                #CIs=32 #CIs>0=9 #CIs<0=23
                min(5)=        3 -> 10        -0.02027
                #CIs=32 #CIs>0=15 #CIs<0=17             33       Singlet-?Sym   47.6919         26.00    0.0117    0.000
                                                                       max(1)=        3 -> 13         0.66569
30       Singlet-?Sym   45.2872         27.38    0.0015    0.000         max(2)=        3 -> 14         0.19729
                max(1)=        5 -> 15         0.69676                max(3)=        3 -> 16         0.09241
                max(2)=        5 -> 11         0.03763                max(4)=        5 -> 15         0.04525
                max(3)=        2 ->  8         0.03419                max(5)=        3 -> 17         0.02739
                max(4)=        5 -> 18         0.02796                min(1)=        3 ->  8        -0.06443
                max(5)=        5 ->  7         0.02605                min(2)=        3 ->  9        -0.01909
                min(1)=        5 -> 12        -0.05371                min(3)=        5 -> 21        -0.00675
                min(2)=        3 -> 13        -0.05278                min(4)=        5 -> 12        -0.00616
                min(3)=        5 -> 10        -0.04635                min(5)=        2 -> 24        -0.00571
                min(4)=        4 -> 15        -0.02539                #CIs=43 #CIs>0=28 #CIs<0=15
                min(5)=        5 -> 20        -0.01536
                #CIs=41 #CIs>0=20 #CIs<0=21             34       Singlet-?Sym   47.6919         26.00    0.0117    0.000
                                                                       max(1)=        3 -> 14         0.66569
31       Singlet-?Sym   45.2872         27.38    0.0015    0.000         max(2)=        3 -> 17         0.09241
                max(1)=        4 -> 15         0.69676                max(3)=        4 -> 15         0.04525
                max(2)=        4 -> 11         0.03763                max(4)=        2 -> 17         0.02205
                max(3)=        2 ->  9         0.03419                max(5)=        4 -> 18         0.02129
                max(4)=        4 -> 18         0.02796                min(1)=        3 -> 13        -0.19729
                max(5)=        4 ->  7         0.02605                min(2)=        3 ->  9        -0.06443
                min(1)=        4 -> 12        -0.05371                min(3)=        3 -> 16        -0.02739
                min(2)=        3 -> 14        -0.05278                min(4)=        5 -> 15        -0.01341
                min(3)=        4 -> 10        -0.04635                min(5)=        2 -> 16        -0.00653
```

```
                #CIs=43 #CIs>0=24 #CIs<0=19             38       Singlet-?Sym   49.0153         25.29    0.0000    0.000

                                                                        max(1)=        5 -> 17         0.49598
35       Singlet-SG   47.7102         25.99    0.1516    0.000         max(2)=        4 -> 13         0.06044
                max(1)=        3 -> 15         0.52662                 max(3)=        5 -> 23         0.01545
                max(2)=        3 -> 19         0.03383                 max(4)=        4 ->  8         0.00967
                max(3)=        3 ->  7         0.03330                 max(5)=        5 -> 27         0.00377
                max(4)=        3 -> 11         0.03280                 min(1)=        4 -> 16        -0.49598
                max(5)=        3 -> 18         0.02923                 min(2)=        5 -> 14        -0.06044
                min(1)=        4 -> 14        -0.28466                 min(3)=        4 -> 24        -0.01545
                min(2)=        5 -> 13        -0.28466                 min(4)=        5 ->  9        -0.00967
                min(3)=        2 -> 10        -0.12197                 min(5)=        4 -> 26        -0.00377
                min(4)=        4 -> 17        -0.09935                 #CIs=10 #CIs>0=5 #CIs<0=5
                min(5)=        5 -> 16        -0.09935
                #CIs=33 #CIs>0=14 #CIs<0=19             39       Singlet-?Sym   50.6494         24.48    0.9291    0.000

                                                                        max(1)=        4 -> 17         0.40917
36       Singlet-?Sym   48.9219         25.34    0.0000    0.000         max(2)=        5 -> 16         0.40917
                max(1)=        4 -> 16         0.49637                 max(3)=        2 -> 12         0.28695
                max(2)=        5 -> 17         0.49637                 max(4)=        2 ->  7         0.12930
                max(3)=        4 -> 24         0.01447                 max(5)=        3 -> 19         0.05461
                max(4)=        5 -> 23         0.01447                 min(1)=        4 -> 14        -0.16281
                max(5)=        4 -> 26         0.00335                 min(2)=        5 -> 13        -0.16281
                min(1)=        4 -> 13        -0.05743                 min(3)=        3 -> 12        -0.06142
                min(2)=        5 -> 14        -0.05743                 min(4)=        2 -> 10        -0.04661
                min(3)=        4 ->  8        -0.00963                 min(5)=        2 -> 11        -0.02511
                min(4)=        5 ->  9        -0.00963                 #CIs=32 #CIs>0=17 #CIs<0=15
                min(5)=        3 -> 22        -0.00082
                #CIs=12 #CIs>0=6 #CIs<0=6               40       Singlet-SG   51.3252         24.16    0.0531    0.000

                                                                        max(1)=        2 -> 12         0.58096
37       Singlet-?Sym   48.9219         25.34    0.0000    0.000         max(2)=        3 -> 15         0.18145
                max(1)=        5 -> 16         0.49637                 max(3)=        2 ->  7         0.17257
                max(2)=        4 -> 14         0.05743                 max(4)=        4 -> 14         0.10663
                max(3)=        5 -> 24         0.01447                 max(5)=        5 -> 13         0.10663
                max(4)=        4 ->  9         0.00963                 min(1)=        4 -> 17        -0.18363
                max(5)=        5 -> 26         0.00335                 min(2)=        5 -> 16        -0.18363
                min(1)=        4 -> 17        -0.49637                 min(3)=        3 -> 19        -0.02596
                min(2)=        5 -> 13        -0.05743                 min(4)=        3 -> 18        -0.02488
                min(3)=        4 -> 23        -0.01447                 min(5)=        2 -> 18        -0.01056
                min(4)=        5 ->  8        -0.00963                 #CIs=32 #CIs>0=21 #CIs<0=11
                min(5)=        4 -> 27        -0.00335
                #CIs=12 #CIs>0=7 #CIs<0=5               41       Singlet-?Sym   53.2054         23.30    0.4536    0.000

                                                                        max(1)=        3 -> 16         0.54376
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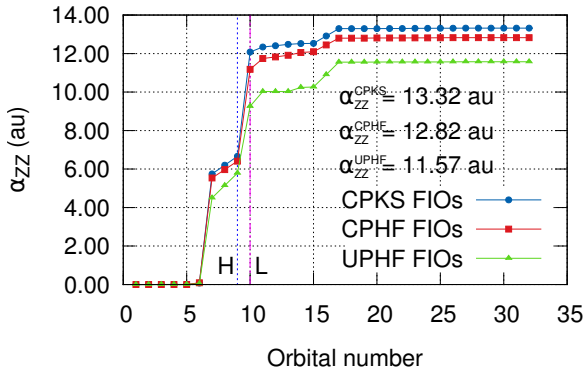
max(2)=        3 -> 17         0.43157                 min(2)=        3 -> 14        -0.05651
max(3)=        2 -> 13         0.03149                 min(3)=        5 -> 18        -0.04060
max(4)=        2 -> 14         0.02499                 min(4)=        2 ->  8        -0.03482
max(5)=        5 -> 21         0.01432                 min(5)=        4 -> 18        -0.03222
min(1)=        3 -> 13        -0.07121                 #CIs=44 #CIs>0=19 #CIs<0=25
```

6 HCl

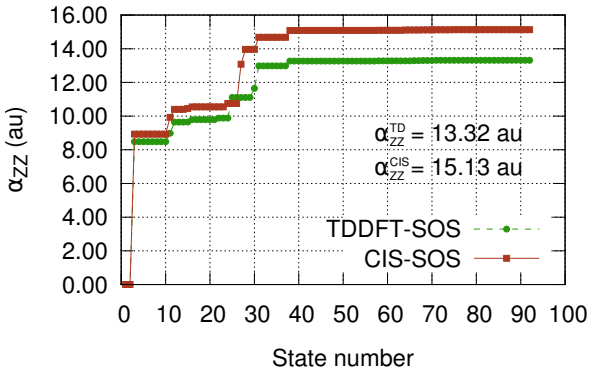
6.1 6-311G(d,p)

6.1.1 Plots

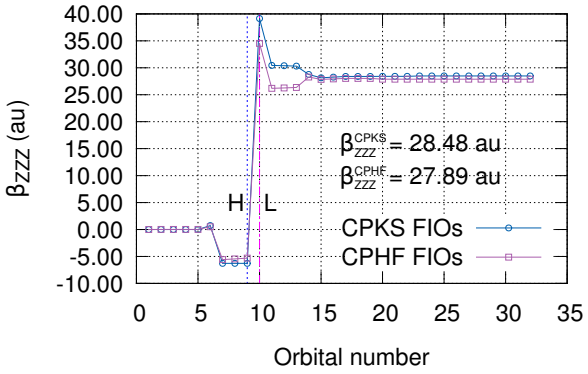
Figure S7: For HCl molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S7a and S7c) or states (SOS approaches, in Plots S7b and S7d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.01 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



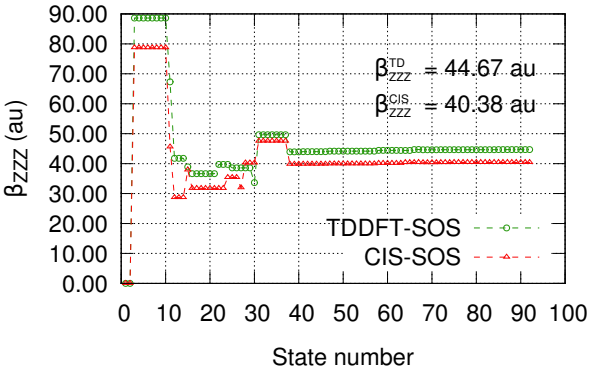
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

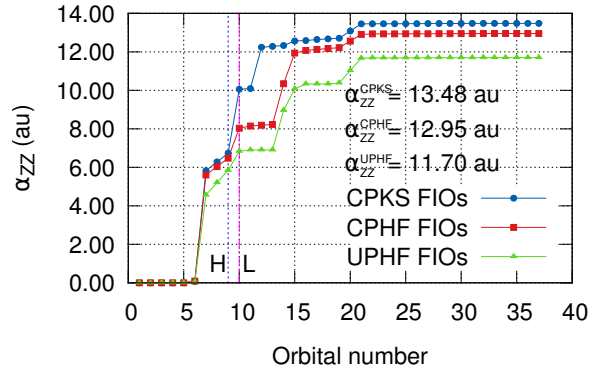


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

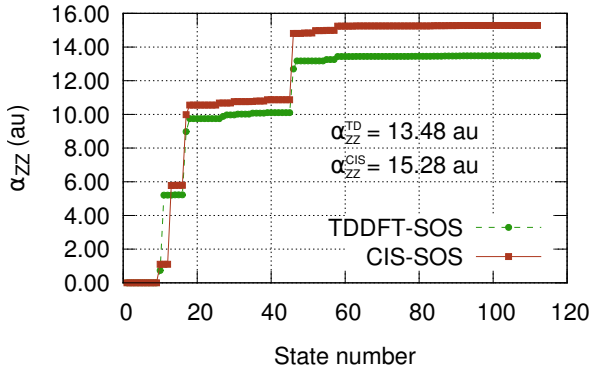
6.2 6-311++G(d,p)

6.2.1 Plots

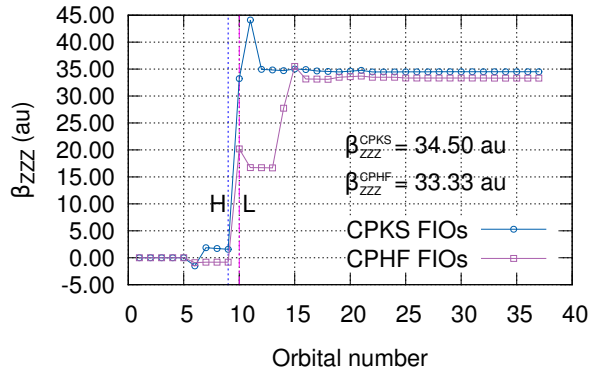
Figure S8: For HCl molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S8a and S8c) or states (SOS approaches, in Plots S8b and S8d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.06 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



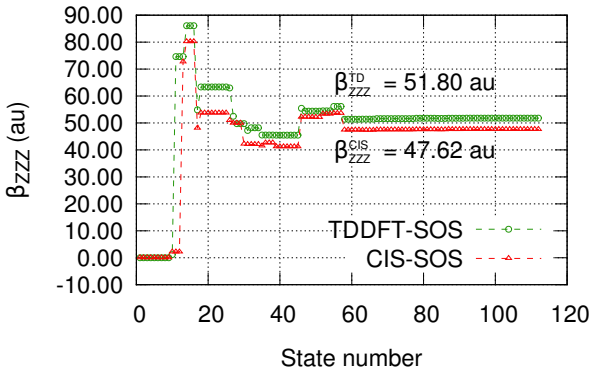
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



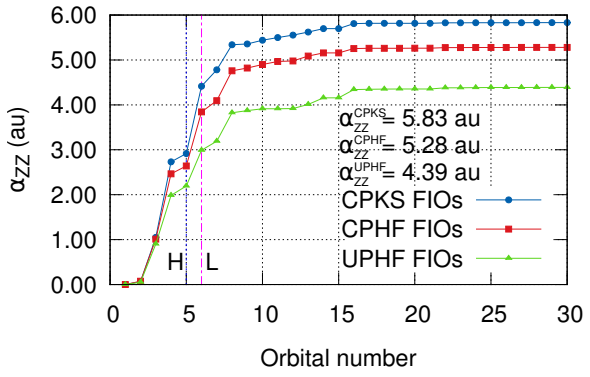
(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

7 H_2O

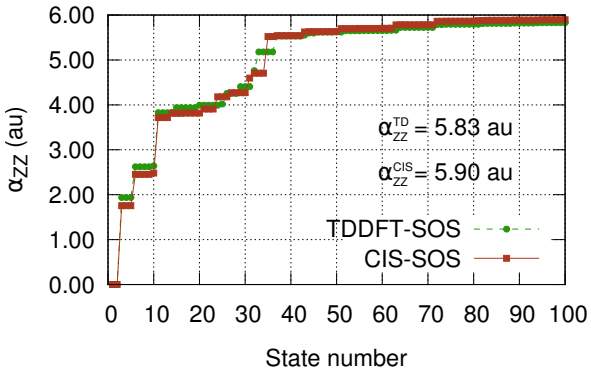
7.1 6-311G(d,p)

7.1.1 Plots

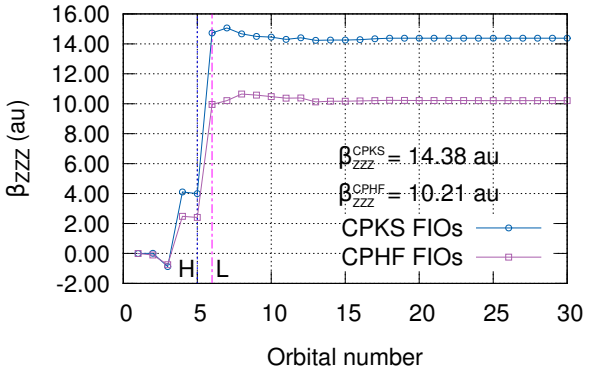
Figure S9: For H_2O molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S9a and S9c) or states (SOS approaches, in Plots S9b and S9d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.02 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



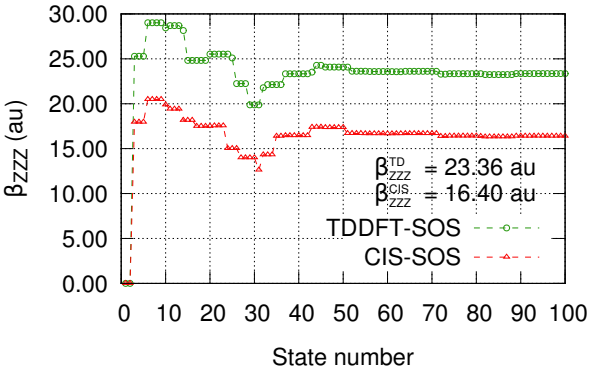
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

7.1.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_          min(4)=        5 -> 11        -0.02154
                                                                        min(5)=        4 -> 13        -0.01319
1        Singlet-B1    7.4450        166.53    0.0265    0.000          #states=32 #states>0=13 #states<0=19
               max(1)=        5 ->  6         0.70615
               max(2)=        5 -> 10         0.01679   4        Singlet-B2   11.7572        105.45    0.0493    0.000
               max(3)=        2 -> 11         0.00495                max(1)=        4 ->  7         0.70420
               max(4)=        5 -> 25         0.00396                max(2)=        4 -> 12         0.03096
               max(5)=        2 -> 22         0.00187                max(3)=        3 ->  9         0.02523
               min(1)=        5 ->  9        -0.03581                max(4)=        4 ->  8         0.02167
               min(2)=        5 -> 13        -0.00974                max(5)=        5 -> 15         0.00949
               min(3)=        3 -> 23        -0.00333                min(1)=        3 ->  6        -0.04529
               min(4)=        5 -> 14        -0.00314                min(2)=        2 ->  7        -0.01200
               min(5)=        4 -> 22        -0.00306                min(3)=        2 ->  8        -0.00448
               #states=21 #states>0=12 #states<0=9                    min(4)=        3 -> 13        -0.00342
                                                                      min(5)=        3 -> 19        -0.00315
2        Singlet-A2    9.3956        131.96    0.0000    0.000          #states=28 #states>0=15 #states<0=13
               max(1)=        5 ->  7         0.70282
               max(2)=        5 ->  8         0.07139   5        Singlet-B2   13.6671         90.72    0.2301    0.000
               max(3)=        5 -> 12         0.03137                max(1)=        3 ->  6         0.70410
               max(4)=        2 -> 15         0.00328                max(2)=        4 ->  7         0.04683
               max(5)=        5 -> 21         0.00186                max(3)=        3 -> 10         0.01555
               min(1)=        3 -> 11        -0.00287                max(4)=        2 ->  8         0.00900
               min(2)=        5 -> 18        -0.00189                max(5)=        2 -> 12         0.00728
               min(3)=        4 -> 15         0.00005                min(1)=        4 ->  8        -0.04276
               min(4)=        3 -> 22         0.00023                min(2)=        3 ->  9        -0.00787
               min(5)=        3 -> 27         0.00029                min(3)=        3 -> 13        -0.00786
               #states=16 #states>0=14 #states<0=2                    min(4)=        2 ->  7        -0.00427
                                                                      min(5)=        3 -> 20        -0.00268
3        Singlet-A1    9.7608        127.02    0.0831    0.000          #states=28 #states>0=14 #states<0=14
               max(1)=        4 ->  6         0.70407
               max(2)=        2 ->  6         0.01169   6        Singlet-A1   16.4798         75.23    0.0836    0.000
               max(3)=        2 -> 10         0.00301                max(1)=        3 ->  7         0.69406
               max(4)=        2 -> 19         0.00177                max(2)=        4 ->  9         0.07754
               max(5)=        2 -> 24         0.00141                max(3)=        4 ->  6         0.05067
               min(1)=        3 ->  7        -0.05111                max(4)=        2 ->  6         0.02311
               min(2)=        3 ->  8        -0.02584                max(5)=        2 -> 10         0.00505
               min(3)=        4 ->  9        -0.02436                min(1)=        5 -> 11        -0.08145
```

```
                min(2)=        4 -> 10        -0.05991                min(4)=        3 -> 19        -0.01154
                min(3)=        2 -> 13        -0.01796                min(5)=        2 -> 12        -0.00636
                min(4)=        2 ->  9        -0.01170                #states=29 #states>0=16 #states<0=13
                min(5)=        4 -> 14        -0.00820
                #states=32 #states>0=13 #states<0=19    10      Singlet-A1   20.1903         61.41    0.0037    0.000
                                                                       max(1)=        4 ->  9         0.69335
7       Singlet-A2   16.4823         75.22    0.0000    0.000          max(2)=        3 ->  8         0.09678
                max(1)=        5 ->  8         0.70335                 max(3)=        5 -> 11         0.04687
                max(2)=        5 -> 17         0.00551                 max(4)=        4 ->  6         0.02446
                max(3)=        2 -> 15         0.00402                 max(5)=        4 -> 10         0.02086
                max(4)=        3 -> 16         0.00152                 min(1)=        3 ->  7        -0.07088
                max(5)=        4 -> 23         0.00146                 min(2)=        2 ->  6        -0.03485
                min(1)=        5 ->  7        -0.07148                 min(3)=        4 -> 19        -0.00907
                min(2)=        5 -> 18        -0.01292                 min(4)=        3 -> 18        -0.00638
                min(3)=        3 -> 11        -0.00279                 min(5)=        2 -> 10        -0.00439
                min(4)=        5 -> 21         0.00007                 #states=33 #states>0=18 #states<0=15
                min(5)=        5 -> 12         0.00027
                #states=16 #states>0=13 #states<0=3     11      Singlet-A1   24.0479         51.56    0.3084    0.000
                                                                       max(1)=        3 ->  8         0.68778
8       Singlet-B1   18.2693         67.86    0.1098    0.000          max(2)=        2 ->  6         0.02708
                max(1)=        5 ->  9         0.70462                 max(3)=        4 -> 13         0.02354
                max(2)=        5 ->  6         0.03664                 max(4)=        4 ->  6         0.01822
                max(3)=        3 -> 15         0.01952                 max(5)=        5 -> 22         0.00870
                max(4)=        5 -> 14         0.01511                 min(1)=        5 -> 11        -0.10529
                max(5)=        4 -> 22         0.00784                 min(2)=        4 ->  9        -0.08643
                min(1)=        5 -> 10        -0.03669                 min(3)=        4 -> 10        -0.06761
                min(2)=        4 -> 16        -0.01422                 min(4)=        4 -> 14        -0.04153
                min(3)=        2 -> 11        -0.01195                 min(5)=        5 -> 16        -0.03280
                min(4)=        4 -> 11        -0.00983                 #states=34 #states>0=17 #states<0=17
                min(5)=        5 -> 25        -0.00862
                #states=21 #states>0=10 #states<0=11    12      Singlet-B2   25.3359         48.94    0.4482    0.000
                                                                       max(1)=        3 ->  9         0.68990
9       Singlet-B2   19.5924         63.28    0.1626    0.000          max(2)=        2 ->  7         0.02512
                max(1)=        4 ->  8         0.69023                 max(3)=        2 -> 18         0.01090
                max(2)=        3 ->  9         0.13979                 max(4)=        4 -> 17         0.01027
                max(3)=        3 ->  6         0.04498                 max(5)=        3 -> 19         0.00553
                max(4)=        5 -> 15         0.02937                 min(1)=        4 ->  8        -0.13780
                max(5)=        3 -> 14         0.01653                 min(2)=        5 -> 15        -0.05403
                min(1)=        2 ->  7        -0.02841                 min(3)=        3 -> 10        -0.03554
                min(2)=        4 ->  7        -0.02381                 min(4)=        3 -> 14        -0.02565
                min(3)=        3 -> 13        -0.01183                 min(5)=        4 ->  7        -0.01947
```

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                #states=29 #states>0=11 #states<0=18     16       Singlet-B1   28.5426         43.44    0.0004    0.000
                                                                        max(1)=        4 -> 11         0.70385
13       Singlet-B1   26.3281         47.09    0.0043    0.000          max(2)=        5 -> 10         0.06474
                max(1)=        5 -> 10         0.70251                 max(3)=        5 ->  9         0.01298
                max(2)=        5 ->  9         0.03551                 max(4)=        3 -> 15         0.00754
                max(3)=        5 -> 14         0.01435                 max(5)=        4 -> 22         0.00601
                max(4)=        5 -> 13         0.01142                 min(1)=        2 -> 11        -0.00748
                max(5)=        3 -> 15         0.01069                 min(2)=        5 -> 13        -0.00654
                min(1)=        4 -> 11        -0.06565                 min(3)=        4 -> 16        -0.00487
                min(2)=        5 ->  6        -0.01448                 min(4)=        5 -> 20        -0.00393
                min(3)=        4 -> 16        -0.01123                 min(5)=        5 -> 25        -0.00340
                min(4)=        2 -> 11        -0.00628                 #states=21 #states>0=12 #states<0=9
                min(5)=        5 -> 20        -0.00469
                #states=21 #states>0=13 #states<0=8      17       Singlet-A2   29.1471         42.54    0.0000    0.000
                                                                        max(1)=        5 -> 12         0.70607
14       Singlet-A1   26.8026         46.26    0.0023    0.000          max(2)=        5 -> 26         0.00594
                max(1)=        2 ->  6         0.55788                 max(3)=        5 -> 29         0.00309
                max(2)=        5 -> 11         0.40820                 max(4)=        4 -> 15         0.00279
                max(3)=        3 ->  8         0.02606                 max(5)=        5 -> 21         0.00213
                max(4)=        3 ->  7         0.01543                 min(1)=        5 ->  7        -0.03122
                max(5)=        5 -> 22         0.00590                 min(2)=        3 -> 11        -0.01859
                min(1)=        4 -> 10        -0.14000                 min(3)=        5 -> 18        -0.00901
                min(2)=        2 ->  9        -0.03125                 min(4)=        5 ->  8        -0.00368
                min(3)=        4 -> 14        -0.01544                 min(5)=        3 -> 16        -0.00133
                min(4)=        2 -> 13        -0.01275                 #states=15 #states>0=8 #states<0=7
                min(5)=        4 -> 13        -0.01143
                #states=34 #states>0=19 #states<0=15     18       Singlet-B2   29.1639         42.51    0.0249    0.000
                                                                        max(1)=        2 ->  7         0.70262
15       Singlet-A1   27.7605         44.66    0.0361    0.000          max(2)=        2 ->  8         0.04474
                max(1)=        4 -> 10         0.45111                 max(3)=        4 ->  8         0.03021
                max(2)=        2 ->  6         0.38926                 max(4)=        5 -> 15         0.02864
                max(3)=        3 -> 12         0.05436                 max(5)=        3 -> 10         0.02379
                max(4)=        4 ->  9         0.03237                 min(1)=        3 -> 13        -0.03709
                max(5)=        2 ->  9         0.01766                 min(2)=        3 ->  9        -0.01426
                min(1)=        5 -> 11        -0.37344                 min(3)=        3 -> 24        -0.00479
                min(2)=        3 ->  8        -0.02482                 min(4)=        3 -> 19        -0.00293
                min(3)=        3 ->  7        -0.02093                 min(5)=        4 -> 21        -0.00257
                min(4)=        4 ->  6        -0.01708                 #states=29 #states>0=19 #states<0=10
                min(5)=        4 -> 14        -0.01236
                #states=33 #states>0=19 #states<0=14     19       Singlet-B2   31.2720         39.65    0.0552    0.000
                                                                        max(1)=        4 -> 12         0.70364
```

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              max(2)=        3 ->  9         0.00621                  max(4)=        3 -> 19         0.00651
              max(3)=        4 -> 26         0.00557                  max(5)=        2 -> 12         0.00643
              max(4)=        4 -> 21         0.00257                  min(1)=        2 ->  8        -0.04556
              max(5)=        4 -> 29         0.00248                  min(2)=        5 -> 15        -0.02289
              min(1)=        3 -> 10        -0.05643                  min(3)=        2 ->  7        -0.01922
              min(2)=        4 ->  7        -0.03140                  min(4)=        3 ->  6        -0.01537
              min(3)=        2 ->  8        -0.02077                  min(5)=        4 ->  8        -0.00929
              min(4)=        4 -> 18        -0.00909                  #states=28 #states>0=13 #states<0=15
              min(5)=        3 -> 13        -0.00785
              #states=29 #states>0=12 #states<0=17    23       Singlet-B1   35.3546         35.07    0.2780    0.000
                                                                      max(1)=        5 -> 13         0.70377
20       Singlet-A1   32.2158         38.49    0.0254    0.000          max(2)=        4 -> 16         0.01793
              max(1)=        4 -> 10         0.44285                  max(3)=        5 ->  6         0.00925
              max(2)=        5 -> 11         0.31756                  max(4)=        4 -> 11         0.00800
              max(3)=        3 ->  8         0.08835                  max(5)=        5 -> 20         0.00343
              max(4)=        3 ->  7         0.08007                  min(1)=        5 -> 14        -0.05491
              max(5)=        3 -> 18         0.01510                  min(2)=        3 -> 15        -0.02904
              min(1)=        3 -> 12        -0.38580                  min(3)=        2 -> 11        -0.01841
              min(2)=        2 ->  6        -0.14060                  min(4)=        5 -> 10        -0.00858
              min(3)=        2 ->  9        -0.10601                  min(5)=        2 -> 16        -0.00635
              min(4)=        4 -> 13        -0.07796                  #states=20 #states>0=9 #states<0=11
              min(5)=        2 -> 13        -0.04564
              #states=34 #states>0=13 #states<0=21    24       Singlet-B2   36.4613         34.00    0.0132    0.000
                                                                      max(1)=        2 ->  8         0.70193
21       Singlet-A2   32.5557         38.08    0.0000    0.000          max(2)=        3 -> 10         0.04063
              max(1)=        3 -> 11         0.70682                  max(3)=        4 -> 12         0.02309
              max(2)=        5 -> 12         0.01868                  max(4)=        3 ->  9         0.01872
              max(3)=        3 -> 16         0.00515                  max(5)=        3 -> 19         0.01043
              max(4)=        3 -> 27         0.00474                  min(1)=        2 ->  7        -0.04660
              max(5)=        2 -> 15         0.00408                  min(2)=        3 -> 13        -0.04061
              min(1)=        5 -> 21        -0.00040                  min(3)=        5 -> 15        -0.02747
              min(2)=        2 -> 23        -0.00013                  min(4)=        3 ->  6        -0.01090
              min(3)=        5 -> 17        -0.00008                  min(5)=        2 -> 12        -0.00575
              min(4)=        5 -> 26         0.00015                  #states=29 #states>0=16 #states<0=13
              min(5)=        5 -> 29         0.00021
              #states=16 #states>0=13 #states<0=3     25       Singlet-A1   36.7454         33.74    0.0155    0.000
                                                                      max(1)=        4 -> 13         0.45863
22       Singlet-B2   32.6057         38.03    0.0715    0.000          max(2)=        2 ->  9         0.37355
              max(1)=        3 -> 10         0.70183                  max(3)=        2 ->  6         0.05708
              max(2)=        4 -> 12         0.05425                  max(4)=        4 -> 14         0.04690
              max(3)=        3 ->  9         0.03306                  max(5)=        2 -> 13         0.02161
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              min(1)=        3 -> 12        -0.36537                 min(3)=        4 -> 18        -0.00602
              min(2)=        4 -> 10        -0.07242                 min(4)=        4 ->  7        -0.00452
              min(3)=        5 -> 11        -0.06040                 min(5)=        2 -> 12        -0.00221
              min(4)=        3 ->  8        -0.03184                 #states=27 #states>0=18 #states<0=9
              min(5)=        2 -> 10        -0.02044
              #states=34 #states>0=18 #states<0=16    29       Singlet-A1   41.5677         29.83    0.1166    0.000
                                                                     max(1)=        3 -> 12         0.43955
26       Singlet-A1   37.7280         32.86    0.1525    0.000          max(2)=        2 ->  9         0.28204
              max(1)=        2 ->  9         0.51484                  max(3)=        5 -> 11         0.24599
              max(2)=        3 ->  8         0.01380                  max(4)=        4 -> 10         0.23623
              max(3)=        2 ->  6         0.01327                  max(5)=        4 -> 13         0.22929
              max(4)=        2 -> 10         0.01316                  min(1)=        2 -> 13        -0.15527
              max(5)=        3 -> 18         0.00515                  min(2)=        4 -> 14        -0.12715
              min(1)=        4 -> 13        -0.47829                  min(3)=        2 ->  6        -0.10825
              min(2)=        3 -> 12        -0.07042                  min(4)=        4 ->  9        -0.04234
              min(3)=        4 -> 10        -0.02153                  min(5)=        2 -> 14        -0.03711
              min(4)=        5 -> 16        -0.01458                  #states=34 #states>0=16 #states<0=18
              min(5)=        4 ->  9        -0.00910
              #states=34 #states>0=16 #states<0=18    30       Singlet-B2   41.7356         29.71    0.2993    0.000
                                                                     max(1)=        3 -> 13         0.49106
27       Singlet-B1   39.9174         31.06    0.1543    0.000          max(2)=        3 -> 14         0.11443
              max(1)=        5 -> 14         0.70186                  max(3)=        2 ->  7         0.04167
              max(2)=        5 -> 13         0.05138                  max(4)=        4 -> 18         0.03530
              max(3)=        4 -> 16         0.03444                  max(5)=        4 ->  8         0.03363
              max(4)=        5 -> 20         0.00563                  min(1)=        5 -> 15        -0.48759
              max(5)=        5 ->  6         0.00309                  min(2)=        2 -> 12        -0.03995
              min(1)=        3 -> 15        -0.05544                  min(3)=        3 ->  9        -0.02908
              min(2)=        5 ->  9        -0.01411                  min(4)=        3 -> 19        -0.02258
              min(3)=        5 -> 10        -0.01302                  min(5)=        4 -> 17        -0.01930
              min(4)=        2 -> 16        -0.00800                  #states=29 #states>0=15 #states<0=14
              min(5)=        5 -> 24        -0.00635
              #states=21 #states>0=10 #states<0=11    31       Singlet-A2   42.5653         29.13    0.0000    0.000
                                                                     max(1)=        4 -> 15         0.70702
28       Singlet-B2   41.0057         30.24    0.1354    0.000          max(2)=        4 -> 23         0.00788
              max(1)=        3 -> 13         0.50218                  max(3)=        3 -> 16         0.00465
              max(2)=        5 -> 15         0.49075                  max(4)=        5 -> 21         0.00433
              max(3)=        2 ->  8         0.04624                  max(5)=        2 -> 23         0.00263
              max(4)=        3 ->  9         0.03435                  min(1)=        2 -> 15        -0.00344
              max(5)=        3 -> 10         0.01399                  min(2)=        5 -> 12        -0.00287
              min(1)=        3 -> 14        -0.05363                  min(3)=        3 -> 11        -0.00231
              min(2)=        4 ->  8        -0.01843                  min(4)=        5 -> 18        -0.00197
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                min(5)=        3 -> 22        -0.00088                max(5)=        5 -> 14         0.01143
                #states=16 #states>0=8 #states<0=8                     min(1)=        4 -> 16        -0.04529
                                                                       min(2)=        5 -> 20        -0.01266
32      Singlet-A1   43.0050         28.83    0.2949    0.000         min(3)=        5 -> 25        -0.00640
                max(1)=        4 -> 14         0.65511                min(4)=        5 ->  6        -0.00352
                max(2)=        3 -> 12         0.07714                min(5)=        5 -> 24        -0.00331
                max(3)=        4 -> 10         0.06582                #states=21 #states>0=15 #states<0=6
                max(4)=        5 -> 11         0.05746
                max(5)=        3 ->  8         0.04229  35      Singlet-B2   46.6248         26.59    0.4669    0.000
                min(1)=        5 -> 16        -0.21775                max(1)=        3 -> 14         0.69181
                min(2)=        2 -> 10        -0.05573                max(2)=        5 -> 15         0.11017
                min(3)=        2 -> 13        -0.05376                max(3)=        2 -> 12         0.06058
                min(4)=        4 ->  9        -0.02003                max(4)=        3 ->  9         0.02927
                min(5)=        3 -> 17        -0.01494                max(5)=        3 -> 19         0.02577
                #states=34 #states>0=19 #states<0=15                   min(1)=        3 -> 13        -0.03756
                                                                       min(2)=        4 -> 18        -0.03331
33      Singlet-A1   46.1901         26.84    0.4031    0.000         min(3)=        4 ->  8        -0.02828
                max(1)=        2 -> 10         0.61488                min(4)=        2 ->  7        -0.01462
                max(2)=        2 -> 13         0.05756                min(5)=        5 -> 23        -0.01454
                max(3)=        4 ->  9         0.01346                #states=29 #states>0=14 #states<0=15
                max(4)=        2 -> 14         0.01293
                max(5)=        4 -> 13         0.01091  36      Singlet-B1   47.1570         26.29    0.0698    0.000
                min(1)=        5 -> 16        -0.33361                max(1)=        3 -> 15         0.58822
                min(2)=        3 -> 12        -0.04863                max(2)=        4 -> 16         0.37646
                min(3)=        4 -> 14        -0.03980                max(3)=        5 -> 14         0.02667
                min(4)=        4 -> 10        -0.03068                max(4)=        5 -> 13         0.01396
                min(5)=        5 -> 11        -0.02834                max(5)=        2 -> 22         0.00588
                #states=34 #states>0=20 #states<0=14                   min(1)=        2 -> 11        -0.10346
                                                                       min(2)=        5 -> 19        -0.02000
34      Singlet-B1   46.3378         26.76    0.3301    0.000         min(3)=        5 ->  9        -0.01160
                max(1)=        2 -> 11         0.68852                min(4)=        4 -> 22        -0.01026
                max(2)=        3 -> 15         0.14998                min(5)=        5 -> 10        -0.00410
                max(3)=        5 -> 13         0.02656                #states=20 #states>0=11 #states<0=9
                max(4)=        5 -> 19         0.01564
```

7.1.3 Main contributions from different excited states at CIS approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_         max(1)=        5 ->  6         0.69518
                                                                       max(2)=        5 -> 10         0.05494
1       Singlet-B1    9.1435        135.60    0.0334    0.000         max(3)=        5 -> 25         0.00722
```

```
              max(4)=        5 -> 19         0.00552                 min(1)=        2 ->  7        -0.01435
              max(5)=        5 -> 24         0.00356                 min(2)=        4 -> 18        -0.00906
              min(1)=        5 ->  9        -0.10597                 min(3)=        3 -> 19        -0.00604
              min(2)=        5 -> 13        -0.04558                 min(4)=        2 ->  8        -0.00597
              min(3)=        5 -> 14        -0.01226                 min(5)=        2 -> 18        -0.00540
              min(4)=        5 -> 20        -0.00711                 #states=28 #states>0=17 #states<0=11
              min(5)=        4 -> 22        -0.00503
              #states=20 #states>0=9 #states<0=11     5       Singlet-B2   15.1582         81.79    0.2558    0.000
                                                                      max(1)=        3 ->  6         0.69771
2       Singlet-A2   10.9976        112.74    0.0000    0.000         max(2)=        3 -> 10         0.05047
              max(1)=        5 ->  7         0.67657                 max(3)=        4 ->  7         0.00692
              max(2)=        5 ->  8         0.17849                 max(4)=        3 -> 19         0.00438
              max(3)=        5 -> 12         0.09969                 max(5)=        5 -> 15         0.00434
              max(4)=        5 -> 21         0.00613                 min(1)=        3 ->  9        -0.08306
              max(5)=        5 -> 29         0.00465                 min(2)=        4 ->  8        -0.03719
              min(1)=        5 -> 18        -0.01808                 min(3)=        3 -> 13        -0.03667
              min(2)=        3 -> 11        -0.00342                 min(4)=        2 ->  7        -0.02717
              min(3)=        2 -> 23        -0.00233                 min(5)=        3 -> 20        -0.00886
              min(4)=        4 -> 15        -0.00054                 #states=27 #states>0=14 #states<0=13
              min(5)=        4 -> 23        -0.00027
              #states=16 #states>0=10 #states<0=6     6       Singlet-A1   18.1618         68.27    0.1038    0.000
                                                                      max(1)=        3 ->  7         0.68186
3       Singlet-A1   11.6285        106.62    0.1068    0.000         max(2)=        3 ->  8         0.08424
              max(1)=        4 ->  6         0.69628                 max(3)=        4 ->  9         0.06440
              max(2)=        4 -> 10         0.01680                 max(4)=        4 ->  6         0.04995
              max(3)=        4 -> 19         0.00583                 max(5)=        3 -> 12         0.04112
              max(4)=        3 -> 18         0.00353                 min(1)=        5 -> 11        -0.11005
              max(5)=        2 -> 19         0.00296                 min(2)=        4 -> 10        -0.08070
              min(1)=        4 ->  9        -0.08738                 min(3)=        2 -> 13        -0.02116
              min(2)=        4 -> 13        -0.04633                 min(4)=        4 -> 14        -0.01121
              min(3)=        3 ->  7        -0.04312                 min(5)=        4 -> 13        -0.00981
              min(4)=        5 -> 11        -0.03734                 #states=32 #states>0=14 #states<0=18
              min(5)=        3 ->  8        -0.03001
              #states=31 #states>0=10 #states<0=21    7       Singlet-A2   18.3499         67.57    0.0000    0.000
                                                                      max(1)=        5 ->  8         0.68094
4       Singlet-B2   13.4601         92.11    0.0774    0.000         max(2)=        5 -> 12         0.02015
              max(1)=        4 ->  7         0.69007                 max(3)=        5 -> 17         0.01640
              max(2)=        4 ->  8         0.11713                 max(4)=        2 -> 15         0.00592
              max(3)=        4 -> 12         0.09317                 max(5)=        5 -> 29         0.00344
              max(4)=        3 ->  9         0.02572                 min(1)=        5 ->  7        -0.18393
              max(5)=        5 -> 15         0.01164                 min(2)=        5 -> 18        -0.04149
```

```
                min(3)=        3 -> 11        -0.00457                min(5)=        3 -> 18        -0.00817
                min(4)=        2 -> 23        -0.00179                #states=32 #states>0=19 #states<0=13
                min(5)=        5 -> 26        -0.00082
                #states=15 #states>0=10 #states<0=5     11       Singlet-A1   25.6809         48.28    0.3664    0.000
                                                                       max(1)=        3 ->  8         0.67337
8       Singlet-B1   20.2860         61.12    0.1541    0.000          max(2)=        2 ->  9         0.03073
                max(1)=        5 ->  9         0.68891                 max(3)=        3 -> 12         0.02558
                max(2)=        5 ->  6         0.11412                 max(4)=        4 -> 13         0.01546
                max(3)=        5 -> 14         0.02534                 max(5)=        4 ->  6         0.01047
                max(4)=        3 -> 15         0.02465                 min(1)=        5 -> 11        -0.14822
                max(5)=        4 -> 22         0.01113                 min(2)=        3 ->  7        -0.11290
                min(1)=        5 -> 10        -0.09559                 min(3)=        4 -> 10        -0.05653
                min(2)=        5 -> 19        -0.02832                 min(4)=        4 ->  9        -0.04766
                min(3)=        4 -> 16        -0.01626                 min(5)=        4 -> 14        -0.04454
                min(4)=        2 -> 11        -0.01490                 #states=31 #states>0=14 #states<0=17
                min(5)=        5 -> 25        -0.01030
                #states=21 #states>0=12 #states<0=9     12       Singlet-B2   27.0060         45.91    0.5800    0.000
                                                                       max(1)=        3 ->  9         0.67812
9       Singlet-B2   21.5272         57.59    0.2114    0.000          max(2)=        3 ->  6         0.08248
                max(1)=        4 ->  8         0.67789                 max(3)=        2 ->  7         0.04611
                max(2)=        3 ->  9         0.13310                 max(4)=        2 ->  8         0.03192
                max(3)=        3 ->  6         0.05162                 max(5)=        2 -> 12         0.02255
                max(4)=        5 -> 15         0.03412                 min(1)=        4 ->  8        -0.13319
                max(5)=        3 -> 14         0.02330                 min(2)=        3 -> 10        -0.08283
                min(1)=        4 ->  7        -0.12491                 min(3)=        5 -> 15        -0.05735
                min(2)=        4 -> 18        -0.02601                 min(4)=        4 -> 12        -0.02643
                min(3)=        2 ->  7        -0.02316                 min(5)=        3 -> 19        -0.01851
                min(4)=        3 -> 13        -0.01782                 #states=27 #states>0=12 #states<0=15
                min(5)=        3 -> 19        -0.01612
                #states=28 #states>0=16 #states<0=12     13       Singlet-B1   27.2615         45.48    0.0033    0.000
                                                                       max(1)=        5 -> 10         0.69414
10      Singlet-A1   22.3378         55.50    0.0065    0.000          max(2)=        5 ->  9         0.10012
                max(1)=        4 ->  9         0.68979                 max(3)=        5 -> 14         0.03855
                max(2)=        4 ->  6         0.09055                 max(4)=        5 -> 13         0.03081
                max(3)=        5 -> 11         0.07695                 max(5)=        5 -> 19         0.01715
                max(4)=        3 ->  8         0.05313                 min(1)=        4 -> 11        -0.05686
                max(5)=        4 -> 14         0.01915                 min(2)=        5 ->  6        -0.03750
                min(1)=        3 ->  7        -0.06763                 min(3)=        4 -> 16        -0.01272
                min(2)=        4 -> 19        -0.03185                 min(4)=        5 -> 20        -0.00870
                min(3)=        2 ->  6        -0.02035                 min(5)=        2 -> 11        -0.00357
                min(4)=        4 -> 10        -0.00954                 #states=21 #states>0=13 #states<0=8
```

```
                                                                max(1)=        2 ->  6         0.60273
14       Singlet-A1   28.2577         43.88    0.0348    0.000          max(2)=        4 -> 10         0.28259
               max(1)=        5 -> 11         0.52721                  max(3)=        5 -> 11         0.15393
               max(2)=        3 ->  8         0.07978                  max(4)=        3 ->  8         0.07180
               max(3)=        2 ->  6         0.05201                  max(5)=        3 ->  7         0.05580
               max(4)=        4 ->  6         0.03192                  min(1)=        2 ->  9        -0.10806
               max(5)=        3 ->  7         0.02822                  min(2)=        3 -> 12        -0.08806
               min(1)=        4 -> 10        -0.44913                  min(3)=        2 -> 13        -0.04101
               min(2)=        4 ->  9        -0.07043                  min(4)=        4 -> 14        -0.02765
               min(3)=        3 -> 12        -0.04264                  min(5)=        5 -> 16        -0.01880
               min(4)=        2 ->  9        -0.03126                  #states=33 #states>0=21 #states<0=12
               min(5)=        4 -> 13        -0.02101
               #states=32 #states>0=14 #states<0=18     18       Singlet-B2   32.9175         37.67    0.0662    0.000
                                                                max(1)=        4 -> 12         0.69403
15       Singlet-B1   29.3023         42.31    0.0020    0.000          max(2)=        2 ->  7         0.07541
               max(1)=        4 -> 11         0.70379                  max(3)=        2 -> 12         0.02083
               max(2)=        5 -> 10         0.05384                  max(4)=        4 -> 26         0.01833
               max(3)=        5 -> 13         0.02516                  max(5)=        4 -> 29         0.01056
               max(4)=        4 -> 27         0.01977                  min(1)=        4 ->  7        -0.08737
               max(5)=        5 ->  9         0.01682                  min(2)=        4 ->  8        -0.03599
               min(1)=        2 -> 22        -0.00480                  min(3)=        3 -> 10        -0.03290
               min(2)=        2 -> 16        -0.00368                  min(4)=        4 -> 18        -0.03150
               min(3)=        5 -> 20        -0.00182                  min(5)=        2 ->  8        -0.01558
               min(4)=        5 -> 25        -0.00170                  #states=25 #states>0=12 #states<0=13
               min(5)=        5 -> 24        -0.00143
               #states=20 #states>0=15 #states<0=5      19       Singlet-A2   33.7332         36.75    0.0000    0.000
                                                                max(1)=        3 -> 11         0.70597
16       Singlet-A2   30.8203         40.23    0.0000    0.000          max(2)=        3 -> 16         0.02493
               max(1)=        5 -> 12         0.69802                  max(3)=        5 -> 12         0.02242
               max(2)=        5 -> 26         0.02110                  max(4)=        3 -> 27         0.01668
               max(3)=        5 -> 29         0.01102                  max(5)=        2 -> 15         0.01003
               max(4)=        5 -> 21         0.01074                  min(1)=        2 -> 23        -0.00610
               max(5)=        2 -> 15         0.00323                  min(2)=        4 -> 15        -0.00465
               min(1)=        5 ->  7        -0.09158                  min(3)=        5 ->  7        -0.00081
               min(2)=        5 ->  8        -0.04745                  min(4)=        5 -> 29        -0.00013
               min(3)=        5 -> 18        -0.03045                  min(5)=        5 -> 26        -0.00011
               min(4)=        3 -> 11        -0.02195                  #states=16 #states>0=11 #states<0=5
               min(5)=        2 -> 23        -0.00255
               #states=16 #states>0=6 #states<0=10      20       Singlet-B2   33.9624         36.51    0.0927    0.000
                                                                max(1)=        3 -> 10         0.65878
17       Singlet-A1   32.1386         38.58    0.0020    0.000          max(2)=        2 ->  7         0.23906
```

```
                max(3)=        3 ->  9         0.06643                 max(5)=        4 -> 16         0.01432
                max(4)=        2 -> 12         0.04028                 min(1)=        3 -> 15        -0.02827
                max(5)=        3 -> 14         0.02538                 min(2)=        5 -> 10        -0.02638
                min(1)=        3 ->  6        -0.03125                 min(3)=        4 -> 11        -0.02333
                min(2)=        3 -> 13        -0.01754                 min(4)=        5 ->  9        -0.01633
                min(3)=        5 -> 15        -0.00884                 min(5)=        5 -> 25        -0.01609
                min(4)=        3 -> 20        -0.00816                 #states=21 #states>0=10 #states<0=11
                min(5)=        2 -> 18        -0.00727
                #states=28 #states>0=16 #states<0=12    24      Singlet-A1   38.6227         32.10    0.1823    0.000
                                                                        max(1)=        4 -> 13         0.68243
21      Singlet-A1   34.4238         36.02    0.0476    0.000          max(2)=        4 -> 14         0.06061
                max(1)=        3 -> 12         0.42831                 max(3)=        4 ->  6         0.04244
                max(2)=        2 ->  6         0.32214                 max(4)=        2 ->  6         0.03135
                max(3)=        2 ->  9         0.04064                 max(5)=        2 -> 10         0.03108
                max(4)=        4 ->  9         0.02431                 min(1)=        3 -> 12        -0.13386
                max(5)=        4 -> 14         0.02087                 min(2)=        4 -> 10        -0.07313
                min(1)=        4 -> 10        -0.34337                 min(3)=        5 -> 11        -0.04533
                min(2)=        5 -> 11        -0.25990                 min(4)=        3 ->  8        -0.02165
                min(3)=        3 ->  8        -0.11649                 min(5)=        4 -> 25        -0.01294
                min(4)=        3 ->  7        -0.09233                 #states=33 #states>0=22 #states<0=11
                min(5)=        3 -> 18        -0.02545
                #states=33 #states>0=20 #states<0=13    25      Singlet-B2   41.7617         29.69    0.0071    0.000
                                                                        max(1)=        2 ->  8         0.68256
22      Singlet-B2   34.5364         35.90    0.0024    0.000          max(2)=        3 -> 10         0.04830
                max(1)=        2 ->  7         0.63055                 max(3)=        4 -> 12         0.03401
                max(2)=        2 ->  8         0.16759                 max(4)=        2 -> 12         0.02331
                max(3)=        2 -> 12         0.06511                 max(5)=        5 -> 15         0.01912
                max(4)=        5 -> 15         0.06019                 min(1)=        2 ->  7        -0.16609
                max(5)=        4 ->  8         0.03692                 min(2)=        2 -> 18        -0.03097
                min(1)=        3 -> 10        -0.22620                 min(3)=        4 -> 17        -0.01476
                min(2)=        3 ->  9        -0.07412                 min(4)=        3 ->  6        -0.01196
                min(3)=        4 -> 12        -0.07274                 min(5)=        3 ->  9        -0.01089
                min(4)=        3 -> 13        -0.03724                 #states=28 #states>0=16 #states<0=12
                min(5)=        2 -> 18        -0.00980
                #states=28 #states>0=16 #states<0=12    26      Singlet-B1   42.0235         29.50    0.1561    0.000
                                                                        max(1)=        5 -> 14         0.70071
23      Singlet-B1   36.5430         33.93    0.3422    0.000          max(2)=        2 -> 11         0.03494
                max(1)=        5 -> 13         0.70240                 max(3)=        4 -> 16         0.03492
                max(2)=        5 ->  6         0.04567                 max(4)=        5 -> 20         0.01183
                max(3)=        2 -> 11         0.03321                 max(5)=        5 ->  6         0.01042
                max(4)=        5 -> 20         0.02217                 min(1)=        3 -> 15        -0.05856
```

```
              min(2)=        5 -> 10        -0.03304                 min(4)=        4 ->  8        -0.04507
              min(3)=        5 ->  9        -0.02867                 min(5)=        4 -> 18        -0.03734
              min(4)=        5 -> 24        -0.02765                 #states=29 #states>0=13 #states<0=16
              min(5)=        2 -> 16        -0.00874
              #states=21 #states>0=9 #states<0=12     30       Singlet-A2   44.8333         27.65    0.0000    0.000
                                                                      max(1)=        4 -> 15         0.70546
27       Singlet-A1   42.1927         29.39    0.0734    0.000          max(2)=        4 -> 23         0.02901
              max(1)=        2 ->  9         0.54454                  max(3)=        3 -> 11         0.00592
              max(2)=        2 ->  6         0.15166                  max(4)=        2 -> 23         0.00589
              max(3)=        4 -> 14         0.08151                  max(5)=        5 -> 21         0.00578
              max(4)=        5 -> 16         0.04288                  min(1)=        2 -> 15        -0.02964
              max(5)=        2 -> 13         0.03024                  min(2)=        3 -> 16        -0.02206
              min(1)=        3 -> 12        -0.34801                  min(3)=        5 -> 12        -0.00231
              min(2)=        4 -> 10        -0.12503                  min(4)=        5 -> 18        -0.00173
              min(3)=        5 -> 11        -0.11016                  min(5)=        3 -> 22        -0.00158
              min(4)=        4 -> 13        -0.10997                  #states=16 #states>0=10 #states<0=6
              min(5)=        2 -> 10        -0.07825
              #states=32 #states>0=15 #states<0=17     31       Singlet-A1   45.0818         27.50    0.2982    0.000
                                                                      max(1)=        4 -> 14         0.47648
28       Singlet-B2   42.6650         29.06    0.0658    0.000          max(2)=        2 -> 13         0.07653
              max(1)=        3 -> 13         0.62289                  max(3)=        2 ->  6         0.04783
              max(2)=        5 -> 15         0.32791                  max(4)=        5 -> 22         0.02402
              max(3)=        2 -> 12         0.04011                  max(5)=        2 -> 20         0.01195
              max(4)=        3 ->  6         0.03196                  min(1)=        2 ->  9        -0.34241
              max(5)=        3 -> 20         0.02091                  min(2)=        3 -> 12        -0.25008
              min(1)=        3 -> 14        -0.01060                  min(3)=        5 -> 11        -0.16084
              min(2)=        3 -> 25        -0.00644                  min(4)=        4 -> 10        -0.15815
              min(3)=        4 ->  8        -0.00507                  min(5)=        5 -> 16        -0.11879
              min(4)=        4 ->  7        -0.00289                  #states=33 #states>0=18 #states<0=15
              min(5)=        2 ->  8        -0.00155
              #states=29 #states>0=20 #states<0=9      32       Singlet-A1   45.7819         27.08    0.1032    0.000
                                                                      max(1)=        4 -> 14         0.44738
29       Singlet-B2   43.9053         28.24    0.4593    0.000          max(2)=        3 -> 12         0.29431
              max(1)=        5 -> 15         0.60140                  max(3)=        2 ->  9         0.25822
              max(2)=        3 ->  9         0.05190                  max(4)=        5 -> 11         0.19824
              max(3)=        3 -> 19         0.02666                  max(5)=        4 -> 10         0.17814
              max(4)=        4 -> 17         0.02440                  min(1)=        5 -> 16        -0.22536
              max(5)=        3 -> 10         0.01691                  min(2)=        2 -> 13        -0.09773
              min(1)=        3 -> 13        -0.31918                  min(3)=        2 ->  6        -0.04880
              min(2)=        3 -> 14        -0.15346                  min(4)=        4 ->  9        -0.03835
              min(3)=        2 ->  7        -0.05055                  min(5)=        2 -> 14        -0.02401
```

```
                #states=32 #states>0=16 #states<0=16

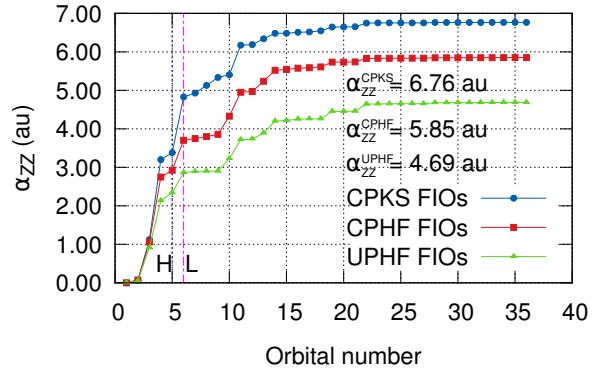
                                                    35       Singlet-A1   49.6651         24.96    0.9053    0.000
33       Singlet-B2   48.5010         25.56    0.6210    0.000          max(1)=        5 -> 16         0.60968
                max(1)=        3 -> 14         0.67999                  max(2)=        4 -> 14         0.19094
                max(2)=        5 -> 15         0.12960                  max(3)=        3 -> 12         0.08274
                max(3)=        2 -> 12         0.08822                  max(4)=        4 -> 10         0.06910
                max(4)=        4 -> 17         0.04585                  max(5)=        3 -> 18         0.05888
                max(5)=        3 -> 19         0.02988                  min(1)=        2 -> 10        -0.25123
                min(1)=        3 -> 13        -0.06053                  min(2)=        2 -> 13        -0.07250
                min(2)=        2 -> 17        -0.04136                  min(3)=        4 -> 19        -0.02658
                min(3)=        4 ->  8        -0.03682                  min(4)=        3 -> 17        -0.02650
                min(4)=        4 -> 18        -0.03179                  min(5)=        4 ->  9        -0.02388
                min(5)=        2 ->  7        -0.03020                  #states=33 #states>0=16 #states<0=17
                #states=28 #states>0=13 #states<0=15

                                                    36       Singlet-B1   50.4783         24.56    0.2298    0.000
34       Singlet-B1   48.9576         25.32    0.6080    0.000          max(1)=        2 -> 11         0.54624
                max(1)=        3 -> 15         0.64380                  max(2)=        2 -> 16         0.02227
                max(2)=        2 -> 11         0.25506                  max(3)=        4 -> 22         0.01636
                max(3)=        4 -> 16         0.12313                  max(4)=        2 -> 27         0.01540
                max(4)=        2 -> 16         0.04626                  max(5)=        5 ->  9         0.00624
                max(5)=        5 -> 14         0.03417                  min(1)=        4 -> 16        -0.42598
                min(1)=        5 -> 19        -0.02506                  min(2)=        3 -> 15        -0.13474
                min(2)=        5 ->  9        -0.01912                  min(3)=        5 -> 13        -0.02236
                min(3)=        4 -> 22        -0.01893                  min(4)=        5 -> 14        -0.01666
                min(4)=        4 -> 11        -0.00794                  min(5)=        3 -> 23        -0.01037
                min(5)=        5 -> 10        -0.00645                  #states=20 #states>0=10 #states<0=10
                #states=20 #states>0=14 #states<0=6
```

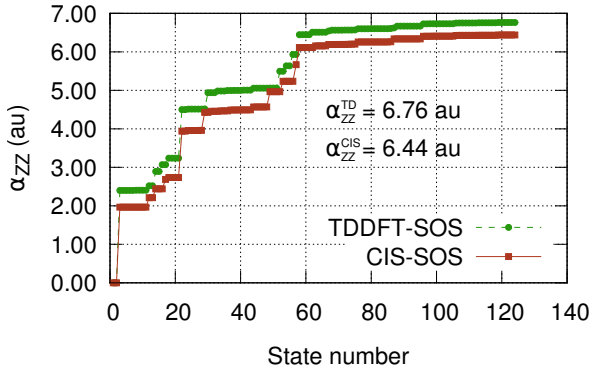
7.2 6-311++G(d,p)

7.2.1 Plots

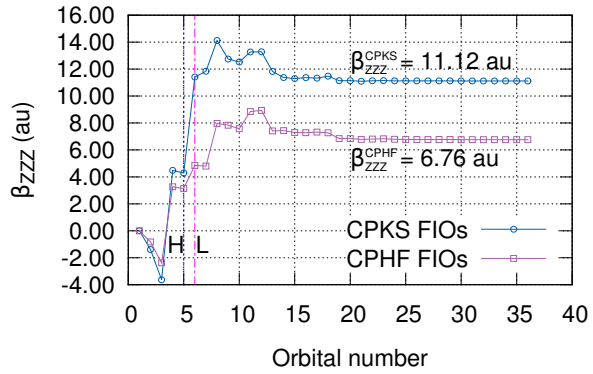
Figure S10: For H_2O molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S10a and S10c) or states (SOS approaches, in Plots S10b and S10d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.01 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



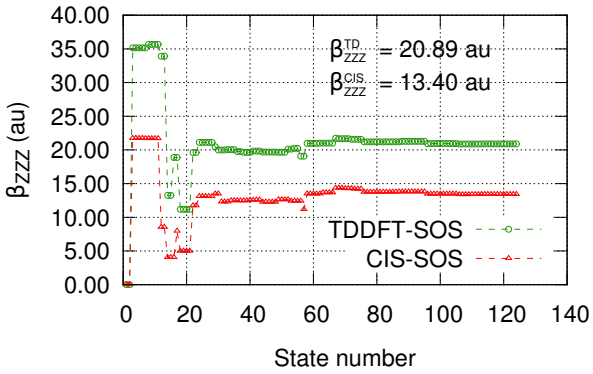
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

7.2.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_
1        Singlet-B1    7.0656        175.48    0.0430    0.000
               max(1)=        5 ->  6         0.69164
               max(2)=        5 ->  9         0.02286
               max(3)=        5 -> 16         0.00818
               max(4)=        2 -> 17         0.00446
               max(5)=        5 -> 31         0.00316
               min(1)=        5 ->  8        -0.14018
               min(2)=        5 -> 13        -0.03766
               min(3)=        5 -> 15        -0.01357
               min(4)=        4 -> 10        -0.00353
               min(5)=        3 -> 29        -0.00260
               #CIs=24 #CIs>0=15 #CIs<0=9
2        Singlet-A2    8.6960        142.58    0.0000    0.000
               max(1)=        5 ->  7         0.68100
               max(2)=        5 -> 11         0.14949
               max(3)=        5 -> 12         0.11540
               max(4)=        5 -> 18         0.00966
               max(5)=        2 -> 20         0.00197
               min(1)=        5 -> 14        -0.02226
               min(2)=        3 -> 10        -0.00261
               min(3)=        3 -> 17        -0.00148
               min(4)=        5 -> 32        -0.00065
               min(5)=        4 -> 20         0.00009
               #CIs=19 #CIs>0=15 #CIs<0=4
3        Singlet-A1    9.1679        135.24    0.0909    0.000
               max(1)=        4 ->  6         0.69150
               max(2)=        4 ->  9         0.05192
               max(3)=        3 -> 14         0.01310
               max(4)=        2 ->  9         0.00593
               max(5)=        2 -> 16         0.00377
               min(1)=        4 ->  8        -0.10644
               min(2)=        5 -> 10        -0.06702
               min(3)=        4 -> 13        -0.04911
               min(4)=        3 -> 11        -0.02173
               min(5)=        3 ->  7        -0.01263
               #CIs=42 #CIs>0=18 #CIs<0=24
4        Singlet-B2   10.7886        114.92    0.0220    0.000
               max(1)=        4 ->  7         0.69084
               max(2)=        4 -> 12         0.11322
               max(3)=        4 -> 11         0.09718
               max(4)=        3 ->  8         0.01419
               max(5)=        3 -> 15         0.01030
               min(1)=        2 -> 11        -0.00694
               min(2)=        3 ->  6        -0.00615
               min(3)=        2 ->  7        -0.00548
               min(4)=        3 -> 19        -0.00384
               min(5)=        3 ->  9        -0.00355
               #CIs=34 #CIs>0=17 #CIs<0=17
5        Singlet-B1   11.3098        109.63    0.0135    0.000
               max(1)=        5 ->  8         0.61707
               max(2)=        5 ->  9         0.31795
               max(3)=        5 ->  6         0.11232
               max(4)=        5 -> 15         0.03812
               max(5)=        3 -> 20         0.00328
               min(1)=        5 -> 13        -0.05807
               min(2)=        5 -> 16        -0.02629
               min(3)=        2 -> 10        -0.00579
               min(4)=        5 -> 19        -0.00305
               min(5)=        4 -> 10        -0.00149
               #CIs=25 #CIs>0=12 #CIs<0=13
6        Singlet-A2   11.6120        106.77    0.0000    0.000
               max(1)=        5 -> 11         0.68564
               max(2)=        5 -> 18         0.00691
               max(3)=        2 -> 20         0.00282
               max(4)=        5 -> 24         0.00274
               max(5)=        5 -> 12         0.00157
               min(1)=        5 ->  7        -0.15347
               min(2)=        5 -> 14        -0.07909
               min(3)=        3 -> 10        -0.00732
               min(4)=        3 -> 17        -0.00117
               min(5)=        5 -> 32        -0.00044
               #CIs=19 #CIs>0=14 #CIs<0=5
7        Singlet-B1   11.8022        105.05    0.0041    0.000
               max(1)=        5 ->  9         0.62946
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              max(2)=        4 -> 10         0.03259                  max(4)=        5 ->  8         0.02209
              max(3)=        2 -> 10         0.00942                  max(5)=        5 ->  6         0.01483
              max(4)=        4 -> 22         0.00487                  min(1)=        5 ->  9        -0.02346
              max(5)=        4 -> 17         0.00454                  min(2)=        5 -> 15        -0.00582
              min(1)=        5 ->  8        -0.30783                  min(3)=        5 -> 16        -0.00465
              min(2)=        5 ->  6        -0.08343                  min(4)=        4 -> 22        -0.00297
              min(3)=        5 -> 15        -0.02764                  min(5)=        5 -> 25        -0.00249
              min(4)=        5 -> 21        -0.00965                  #CIs=26 #CIs>0=14 #CIs<0=12
              min(5)=        5 -> 16        -0.00793
              #CIs=25 #CIs>0=11 #CIs<0=14            11       Singlet-A2   13.5756         91.33    0.0000    0.000
                                                                      max(1)=        5 -> 12         0.69638
8       Singlet-A1   12.0620        102.79    0.0003    0.000        max(2)=        5 -> 18         0.03155
              max(1)=        5 -> 10         0.65817                  max(3)=        2 -> 20         0.00131
              max(2)=        4 ->  9         0.16900                  max(4)=        5 -> 24         0.00105
              max(3)=        4 ->  8         0.14652                  max(5)=        5 -> 23         0.00070
              max(4)=        4 ->  6         0.06805                  min(1)=        5 ->  7        -0.11265
              max(5)=        4 -> 15         0.01365                  min(2)=        5 -> 11        -0.02965
              min(1)=        3 ->  7        -0.07841                  min(3)=        5 -> 14        -0.02148
              min(2)=        3 -> 11        -0.04695                  min(4)=        3 -> 10        -0.00598
              min(3)=        3 -> 12        -0.04549                  min(5)=        3 -> 17        -0.00171
              min(4)=        4 -> 13        -0.03117                  #CIs=19 #CIs>0=13 #CIs<0=6
              min(5)=        4 -> 16        -0.02145
              #CIs=42 #CIs>0=17 #CIs<0=25            12       Singlet-A1   13.6771         90.65    0.0098    0.000
                                                                      max(1)=        4 ->  8         0.62303
9       Singlet-B2   13.0950         94.68    0.1017    0.000        max(2)=        3 ->  7         0.12860
              max(1)=        3 ->  6         0.68118                  max(3)=        4 ->  6         0.11331
              max(2)=        4 ->  7         0.02655                  max(4)=        3 -> 11         0.06724
              max(3)=        4 -> 14         0.02207                  max(5)=        4 -> 15         0.04742
              max(4)=        3 -> 16         0.00659                  min(1)=        4 ->  9        -0.26220
              max(5)=        4 -> 12         0.00530                  min(2)=        5 -> 10        -0.06239
              min(1)=        4 -> 11        -0.13326                  min(3)=        4 -> 13        -0.03142
              min(2)=        3 ->  8        -0.12410                  min(4)=        3 -> 14        -0.02107
              min(3)=        3 -> 13        -0.03958                  min(5)=        2 ->  6        -0.01224
              min(4)=        2 -> 14        -0.00697                  #CIs=41 #CIs>0=26 #CIs<0=15
              min(5)=        2 ->  7        -0.00589
              #CIs=35 #CIs>0=22 #CIs<0=13            13       Singlet-B1   13.9251         89.04    0.2295    0.000
                                                                      max(1)=        5 -> 13         0.69411
10      Singlet-B1   13.4847         91.94    0.0043    0.000        max(2)=        5 ->  6         0.04408
              max(1)=        4 -> 10         0.69627                  max(3)=        5 ->  8         0.04008
              max(2)=        5 -> 13         0.11539                  max(4)=        5 ->  9         0.03321
              max(3)=        4 -> 17         0.02519                  max(5)=        2 -> 10         0.00825
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              min(1)=        4 -> 10        -0.11588              min(3)=        4 ->  9        -0.18768
              min(2)=        3 -> 20        -0.00829              min(4)=        3 -> 11        -0.03757
              min(3)=        4 -> 17        -0.00549              min(5)=        5 -> 17        -0.03005
              min(4)=        2 -> 17        -0.00266              #CIs=41 #CIs>0=12 #CIs<0=29
              min(5)=        5 -> 16        -0.00254
              #CIs=26 #CIs>0=13 #CIs<0=13            17      Singlet-B2   15.5247         79.86    0.0042    0.000
                                                                   max(1)=        4 -> 12         0.69625
14      Singlet-A1   14.0017         88.55    0.0329    0.000   max(2)=        4 -> 18         0.03145
              max(1)=        4 ->  9         0.58843              max(3)=        3 ->  9         0.01360
              max(2)=        3 ->  7         0.30091              max(4)=        4 -> 11         0.00900
              max(3)=        4 ->  8         0.16055              max(5)=        2 -> 14         0.00268
              max(4)=        3 -> 11         0.10152              min(1)=        4 ->  7        -0.11596
              max(5)=        3 -> 12         0.08715              min(2)=        4 -> 14        -0.01951
              min(1)=        5 -> 10        -0.13441              min(3)=        3 -> 13        -0.01010
              min(2)=        4 ->  6        -0.02044              min(4)=        2 -> 11        -0.00717
              min(3)=        3 -> 14        -0.01773              min(5)=        3 -> 19        -0.00186
              min(4)=        4 -> 15        -0.00766              #CIs=35 #CIs>0=20 #CIs<0=15
              min(5)=        2 ->  6        -0.00742
              #CIs=43 #CIs>0=25 #CIs<0=18            18      Singlet-A1   15.8050         78.45    0.0187    0.000
                                                                   max(1)=        4 -> 13         0.64318
15      Singlet-B2   14.0754         88.09    0.1941    0.000   max(2)=        3 ->  7         0.24176
              max(1)=        4 -> 11         0.68254              max(3)=        5 -> 10         0.08461
              max(2)=        3 ->  6         0.14328              max(4)=        4 ->  6         0.05523
              max(3)=        3 ->  8         0.03412              max(5)=        3 -> 14         0.01957
              max(4)=        3 -> 15         0.03244              min(1)=        4 ->  9        -0.10020
              max(5)=        5 -> 20         0.01551              min(2)=        4 ->  8        -0.05266
              min(1)=        4 ->  7        -0.09217              min(3)=        3 -> 12        -0.04434
              min(2)=        4 -> 14        -0.02779              min(4)=        3 -> 11        -0.04188
              min(3)=        3 -> 13        -0.02701              min(5)=        4 -> 16        -0.02236
              min(4)=        4 -> 12        -0.02574              #CIs=43 #CIs>0=17 #CIs<0=26
              min(5)=        3 ->  9        -0.02356
              #CIs=35 #CIs>0=17 #CIs<0=18            19      Singlet-B2   17.4650         70.99    0.0202    0.000
                                                                   max(1)=        3 ->  8         0.55587
16      Singlet-A1   15.4623         80.18    0.0190    0.000   max(2)=        3 ->  9         0.42179
              max(1)=        3 ->  7         0.57066              max(3)=        3 ->  6         0.08974
              max(2)=        5 -> 10         0.14238              max(4)=        3 -> 15         0.02027
              max(3)=        3 -> 14         0.02853              max(5)=        2 -> 11         0.00993
              max(4)=        4 -> 15         0.01668              min(1)=        3 -> 13        -0.04292
              max(5)=        2 ->  6         0.00983              min(2)=        4 -> 11        -0.03631
              min(1)=        4 -> 13        -0.28149              min(3)=        3 -> 16        -0.02610
              min(2)=        4 ->  8        -0.18966              min(4)=        4 -> 14        -0.02418
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                min(5)=        4 -> 12        -0.01091
                #CIs=35 #CIs>0=14 #CIs<0=21            23       Singlet-B2   19.6757         63.01    0.0093    0.000
                                                                       max(1)=        3 -> 13         0.70202
20       Singlet-A2   17.5250         70.75    0.0000    0.000        max(2)=        3 ->  6         0.04877
                max(1)=        3 -> 10         0.70641                max(3)=        4 -> 14         0.04441
                max(2)=        3 -> 17         0.02690                max(4)=        3 ->  8         0.03327
                max(3)=        5 -> 14         0.01239                max(5)=        3 ->  9         0.02172
                max(4)=        5 -> 11         0.00898                min(1)=        2 ->  7        -0.01917
                max(5)=        5 -> 12         0.00697                min(2)=        2 -> 11        -0.01135
                min(1)=        5 -> 18        -0.00213                min(3)=        3 -> 16        -0.00612
                min(2)=        5 -> 24        -0.00087                min(4)=        3 -> 15        -0.00505
                min(3)=        4 -> 20        -0.00015                min(5)=        4 -> 24        -0.00454
                min(4)=        5 -> 32         0.00008                #CIs=36 #CIs>0=22 #CIs<0=14
                min(5)=        5 -> 23         0.00010
                #CIs=18 #CIs>0=15 #CIs<0=3             24       Singlet-A1   20.4158         60.73    0.0023    0.000
                                                                       max(1)=        3 -> 12         0.66605
21       Singlet-B2   18.0920         68.53    0.2259    0.000        max(2)=        3 -> 11         0.15716
                max(1)=        3 ->  9         0.56528                max(3)=        5 -> 10         0.08316
                max(2)=        4 -> 11         0.05752                max(4)=        4 -> 15         0.05918
                max(3)=        4 -> 14         0.04931                max(5)=        2 ->  6         0.04360
                max(4)=        5 -> 20         0.01222                min(1)=        4 ->  8        -0.07218
                max(5)=        4 -> 18         0.00590                min(2)=        4 ->  9        -0.06693
                min(1)=        3 ->  8        -0.41187                min(3)=        5 -> 17        -0.06213
                min(2)=        3 ->  6        -0.06941                min(4)=        4 -> 16        -0.04766
                min(3)=        2 -> 14        -0.00990                min(5)=        3 ->  7        -0.03684
                min(4)=        3 -> 15        -0.00868                #CIs=43 #CIs>0=17 #CIs<0=26
                min(5)=        4 -> 12        -0.00862
                #CIs=36 #CIs>0=19 #CIs<0=17            25       Singlet-A2   20.6769         59.96    0.0000    0.000
                                                                       max(1)=        5 -> 14         0.70166
22       Singlet-A1   18.4329         67.26    0.1931    0.000        max(2)=        5 -> 11         0.08102
                max(1)=        3 -> 11         0.66976                max(3)=        5 -> 12         0.02532
                max(2)=        5 -> 10         0.05992                max(4)=        5 -> 27         0.00207
                max(3)=        4 -> 15         0.03792                max(5)=        5 ->  7         0.00089
                max(4)=        2 ->  9         0.02021                min(1)=        5 -> 24        -0.01531
                max(5)=        4 ->  6         0.01630                min(2)=        3 -> 10        -0.01361
                min(1)=        3 -> 12        -0.18873                min(3)=        5 -> 23        -0.00462
                min(2)=        4 ->  8        -0.07682                min(4)=        2 -> 20        -0.00418
                min(3)=        4 ->  9        -0.04611                min(5)=        5 -> 18        -0.00409
                min(4)=        5 -> 17        -0.03060                #CIs=19 #CIs>0=7 #CIs<0=12
                min(5)=        4 -> 16        -0.02777
                #CIs=42 #CIs>0=17 #CIs<0=25            26       Singlet-B1   22.2915         55.62    0.0437    0.000
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                max(1)=        5 -> 15         0.70375                 max(3)=        4 -> 15         0.05628
                max(2)=        3 -> 20         0.01885                 max(4)=        3 -> 18         0.03052
                max(3)=        5 -> 21         0.01033                 max(5)=        4 -> 16         0.02993
                max(4)=        5 ->  9         0.00756                 min(1)=        3 -> 14        -0.13429
                max(5)=        4 -> 28         0.00736                 min(2)=        2 ->  8        -0.07799
                min(1)=        5 ->  8        -0.04895                 min(3)=        3 -> 12        -0.03442
                min(2)=        2 -> 10        -0.02729                 min(4)=        2 -> 13        -0.03160
                min(3)=        5 -> 16        -0.02487                 min(5)=        3 -> 11        -0.02429
                min(4)=        4 -> 22        -0.01332                 #CIs=42 #CIs>0=23 #CIs<0=19
                min(5)=        5 -> 19        -0.01326
                #CIs=26 #CIs>0=13 #CIs<0=13            30       Singlet-A1   28.0269         44.24    0.1489    0.000
                                                                        max(1)=        3 -> 14         0.67325
27       Singlet-B2   23.5321         52.69    0.0937    0.000          max(2)=        2 ->  6         0.11931
                max(1)=        4 -> 14         0.69115                 max(3)=        5 -> 17         0.09799
                max(2)=        3 ->  8         0.05384                 max(4)=        4 -> 15         0.08717
                max(3)=        4 -> 11         0.03163                 max(5)=        4 -> 16         0.08316
                max(4)=        2 ->  7         0.02634                 min(1)=        2 ->  9        -0.05739
                max(5)=        2 -> 12         0.02287                 min(2)=        4 -> 19        -0.03710
                min(1)=        3 -> 15        -0.11070                 min(3)=        5 -> 10        -0.02311
                min(2)=        3 -> 13        -0.04548                 min(4)=        3 ->  7        -0.01165
                min(3)=        5 -> 20        -0.03452                 min(5)=        4 ->  6        -0.01067
                min(4)=        3 ->  9        -0.02616                 #CIs=42 #CIs>0=27 #CIs<0=15
                min(5)=        4 -> 18        -0.01290
                #CIs=36 #CIs>0=17 #CIs<0=19            31       Singlet-B2   28.1815         43.99    0.0104    0.000
                                                                        max(1)=        2 ->  7         0.69014
28       Singlet-A1   24.2153         51.20    0.0000    0.000          max(2)=        2 -> 11         0.11997
                max(1)=        4 -> 15         0.69087                 max(3)=        2 -> 12         0.08383
                max(2)=        2 ->  8         0.04154                 max(4)=        3 -> 13         0.02229
                max(3)=        5 -> 17         0.02851                 max(5)=        5 -> 20         0.01823
                max(4)=        4 ->  9         0.02817                 min(1)=        4 -> 14        -0.02827
                max(5)=        4 -> 16         0.01398                 min(2)=        3 -> 19        -0.01307
                min(1)=        3 -> 14        -0.08550                 min(3)=        4 -> 24        -0.00683
                min(2)=        2 ->  6        -0.07527                 min(4)=        2 -> 14        -0.00596
                min(3)=        3 -> 11        -0.04622                 min(5)=        3 -> 21        -0.00495
                min(4)=        4 ->  8        -0.03836                 #CIs=35 #CIs>0=20 #CIs<0=15
                min(5)=        3 -> 12        -0.03810
                #CIs=43 #CIs>0=20 #CIs<0=23            32       Singlet-B2   29.1611         42.52    0.2181    0.000
                                                                        max(1)=        3 -> 15         0.69228
29       Singlet-A1   26.6660         46.50    0.0018    0.000          max(2)=        4 -> 14         0.10962
                max(1)=        2 ->  6         0.68062                 max(3)=        3 -> 19         0.01872
                max(2)=        5 -> 17         0.06049                 max(4)=        2 -> 14         0.01758
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                max(5)=        2 -> 12         0.01287                 min(2)=        2 -> 14        -0.05116
                min(1)=        5 -> 20        -0.06585                 min(3)=        2 -> 12        -0.02835
                min(2)=        2 -> 11        -0.03616                 min(4)=        3 ->  9        -0.01142
                min(3)=        3 -> 16        -0.03299                 min(5)=        3 -> 19        -0.01000
                min(4)=        4 -> 18        -0.02836                 #CIs=36 #CIs>0=16 #CIs<0=20
                min(5)=        4 -> 11        -0.02567
                #CIs=36 #CIs>0=16 #CIs<0=20            36       Singlet-A1   31.2155         39.72    0.0052    0.000
                                                                        max(1)=        2 ->  8         0.48330
33       Singlet-A1   31.0251         39.96    0.0189    0.000          max(2)=        2 ->  6         0.07753
                max(1)=        2 ->  9         0.50775                 max(3)=        2 -> 15         0.03221
                max(2)=        2 ->  8         0.48135                 max(4)=        5 -> 10         0.02209
                max(3)=        2 ->  6         0.04451                 max(5)=        2 -> 16         0.00798
                max(4)=        3 -> 14         0.02177                 min(1)=        2 ->  9        -0.48189
                max(5)=        4 -> 21         0.02010                 min(2)=        5 -> 17        -0.12505
                min(1)=        2 -> 13        -0.06626                 min(3)=        2 -> 13        -0.06967
                min(2)=        5 -> 17        -0.03361                 min(4)=        4 -> 16        -0.05388
                min(3)=        3 -> 18        -0.01986                 min(5)=        3 -> 14        -0.04433
                min(4)=        4 -> 19        -0.01932                 #CIs=42 #CIs>0=17 #CIs<0=25
                min(5)=        2 -> 16        -0.01669
                #CIs=41 #CIs>0=19 #CIs<0=22            37       Singlet-B1   32.4479         38.21    0.0142    0.000
                                                                        max(1)=        5 -> 16         0.70243
34       Singlet-B1   31.0404         39.94    0.0142    0.000          max(2)=        2 -> 10         0.05137
                max(1)=        2 -> 10         0.70335                 max(3)=        5 -> 15         0.02635
                max(2)=        5 -> 15         0.02498                 max(4)=        5 -> 21         0.02185
                max(3)=        2 -> 17         0.01805                 max(5)=        5 ->  8         0.01975
                max(4)=        3 -> 20         0.01732                 min(1)=        4 -> 17        -0.03193
                max(5)=        4 -> 28         0.00653                 min(2)=        4 -> 22        -0.01620
                min(1)=        5 -> 16        -0.05427                 min(3)=        5 -> 19        -0.01409
                min(2)=        4 -> 17        -0.01899                 min(4)=        5 -> 25        -0.01100
                min(3)=        5 -> 19        -0.01674                 min(5)=        5 ->  6        -0.00410
                min(4)=        4 -> 22        -0.01368                 #CIs=26 #CIs>0=16 #CIs<0=10
                min(5)=        5 -> 25        -0.01011
                #CIs=26 #CIs>0=14 #CIs<0=12            38       Singlet-A1   33.0038         37.57    0.0017    0.000
                                                                        max(1)=        2 -> 13         0.68721
35       Singlet-B2   31.1819         39.76    0.0149    0.000          max(2)=        2 ->  8         0.06590
                max(1)=        2 -> 11         0.69297                 max(3)=        2 ->  6         0.05231
                max(2)=        3 -> 15         0.04061                 max(4)=        2 ->  9         0.01416
                max(3)=        3 -> 13         0.00939                 max(5)=        2 -> 21         0.01130
                max(4)=        4 -> 12         0.00818                 min(1)=        5 -> 17        -0.13902
                max(5)=        3 -> 16         0.00701                 min(2)=        4 -> 16        -0.02383
                min(1)=        2 ->  7        -0.11893                 min(3)=        2 -> 15        -0.01242
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                min(4)=        2 -> 16        -0.00477                #CIs=42 #CIs>0=25 #CIs<0=17
                min(5)=        3 -> 14        -0.00464
                #CIs=41 #CIs>0=23 #CIs<0=18            42       Singlet-B1   35.4521         34.97    0.0039    0.000
                                                                      max(1)=        4 -> 17         0.70538
39       Singlet-B2   33.1201         37.43    0.0247    0.000          max(2)=        5 -> 16         0.02975
                max(1)=        2 -> 12         0.69911                max(3)=        2 -> 10         0.02052
                max(2)=        5 -> 20         0.02937                max(4)=        3 -> 20         0.00947
                max(3)=        3 -> 16         0.02180                max(5)=        5 -> 15         0.00740
                max(4)=        2 -> 18         0.02088                min(1)=        4 -> 10        -0.02562
                max(5)=        4 -> 18         0.01152                min(2)=        5 -> 19        -0.01421
                min(1)=        2 ->  7        -0.08959                min(3)=        5 -> 25        -0.00616
                min(2)=        3 -> 19        -0.01843                min(4)=        4 -> 22        -0.00592
                min(3)=        4 -> 14        -0.01818                min(5)=        2 -> 17        -0.00464
                min(4)=        2 -> 14        -0.01467                #CIs=26 #CIs>0=13 #CIs<0=13
                min(5)=        3 -> 21        -0.00984
                #CIs=36 #CIs>0=19 #CIs<0=17            43       Singlet-B2   35.7803         34.65    0.0613    0.000
                                                                      max(1)=        4 -> 18         0.70366
40       Singlet-A2   33.7225         36.77    0.0000    0.000          max(2)=        2 -> 14         0.02139
                max(1)=        5 -> 18         0.70618                max(3)=        3 -> 15         0.02050
                max(2)=        5 -> 24         0.00677                max(4)=        4 -> 14         0.01553
                max(3)=        5 -> 14         0.00629                max(5)=        3 -> 19         0.01126
                max(4)=        4 -> 20         0.00435                min(1)=        5 -> 20        -0.03337
                max(5)=        5 -> 35         0.00314                min(2)=        3 -> 16        -0.03210
                min(1)=        5 -> 12        -0.03253                min(3)=        4 -> 12        -0.03179
                min(2)=        5 -> 11        -0.00695                min(4)=        4 -> 11        -0.01158
                min(3)=        5 -> 32        -0.00639                min(5)=        3 -> 21        -0.00842
                min(4)=        3 -> 17        -0.00615                #CIs=36 #CIs>0=14 #CIs<0=22
                min(5)=        5 ->  7        -0.00278
                #CIs=18 #CIs>0=9 #CIs<0=9              44       Singlet-A1   37.5457         33.02    0.0323    0.000
                                                                      max(1)=        4 -> 16         0.41222
41       Singlet-A1   34.0157         36.45    0.0040    0.000          max(2)=        5 -> 17         0.35178
                max(1)=        4 -> 16         0.49937                max(3)=        2 ->  8         0.07722
                max(2)=        3 -> 18         0.04544                max(4)=        2 -> 13         0.07503
                max(3)=        2 -> 15         0.02644                max(5)=        3 -> 12         0.05513
                max(4)=        4 ->  9         0.01845                min(1)=        3 -> 18        -0.39272
                max(5)=        4 -> 19         0.01842                min(2)=        2 -> 15        -0.12480
                min(1)=        5 -> 17        -0.48584                min(3)=        3 -> 14        -0.08672
                min(2)=        2 -> 13        -0.07454                min(4)=        4 -> 21        -0.08143
                min(3)=        2 ->  8        -0.06545                min(5)=        2 ->  9        -0.05149
                min(4)=        3 -> 12        -0.01306                #CIs=43 #CIs>0=20 #CIs<0=23
                min(5)=        5 -> 22        -0.00944
```

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45       Singlet-B2   38.5945         32.12    0.0351    0.000          max(2)=        5 -> 20         0.12263
               max(1)=        3 -> 16         0.69627                  max(3)=        2 -> 11         0.05122
               max(2)=        2 -> 14         0.04436                  max(4)=        3 -> 19         0.03177
               max(3)=        4 -> 18         0.02423                  max(5)=        3 -> 21         0.01376
               max(4)=        3 -> 15         0.02209                  min(1)=        3 -> 16        -0.02499
               max(5)=        3 ->  9         0.01842                  min(2)=        4 -> 18        -0.01663
               min(1)=        5 -> 20        -0.10355                  min(3)=        4 -> 23        -0.00809
               min(2)=        2 -> 12        -0.01691                  min(4)=        3 ->  8        -0.00765
               min(3)=        3 -> 25        -0.00895                  min(5)=        3 -> 15        -0.00739
               min(4)=        5 -> 29        -0.00581                  #CIs=36 #CIs>0=20 #CIs<0=16
               min(5)=        2 -> 11        -0.00513
               #CIs=35 #CIs>0=21 #CIs<0=14             49       Singlet-A1   41.2289         30.07    0.0012    0.000
                                                                        max(1)=        2 -> 15         0.56383
46       Singlet-A2   39.4993         31.39    0.0000    0.000          max(2)=        4 -> 19         0.11671
               max(1)=        3 -> 17         0.70647                  max(3)=        4 -> 21         0.09107
               max(2)=        4 -> 20         0.00927                  max(4)=        2 ->  9         0.02576
               max(3)=        5 -> 18         0.00629                  max(5)=        3 -> 14         0.02202
               max(4)=        3 -> 33         0.00532                  min(1)=        3 -> 18        -0.37659
               max(5)=        2 -> 20         0.00429                  min(2)=        4 -> 16        -0.08335
               min(1)=        3 -> 10        -0.02688                  min(3)=        5 -> 17        -0.07448
               min(2)=        5 -> 14        -0.00138                  min(4)=        2 ->  8        -0.05965
               min(3)=        5 -> 24        -0.00136                  min(5)=        2 -> 16        -0.01841
               min(4)=        2 -> 29        -0.00080                  #CIs=43 #CIs>0=21 #CIs<0=22
               min(5)=        5 -> 27        -0.00069
               #CIs=18 #CIs>0=11 #CIs<0=7              50       Singlet-B2   41.7194         29.72    0.5302    0.000
                                                                        max(1)=        5 -> 20         0.67217
47       Singlet-B1   40.1338         30.89    0.0075    0.000          max(2)=        3 -> 19         0.11765
               max(1)=        5 -> 19         0.70576                  max(3)=        3 -> 16         0.10033
               max(2)=        3 -> 20         0.02474                  max(4)=        3 -> 15         0.06251
               max(3)=        2 -> 10         0.01663                  max(5)=        4 -> 14         0.04309
               max(4)=        5 -> 15         0.01360                  min(1)=        2 -> 14        -0.11772
               max(5)=        4 -> 17         0.01259                  min(2)=        2 -> 12        -0.03085
               min(1)=        4 -> 22        -0.01680                  min(3)=        3 -> 25        -0.02362
               min(2)=        2 -> 17        -0.00763                  min(4)=        2 -> 11        -0.01857
               min(3)=        5 -> 13        -0.00665                  min(5)=        4 -> 11        -0.01815
               min(4)=        5 -> 25        -0.00662                  #CIs=36 #CIs>0=18 #CIs<0=18
               min(5)=        5 -> 21        -0.00489
               #CIs=26 #CIs>0=16 #CIs<0=10             51       Singlet-A2   42.6302         29.08    0.0000    0.000
                                                                        max(1)=        4 -> 20         0.70696
48       Singlet-B2   40.4690         30.64    0.0014    0.000          max(2)=        4 -> 29         0.00786
               max(1)=        2 -> 14         0.69266                  max(3)=        3 -> 22         0.00383
```

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              max(4)=        5 -> 27         0.00372                 min(1)=        4 -> 19        -0.32640
              max(5)=        2 -> 29         0.00263                 min(2)=        3 -> 18        -0.28620
              min(1)=        3 -> 17        -0.00923                 min(3)=        2 -> 15        -0.23623
              min(2)=        5 -> 18        -0.00446                 min(4)=        5 -> 22        -0.19379
              min(3)=        2 -> 20        -0.00364                 min(5)=        5 -> 17        -0.14837
              min(4)=        3 -> 28        -0.00089                 #CIs=43 #CIs>0=19 #CIs<0=24
              min(5)=        5 -> 11        -0.00020
              #CIs=19 #CIs>0=13 #CIs<0=6             55      Singlet-B2   47.0241         26.37    0.3558    0.000
                                                                max(1)=        3 -> 19         0.69344
52      Singlet-A1   42.7663         28.99    0.3607    0.000  max(2)=        3 -> 25         0.02654
              max(1)=        4 -> 19         0.56600                 max(3)=        2 -> 12         0.02362
              max(2)=        4 -> 21         0.13569                 max(4)=        5 -> 29         0.02103
              max(3)=        5 -> 22         0.12699                 max(5)=        4 -> 11         0.01362
              max(4)=        3 -> 14         0.05355                 min(1)=        5 -> 20        -0.11068
              max(5)=        4 -> 15         0.02704                 min(2)=        4 -> 24        -0.03698
              min(1)=        2 -> 15        -0.29279                 min(3)=        2 -> 18        -0.03028
              min(2)=        3 -> 18        -0.17426                 min(4)=        4 -> 14        -0.03028
              min(3)=        5 -> 17        -0.10626                 min(5)=        3 -> 15        -0.02499
              min(4)=        4 -> 16        -0.10144                 #CIs=35 #CIs>0=18 #CIs<0=17
              min(5)=        2 -> 13        -0.03243
              #CIs=43 #CIs>0=22 #CIs<0=21            56      Singlet-A1   47.2714         26.23    0.2965    0.000
                                                                max(1)=        5 -> 22         0.39363
53      Singlet-B1   43.7573         28.33    0.1145    0.000  max(2)=        2 -> 21         0.08596
              max(1)=        5 -> 21         0.69212                 max(3)=        2 ->  6         0.03560
              max(2)=        4 -> 22         0.05676                 max(4)=        4 ->  8         0.02839
              max(3)=        5 -> 19         0.01071                 max(5)=        2 -> 19         0.02639
              max(4)=        2 -> 10         0.00865                 min(1)=        4 -> 21        -0.37873
              max(5)=        5 ->  9         0.00562                 min(2)=        3 -> 18        -0.25834
              min(1)=        3 -> 20        -0.12990                 min(3)=        4 -> 19        -0.21687
              min(2)=        5 -> 16        -0.01665                 min(4)=        5 -> 17        -0.17170
              min(3)=        2 -> 17        -0.01244                 min(5)=        4 -> 16        -0.13721
              min(4)=        2 -> 22        -0.01132                 #CIs=43 #CIs>0=21 #CIs<0=22
              min(5)=        5 -> 13        -0.00623
              #CIs=25 #CIs>0=11 #CIs<0=14            57      Singlet-B1   47.3841         26.17    0.1902    0.000
                                                                max(1)=        3 -> 20         0.62026
54      Singlet-A1   44.5993         27.80    0.1259    0.000  max(2)=        4 -> 22         0.32582
              max(1)=        4 -> 21         0.41260                 max(3)=        5 -> 21         0.08981
              max(2)=        2 -> 21         0.04761                 max(4)=        5 -> 25         0.01873
              max(3)=        2 ->  6         0.03356                 max(5)=        2 -> 17         0.00921
              max(4)=        5 -> 28         0.02047                 min(1)=        5 -> 15        -0.01313
              max(5)=        4 -> 15         0.01940                 min(2)=        5 -> 19        -0.01227
```

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                min(3)=        5 -> 16        -0.01145                max(4)=        2 -> 12         0.01002
                min(4)=        4 -> 28        -0.01091                max(5)=        4 -> 18         0.00934
                min(5)=        2 -> 10        -0.00900                min(1)=        2 -> 18        -0.01892
                #CIs=26 #CIs>0=14 #CIs<0=12                           min(2)=        2 -> 14        -0.01674
                                                                      min(3)=        3 -> 13        -0.01336
58      Singlet-A1   47.9076         25.88    0.5313    0.000        min(4)=        4 -> 24        -0.00861
                max(1)=        5 -> 22         0.53441               min(5)=        4 -> 23        -0.00630
                max(2)=        4 -> 21         0.38508               #CIs=36 #CIs>0=18 #CIs<0=18
                max(3)=        4 -> 16         0.11178
                max(4)=        3 -> 18         0.11138   60      Singlet-B1   49.7636         24.91    1.0406    0.000
                max(5)=        5 -> 17         0.10545               max(1)=        4 -> 22         0.62129
                min(1)=        4 -> 19        -0.08636               max(2)=        5 -> 15         0.02381
                min(2)=        3 -> 14        -0.07030               max(3)=        5 -> 16         0.02359
                min(3)=        3 -> 24        -0.06036               max(4)=        5 -> 19         0.02200
                min(4)=        2 -> 21        -0.05737               max(5)=        2 -> 10         0.02083
                min(5)=        2 -> 19        -0.04030               min(1)=        3 -> 20        -0.30623
                #CIs=43 #CIs>0=23 #CIs<0=20                           min(2)=        5 -> 21        -0.10925
                                                                      min(3)=        5 -> 25        -0.05403
59      Singlet-B2   49.5809         25.01    0.0571    0.000        min(4)=        2 -> 17        -0.05347
                max(1)=        3 -> 21         0.70602               min(5)=        5 -> 31        -0.01513
                max(2)=        3 -> 15         0.01107               #CIs=26 #CIs>0=14 #CIs<0=12
                max(3)=        5 -> 20         0.01099
```

7.2.3 Main contributions from different excited states at CIS approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_   Singlet-A2   10.4507        118.64    0.0000    0.000
                                                                      max(1)=        5 ->  7         0.60811
1       Singlet-B1    8.7596        141.54    0.0459    0.000        max(2)=        5 -> 11         0.20221
                max(1)=        5 ->  6         0.62825               max(3)=        5 -> 17         0.04983
                max(2)=        5 -> 10         0.28394               max(4)=        5 -> 24         0.01332
                max(3)=        5 -> 16         0.03734               max(5)=        5 -> 35         0.00357
                max(4)=        5 -> 31         0.00598               min(1)=        5 -> 12        -0.27743
                max(5)=        5 -> 30         0.00246               min(2)=        5 -> 14        -0.09814
                min(1)=        5 -> 13        -0.11236               min(3)=        5 -> 32        -0.00334
                min(2)=        5 ->  8        -0.07639               min(4)=        3 -> 18        -0.00240
                min(3)=        5 -> 15        -0.06447               min(5)=        2 -> 29        -0.00173
                min(4)=        5 -> 21        -0.01729               #CIs=18 #CIs>0=9 #CIs<0=9
                min(5)=        4 ->  9        -0.01212
                #CIs=23 #CIs>0=8 #CIs<0=15             3       Singlet-A1   11.0021        112.69    0.1071    0.000
                                                                      max(1)=        4 ->  6         0.63565
```

```
              max(2)=        4 -> 10         0.25113                  max(4)=        5 -> 23         0.00384
              max(3)=        4 -> 16         0.01739                  max(5)=        2 -> 20         0.00373
              max(4)=        3 -> 14         0.01514                  min(1)=        5 ->  7        -0.30191
              max(5)=        3 -> 12         0.01127                  min(2)=        5 -> 14        -0.18242
              min(1)=        4 -> 13        -0.12227                  min(3)=        5 -> 12        -0.16008
              min(2)=        5 ->  9        -0.11528                  min(4)=        3 ->  9        -0.00832
              min(3)=        4 -> 15        -0.04943                  min(5)=        3 -> 18        -0.00221
              min(4)=        5 -> 18        -0.02191                  #CIs=17 #CIs>0=9 #CIs<0=8
              min(5)=        4 -> 21        -0.01857
              #CIs=40 #CIs>0=15 #CIs<0=25            7       Singlet-A1   13.5680         91.38    0.0000    0.000
                                                                      max(1)=        5 ->  9         0.64194
4       Singlet-B2   12.6684         97.87    0.0363    0.000          max(2)=        4 ->  8         0.21415
              max(1)=        4 ->  7         0.63875                  max(3)=        4 ->  6         0.13264
              max(2)=        4 -> 11         0.13686                  max(4)=        3 -> 12         0.07246
              max(3)=        4 -> 17         0.04438                  max(5)=        5 -> 18         0.04609
              max(4)=        3 ->  6         0.01718                  min(1)=        3 ->  7        -0.06950
              max(5)=        3 -> 15         0.01369                  min(2)=        4 -> 13        -0.05154
              min(1)=        4 -> 12        -0.25881                  min(3)=        4 -> 10        -0.05098
              min(2)=        4 -> 14        -0.05847                  min(4)=        3 -> 11        -0.04809
              min(3)=        2 -> 11        -0.01115                  min(5)=        4 -> 16        -0.04084
              min(4)=        2 ->  7        -0.00633                  #CIs=40 #CIs>0=19 #CIs<0=21
              min(5)=        3 -> 19        -0.00587
              #CIs=34 #CIs>0=22 #CIs<0=12            8       Singlet-B1   13.7253         90.33    0.0028    0.000
                                                                      max(1)=        5 -> 10         0.54474
5       Singlet-B1   12.8836         96.23    0.0149    0.000          max(2)=        5 ->  8         0.36545
              max(1)=        5 ->  8         0.58332                  max(3)=        4 ->  9         0.01396
              max(2)=        5 ->  6         0.20488                  max(4)=        2 ->  9         0.00619
              max(3)=        5 -> 15         0.07595                  max(5)=        4 -> 22         0.00579
              max(4)=        5 -> 25         0.00777                  min(1)=        5 ->  6        -0.22749
              max(5)=        3 -> 20         0.00309                  min(2)=        5 -> 15        -0.09445
              min(1)=        5 -> 10        -0.30898                  min(3)=        5 -> 13        -0.08680
              min(2)=        5 -> 13        -0.09950                  min(4)=        5 -> 21        -0.03115
              min(3)=        5 -> 16        -0.07870                  min(5)=        3 -> 20        -0.00753
              min(4)=        4 ->  9        -0.01301                  #CIs=23 #CIs>0=10 #CIs<0=13
              min(5)=        5 -> 19        -0.00819
              #CIs=23 #CIs>0=12 #CIs<0=11            9       Singlet-B2   14.7596         84.00    0.1622    0.000
                                                                      max(1)=        3 ->  6         0.63000
6       Singlet-A2   13.4466         92.20    0.0000    0.000          max(2)=        3 -> 10         0.27294
              max(1)=        5 -> 11         0.59031                  max(3)=        3 -> 16         0.03319
              max(2)=        5 -> 17         0.03236                  max(4)=        4 -> 14         0.03221
              max(3)=        5 -> 24         0.01770                  max(5)=        2 -> 12         0.01593
```

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              min(1)=        3 -> 13        -0.10156                 min(3)=        4 -> 10        -0.10060
              min(2)=        3 ->  8        -0.09205                 min(4)=        3 -> 14        -0.06389
              min(3)=        4 -> 11        -0.06495                 min(5)=        4 -> 13        -0.06292
              min(4)=        3 -> 15        -0.04971                 #CIs=41 #CIs>0=21 #CIs<0=20
              min(5)=        2 ->  7        -0.01485
              #CIs=32 #CIs>0=16 #CIs<0=16            13       Singlet-B1   15.7335         78.80    0.2358    0.000
                                                                     max(1)=        5 -> 13         0.66537
10       Singlet-B1   14.9661         82.84    0.0090    0.000     max(2)=        5 ->  8         0.11575
              max(1)=        4 ->  9         0.68307                 max(3)=        5 ->  6         0.09714
              max(2)=        5 -> 13         0.15723                 max(4)=        5 -> 10         0.06988
              max(3)=        4 -> 18         0.07561                 max(5)=        5 -> 21         0.02661
              max(4)=        5 ->  6         0.04301                 min(1)=        4 ->  9        -0.16392
              max(5)=        5 ->  8         0.02926                 min(2)=        5 -> 15        -0.02624
              min(1)=        5 -> 15        -0.00824                 min(3)=        4 -> 18        -0.01546
              min(2)=        5 -> 16        -0.00811                 min(4)=        3 -> 20        -0.00964
              min(3)=        2 -> 28        -0.00230                 min(5)=        5 -> 16        -0.00954
              min(4)=        2 -> 22        -0.00155                 #CIs=25 #CIs>0=14 #CIs<0=11
              min(5)=        5 -> 31        -0.00138
              #CIs=23 #CIs>0=15 #CIs<0=8             14       Singlet-A1   15.7372         78.78    0.0256    0.000
                                                                     max(1)=        4 -> 10         0.61650
11       Singlet-A2   15.4066         80.47    0.0000    0.000     max(2)=        4 ->  8         0.17460
              max(1)=        5 -> 12         0.61191                 max(3)=        3 -> 12         0.03133
              max(2)=        5 -> 11         0.28172                 max(4)=        5 ->  9         0.02985
              max(3)=        5 ->  7         0.19743                 max(5)=        3 -> 14         0.01904
              max(4)=        5 -> 14         0.03333                 min(1)=        4 ->  6        -0.25581
              max(5)=        5 -> 32         0.00278                 min(2)=        4 -> 15        -0.11425
              min(1)=        5 -> 17        -0.07776                 min(3)=        3 ->  7        -0.06418
              min(2)=        5 -> 24        -0.00666                 min(4)=        4 -> 13        -0.05068
              min(3)=        5 -> 35        -0.00234                 min(5)=        3 -> 11        -0.03187
              min(4)=        5 -> 27        -0.00122                 #CIs=40 #CIs>0=15 #CIs<0=25
              min(5)=        5 -> 23        -0.00080
              #CIs=19 #CIs>0=11 #CIs<0=8             15       Singlet-B2   15.9655         77.66    0.1960    0.000
                                                                     max(1)=        4 -> 11         0.63282
12       Singlet-A1   15.5618         79.67    0.0269    0.000     max(2)=        3 ->  6         0.07824
              max(1)=        4 ->  8         0.57809                 max(3)=        3 -> 15         0.03434
              max(2)=        3 ->  7         0.27203                 max(4)=        4 -> 17         0.03209
              max(3)=        3 -> 11         0.12373                 max(5)=        5 -> 20         0.01834
              max(4)=        3 -> 17         0.04115                 min(1)=        4 ->  7        -0.21697
              max(5)=        4 ->  6         0.02522                 min(2)=        4 -> 12        -0.15568
              min(1)=        3 -> 12        -0.17389                 min(3)=        4 -> 14        -0.13131
              min(2)=        5 ->  9        -0.15136                 min(4)=        3 -> 13        -0.02863
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                min(5)=        3 ->  8        -0.01830
                #CIs=34 #CIs>0=20 #CIs<0=14            19       Singlet-A2   19.1457         64.76    0.0000    0.000
                                                                       max(1)=        3 ->  9         0.70306
16       Singlet-B2   17.3383         71.51    0.0078    0.000          max(2)=        3 -> 18         0.07278
                max(1)=        4 -> 12         0.62464                 max(3)=        5 -> 14         0.01330
                max(2)=        4 -> 11         0.23694                 max(4)=        5 -> 11         0.01184
                max(3)=        4 ->  7         0.21147                 max(5)=        3 -> 22         0.00346
                max(4)=        4 -> 14         0.04462                 min(1)=        5 -> 12        -0.00607
                max(5)=        3 ->  6         0.01592                 min(2)=        5 -> 17        -0.00354
                min(1)=        4 -> 17        -0.07803                 min(3)=        2 -> 29        -0.00273
                min(2)=        3 -> 10        -0.01703                 min(4)=        5 ->  7        -0.00200
                min(3)=        4 -> 24        -0.00764                 min(5)=        5 -> 24        -0.00164
                min(4)=        3 ->  8        -0.00527                 #CIs=18 #CIs>0=11 #CIs<0=7
                min(5)=        2 ->  7        -0.00513
                #CIs=34 #CIs>0=17 #CIs<0=17            20       Singlet-B2   19.1730         64.67    0.0189    0.000
                                                                       max(1)=        3 ->  8         0.56403
17       Singlet-A1   17.5348         70.71    0.0339    0.000          max(2)=        3 ->  6         0.22703
                max(1)=        4 -> 13         0.40651                 max(3)=        3 -> 15         0.06856
                max(2)=        4 ->  8         0.24658                 max(4)=        2 -> 11         0.01950
                max(3)=        4 ->  6         0.07958                 max(5)=        3 -> 25         0.00519
                max(4)=        3 -> 12         0.06108                 min(1)=        3 -> 10        -0.33970
                max(5)=        4 -> 21         0.03034                 min(2)=        3 -> 16        -0.06965
                min(1)=        3 ->  7        -0.49006                 min(3)=        3 -> 13        -0.06051
                min(2)=        5 ->  9        -0.12969                 min(4)=        4 -> 11        -0.02918
                min(3)=        4 -> 10        -0.05437                 min(5)=        4 -> 14        -0.01775
                min(4)=        4 -> 15        -0.02547                 #CIs=33 #CIs>0=12 #CIs<0=21
                min(5)=        3 -> 14        -0.01832
                #CIs=41 #CIs>0=26 #CIs<0=15            21       Singlet-B2   19.9832         62.04    0.2440    0.000
                                                                       max(1)=        3 -> 10         0.53455
18       Singlet-A1   17.6965         70.06    0.0067    0.000          max(2)=        3 ->  8         0.40143
                max(1)=        4 -> 13         0.55292                 max(3)=        4 -> 11         0.05526
                max(2)=        3 ->  7         0.38995                 max(4)=        4 -> 14         0.03717
                max(3)=        4 -> 10         0.12599                 max(5)=        2 -> 12         0.02623
                max(4)=        5 ->  9         0.10510                 min(1)=        3 ->  6        -0.18860
                max(5)=        4 ->  6         0.07262                 min(2)=        3 -> 13        -0.07845
                min(1)=        4 ->  8        -0.07216                 min(3)=        3 -> 15        -0.07263
                min(2)=        5 -> 18        -0.02929                 min(4)=        3 -> 21        -0.01982
                min(3)=        4 -> 15        -0.02700                 min(5)=        3 -> 19        -0.01230
                min(4)=        4 -> 16        -0.02532                 #CIs=34 #CIs>0=19 #CIs<0=15
                min(5)=        3 -> 17        -0.02259
                #CIs=41 #CIs>0=14 #CIs<0=27            22       Singlet-A1   20.4429         60.65    0.2268    0.000
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              max(1)=        3 -> 11         0.68186                  max(3)=        5 -> 27         0.00309
              max(2)=        5 ->  9         0.07609                  max(4)=        2 -> 29         0.00193
              max(3)=        4 -> 10         0.05693                  max(5)=        3 -> 18         0.00032
              max(4)=        4 -> 15         0.02959                  min(1)=        5 -> 12        -0.12071
              max(5)=        2 ->  8         0.02204                  min(2)=        5 -> 24        -0.04651
              min(1)=        4 ->  8        -0.08372                  min(3)=        5 -> 17        -0.02464
              min(2)=        3 -> 14        -0.08321                  min(4)=        3 ->  9        -0.01695
              min(3)=        3 ->  7        -0.07509                  min(5)=        5 -> 23        -0.01273
              min(4)=        5 -> 18        -0.04105                  #CIs=17 #CIs>0=5 #CIs<0=12
              min(5)=        4 -> 16        -0.03452
              #CIs=38 #CIs>0=19 #CIs<0=19            26       Singlet-B1   24.5042         50.60    0.0603    0.000
                                                                      max(1)=        5 -> 15         0.68640
23       Singlet-B2   21.5936         57.42    0.0146    0.000         max(2)=        5 -> 10         0.14307
              max(1)=        3 -> 13         0.68816                  max(3)=        5 -> 25         0.03245
              max(2)=        3 ->  6         0.09417                  max(4)=        3 -> 20         0.02433
              max(3)=        3 ->  8         0.08007                  max(5)=        5 -> 21         0.01642
              max(4)=        3 -> 10         0.06493                  min(1)=        5 -> 16        -0.06138
              max(5)=        4 -> 14         0.05304                  min(2)=        5 -> 19        -0.02465
              min(1)=        3 -> 15        -0.02446                  min(3)=        5 ->  8        -0.02375
              min(2)=        4 -> 12        -0.01946                  min(4)=        2 ->  9        -0.01869
              min(3)=        3 -> 16        -0.01646                  min(5)=        4 -> 22        -0.01563
              min(4)=        2 ->  7        -0.01594                  #CIs=26 #CIs>0=14 #CIs<0=12
              min(5)=        2 -> 11        -0.01132
              #CIs=34 #CIs>0=21 #CIs<0=13            27       Singlet-B2   25.6603         48.32    0.1368    0.000
                                                                      max(1)=        4 -> 14         0.67300
24       Singlet-A1   22.3774         55.41    0.0038    0.000         max(2)=        4 -> 11         0.12166
              max(1)=        3 -> 12         0.66336                  max(3)=        3 -> 19         0.02661
              max(2)=        3 ->  7         0.13470                  max(4)=        2 -> 17         0.01065
              max(3)=        4 ->  8         0.09661                  max(5)=        2 -> 23         0.00927
              max(4)=        5 -> 18         0.07963                  min(1)=        4 -> 12        -0.09940
              max(5)=        4 -> 16         0.05954                  min(2)=        3 -> 15        -0.09057
              min(1)=        5 ->  9        -0.09606                  min(3)=        3 -> 10        -0.07164
              min(2)=        4 -> 10        -0.05992                  min(4)=        3 -> 13        -0.05417
              min(3)=        4 -> 15        -0.05554                  min(5)=        5 -> 20        -0.04144
              min(4)=        4 -> 13        -0.04185                  #CIs=35 #CIs>0=13 #CIs<0=22
              min(5)=        3 -> 17        -0.03172
              #CIs=38 #CIs>0=24 #CIs<0=14            28       Singlet-A1   26.6329         46.55    0.0014    0.000
                                                                      max(1)=        4 -> 15         0.68371
25       Singlet-A2   22.6302         54.79    0.0000    0.000         max(2)=        4 -> 10         0.12677
              max(1)=        5 -> 14         0.67160                  max(3)=        3 -> 12         0.06187
              max(2)=        5 -> 11         0.17631                  max(4)=        5 -> 18         0.05178
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                max(5)=        4 -> 25         0.03802                 min(2)=        2 ->  8        -0.07473
                min(1)=        3 -> 14        -0.05265                 min(3)=        2 -> 15        -0.05155
                min(2)=        3 -> 11        -0.04027                 min(4)=        5 ->  9        -0.01481
                min(3)=        5 ->  9        -0.02890                 min(5)=        3 -> 12        -0.00787
                min(4)=        2 ->  6        -0.02531                 #CIs=40 #CIs>0=25 #CIs<0=15
                min(5)=        2 -> 10        -0.02402
                #CIs=41 #CIs>0=25 #CIs<0=16             32       Singlet-B1   33.7190         36.77    0.0231    0.000
                                                                        max(1)=        5 -> 16         0.69503
29       Singlet-A1   29.8738         41.50    0.1879    0.000          max(2)=        5 ->  8         0.07297
                max(1)=        3 -> 14         0.67362                 max(3)=        5 -> 15         0.06835
                max(2)=        5 -> 18         0.10998                 max(4)=        5 -> 21         0.04478
                max(3)=        3 -> 11         0.10357                 max(5)=        3 -> 20         0.01845
                max(4)=        4 -> 16         0.08018                 min(1)=        4 -> 18        -0.03435
                max(5)=        4 -> 15         0.05176                 min(2)=        5 -> 10        -0.03166
                min(1)=        3 -> 12        -0.05647                 min(3)=        5 -> 19        -0.02252
                min(2)=        4 -> 19        -0.04197                 min(4)=        2 ->  9        -0.02200
                min(3)=        2 ->  6        -0.03766                 min(5)=        5 -> 25        -0.01923
                min(4)=        5 ->  9        -0.03354                 #CIs=25 #CIs>0=13 #CIs<0=12
                min(5)=        3 -> 24        -0.02298
                #CIs=41 #CIs>0=20 #CIs<0=21             33       Singlet-B2   33.7757         36.71    0.0051    0.000
                                                                        max(1)=        2 ->  7         0.62408
30       Singlet-B2   31.0714         39.90    0.3134    0.000          max(2)=        2 -> 11         0.19587
                max(1)=        3 -> 15         0.67968                 max(3)=        5 -> 20         0.03739
                max(2)=        3 -> 10         0.11095                 max(4)=        2 -> 17         0.03603
                max(3)=        4 -> 14         0.09533                 max(5)=        4 -> 17         0.02389
                max(4)=        2 -> 11         0.03544                 min(1)=        2 -> 12        -0.24456
                max(5)=        3 -> 25         0.02414                 min(2)=        2 -> 14        -0.07528
                min(1)=        5 -> 20        -0.06814                 min(3)=        3 -> 15        -0.03298
                min(2)=        3 -> 16        -0.06393                 min(4)=        4 -> 14        -0.01900
                min(3)=        4 -> 17        -0.04064                 min(5)=        3 -> 19        -0.01492
                min(4)=        2 -> 14        -0.03912                 #CIs=33 #CIs>0=20 #CIs<0=13
                min(5)=        2 -> 12        -0.02993
                #CIs=35 #CIs>0=14 #CIs<0=21             34       Singlet-A1   35.2181         35.20    0.0045    0.000
                                                                        max(1)=        5 -> 18         0.50657
31       Singlet-A1   32.0963         38.63    0.0133    0.000          max(2)=        2 -> 10         0.06044
                max(1)=        2 ->  6         0.63250                 max(3)=        2 ->  8         0.02961
                max(2)=        2 -> 10         0.25262                 max(4)=        4 -> 10         0.02261
                max(3)=        5 -> 18         0.11731                 max(5)=        5 -> 22         0.01503
                max(4)=        4 -> 16         0.05076                 min(1)=        4 -> 16        -0.46822
                max(5)=        4 -> 15         0.02899                 min(2)=        2 ->  6        -0.07725
                min(1)=        2 -> 13        -0.09101                 min(3)=        4 ->  8        -0.05520
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                min(4)=        5 ->  9        -0.04700                #CIs=41 #CIs>0=17 #CIs<0=24
                min(5)=        4 -> 15        -0.04417
                #CIs=41 #CIs>0=15 #CIs<0=26            38       Singlet-B2   36.7336         33.75    0.0025    0.000
                                                                      max(1)=        2 -> 11         0.60576
35       Singlet-A2   35.2394         35.18    0.0000    0.000      max(2)=        5 -> 20         0.03480
                max(1)=        5 -> 17         0.69875              max(3)=        2 -> 17         0.02424
                max(2)=        5 -> 12         0.09127              max(4)=        4 -> 17         0.01709
                max(3)=        5 -> 14         0.04491              max(5)=        2 -> 24         0.01095
                max(4)=        5 -> 24         0.02428              min(1)=        2 ->  7        -0.27770
                max(5)=        5 -> 35         0.01103              min(2)=        2 -> 14        -0.16780
                min(1)=        5 -> 32        -0.02214              min(3)=        2 -> 12        -0.15289
                min(2)=        5 ->  7        -0.00764              min(4)=        3 -> 15        -0.03692
                min(3)=        5 -> 23        -0.00494              min(5)=        4 -> 14        -0.01345
                min(4)=        5 -> 11        -0.00440              #CIs=34 #CIs>0=18 #CIs<0=16
                min(5)=        3 -> 18        -0.00405
                #CIs=19 #CIs>0=9 #CIs<0=10             39       Singlet-B1   36.7426         33.74    0.0034    0.000
                                                                      max(1)=        4 -> 18         0.70094
36       Singlet-B1   36.3011         34.15    0.0120    0.000      max(2)=        5 -> 16         0.03300
                max(1)=        2 ->  9         0.70076              max(3)=        5 -> 21         0.02603
                max(2)=        2 -> 18         0.07258              max(4)=        4 -> 33         0.02090
                max(3)=        3 -> 20         0.02840              max(5)=        4 -> 22         0.00959
                max(4)=        5 -> 15         0.02030              min(1)=        4 ->  9        -0.07683
                max(5)=        5 -> 16         0.01944              min(2)=        2 ->  9        -0.01632
                min(1)=        5 -> 19        -0.02955              min(3)=        2 -> 22        -0.00614
                min(2)=        5 -> 13        -0.01969              min(4)=        2 -> 28        -0.00459
                min(3)=        4 -> 22        -0.01891              min(5)=        5 -> 25        -0.00272
                min(4)=        5 -> 25        -0.00675              #CIs=25 #CIs>0=15 #CIs<0=10
                min(5)=        5 -> 26        -0.00375
                #CIs=25 #CIs>0=15 #CIs<0=10             40       Singlet-A1   36.8413         33.65    0.0009    0.000
                                                                      max(1)=        2 -> 10         0.48932
37       Singlet-A1   36.3670         34.09    0.0146    0.000      max(2)=        2 ->  8         0.42051
                max(1)=        2 ->  8         0.54910              max(3)=        4 -> 16         0.18502
                max(2)=        2 ->  6         0.21403              max(4)=        5 -> 18         0.05012
                max(3)=        2 -> 15         0.07624              max(5)=        4 ->  8         0.01275
                max(4)=        3 -> 14         0.02803              min(1)=        2 ->  6        -0.17958
                max(5)=        5 -> 22         0.02250              min(2)=        2 -> 15        -0.10338
                min(1)=        2 -> 10        -0.35381              min(3)=        3 -> 17        -0.03379
                min(2)=        2 -> 13        -0.08652              min(4)=        3 -> 12        -0.02664
                min(3)=        4 -> 16        -0.06861              min(5)=        3 -> 14        -0.02294
                min(4)=        2 -> 16        -0.06769              #CIs=41 #CIs>0=20 #CIs<0=21
                min(5)=        4 -> 19        -0.03761
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41       Singlet-B2   37.3220         33.22    0.0788    0.000          max(2)=        2 -> 10         0.17107
               max(1)=        4 -> 17         0.69084                  max(3)=        3 -> 14         0.11480
               max(2)=        4 -> 12         0.08905                  max(4)=        3 -> 12         0.08843
               max(3)=        4 -> 14         0.04788                  max(5)=        2 -> 15         0.05726
               max(4)=        3 -> 15         0.02994                  min(1)=        4 -> 16        -0.33921
               max(5)=        4 -> 24         0.02886                  min(2)=        5 -> 18        -0.28845
               min(1)=        2 -> 12        -0.06707                  min(3)=        2 -> 13        -0.19975
               min(2)=        2 -> 11        -0.04041                  min(4)=        2 ->  6        -0.02528
               min(3)=        2 ->  7        -0.03286                  min(5)=        4 -> 19        -0.02193
               min(4)=        5 -> 20        -0.03012                  #CIs=43 #CIs>0=20 #CIs<0=23
               min(5)=        3 -> 16        -0.02383
               #CIs=34 #CIs>0=18 #CIs<0=16            45       Singlet-B2   40.2449         30.81    0.0363    0.000
                                                                       max(1)=        3 -> 16         0.67925
42       Singlet-A1   38.6380         32.09    0.0004    0.000          max(2)=        2 -> 12         0.08501
               max(1)=        2 -> 13         0.65792                  max(3)=        3 -> 15         0.06213
               max(2)=        2 ->  6         0.10789                  max(4)=        3 ->  8         0.06127
               max(3)=        2 ->  8         0.08301                  max(5)=        2 -> 14         0.04825
               max(4)=        3 -> 17         0.08141                  min(1)=        5 -> 20        -0.12247
               max(5)=        2 -> 10         0.06139                  min(2)=        3 -> 10        -0.03104
               min(1)=        4 -> 16        -0.14605                  min(3)=        3 -> 19        -0.02494
               min(2)=        5 -> 18        -0.11390                  min(4)=        3 -> 25        -0.01723
               min(3)=        2 -> 15        -0.03051                  min(5)=        2 -> 23        -0.01113
               min(4)=        2 -> 16        -0.01659                  #CIs=33 #CIs>0=22 #CIs<0=11
               min(5)=        5 -> 22        -0.00695
               #CIs=40 #CIs>0=22 #CIs<0=18            46       Singlet-A2   41.1640         30.12    0.0000    0.000
                                                                       max(1)=        3 -> 18         0.70202
43       Singlet-B2   38.7323         32.01    0.0186    0.000          max(2)=        3 -> 22         0.02970
               max(1)=        2 -> 12         0.61562                  max(3)=        3 -> 33         0.01811
               max(2)=        2 -> 11         0.24977                  max(4)=        2 -> 20         0.01617
               max(3)=        2 ->  7         0.17607                  max(5)=        5 -> 17         0.00505
               max(4)=        4 -> 17         0.07587                  min(1)=        3 ->  9        -0.07295
               max(5)=        2 -> 14         0.03229                  min(2)=        4 -> 20        -0.01673
               min(1)=        3 -> 16        -0.10873                  min(3)=        2 -> 29        -0.00612
               min(2)=        2 -> 17        -0.07037                  min(4)=        5 -> 14        -0.00225
               min(3)=        5 -> 20        -0.04642                  min(5)=        5 -> 24        -0.00196
               min(4)=        3 ->  8        -0.02409                  #CIs=19 #CIs>0=11 #CIs<0=8
               min(5)=        3 -> 10        -0.01045
               #CIs=35 #CIs>0=18 #CIs<0=17            47       Singlet-B1   41.8180         29.65    0.0008    0.000
                                                                       max(1)=        5 -> 19         0.70008
44       Singlet-A1   39.7527         31.19    0.0544    0.000          max(2)=        5 -> 21         0.07691
               max(1)=        3 -> 17         0.44723                  max(3)=        2 ->  9         0.02496
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              max(4)=        5 -> 30         0.02382                  min(1)=        2 -> 20        -0.02901
              max(5)=        5 -> 15         0.02338                  min(2)=        3 -> 22        -0.01984
              min(1)=        5 -> 13        -0.02516                  min(3)=        5 -> 17        -0.00404
              min(2)=        4 -> 22        -0.01895                  min(4)=        3 -> 28        -0.00155
              min(3)=        5 -> 31        -0.00968                  min(5)=        5 -> 12        -0.00091
              min(4)=        4 -> 18        -0.00468                  #CIs=19 #CIs>0=12 #CIs<0=7
              min(5)=        2 -> 28        -0.00075
              #CIs=26 #CIs>0=19 #CIs<0=7              51       Singlet-B1   44.9045         27.61    0.1799    0.000
                                                                      max(1)=        5 -> 21         0.68550
48       Singlet-B2   43.8497         28.27    0.6294    0.000      max(2)=        4 -> 22         0.05002
              max(1)=        5 -> 20         0.67056                  max(3)=        2 -> 18         0.03913
              max(2)=        3 -> 19         0.14827                  max(4)=        5 -> 10         0.02225
              max(3)=        3 -> 16         0.10714                  max(5)=        5 -> 26         0.01838
              max(4)=        3 -> 15         0.07597                  min(1)=        3 -> 20        -0.12507
              max(5)=        2 -> 12         0.06606                  min(2)=        5 -> 19        -0.07038
              min(1)=        3 -> 25        -0.02200                  min(3)=        5 -> 16        -0.04001
              min(2)=        2 -> 11        -0.01317                  min(4)=        5 -> 13        -0.03376
              min(3)=        4 -> 11        -0.01310                  min(5)=        4 -> 18        -0.02401
              min(4)=        3 -> 10        -0.01190                  #CIs=26 #CIs>0=11 #CIs<0=15
              min(5)=        2 -> 24        -0.01105
              #CIs=35 #CIs>0=23 #CIs<0=12             52       Singlet-B2   45.9796         26.97    0.0095    0.000
                                                                      max(1)=        2 -> 14         0.67546
49       Singlet-A1   44.4780         27.88    0.3529    0.000      max(2)=        2 -> 11         0.16138
              max(1)=        4 -> 19         0.54294                  max(3)=        3 -> 15         0.01766
              max(2)=        4 -> 21         0.28533                  max(4)=        4 -> 23         0.01378
              max(3)=        5 -> 22         0.10177                  max(5)=        3 -> 10         0.01057
              max(4)=        2 -> 15         0.05775                  min(1)=        2 -> 12        -0.10786
              max(5)=        3 -> 14         0.04971                  min(2)=        3 -> 16        -0.04498
              min(1)=        3 -> 17        -0.25462                  min(3)=        2 -> 24        -0.03800
              min(2)=        4 -> 16        -0.13944                  min(4)=        2 -> 17        -0.02782
              min(3)=        5 -> 18        -0.12350                  min(5)=        4 -> 17        -0.02396
              min(4)=        4 -> 13        -0.03786                  #CIs=35 #CIs>0=17 #CIs<0=18
              min(5)=        2 -> 13        -0.03007
              #CIs=43 #CIs>0=24 #CIs<0=19             53       Singlet-A1   46.2190         26.83    0.2598    0.000
                                                                      max(1)=        2 -> 15         0.35197
50       Singlet-A2   44.8370         27.65    0.0000    0.000      max(2)=        4 -> 21         0.26330
              max(1)=        4 -> 20         0.70534                  max(3)=        2 -> 10         0.14142
              max(2)=        4 -> 29         0.02893                  max(4)=        2 -> 25         0.02856
              max(3)=        3 -> 18         0.01829                  max(5)=        5 -> 28         0.02179
              max(4)=        2 -> 29         0.00567                  min(1)=        4 -> 19        -0.36965
              max(5)=        5 -> 27         0.00497                  min(2)=        3 -> 17        -0.30946
```

```
              min(3)=        5 -> 22        -0.13103                  min(5)=        5 -> 16        -0.01731
              min(4)=        4 -> 16        -0.12079                  #CIs=25 #CIs>0=16 #CIs<0=9
              min(5)=        5 -> 18        -0.12025
              #CIs=43 #CIs>0=15 #CIs<0=28            57       Singlet-A1   50.0322         24.78    0.4859    0.000
                                                                max(1)=        5 -> 22         0.46561
54       Singlet-A1   47.2925         26.22    0.0000    0.000 max(2)=        2 -> 21         0.04067
              max(1)=        2 -> 15         0.46747           max(3)=        3 -> 12         0.02742
              max(2)=        5 -> 22         0.23390           max(4)=        2 -> 10         0.02302
              max(3)=        4 -> 19         0.11667           max(5)=        4 ->  8         0.02256
              max(4)=        2 -> 10         0.09411           min(1)=        2 -> 15        -0.31405
              max(5)=        2 -> 25         0.02660           min(2)=        3 -> 17        -0.25054
              min(1)=        4 -> 21        -0.44386           min(3)=        4 -> 19        -0.19490
              min(2)=        3 -> 17        -0.04212           min(4)=        5 -> 18        -0.18599
              min(3)=        2 -> 16        -0.03702           min(5)=        4 -> 16        -0.13515
              min(4)=        5 -> 28        -0.02029           #CIs=41 #CIs>0=18 #CIs<0=23
              min(5)=        2 -> 19        -0.02012
              #CIs=41 #CIs>0=21 #CIs<0=20            58       Singlet-A1   50.7723         24.42    0.5155    0.000
                                                                max(1)=        5 -> 22         0.43907
55       Singlet-B2   48.8593         25.38    0.5023    0.000 max(2)=        4 -> 21         0.35649
              max(1)=        3 -> 19         0.68478           max(3)=        3 -> 17         0.22464
              max(2)=        2 -> 23         0.04075           max(4)=        5 -> 18         0.17803
              max(3)=        3 -> 25         0.03362           max(5)=        4 -> 16         0.17007
              max(4)=        5 -> 29         0.01928           min(1)=        3 -> 14        -0.08888
              max(5)=        4 -> 11         0.01174           min(2)=        4 -> 19        -0.07601
              min(1)=        5 -> 20        -0.13906           min(3)=        3 -> 24        -0.06889
              min(2)=        4 -> 14        -0.04404           min(4)=        2 -> 21        -0.06757
              min(3)=        2 -> 12        -0.03880           min(5)=        4 -> 15        -0.04139
              min(4)=        4 -> 24        -0.03616           #CIs=42 #CIs>0=21 #CIs<0=21
              min(5)=        4 -> 23        -0.03296
              #CIs=36 #CIs>0=18 #CIs<0=18            59       Singlet-B2   51.1934         24.22    0.0677    0.000
                                                                max(1)=        3 -> 21         0.70073
56       Singlet-B1   49.3378         25.13    0.4700    0.000 max(2)=        2 -> 17         0.07312
              max(1)=        3 -> 20         0.66290           max(3)=        3 -> 19         0.02049
              max(2)=        4 -> 22         0.20740           max(4)=        3 -> 26         0.01697
              max(3)=        5 -> 21         0.09872           max(5)=        3 -> 10         0.01441
              max(4)=        2 -> 18         0.04911           min(1)=        3 -> 13        -0.03992
              max(5)=        2 -> 22         0.03564           min(2)=        3 -> 16        -0.01375
              min(1)=        5 -> 15        -0.02663           min(3)=        4 -> 24        -0.01175
              min(2)=        2 ->  9        -0.02621           min(4)=        3 -> 30        -0.01132
              min(3)=        5 -> 19        -0.02558           min(5)=        2 -> 14        -0.00923
              min(4)=        4 -> 28        -0.02424           #CIs=34 #CIs>0=18 #CIs<0=16
```

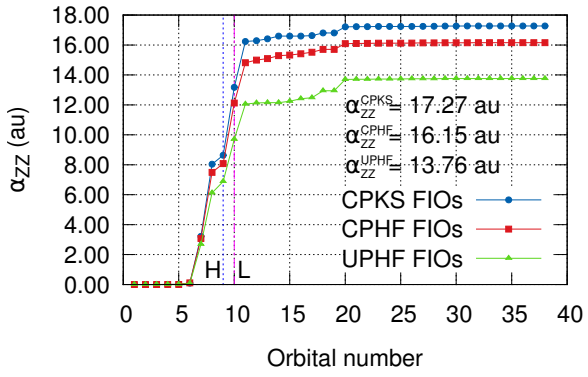
						min(1)=	3 -> 20	-0.18318
60	Singlet-B1	52.1627	23.77	1.0346	0.000	min(2)=	2 -> 18	-0.09098
	max(1)=	4 -> 22	0.66714			min(3)=	5 -> 21	-0.07206
	max(2)=	2 -> 9	0.03462			min(4)=	5 -> 25	-0.05107
	max(3)=	5 -> 19	0.03070			min(5)=	2 -> 22	-0.02926
	max(4)=	5 -> 15	0.03017			#CIs=25 #CIs>0=10 #CIs<0=15		
	max(5)=	5 -> 16	0.02095					

8 H_2S

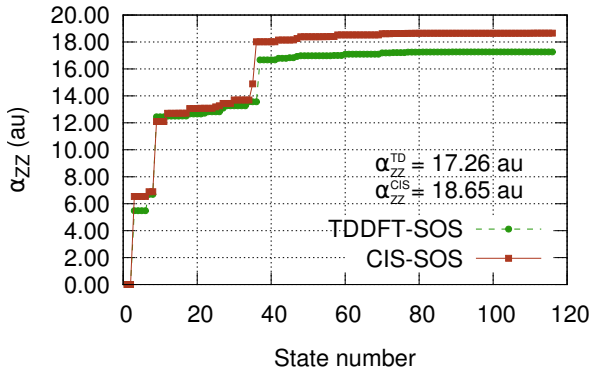
8.1 6-311G(d,p)

8.1.1 Plots

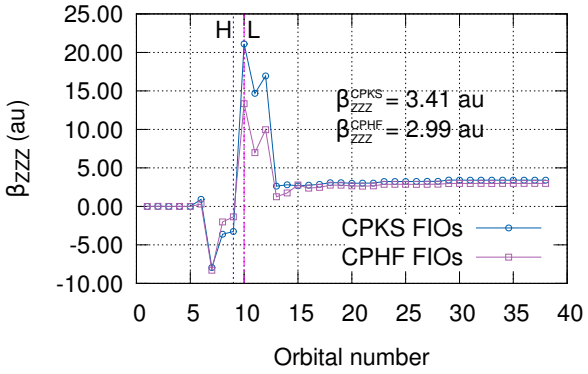
Figure S11: For H_2S molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S11a and S11c) or states (SOS approaches, in Plots S11b and S11d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.02 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



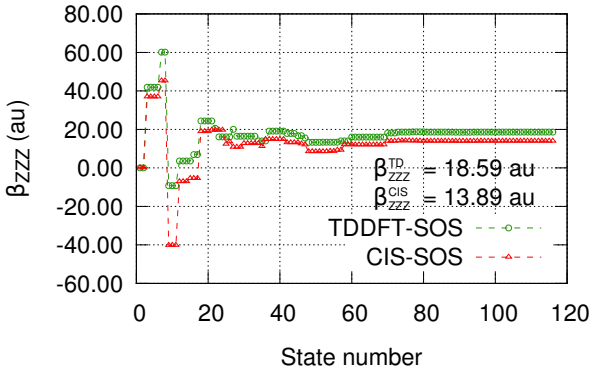
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

8.1.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_          min(4)=        8 -> 17        -0.01946
                                                                        min(5)=        6 -> 13        -0.00962
1        Singlet-A2    6.1367        202.04    0.0000    0.000          #states=38 #states>0=18 #states<0=20
               max(1)=        9 -> 11         0.70435
               max(2)=        9 -> 12         0.01945   4        Singlet-A2    9.9884        124.13    0.0000    0.000
               max(3)=        6 -> 19         0.00681                max(1)=        9 -> 12         0.70624
               max(4)=        8 -> 19         0.00487                max(2)=        9 -> 22         0.01446
               max(5)=        9 -> 27         0.00433                max(3)=        9 -> 27         0.00631
               min(1)=        9 -> 16        -0.06033                max(4)=        6 -> 19         0.00130
               min(2)=        9 -> 22        -0.00621                max(5)=        9 -> 33         0.00092
               min(3)=        7 -> 14        -0.00595                min(1)=        9 -> 16        -0.02251
               min(4)=        8 -> 24        -0.00203                min(2)=        9 -> 11        -0.02132
               min(5)=        9 -> 23        -0.00147                min(3)=        9 -> 23        -0.00530
               #states=18 #states>0=7 #states<0=11                    min(4)=        7 -> 14        -0.00178
                                                                      min(5)=        9 -> 31        -0.00140
2        Singlet-B1    6.6231        187.20    0.0300    0.000          #states=18 #states>0=8 #states<0=10
               max(1)=        9 -> 10         0.70574
               max(2)=        9 -> 13         0.03158   5        Singlet-B2   10.0186        123.75    0.1793    0.000
               max(3)=        9 -> 21         0.01189                max(1)=        8 -> 11         0.68078
               max(4)=        6 -> 20         0.00537                max(2)=        7 -> 15         0.06911
               max(5)=        6 -> 14         0.00458                max(3)=        9 -> 19         0.05056
               min(1)=        9 -> 17        -0.02781                max(4)=        8 -> 12         0.01396
               min(2)=        9 -> 15        -0.02244                max(5)=        7 -> 13         0.01266
               min(3)=        7 -> 19        -0.01261                min(1)=        7 -> 10        -0.17246
               min(4)=        8 -> 20        -0.01088                min(2)=        7 -> 18        -0.02942
               min(5)=        6 -> 25        -0.00237                min(3)=        6 -> 12        -0.01983
               #states=24 #states>0=15 #states<0=9                    min(4)=        6 -> 11        -0.01752
                                                                      min(5)=        8 -> 16        -0.00745
3        Singlet-A1    9.6042        129.09    0.2275    0.000          #states=32 #states>0=13 #states<0=19
               max(1)=        8 -> 10         0.69458
               max(2)=        7 -> 16         0.02000   6        Singlet-B1   10.4787        118.32    0.0018    0.000
               max(3)=        8 -> 18         0.00994                max(1)=        9 -> 13         0.70295
               max(4)=        6 -> 10         0.00497                max(2)=        8 -> 20         0.01254
               max(5)=        9 -> 20         0.00479                max(3)=        9 -> 26         0.00918
               min(1)=        7 -> 11        -0.11848                max(4)=        7 -> 19         0.00867
               min(2)=        9 -> 14        -0.04780                max(5)=        9 -> 17         0.00596
               min(3)=        8 -> 15        -0.02088                min(1)=        9 -> 15        -0.06523
```

```
                min(2)=        9 -> 10        -0.03329                min(4)=        8 -> 26        -0.01105
                min(3)=        8 -> 14        -0.01451                min(5)=        6 -> 18        -0.00936
                min(4)=        6 -> 14        -0.00557                #states=37 #states>0=18 #states<0=19
                min(5)=        9 -> 18        -0.00470
                #states=24 #states>0=9 #states<0=15     10       Singlet-B2   12.8738         96.31    0.0017    0.000
                                                                       max(1)=        8 -> 12         0.69998
7       Singlet-A1   11.6289        106.62    0.0737    0.000          max(2)=        8 -> 22         0.01414
                max(1)=        9 -> 14         0.61895                 max(3)=        8 -> 27         0.00623
                max(2)=        6 -> 10         0.03172                 max(4)=        6 -> 12         0.00507
                max(3)=        7 -> 16         0.00892                 max(5)=        7 -> 30         0.00196
                max(4)=        6 -> 13         0.00629                 min(1)=        7 -> 10        -0.07937
                max(5)=        7 -> 23         0.00425                 min(2)=        8 -> 16        -0.03786
                min(1)=        7 -> 11        -0.27824                 min(3)=        8 -> 11        -0.03488
                min(2)=        8 -> 13        -0.16962                 min(4)=        6 -> 11        -0.02195
                min(3)=        7 -> 12        -0.09646                 min(5)=        7 -> 13        -0.01772
                min(4)=        9 -> 20        -0.01636                 #states=32 #states>0=14 #states<0=18
                min(5)=        8 -> 18        -0.01285
                #states=38 #states>0=17 #states<0=21     11       Singlet-B1   13.8437         89.56    0.1843    0.000
                                                                       max(1)=        8 -> 14         0.53160
8       Singlet-B2   12.0005        103.32    0.6273    0.000          max(2)=        6 -> 14         0.01835
                max(1)=        7 -> 10         0.67641                 max(3)=        9 -> 18         0.01185
                max(2)=        8 -> 11         0.16647                 max(4)=        6 -> 20         0.00908
                max(3)=        8 -> 12         0.08854                 max(5)=        9 -> 21         0.00544
                max(4)=        7 -> 13         0.07661                 min(1)=        9 -> 15        -0.46366
                max(5)=        8 -> 16         0.02615                 min(2)=        9 -> 13        -0.03214
                min(1)=        6 -> 12        -0.02424                 min(3)=        7 -> 19        -0.02046
                min(2)=        7 -> 18        -0.01118                 min(4)=        8 -> 20        -0.01625
                min(3)=        6 -> 16        -0.01006                 min(5)=        9 -> 10        -0.01399
                min(4)=        7 -> 17        -0.00643                 #states=24 #states>0=15 #states<0=9
                min(5)=        7 -> 21        -0.00473
                #states=31 #states>0=15 #states<0=16     12       Singlet-A1   13.9451         88.91    0.0047    0.000
                                                                       max(1)=        8 -> 13         0.59095
9       Singlet-A1   12.5786         98.57    0.4102    0.000          max(2)=        9 -> 14         0.19051
                max(1)=        7 -> 11         0.60443                 max(3)=        7 -> 11         0.15729
                max(2)=        9 -> 14         0.20389                 max(4)=        8 -> 15         0.10214
                max(3)=        8 -> 10         0.11705                 max(5)=        8 -> 10         0.04891
                max(4)=        8 -> 15         0.10692                 min(1)=        7 -> 12        -0.27337
                max(5)=        9 -> 20         0.03894                 min(2)=        8 -> 17        -0.01889
                min(1)=        8 -> 13        -0.25663                 min(3)=        6 -> 17        -0.01885
                min(2)=        6 -> 13        -0.02392                 min(4)=        7 -> 22        -0.01405
                min(3)=        6 -> 17        -0.01535                 min(5)=        7 -> 27        -0.00716
```

```
                #states=38 #states>0=20 #states<0=18     16       Singlet-A1   16.0085         77.45    0.0009    0.000

                                                                        max(1)=        7 -> 12         0.57083
13       Singlet-B1   14.1832         87.42    0.2250    0.000          max(2)=        8 -> 15         0.35091
                max(1)=        9 -> 15         0.52432                 max(3)=        8 -> 13         0.17185
                max(2)=        8 -> 14         0.46534                 max(4)=        9 -> 14         0.12202
                max(3)=        9 -> 13         0.05775                 max(5)=        6 -> 10         0.04200
                max(4)=        8 -> 20         0.04483                 min(1)=        8 -> 18        -0.03258
                max(5)=        7 -> 19         0.04155                 min(2)=        9 -> 20        -0.03232
                min(1)=        6 -> 14        -0.02960                 min(3)=        8 -> 17        -0.03155
                min(2)=        9 -> 21        -0.02388                 min(4)=        7 -> 11        -0.03142
                min(3)=        9 -> 18        -0.01792                 min(5)=        7 -> 16        -0.02785
                min(4)=        6 -> 20        -0.00930                 #states=38 #states>0=20 #states<0=18
                min(5)=        9 -> 30        -0.00580
                #states=24 #states>0=11 #states<0=13     17       Singlet-A2   16.2374         76.36    0.0000    0.000

                                                                        max(1)=        7 -> 14         0.70622
14       Singlet-A2   15.1065         82.07    0.0000    0.000          max(2)=        8 -> 19         0.00674
                max(1)=        9 -> 16         0.70316                 max(3)=        7 -> 28         0.00559
                max(2)=        9 -> 11         0.05995                 max(4)=        9 -> 11         0.00308
                max(3)=        7 -> 14         0.03194                 max(5)=        6 -> 19         0.00217
                max(4)=        9 -> 12         0.02405                 min(1)=        9 -> 16        -0.03229
                max(5)=        9 -> 22         0.01650                 min(2)=        7 -> 25        -0.01078
                min(1)=        9 -> 27        -0.00944                 min(3)=        8 -> 24        -0.00266
                min(2)=        6 -> 19        -0.00574                 min(4)=        9 -> 22        -0.00226
                min(3)=        8 -> 19        -0.00331                 min(5)=        9 -> 31        -0.00055
                min(4)=        9 -> 31        -0.00254                 #states=18 #states>0=11 #states<0=7
                min(5)=        6 -> 24        -0.00074
                #states=18 #states>0=13 #states<0=5      18       Singlet-A1   17.0906         72.55    0.0194    0.000

                                                                        max(1)=        8 -> 15         0.58020
15       Singlet-B2   15.7316         78.81    0.0116    0.000          max(2)=        6 -> 17         0.04129
                max(1)=        7 -> 13         0.69939                 max(3)=        8 -> 21         0.02503
                max(2)=        6 -> 12         0.01545                 max(4)=        6 -> 15         0.01573
                max(3)=        7 -> 18         0.01182                 max(5)=        9 -> 28         0.01180
                max(4)=        7 -> 21         0.00913                 min(1)=        7 -> 12        -0.29133
                max(5)=        7 -> 26         0.00771                 min(2)=        8 -> 13        -0.15155
                min(1)=        7 -> 10        -0.07038                 min(3)=        9 -> 14        -0.12753
                min(2)=        7 -> 15        -0.04752                 min(4)=        7 -> 11        -0.11240
                min(3)=        6 -> 11        -0.03661                 min(5)=        6 -> 10        -0.11115
                min(4)=        8 -> 16        -0.02901                 #states=38 #states>0=15 #states<0=23
                min(5)=        8 -> 11        -0.02597
                #states=30 #states>0=15 #states<0=15     19       Singlet-B2   18.0971         68.51    0.0232    0.000

                                                                        max(1)=        8 -> 16         0.61322
```

```
              max(2)=        6 -> 11         0.04170                  max(4)=        8 -> 21         0.01715
              max(3)=        8 -> 11         0.03744                  max(5)=        7 -> 22         0.01384
              max(4)=        8 -> 12         0.03230                  min(1)=        7 -> 16        -0.47757
              max(5)=        7 -> 17         0.01941                  min(2)=        7 -> 12        -0.03569
              min(1)=        7 -> 15        -0.34197                  min(3)=        9 -> 14        -0.03327
              min(2)=        9 -> 19        -0.03225                  min(4)=        6 -> 15        -0.02372
              min(3)=        7 -> 10        -0.02769                  min(5)=        8 -> 13        -0.01713
              min(4)=        6 -> 16        -0.01199                  #states=38 #states>0=15 #states<0=23
              min(5)=        7 -> 21        -0.01034
              #states=31 #states>0=23 #states<0=8     23       Singlet-A1   21.2525         58.34    0.0214    0.000
                                                                      max(1)=        7 -> 16         0.49341
20       Singlet-B2   19.5598         63.39    0.0830    0.000          max(2)=        6 -> 10         0.47400
              max(1)=        7 -> 15         0.58455                  max(3)=        8 -> 15         0.10782
              max(2)=        8 -> 16         0.32830                  max(4)=        7 -> 22         0.04069
              max(3)=        7 -> 18         0.05383                  max(5)=        8 -> 21         0.03142
              max(4)=        7 -> 13         0.03794                  min(1)=        9 -> 14        -0.07346
              max(5)=        6 -> 12         0.03001                  min(2)=        9 -> 20        -0.05997
              min(1)=        9 -> 19        -0.16164                  min(3)=        8 -> 13        -0.05383
              min(2)=        6 -> 11        -0.12849                  min(4)=        7 -> 12        -0.05275
              min(3)=        8 -> 11        -0.05179                  min(5)=        8 -> 18        -0.04297
              min(4)=        7 -> 17        -0.03429                  #states=38 #states>0=19 #states<0=19
              min(5)=        7 -> 10        -0.01490
              #states=32 #states>0=19 #states<0=13     24       Singlet-B1   21.8636         56.71    0.3016    0.000
                                                                      max(1)=        9 -> 17         0.67575
21       Singlet-B2   20.3042         61.06    0.0094    0.000          max(2)=        9 -> 18         0.19922
              max(1)=        6 -> 11         0.68751                  max(3)=        9 -> 10         0.02422
              max(2)=        7 -> 15         0.13685                  max(4)=        6 -> 14         0.01559
              max(3)=        7 -> 13         0.04474                  max(5)=        9 -> 32         0.00271
              max(4)=        6 -> 12         0.03842                  min(1)=        8 -> 20        -0.04252
              max(5)=        9 -> 19         0.03037                  min(2)=        7 -> 19        -0.02921
              min(1)=        7 -> 17        -0.04419                  min(3)=        9 -> 15        -0.01561
              min(2)=        6 -> 16        -0.02137                  min(4)=        8 -> 14        -0.00465
              min(3)=        7 -> 10        -0.01536                  min(5)=        9 -> 30        -0.00454
              min(4)=        7 -> 18        -0.01416                  #states=24 #states>0=12 #states<0=12
              min(5)=        9 -> 24        -0.01097
              #states=32 #states>0=17 #states<0=15     25       Singlet-B1   22.2070         55.83    0.0836    0.000
                                                                      max(1)=        9 -> 18         0.67695
22       Singlet-A1   20.5496         60.33    0.0112    0.000          max(2)=        8 -> 20         0.03182
              max(1)=        6 -> 10         0.50437                  max(3)=        7 -> 19         0.03053
              max(2)=        9 -> 20         0.10029                  max(4)=        9 -> 15         0.02227
              max(3)=        8 -> 18         0.06266                  max(5)=        9 -> 26         0.00924
```

```
                min(1)=        9 -> 17        -0.19484                min(3)=        6 -> 17        -0.01203
                min(2)=        6 -> 14        -0.03095                min(4)=        7 -> 11        -0.01009
                min(3)=        9 -> 21        -0.01205                min(5)=        8 -> 29        -0.00957
                min(4)=        9 -> 10        -0.00936                #states=38 #states>0=23 #states<0=15
                min(5)=        9 -> 29        -0.00813
                #states=23 #states>0=12 #states<0=11    29      Singlet-A1   24.7506         50.09    0.0000    0.000
                                                                       max(1)=        9 -> 20         0.49323
26      Singlet-B2   23.7240         52.26    0.1220    0.000          max(2)=        7 -> 16         0.03197
                max(1)=        6 -> 12         0.66374                 max(3)=        8 -> 29         0.00782
                max(2)=        9 -> 19         0.23393                 max(4)=        7 -> 12         0.00622
                max(3)=        8 -> 16         0.01576                 max(5)=        6 -> 18         0.00606
                max(4)=        7 -> 10         0.01525                 min(1)=        8 -> 18        -0.47452
                max(5)=        7 -> 15         0.00961                 min(2)=        6 -> 13        -0.17269
                min(1)=        6 -> 11        -0.05142                 min(3)=        7 -> 11        -0.01531
                min(2)=        7 -> 17        -0.03099                 min(4)=        7 -> 22        -0.01018
                min(3)=        6 -> 16        -0.01705                 min(5)=        6 -> 15        -0.01010
                min(4)=        7 -> 13        -0.00840                 #states=38 #states>0=19 #states<0=19
                min(5)=        6 -> 23        -0.00388
                #states=30 #states>0=14 #states<0=16    30      Singlet-B1   24.9203         49.75    0.0307    0.000
                                                                       max(1)=        6 -> 14         0.69740
27      Singlet-A1   24.1684         51.30    0.0713    0.000          max(2)=        8 -> 20         0.08917
                max(1)=        6 -> 13         0.66567                 max(3)=        7 -> 19         0.04966
                max(2)=        9 -> 20         0.21278                 max(4)=        9 -> 15         0.02526
                max(3)=        7 -> 16         0.02262                 max(5)=        9 -> 18         0.01986
                max(4)=        8 -> 15         0.00974                 min(1)=        9 -> 21        -0.04497
                max(5)=        7 -> 11         0.00833                 min(2)=        9 -> 17        -0.01376
                min(1)=        8 -> 17        -0.09676                 min(3)=        6 -> 25        -0.00751
                min(2)=        6 -> 10        -0.02571                 min(4)=        6 -> 20        -0.00558
                min(3)=        8 -> 18        -0.01770                 min(5)=        9 -> 10        -0.00209
                min(4)=        8 -> 13        -0.01367                 #states=23 #states>0=12 #states<0=11
                min(5)=        7 -> 12        -0.01223
                #states=37 #states>0=17 #states<0=20    31      Singlet-B2   25.4755         48.67    0.1569    0.000
                                                                       max(1)=        9 -> 19         0.56675
28      Singlet-A1   24.5575         50.49    0.0459    0.000          max(2)=        7 -> 18         0.34208
                max(1)=        8 -> 17         0.69727                 max(3)=        7 -> 15         0.08740
                max(2)=        6 -> 13         0.09223                 max(4)=        8 -> 16         0.07374
                max(3)=        6 -> 15         0.04423                 max(5)=        7 -> 17         0.02337
                max(4)=        9 -> 20         0.03082                 min(1)=        6 -> 12        -0.20767
                max(5)=        8 -> 13         0.02263                 min(2)=        8 -> 11        -0.04970
                min(1)=        8 -> 18        -0.01368                 min(3)=        8 -> 22        -0.04715
                min(2)=        8 -> 21        -0.01333                 min(4)=        6 -> 11        -0.02610
```

```
              min(5)=        7 -> 21        -0.02543                #states=38 #states>0=18 #states<0=20
              #states=32 #states>0=17 #states<0=15
                                                    35       Singlet-B1   27.9258         44.40    0.1019    0.000
32       Singlet-A2   26.7526         46.34    0.0000    0.000          max(1)=        8 -> 20         0.61956
              max(1)=        8 -> 19         0.70688                 max(2)=        9 -> 21         0.18128
              max(2)=        9 -> 16         0.00406                 max(3)=        9 -> 17         0.02993
              max(3)=        9 -> 33         0.00140                 max(4)=        7 -> 24         0.00588
              max(4)=        9 -> 31         0.00038                 max(5)=        9 -> 30         0.00514
              max(5)=        7 -> 28         0.00036                 min(1)=        7 -> 19        -0.28231
              min(1)=        8 -> 24        -0.01287                 min(2)=        6 -> 14        -0.04621
              min(2)=        6 -> 19        -0.00908                 min(3)=        9 -> 15        -0.01943
              min(3)=        7 -> 14        -0.00658                 min(4)=        9 -> 13        -0.01017
              min(4)=        9 -> 22        -0.00466                 min(5)=        6 -> 20        -0.00921
              min(5)=        9 -> 11        -0.00458                 #states=24 #states>0=12 #states<0=12
              #states=18 #states>0=8 #states<0=10
                                                    36       Singlet-B2   29.0272         42.71    0.1140    0.000
33       Singlet-B2   26.8165         46.23    0.0452    0.000          max(1)=        6 -> 16         0.66893
              max(1)=        7 -> 17         0.70232                 max(2)=        7 -> 18         0.19743
              max(2)=        7 -> 15         0.04531                 max(3)=        6 -> 12         0.04546
              max(3)=        6 -> 12         0.03676                 max(4)=        6 -> 11         0.03486
              max(4)=        6 -> 11         0.03374                 max(5)=        8 -> 11         0.02117
              max(5)=        6 -> 16         0.02727                 min(1)=        9 -> 19        -0.07958
              min(1)=        7 -> 18        -0.02880                 min(2)=        7 -> 15        -0.04690
              min(2)=        8 -> 11        -0.01260                 min(3)=        8 -> 16        -0.02622
              min(3)=        7 -> 26        -0.00957                 min(4)=        7 -> 17        -0.01932
              min(4)=        9 -> 24        -0.00946                 min(5)=        7 -> 30        -0.00900
              min(5)=        8 -> 27        -0.00576                 #states=32 #states>0=16 #states<0=16
              #states=31 #states>0=16 #states<0=15
                                                    37       Singlet-A1   29.2152         42.44    1.1948    0.000
34       Singlet-A1   27.1740         45.63    0.0998    0.000          max(1)=        8 -> 18         0.48614
              max(1)=        6 -> 15         0.68058                 max(2)=        9 -> 20         0.41615
              max(2)=        6 -> 13         0.03474                 max(3)=        6 -> 15         0.17512
              max(3)=        6 -> 10         0.02516                 max(4)=        7 -> 16         0.11052
              max(4)=        7 -> 11         0.02156                 max(5)=        8 -> 15         0.07562
              max(5)=        9 -> 14         0.01439                 min(1)=        6 -> 13        -0.12333
              min(1)=        8 -> 18        -0.13433                 min(2)=        7 -> 22        -0.11234
              min(2)=        9 -> 20        -0.09980                 min(3)=        8 -> 21        -0.10008
              min(3)=        7 -> 16        -0.04832                 min(4)=        7 -> 11        -0.07222
              min(4)=        8 -> 17        -0.04678                 min(5)=        8 -> 10        -0.02790
              min(5)=        6 -> 17        -0.03046                 #states=38 #states>0=23 #states<0=15
```

8.1.3 Main contributions from different excited states at CIS approach

#_exc.st	___symm___	Exc.E	Osc._Strength	___f___	_<S**2>_						
						min(5)=	8 -> 21	-0.01313			
						#states=36 #states>0=20 #states<0=16					
1	Singlet-A2	6.5858	188.26	0.0000	0.000						
	max(1)=	9 -> 11	0.68473			4	Singlet-A2	10.6850	116.04	0.0000	0.000
	max(2)=	9 -> 16	0.15654				max(1)=	9 -> 12	0.69713		
	max(3)=	9 -> 22	0.01734				max(2)=	9 -> 11	0.09295		
	max(4)=	7 -> 14	0.00533				max(3)=	9 -> 22	0.03202		
	max(5)=	7 -> 20	0.00230				max(4)=	9 -> 28	0.01743		
	min(1)=	9 -> 12	-0.07773				max(5)=	9 -> 23	0.01093		
	min(2)=	9 -> 28	-0.01399				min(1)=	9 -> 16	-0.06207		
	min(3)=	9 -> 23	-0.00575				min(2)=	8 -> 19	-0.00429		
	min(4)=	6 -> 24	-0.00478				min(3)=	6 -> 19	-0.00422		
	min(5)=	6 -> 19	-0.00109				min(4)=	7 -> 20	-0.00136		
	#states=17 #states>0=10 #states<0=7						min(5)=	7 -> 27	-0.00045		
							#states=16 #states>0=10 #states<0=6				
2	Singlet-B1	7.3651	168.34	0.0293	0.000						
	max(1)=	9 -> 10	0.69679			5	Singlet-B2	10.8617	114.15	0.3294	0.000
	max(2)=	9 -> 13	0.05818				max(1)=	8 -> 11	0.68080		
	max(3)=	9 -> 21	0.00753				max(2)=	7 -> 10	0.10204		
	max(4)=	9 -> 26	0.00566				max(3)=	8 -> 16	0.09296		
	max(5)=	9 -> 29	0.00506				max(4)=	7 -> 18	0.04348		
	min(1)=	9 -> 15	-0.07391				max(5)=	6 -> 12	0.02506		
	min(2)=	9 -> 17	-0.06890				min(1)=	7 -> 15	-0.07779		
	min(3)=	8 -> 20	-0.01783				min(2)=	8 -> 12	-0.06554		
	min(4)=	7 -> 19	-0.01594				min(3)=	9 -> 19	-0.05814		
	min(5)=	8 -> 14	-0.01013				min(4)=	7 -> 13	-0.02140		
	#states=22 #states>0=14 #states<0=8						min(5)=	7 -> 21	-0.01613		
							#states=30 #states>0=15 #states<0=15				
3	Singlet-A1	10.4995	118.09	0.3246	0.000						
	max(1)=	8 -> 10	0.68556			6	Singlet-B1	11.1118	111.58	0.0028	0.000
	max(2)=	7 -> 11	0.05393				max(1)=	9 -> 13	0.69731		
	max(3)=	7 -> 12	0.03096				max(2)=	9 -> 26	0.02083		
	max(4)=	7 -> 16	0.02463				max(3)=	9 -> 29	0.01391		
	max(5)=	8 -> 18	0.00874				max(4)=	9 -> 21	0.01318		
	min(1)=	9 -> 14	-0.12854				max(5)=	7 -> 19	0.00973		
	min(2)=	8 -> 15	-0.06941				min(1)=	9 -> 15	-0.08998		
	min(3)=	8 -> 17	-0.05471				min(2)=	9 -> 10	-0.06723		
	min(4)=	6 -> 13	-0.02563				min(3)=	9 -> 18	-0.00870		

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              min(4)=        6 -> 20        -0.00619                #states=34 #states>0=15 #states<0=19
              min(5)=        6 -> 14        -0.00349
              #states=21 #states>0=10 #states<0=11     10       Singlet-B2   13.8864         89.28    0.0418    0.000
                                                                     max(1)=        8 -> 12         0.65398
7       Singlet-A1   12.2558        101.16    0.0237    0.000          max(2)=        8 -> 11         0.10921
              max(1)=        9 -> 14         0.62332                 max(3)=        7 -> 15         0.03021
              max(2)=        7 -> 11         0.22171                 max(4)=        8 -> 22         0.02657
              max(3)=        8 -> 10         0.10470                 max(5)=        6 -> 11         0.02025
              max(4)=        7 -> 16         0.04615                 min(1)=        7 -> 10        -0.22128
              max(5)=        6 -> 10         0.02881                 min(2)=        8 -> 16        -0.08366
              min(1)=        8 -> 13        -0.18241                 min(3)=        7 -> 13        -0.03516
              min(2)=        7 -> 12        -0.11430                 min(4)=        9 -> 19        -0.01324
              min(3)=        7 -> 22        -0.01541                 min(5)=        6 -> 16        -0.01071
              min(4)=        8 -> 21        -0.01539                 #states=26 #states>0=15 #states<0=11
              min(5)=        9 -> 20        -0.01534
              #states=33 #states>0=14 #states<0=19     11       Singlet-B1   14.6024         84.91    0.1917    0.000
                                                                     max(1)=        8 -> 14         0.57188
8       Singlet-B2   12.8484         96.50    0.6319    0.000          max(2)=        6 -> 14         0.02306
              max(1)=        7 -> 10         0.64037                 max(3)=        8 -> 27         0.01721
              max(2)=        8 -> 12         0.23951                 max(4)=        9 -> 18         0.01592
              max(3)=        7 -> 13         0.13566                 max(5)=        9 -> 17         0.00826
              max(4)=        6 -> 11         0.01482                 min(1)=        9 -> 15        -0.40764
              max(5)=        9 -> 19         0.01473                 min(2)=        9 -> 13        -0.05529
              min(1)=        8 -> 11        -0.07216                 min(3)=        9 -> 10        -0.03066
              min(2)=        7 -> 15        -0.05929                 min(4)=        8 -> 20        -0.02703
              min(3)=        6 -> 12        -0.05095                 min(5)=        7 -> 19        -0.01888
              min(4)=        7 -> 17        -0.04315                 #states=22 #states>0=11 #states<0=11
              min(5)=        6 -> 22        -0.01094
              #states=29 #states>0=15 #states<0=14     12       Singlet-A1   15.0230         82.53    0.0624    0.000
                                                                     max(1)=        8 -> 13         0.55913
9       Singlet-A1   13.6624         90.75    0.4377    0.000          max(2)=        9 -> 14         0.20483
              max(1)=        7 -> 11         0.58606                 max(3)=        8 -> 15         0.11323
              max(2)=        8 -> 13         0.33292                 max(4)=        8 -> 10         0.07694
              max(3)=        7 -> 16         0.07521                 max(5)=        9 -> 20         0.02621
              max(4)=        6 -> 13         0.02747                 min(1)=        7 -> 11        -0.27715
              max(5)=        8 -> 26         0.01669                 min(2)=        7 -> 12        -0.21317
              min(1)=        9 -> 14        -0.11802                 min(3)=        7 -> 22        -0.02652
              min(2)=        8 -> 15        -0.09211                 min(4)=        6 -> 17        -0.02498
              min(3)=        7 -> 12        -0.08284                 min(5)=        8 -> 17        -0.02454
              min(4)=        8 -> 10        -0.07602                 #states=36 #states>0=17 #states<0=19
              min(5)=        9 -> 20        -0.04588
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13       Singlet-B1   15.0892         82.17    0.3497    0.000          max(2)=        8 -> 11         0.03639
               max(1)=        9 -> 15         0.55687                  max(3)=        6 -> 11         0.02536
               max(2)=        8 -> 14         0.41379                  max(4)=        7 -> 21         0.02406
               max(3)=        9 -> 13         0.07593                  max(5)=        7 -> 18         0.01921
               max(4)=        9 -> 10         0.06299                  min(1)=        7 -> 10        -0.13665
               max(5)=        8 -> 20         0.05776                  min(2)=        9 -> 19        -0.03357
               min(1)=        6 -> 14        -0.03077                  min(3)=        7 -> 15        -0.02057
               min(2)=        9 -> 18        -0.02858                  min(4)=        8 -> 12        -0.01820
               min(3)=        8 -> 25        -0.01303                  min(5)=        8 -> 16        -0.01751
               min(4)=        9 -> 30        -0.01140                  #states=27 #states>0=14 #states<0=13
               min(5)=        6 -> 20        -0.01103
               #states=22 #states>0=10 #states<0=12     17       Singlet-A2   17.5608         70.60    0.0000    0.000
                                                                       max(1)=        7 -> 14         0.70593
14       Singlet-A2   16.1920         76.57    0.0000    0.000          max(2)=        7 -> 27         0.01561
               max(1)=        9 -> 16         0.68481                  max(3)=        7 -> 20         0.01196
               max(2)=        9 -> 12         0.07979                  max(4)=        8 -> 19         0.01105
               max(3)=        9 -> 22         0.03659                  max(5)=        6 -> 24         0.00488
               max(4)=        7 -> 14         0.02087                  min(1)=        7 -> 25        -0.02345
               max(5)=        7 -> 20         0.00394                  min(2)=        9 -> 16        -0.02107
               min(1)=        9 -> 11        -0.14917                  min(3)=        6 -> 19        -0.00897
               min(2)=        9 -> 28        -0.02258                  min(4)=        9 -> 12        -0.00511
               min(3)=        9 -> 23        -0.00754                  min(5)=        8 -> 24        -0.00412
               min(4)=        9 -> 31        -0.00508                  #states=16 #states>0=6 #states<0=10
               min(5)=        6 -> 24        -0.00448
               #states=15 #states>0=9 #states<0=6       18       Singlet-A1   18.4442         67.22    0.0525    0.000
                                                                       max(1)=        8 -> 15         0.52475
15       Singlet-A1   17.0097         72.89    0.0006    0.000          max(2)=        7 -> 11         0.09097
               max(1)=        7 -> 12         0.52676                  max(3)=        8 -> 21         0.04737
               max(2)=        8 -> 15         0.42837                  max(4)=        6 -> 17         0.04519
               max(3)=        8 -> 13         0.12368                  max(5)=        8 -> 10         0.03741
               max(4)=        7 -> 11         0.08776                  min(1)=        7 -> 12        -0.39045
               max(5)=        9 -> 14         0.08552                  min(2)=        9 -> 14        -0.15935
               min(1)=        8 -> 18        -0.04567                  min(3)=        8 -> 13        -0.15024
               min(2)=        9 -> 20        -0.04442                  min(4)=        7 -> 16        -0.07807
               min(3)=        8 -> 17        -0.03462                  min(5)=        8 -> 18        -0.03334
               min(4)=        6 -> 15        -0.02922                  #states=37 #states>0=18 #states<0=19
               min(5)=        6 -> 17        -0.01887
               #states=36 #states>0=22 #states<0=14     19       Singlet-B2   19.4648         63.70    0.0170    0.000
                                                                       max(1)=        8 -> 16         0.61072
16       Singlet-B2   17.1768         72.18    0.0381    0.000          max(2)=        8 -> 12         0.08582
               max(1)=        7 -> 13         0.68943                  max(3)=        7 -> 18         0.04182
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              max(4)=        7 -> 17         0.02978                 min(1)=        8 -> 20        -0.03162
              max(5)=        8 -> 22         0.02737                 min(2)=        7 -> 19        -0.02276
              min(1)=        7 -> 15        -0.31294                 min(3)=        8 -> 14        -0.01301
              min(2)=        8 -> 11        -0.10696                 min(4)=        9 -> 30        -0.01280
              min(3)=        7 -> 10        -0.06738                 min(5)=        9 -> 21        -0.01019
              min(4)=        9 -> 19        -0.03711                 #states=23 #states>0=13 #states<0=10
              min(5)=        7 -> 21        -0.01860
              #states=29 #states>0=17 #states<0=12    23      Singlet-B2   23.2098         53.42    0.0020    0.000
                                                                      max(1)=        6 -> 11         0.67826
20      Singlet-B2   20.6228         60.12    0.1434    0.000         max(2)=        6 -> 16         0.13457
              max(1)=        7 -> 15         0.60606                 max(3)=        7 -> 15         0.04326
              max(2)=        8 -> 16         0.30223                 max(4)=        7 -> 17         0.01610
              max(3)=        7 -> 10         0.04738                 max(5)=        8 -> 16         0.01539
              max(4)=        7 -> 18         0.03086                 min(1)=        9 -> 19        -0.10357
              max(5)=        7 -> 13         0.02800                 min(2)=        6 -> 12        -0.08538
              min(1)=        9 -> 19        -0.17337                 min(3)=        7 -> 13        -0.02411
              min(2)=        6 -> 11        -0.06650                 min(4)=        8 -> 12        -0.02148
              min(3)=        7 -> 17        -0.02562                 min(5)=        6 -> 28        -0.00878
              min(4)=        6 -> 16        -0.01600                 #states=31 #states>0=14 #states<0=17
              min(5)=        7 -> 26        -0.01249
              #states=31 #states>0=19 #states<0=12    24      Singlet-B1   23.2490         53.33    0.0912    0.000
                                                                      max(1)=        9 -> 18         0.67944
21      Singlet-A1   22.4841         55.14    0.0044    0.000         max(2)=        8 -> 20         0.04431
              max(1)=        7 -> 16         0.66850                 max(3)=        7 -> 19         0.03545
              max(2)=        8 -> 15         0.05812                 max(4)=        9 -> 15         0.02539
              max(3)=        8 -> 17         0.03117                 max(5)=        9 -> 26         0.02399
              max(4)=        7 -> 22         0.02442                 min(1)=        9 -> 17        -0.17843
              max(5)=        6 -> 21         0.01377                 min(2)=        6 -> 14        -0.03089
              min(1)=        9 -> 20        -0.14210                 min(3)=        9 -> 29        -0.02282
              min(2)=        6 -> 10        -0.10299                 min(4)=        9 -> 10        -0.01832
              min(3)=        7 -> 11        -0.07685                 min(5)=        9 -> 21        -0.00711
              min(4)=        8 -> 18        -0.06477                 #states=22 #states>0=12 #states<0=10
              min(5)=        6 -> 13        -0.04927
              #states=36 #states>0=17 #states<0=19    25      Singlet-A1   23.9737         51.72    0.0267    0.000
                                                                      max(1)=        6 -> 10         0.68376
22      Singlet-B1   22.9087         54.12    0.2891    0.000         max(2)=        7 -> 16         0.10727
              max(1)=        9 -> 17         0.67671                 max(3)=        6 -> 13         0.04879
              max(2)=        9 -> 18         0.18405                 max(4)=        8 -> 15         0.03537
              max(3)=        9 -> 10         0.06453                 max(5)=        9 -> 20         0.03218
              max(4)=        6 -> 14         0.04007                 min(1)=        6 -> 15        -0.10323
              max(5)=        6 -> 27         0.01071                 min(2)=        9 -> 14        -0.04660
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              min(3)=        6 -> 17        -0.02330                min(5)=        7 -> 29        -0.00514
              min(4)=        9 -> 25        -0.02039                #states=30 #states>0=20 #states<0=10
              min(5)=        8 -> 13        -0.01979
              #states=38 #states>0=22 #states<0=16    29       Singlet-B2   27.3836         45.28    0.0244    0.000
                                                                     max(1)=        6 -> 12         0.60364
26       Singlet-A1   25.7829         48.09    0.0246    0.000          max(2)=        6 -> 11         0.04340
              max(1)=        9 -> 20         0.53598                 max(3)=        7 -> 10         0.04293
              max(2)=        8 -> 17         0.06690                 max(4)=        8 -> 22         0.02612
              max(3)=        7 -> 16         0.06441                 max(5)=        6 -> 23         0.01654
              max(4)=        6 -> 13         0.03817                 min(1)=        9 -> 19        -0.25686
              max(5)=        8 -> 29         0.01684                 min(2)=        7 -> 18        -0.24109
              min(1)=        8 -> 18        -0.44563                 min(3)=        7 -> 15        -0.05294
              min(2)=        6 -> 10        -0.04562                 min(4)=        8 -> 11        -0.03880
              min(3)=        9 -> 25        -0.02035                 min(5)=        7 -> 17        -0.02469
              min(4)=        8 -> 26        -0.01530                 #states=30 #states>0=16 #states<0=14
              min(5)=        6 -> 15        -0.01423
              #states=32 #states>0=18 #states<0=14    30       Singlet-A1   27.4499         45.17    0.0862    0.000
                                                                     max(1)=        6 -> 13         0.68473
27       Singlet-A1   25.9826         47.72    0.0525    0.000          max(2)=        8 -> 18         0.12654
              max(1)=        8 -> 17         0.69735                 max(3)=        7 -> 16         0.06091
              max(2)=        8 -> 10         0.06052                 max(4)=        9 -> 20         0.04956
              max(3)=        8 -> 18         0.05459                 max(5)=        6 -> 15         0.03752
              max(4)=        6 -> 13         0.03048                 min(1)=        6 -> 10        -0.05575
              max(5)=        8 -> 15         0.02624                 min(2)=        8 -> 17        -0.03697
              min(1)=        9 -> 20        -0.04304                 min(3)=        7 -> 11        -0.01656
              min(2)=        7 -> 16        -0.03203                 min(4)=        8 -> 13        -0.01134
              min(3)=        7 -> 22        -0.01743                 min(5)=        9 -> 14        -0.00505
              min(4)=        6 -> 15        -0.01633                 #states=33 #states>0=19 #states<0=14
              min(5)=        8 -> 21        -0.01496
              #states=36 #states>0=21 #states<0=15    31       Singlet-A2   27.8593         44.50    0.0000    0.000
                                                                     max(1)=        8 -> 19         0.70440
28       Singlet-B2   26.2831         47.17    0.4513    0.000          max(2)=        7 -> 20         0.04287
              max(1)=        9 -> 19         0.54843                 max(3)=        7 -> 25         0.00611
              max(2)=        6 -> 12         0.32973                 max(4)=        9 -> 12         0.00475
              max(3)=        7 -> 18         0.22509                 max(5)=        7 -> 27         0.00348
              max(4)=        6 -> 11         0.12144                 min(1)=        8 -> 24        -0.03651
              max(5)=        7 -> 15         0.11253                 min(2)=        6 -> 24        -0.01568
              min(1)=        8 -> 22        -0.04148                 min(3)=        9 -> 22        -0.01320
              min(2)=        9 -> 24        -0.02500                 min(4)=        7 -> 14        -0.01187
              min(3)=        7 -> 21        -0.02163                 min(5)=        9 -> 16        -0.00359
              min(4)=        6 -> 16        -0.01091                 #states=17 #states>0=9 #states<0=8
```
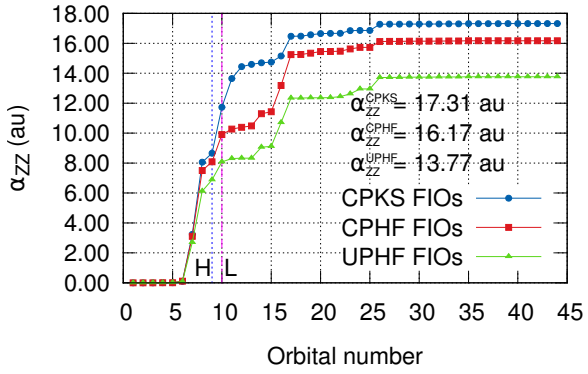

32	Singlet-B1	28.0712	44.17	0.0259	35.000	Singlet-A1	29.9642	41.38	0.4855	0.000	
	max(1)=	6 -> 14	0.68642			max(1)=	6 -> 15	0.58105			
	max(2)=	8 -> 20	0.15865			max(2)=	6 -> 10	0.09990			
	max(3)=	9 -> 15	0.02140			max(3)=	7 -> 22	0.06465			
	max(4)=	7 -> 19	0.02128			max(4)=	6 -> 13	0.05422			
	max(5)=	9 -> 21	0.01638			max(5)=	8 -> 21	0.04070			
	min(1)=	9 -> 17	-0.03629			min(1)=	8 -> 18	-0.29450			
	min(2)=	6 -> 25	-0.02477			min(2)=	9 -> 20	-0.21756			
	min(3)=	9 -> 29	-0.00325			min(3)=	7 -> 16	-0.05893			
	min(4)=	9 -> 32	-0.00303			min(4)=	6 -> 17	-0.03765			
	min(5)=	8 -> 14	-0.00287			min(5)=	8 -> 15	-0.03296			
	#states=23 #states>0=13 #states<0=10					#states=34 #states>0=14 #states<0=20					
33	Singlet-B2	28.5619	43.41	0.0609	36.000	Singlet-A1	30.9514	40.06	1.3451	0.000	
	max(1)=	7 -> 17	0.69996			max(1)=	8 -> 18	0.41756			
	max(2)=	7 -> 10	0.05014			max(2)=	6 -> 15	0.38131			
	max(3)=	7 -> 15	0.02719			max(3)=	9 -> 20	0.35079			
	max(4)=	6 -> 22	0.02123			max(4)=	7 -> 16	0.11347			
	max(5)=	7 -> 13	0.01600			max(5)=	8 -> 15	0.06554			
	min(1)=	6 -> 16	-0.04718			min(1)=	6 -> 13	-0.13218			
	min(2)=	7 -> 18	-0.04293			min(2)=	8 -> 21	-0.08333			
	min(3)=	7 -> 21	-0.02361			min(3)=	7 -> 22	-0.07424			
	min(4)=	7 -> 26	-0.01829			min(4)=	8 -> 10	-0.02169			
	min(5)=	7 -> 30	-0.01100			min(5)=	9 -> 25	-0.01317			
	#states=32 #states>0=13 #states<0=19					#states=37 #states>0=22 #states<0=15					
34	Singlet-B1	29.4276	42.13	0.1699	37.000	Singlet-B2	31.3201	39.59	1.2738	0.000	
	max(1)=	8 -> 20	0.61419			max(1)=	7 -> 18	0.60533			
	max(2)=	9 -> 21	0.18257			max(2)=	8 -> 22	0.13722			
	max(3)=	9 -> 17	0.03571			max(3)=	6 -> 12	0.10996			
	max(4)=	6 -> 20	0.01659			max(4)=	7 -> 21	0.10270			
	max(5)=	7 -> 24	0.01149			max(5)=	7 -> 17	0.03935			
	min(1)=	7 -> 19	-0.25679			min(1)=	9 -> 19	-0.26592			
	min(2)=	6 -> 14	-0.13514			min(2)=	8 -> 16	-0.10003			
	min(3)=	9 -> 15	-0.04316			min(3)=	8 -> 11	-0.06017			
	min(4)=	8 -> 25	-0.02617			min(4)=	7 -> 15	-0.05849			
	min(5)=	9 -> 18	-0.01927			min(5)=	7 -> 13	-0.03313			
	#states=22 #states>0=9 #states<0=13					#states=32 #states>0=15 #states<0=17					

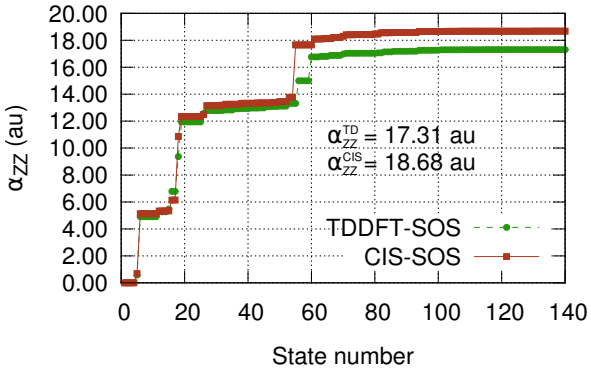
8.2 6-311++G(d,p)

8.2.1 Plots

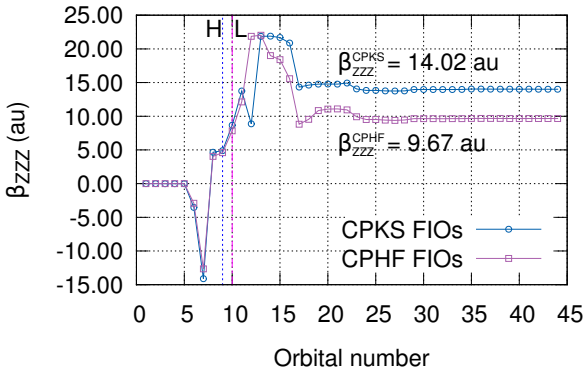
Figure S12: For H_2S molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S12a and S12c) or states (SOS approaches, in Plots S12b and S12d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.02 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



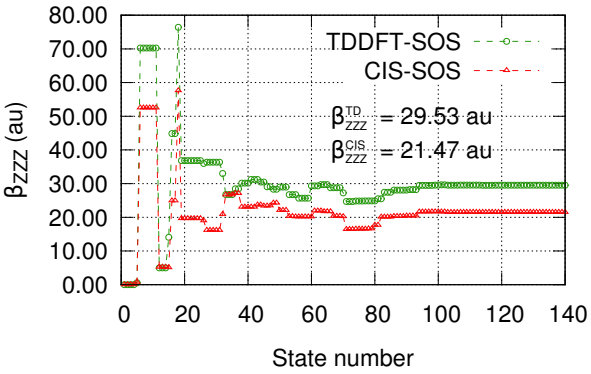
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

8.2.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

#_exc.st	___symm___	Exc.E	Osc._Strength	___f___	_<S**2>_
1	Singlet-B1	5.8982	210.21	0.0486	0.000
	max(1)=	9 -> 10	0.68903		
	max(2)=	9 -> 19	0.01598		
	max(3)=	9 -> 27	0.00650		
	max(4)=	6 -> 26	0.00364		
	max(5)=	6 -> 20	0.00262		
	min(1)=	9 -> 17	-0.15758		
	min(2)=	9 -> 24	-0.01260		
	min(3)=	9 -> 13	-0.01112		
	min(4)=	9 -> 15	-0.00952		
	min(5)=	7 -> 25	-0.00731		
	#CIs=30 #CIs>0=16 #CIs<0=13				
2	Singlet-A2	6.0526	204.84	0.0000	0.000
	max(1)=	9 -> 11	0.57869		
	max(2)=	9 -> 22	0.03410		
	max(3)=	6 -> 25	0.00671		
	max(4)=	8 -> 25	0.00469		
	max(5)=	9 -> 34	0.00412		
	min(1)=	9 -> 12	-0.35211		
	min(2)=	9 -> 16	-0.20026		
	min(3)=	9 -> 28	-0.00594		
	min(4)=	7 -> 14	-0.00450		
	min(5)=	7 -> 20	-0.00368		
	#CIs=21 #CIs>0=7 #CIs<0=13				
3	Singlet-A2	7.6518	162.03	0.0000	0.000
	max(1)=	9 -> 12	0.60616		
	max(2)=	9 -> 11	0.35473		
	max(3)=	9 -> 18	0.07047		
	max(4)=	7 -> 14	0.00394		
	max(5)=	9 -> 28	0.00377		
	min(1)=	9 -> 16	-0.04178		
	min(2)=	8 -> 25	-0.00083		
	min(3)=	7 -> 26	-0.00060		
	min(4)=	7 -> 20	-0.00059		
	min(5)=	9 -> 29	-0.00059		
	#CIs=21 #CIs>0=8 #CIs<0=12				
4	Singlet-B1	7.9539	155.88	0.0256	0.000
	max(1)=	9 -> 13	0.70336		
	max(2)=	9 -> 19	0.06468		
	max(3)=	9 -> 15	0.01784		
	max(4)=	8 -> 14	0.01081		
	max(5)=	9 -> 10	0.00506		
	min(1)=	9 -> 17	-0.02254		
	min(2)=	9 -> 21	-0.01111		
	min(3)=	8 -> 33	-0.00125		
	min(4)=	7 -> 30	-0.00120		
	min(5)=	8 -> 20	-0.00110		
	#CIs=30 #CIs>0=16 #CIs<0=13				
5	Singlet-A1	8.2969	149.44	0.0178	0.000
	max(1)=	9 -> 14	0.68625		
	max(2)=	8 -> 17	0.03457		
	max(3)=	9 -> 20	0.02136		
	max(4)=	7 -> 16	0.01664		
	max(5)=	8 -> 15	0.01396		
	min(1)=	8 -> 10	-0.15111		
	min(2)=	8 -> 13	-0.03731		
	min(3)=	7 -> 11	-0.03663		
	min(4)=	8 -> 19	-0.02913		
	min(5)=	7 -> 18	-0.02161		
	#CIs=49 #CIs>0=19 #CIs<0=26				
6	Singlet-A1	8.8803	139.62	0.1538	0.000
	max(1)=	8 -> 10	0.67052		
	max(2)=	9 -> 14	0.14764		
	max(3)=	7 -> 12	0.04488		
	max(4)=	7 -> 16	0.03557		
	max(5)=	8 -> 23	0.00473		
	min(1)=	8 -> 17	-0.13181		
	min(2)=	8 -> 13	-0.06874		
	min(3)=	7 -> 11	-0.05081		
	min(4)=	8 -> 15	-0.01756		
	min(5)=	6 -> 13	-0.01121		
	#CIs=49 #CIs>0=21 #CIs<0=25				
7	Singlet-B1	9.2056	134.68	0.2307	0.000
	max(1)=	9 -> 15	0.52238		

```
              max(2)=        8 -> 14         0.04840                  max(4)=        7 -> 21         0.03816
              max(3)=        6 -> 14         0.02020                  max(5)=        8 -> 18         0.02081
              max(4)=        9 -> 27         0.01406                  min(1)=        8 -> 12        -0.22991
              max(5)=        9 -> 19         0.01366                  min(2)=        8 -> 16        -0.11479
              min(1)=        9 -> 17        -0.46048                  min(3)=        7 -> 10        -0.10629
              min(2)=        9 -> 10        -0.09871                  min(4)=        7 -> 15        -0.05686
              min(3)=        9 -> 13        -0.02946                  min(5)=        7 -> 23        -0.02450
              min(4)=        9 -> 24        -0.02694                  #CIs=40 #CIs>0=13 #CIs<0=26
              min(5)=        7 -> 25        -0.02588
              #CIs=30 #CIs>0=18 #CIs<0=11            11      Singlet-B2   10.5179       117.88    0.0059    0.000
                                                                max(1)=        8 -> 12         0.60674
8       Singlet-A2    9.6474       128.52    0.0000    0.000  max(2)=        8 -> 11         0.25824
              max(1)=        9 -> 16         0.67365                  max(3)=        7 -> 10         0.23335
              max(2)=        9 -> 11         0.19582                  max(4)=        8 -> 18         0.06229
              max(3)=        7 -> 14         0.00404                  max(5)=        7 -> 15         0.01864
              max(4)=        9 -> 28         0.00257                  min(1)=        7 -> 17        -0.06214
              max(5)=        7 -> 20         0.00191                  min(2)=        8 -> 16        -0.04493
              min(1)=        9 -> 12        -0.06665                  min(3)=        9 -> 25        -0.01410
              min(2)=        9 -> 22        -0.05697                  min(4)=        7 -> 21        -0.00973
              min(3)=        9 -> 18        -0.01235                  min(5)=        6 -> 11        -0.00838
              min(4)=        6 -> 25        -0.00306                  #CIs=40 #CIs>0=26 #CIs<0=13
              min(5)=        8 -> 25        -0.00197
              #CIs=21 #CIs>0=12 #CIs<0=9             12      Singlet-A1   10.9492       113.24    0.0197    0.000
                                                                max(1)=        8 -> 13         0.69040
9       Singlet-B1    9.7084       127.71    0.1949    0.000  max(2)=        7 -> 16         0.04826
              max(1)=        9 -> 17         0.50990                  max(3)=        8 -> 10         0.04678
              max(2)=        9 -> 15         0.47159                  max(4)=        9 -> 14         0.04365
              max(3)=        9 -> 10         0.12419                  max(5)=        8 -> 19         0.03299
              max(4)=        8 -> 14         0.03115                  min(1)=        7 -> 11        -0.11307
              max(5)=        9 -> 24         0.01831                  min(2)=        9 -> 20        -0.04585
              min(1)=        9 -> 19        -0.02227                  min(3)=        7 -> 18        -0.02607
              min(2)=        8 -> 26        -0.00575                  min(4)=        8 -> 17        -0.01265
              min(3)=        7 -> 25        -0.00415                  min(5)=        6 -> 17        -0.00987
              min(4)=        6 -> 33        -0.00191                  #CIs=49 #CIs>0=22 #CIs<0=25
              min(5)=        7 -> 30        -0.00183
              #CIs=30 #CIs>0=18 #CIs<0=11            13      Singlet-B1   11.0167       112.54    0.0021    0.000
                                                                max(1)=        8 -> 14         0.70172
10      Singlet-B2    9.8143       126.33    0.1778    0.000  max(2)=        8 -> 20         0.06167
              max(1)=        8 -> 11         0.63905                  max(3)=        8 -> 26         0.01052
              max(2)=        7 -> 17         0.08951                  max(4)=        9 -> 17         0.00758
              max(3)=        9 -> 25         0.04407                  max(5)=        7 -> 25         0.00750
```

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                min(1)=        9 -> 15        -0.05793                min(3)=        8 -> 19        -0.04425
                min(2)=        9 -> 19        -0.01041                min(4)=        7 -> 16        -0.02658
                min(3)=        9 -> 13        -0.00811                min(5)=        7 -> 22        -0.01615
                min(4)=        9 -> 27        -0.00702                #CIs=49 #CIs>0=21 #CIs<0=24
                min(5)=        6 -> 20        -0.00316
                #CIs=29 #CIs>0=12 #CIs<0=17            17       Singlet-B2   12.6563         97.96    0.0166    0.000
                                                                       max(1)=        8 -> 16         0.67087
14       Singlet-B2   11.0882        111.82    0.3185    0.000          max(2)=        8 -> 11         0.13550
                max(1)=        7 -> 10         0.64118                 max(3)=        7 -> 15         0.12728
                max(2)=        8 -> 16         0.06923                 max(4)=        7 -> 13         0.04392
                max(3)=        7 -> 13         0.04248                 max(5)=        7 -> 23         0.01539
                max(4)=        8 -> 11         0.03188                 min(1)=        7 -> 10        -0.07585
                max(5)=        7 -> 19         0.02242                 min(2)=        7 -> 17        -0.05973
                min(1)=        8 -> 12        -0.26208                 min(3)=        8 -> 22        -0.04188
                min(2)=        7 -> 17        -0.09176                 min(4)=        7 -> 21        -0.03211
                min(3)=        7 -> 15        -0.05217                 min(5)=        9 -> 25        -0.02836
                min(4)=        6 -> 12        -0.01974                 #CIs=40 #CIs>0=24 #CIs<0=14
                min(5)=        8 -> 22        -0.01522
                #CIs=40 #CIs>0=17 #CIs<0=21            18       Singlet-A1   12.7242         97.44    0.1884    0.000
                                                                       max(1)=        8 -> 17         0.51398
15       Singlet-A1   11.5622        107.23    0.0120    0.000          max(2)=        8 -> 15         0.11440
                max(1)=        8 -> 15         0.48634                 max(3)=        9 -> 20         0.10137
                max(2)=        7 -> 12         0.16657                 max(4)=        8 -> 10         0.08893
                max(3)=        7 -> 16         0.08374                 max(5)=        8 -> 19         0.02376
                max(4)=        9 -> 20         0.03104                 min(1)=        7 -> 11        -0.41039
                max(5)=        6 -> 23         0.00855                 min(2)=        7 -> 12        -0.17228
                min(1)=        7 -> 11        -0.33339                 min(3)=        8 -> 13        -0.05511
                min(2)=        8 -> 17        -0.31842                 min(4)=        9 -> 14        -0.03861
                min(3)=        8 -> 10        -0.08846                 min(5)=        7 -> 18        -0.02366
                min(4)=        8 -> 13        -0.05918                 #CIs=49 #CIs>0=30 #CIs<0=17
                min(5)=        8 -> 21        -0.04967
                #CIs=49 #CIs>0=26 #CIs<0=19            19       Singlet-A1   13.0752         94.82    0.1975    0.000
                                                                       max(1)=        7 -> 12         0.65892
16       Singlet-A1   12.2341        101.34    0.0886    0.000          max(2)=        8 -> 17         0.24161
                max(1)=        8 -> 15         0.49266                 max(3)=        7 -> 18         0.02474
                max(2)=        7 -> 11         0.42887                 max(4)=        7 -> 22         0.02070
                max(3)=        8 -> 17         0.22323                 max(5)=        6 -> 13         0.01790
                max(4)=        8 -> 10         0.09374                 min(1)=        8 -> 15        -0.05552
                max(5)=        8 -> 13         0.06661                 min(2)=        9 -> 26        -0.04094
                min(1)=        9 -> 20        -0.05449                 min(3)=        8 -> 23        -0.03548
                min(2)=        7 -> 12        -0.04808                 min(4)=        7 -> 16        -0.02258
```

```
                min(5)=        6 -> 10        -0.01628
                #CIs=49 #CIs>0=24 #CIs<0=23            23       Singlet-B2   14.3271         86.54    0.1762    0.000
                                                                       max(1)=        7 -> 15         0.67788
20       Singlet-B2   13.1913         93.99    0.0003    0.000          max(2)=        9 -> 25         0.04252
                max(1)=        7 -> 13         0.69749                 max(3)=        8 -> 18         0.03449
                max(2)=        7 -> 15         0.06639                 max(4)=        7 -> 10         0.03337
                max(3)=        7 -> 19         0.05696                 max(5)=        8 -> 11         0.02817
                max(4)=        6 -> 12         0.01613                 min(1)=        8 -> 16        -0.13150
                max(5)=        7 -> 24         0.00914                 min(2)=        7 -> 17        -0.08719
                min(1)=        8 -> 16        -0.05932                 min(3)=        7 -> 13        -0.07336
                min(2)=        7 -> 10        -0.03602                 min(4)=        8 -> 12        -0.06517
                min(3)=        8 -> 11        -0.01658                 min(5)=        8 -> 22        -0.02567
                min(4)=        6 -> 11        -0.01500                 #CIs=40 #CIs>0=16 #CIs<0=23
                min(5)=        7 -> 21        -0.01194
                #CIs=40 #CIs>0=20 #CIs<0=18            24       Singlet-B1   14.7434         84.09    0.0012    0.000
                                                                       max(1)=        9 -> 19         0.70081
21       Singlet-A2   13.2692         93.44    0.0000    0.000          max(2)=        9 -> 17         0.03232
                max(1)=        7 -> 14         0.70381                 max(3)=        8 -> 26         0.01141
                max(2)=        7 -> 20         0.06498                 max(4)=        8 -> 14         0.01006
                max(3)=        8 -> 25         0.00308                 max(5)=        9 -> 32         0.00952
                max(4)=        9 -> 11         0.00167                 min(1)=        9 -> 13        -0.06448
                max(5)=        6 -> 25         0.00143                 min(2)=        9 -> 21        -0.05497
                min(1)=        9 -> 18        -0.01854                 min(3)=        9 -> 10        -0.00980
                min(2)=        9 -> 16        -0.00596                 min(4)=        8 -> 20        -0.00731
                min(3)=        9 -> 22        -0.00457                 min(5)=        9 -> 23        -0.00542
                min(4)=        9 -> 12        -0.00373                 #CIs=30 #CIs>0=13 #CIs<0=16
                min(5)=        7 -> 31        -0.00189
                #CIs=21 #CIs>0=10 #CIs<0=11            25       Singlet-B2   14.8207         83.66    0.2415    0.000
                                                                       max(1)=        7 -> 17         0.68011
22       Singlet-A2   14.0257         88.40    0.0000    0.000          max(2)=        7 -> 10         0.11999
                max(1)=        9 -> 18         0.70263                 max(3)=        7 -> 15         0.10272
                max(2)=        9 -> 28         0.02116                 max(4)=        8 -> 16         0.06026
                max(3)=        7 -> 14         0.01835                 max(5)=        8 -> 12         0.04516
                max(4)=        9 -> 22         0.01648                 min(1)=        8 -> 18        -0.04790
                max(5)=        9 -> 16         0.01646                 min(2)=        7 -> 19        -0.04553
                min(1)=        9 -> 12        -0.06344                 min(3)=        8 -> 11        -0.03767
                min(2)=        9 -> 11        -0.02973                 min(4)=        9 -> 25        -0.02276
                min(3)=        9 -> 29        -0.00574                 min(5)=        6 -> 11        -0.01335
                min(4)=        8 -> 25        -0.00106                 #CIs=40 #CIs>0=24 #CIs<0=14
                min(5)=        7 -> 26        -0.00058
                #CIs=21 #CIs>0=13 #CIs<0=7             26       Singlet-A1   14.9766         82.79    0.0638    0.000
```

```
              max(1)=        7 -> 16         0.65021                  max(3)=        8 -> 26         0.03964
              max(2)=        9 -> 20         0.24160                  max(4)=        7 -> 25         0.03951
              max(3)=        7 -> 11         0.10509                  max(5)=        9 -> 24         0.01438
              max(4)=        8 -> 17         0.03890                  min(1)=        8 -> 20        -0.14525
              max(5)=        8 -> 19         0.01197                  min(2)=        9 -> 23        -0.03525
              min(1)=        8 -> 15        -0.03894                  min(3)=        9 -> 17        -0.02302
              min(2)=        7 -> 18        -0.03694                  min(4)=        6 -> 20        -0.02017
              min(3)=        9 -> 26        -0.02848                  min(5)=        6 -> 14        -0.01276
              min(4)=        7 -> 22        -0.02824                  #CIs=30 #CIs>0=11 #CIs<0=18
              min(5)=        8 -> 21        -0.02399
              #CIs=49 #CIs>0=25 #CIs<0=22            30       Singlet-A2   17.8273         69.55    0.0000    0.000
                                                                      max(1)=        9 -> 22         0.70340
27       Singlet-A1   15.9097         77.93    0.0247    0.000       max(2)=        9 -> 16         0.06399
              max(1)=        9 -> 20         0.60937                  max(3)=        9 -> 12         0.01214
              max(2)=        8 -> 13         0.07183                  max(4)=        9 -> 34         0.01176
              max(3)=        6 -> 10         0.06076                  max(5)=        7 -> 14         0.00567
              max(4)=        7 -> 11         0.05139                  min(1)=        9 -> 18        -0.01720
              max(5)=        8 -> 21         0.04047                  min(2)=        9 -> 28        -0.01491
              min(1)=        7 -> 16        -0.22814                  min(3)=        9 -> 11        -0.01220
              min(2)=        8 -> 19        -0.22674                  min(4)=        7 -> 20        -0.01049
              min(3)=        7 -> 18        -0.06616                  min(5)=        7 -> 26        -0.00272
              min(4)=        8 -> 17        -0.05976                  #CIs=21 #CIs>0=9 #CIs<0=12
              min(5)=        6 -> 17        -0.05210
              #CIs=49 #CIs>0=24 #CIs<0=23            31       Singlet-B1   18.0813         68.57    0.0000    0.000
                                                                      max(1)=        8 -> 20         0.68834
28       Singlet-B2   16.9734         73.05    0.0039    0.000       max(2)=        9 -> 21         0.14149
              max(1)=        8 -> 18         0.69812                  max(3)=        8 -> 26         0.02785
              max(2)=        8 -> 22         0.04963                  max(4)=        7 -> 25         0.02197
              max(3)=        7 -> 17         0.04650                  max(5)=        9 -> 19         0.01889
              max(4)=        8 -> 16         0.03778                  min(1)=        8 -> 14        -0.06142
              max(5)=        6 -> 12         0.01871                  min(2)=        9 -> 27        -0.01308
              min(1)=        8 -> 12        -0.04793                  min(3)=        6 -> 14        -0.01257
              min(2)=        8 -> 11        -0.03995                  min(4)=        9 -> 23        -0.01196
              min(3)=        9 -> 25        -0.02765                  min(5)=        6 -> 20        -0.00896
              min(4)=        7 -> 15        -0.02096                  #CIs=30 #CIs>0=11 #CIs<0=17
              min(5)=        7 -> 21        -0.01447
              #CIs=40 #CIs>0=25 #CIs<0=14            32       Singlet-A1   18.2286         68.02    0.0018    0.000
                                                                      max(1)=        8 -> 19         0.61500
29       Singlet-B1   17.5688         70.57    0.0355    0.000       max(2)=        9 -> 20         0.14251
              max(1)=        9 -> 21         0.68589                  max(3)=        6 -> 10         0.12815
              max(2)=        9 -> 19         0.05225                  max(4)=        7 -> 11         0.05560
```

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                max(5)=        9 -> 26         0.04037                 min(2)=        6 -> 16        -0.12975
                min(1)=        7 -> 18        -0.25068                 min(3)=        7 -> 21        -0.07301
                min(2)=        7 -> 16        -0.08313                 min(4)=        8 -> 22        -0.04728
                min(3)=        6 -> 17        -0.06013                 min(5)=        7 -> 13        -0.01771
                min(4)=        7 -> 22        -0.04421                 #CIs=40 #CIs>0=22 #CIs<0=17
                min(5)=        8 -> 24        -0.02106
                #CIs=49 #CIs>0=24 #CIs<0=23            36       Singlet-A1   20.2517         61.22    0.0156    0.000
                                                                       max(1)=        8 -> 21         0.54533
33       Singlet-A1   19.4951         63.60    0.0055    0.000       max(2)=        7 -> 18         0.32908
                max(1)=        6 -> 10         0.53889                 max(3)=        8 -> 19         0.13990
                max(2)=        7 -> 18         0.43630                 max(4)=        9 -> 20         0.08104
                max(3)=        8 -> 19         0.05832                 max(5)=        8 -> 15         0.02891
                max(4)=        6 -> 19         0.01720                 min(1)=        6 -> 10        -0.23773
                max(5)=        9 -> 20         0.01635                 min(2)=        8 -> 23        -0.06646
                min(1)=        6 -> 17        -0.10752                 min(3)=        9 -> 26        -0.06539
                min(2)=        8 -> 21        -0.04274                 min(4)=        8 -> 17        -0.02542
                min(3)=        6 -> 15        -0.01735                 min(5)=        8 -> 24        -0.02512
                min(4)=        8 -> 24        -0.01223                 #CIs=49 #CIs>0=23 #CIs<0=24
                min(5)=        6 -> 24        -0.01051
                #CIs=49 #CIs>0=25 #CIs<0=22            37       Singlet-A2   20.3124         61.04    0.0000    0.000
                                                                       max(1)=        7 -> 20         0.70381
34       Singlet-B2   19.9154         62.26    0.0003    0.000       max(2)=        9 -> 22         0.01113
                max(1)=        7 -> 19         0.58715                 max(3)=        7 -> 33         0.01006
                max(2)=        6 -> 12         0.19697                 max(4)=        8 -> 25         0.00891
                max(3)=        6 -> 16         0.09757                 max(5)=        7 -> 26         0.00337
                max(4)=        8 -> 22         0.03689                 min(1)=        7 -> 14        -0.06501
                max(5)=        7 -> 17         0.02564                 min(2)=        7 -> 31        -0.00971
                min(1)=        6 -> 11        -0.31257                 min(3)=        8 -> 30        -0.00232
                min(2)=        7 -> 13        -0.05920                 min(4)=        9 -> 16        -0.00139
                min(3)=        9 -> 25        -0.05005                 min(5)=        9 -> 28        -0.00134
                min(4)=        8 -> 28        -0.01000                 #CIs=21 #CIs>0=11 #CIs<0=10
                min(5)=        7 -> 10        -0.00931
                #CIs=40 #CIs>0=25 #CIs<0=14            38       Singlet-A1   20.7269         59.82    0.0042    0.000
                                                                       max(1)=        8 -> 21         0.42217
35       Singlet-B2   20.1768         61.45    0.0024    0.000       max(2)=        6 -> 10         0.34541
                max(1)=        6 -> 11         0.52639                 max(3)=        7 -> 22         0.12348
                max(2)=        7 -> 19         0.37951                 max(4)=        7 -> 16         0.05906
                max(3)=        9 -> 25         0.04692                 max(5)=        6 -> 17         0.05689
                max(4)=        7 -> 17         0.04440                 min(1)=        7 -> 18        -0.34743
                max(5)=        8 -> 18         0.02637                 min(2)=        8 -> 19        -0.18501
                min(1)=        6 -> 12        -0.21867                 min(3)=        9 -> 20        -0.13622
```

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               min(4)=        9 -> 14        -0.02604                #CIs=40 #CIs>0=14 #CIs<0=25
               min(5)=        7 -> 11        -0.02602
               #CIs=49 #CIs>0=20 #CIs<0=27            40       Singlet-B2   21.4084         57.91    0.0128    0.000
                                                                      max(1)=        6 -> 12         0.63167
39       Singlet-B2   20.9056         59.31    0.0121    0.000          max(2)=        6 -> 11         0.29795
               max(1)=        8 -> 22         0.68155                 max(3)=        7 -> 21         0.06648
               max(2)=        7 -> 21         0.12328                 max(4)=        6 -> 18         0.05626
               max(3)=        9 -> 25         0.10096                 max(5)=        9 -> 25         0.03342
               max(4)=        6 -> 11         0.05097                 min(1)=        7 -> 19        -0.04818
               max(5)=        8 -> 16         0.04340                 min(2)=        6 -> 16        -0.01384
               min(1)=        8 -> 18        -0.03953                 min(3)=        7 -> 17        -0.00918
               min(2)=        7 -> 23        -0.03637                 min(4)=        8 -> 18        -0.00881
               min(3)=        6 -> 12        -0.03596                 min(5)=        7 -> 23        -0.00781
               min(4)=        7 -> 17        -0.02271                 #CIs=40 #CIs>0=17 #CIs<0=22
               min(5)=        6 -> 16        -0.00929
```

8.2.3 Main contributions from different excited states at CIS approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_          min(2)=        9 -> 15        -0.13769
                                                                       min(3)=        9 -> 23        -0.03448
1        Singlet-A2    6.5227        190.08    0.0000    0.000          min(4)=        9 -> 21        -0.02287
               max(1)=        9 -> 16         0.49044                 min(5)=        9 -> 12        -0.02260
               max(2)=        9 -> 14         0.42062                 #CIs=29 #CIs>0=12 #CIs<0=17
               max(3)=        9 -> 28         0.01715
               max(4)=        9 -> 18         0.00637   3       Singlet-A2    8.3178        149.06    0.0000    0.000
               max(5)=        7 -> 13         0.00368                 max(1)=        9 -> 11         0.58575
               min(1)=        9 -> 11        -0.26651                 max(2)=        9 -> 14         0.37733
               min(2)=        9 -> 22        -0.10463                 max(3)=        9 -> 18         0.11920
               min(3)=        9 -> 34        -0.01290                 max(4)=        9 -> 28         0.01130
               min(4)=        6 -> 30        -0.00472                 max(5)=        9 -> 22         0.01018
               min(5)=        7 -> 31        -0.00163                 min(1)=        9 -> 16        -0.00501
               #CIs=21 #CIs>0=14 #CIs<0=7                             min(2)=        9 -> 29        -0.00210
                                                                       min(3)=        6 -> 25        -0.00208
2        Singlet-B1    6.5983        187.90    0.0563    0.000          min(4)=        8 -> 25        -0.00197
               max(1)=        9 -> 10         0.64239                 min(5)=        9 -> 37        -0.00070
               max(2)=        9 -> 19         0.03151                 #CIs=21 #CIs>0=11 #CIs<0=10
               max(3)=        9 -> 35         0.00433
               max(4)=        9 -> 36         0.00367   4       Singlet-B1    8.5688        144.69    0.0265    0.000
               max(5)=        9 -> 27         0.00300                 max(1)=        9 -> 12         0.69437
               min(1)=        9 -> 17        -0.25373                 max(2)=        9 -> 19         0.11305
```

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                max(3)=        9 -> 15         0.04461                 max(5)=        8 -> 26         0.03578
                max(4)=        8 -> 13         0.01975                 min(1)=        9 -> 15        -0.42620
                max(5)=        9 -> 10         0.01443                 min(2)=        8 -> 13        -0.06070
                min(1)=        9 -> 17        -0.03483                 min(3)=        6 -> 13        -0.02250
                min(2)=        9 -> 21        -0.03267                 min(4)=        8 -> 20        -0.02142
                min(3)=        8 -> 26        -0.00181                 min(5)=        6 -> 20        -0.01703
                min(4)=        6 -> 26        -0.00178                 #CIs=30 #CIs>0=13 #CIs<0=16
                min(5)=        6 -> 31        -0.00132
                #CIs=29 #CIs>0=17 #CIs<0=12            8       Singlet-A2   10.3408        119.90    0.0000    0.000
                                                                       max(1)=        9 -> 14         0.41724
5       Singlet-A1    8.8869        139.51    0.0244    0.000          max(2)=        9 -> 22         0.10332
                max(1)=        9 -> 13         0.67763                 max(3)=        9 -> 18         0.01948
                max(2)=        9 -> 20         0.07299                 max(4)=        9 -> 34         0.00695
                max(3)=        8 -> 17         0.06454                 max(5)=        6 -> 30         0.00228
                max(4)=        8 -> 15         0.04310                 min(1)=        9 -> 16        -0.48695
                max(5)=        7 -> 16         0.03246                 min(2)=        9 -> 11        -0.27861
                min(1)=        8 -> 10        -0.15225                 min(3)=        9 -> 28        -0.00698
                min(2)=        7 -> 11        -0.03594                 min(4)=        9 -> 29        -0.00195
                min(3)=        8 -> 19        -0.03526                 min(5)=        7 -> 26        -0.00131
                min(4)=        7 -> 18        -0.02893                 #CIs=21 #CIs>0=8 #CIs<0=13
                min(5)=        8 -> 12        -0.02525
                #CIs=47 #CIs>0=26 #CIs<0=21            9       Singlet-B1   10.3860        119.38    0.1375    0.000
                                                                       max(1)=        9 -> 15         0.53651
6       Singlet-A1    9.8106        126.38    0.1925    0.000          max(2)=        9 -> 17         0.36504
                max(1)=        8 -> 10         0.62489                 max(3)=        9 -> 10         0.26624
                max(2)=        9 -> 13         0.16583                 max(4)=        9 -> 23         0.05257
                max(3)=        7 -> 16         0.03883                 max(5)=        8 -> 13         0.03795
                max(4)=        7 -> 14         0.03184                 min(1)=        9 -> 19        -0.03860
                max(5)=        7 -> 11         0.00980                 min(2)=        9 -> 12        -0.01574
                min(1)=        8 -> 17        -0.22837                 min(3)=        9 -> 35        -0.00415
                min(2)=        8 -> 15        -0.12285                 min(4)=        9 -> 32        -0.00260
                min(3)=        8 -> 12        -0.09917                 min(5)=        9 -> 36        -0.00256
                min(4)=        8 -> 23        -0.02744                 #CIs=30 #CIs>0=20 #CIs<0=9
                min(5)=        6 -> 12        -0.01853
                #CIs=47 #CIs>0=19 #CIs<0=28           10       Singlet-B2   10.6813        116.08    0.3373    0.000
                                                                       max(1)=        8 -> 16         0.40596
7       Singlet-B1    9.9124        125.08    0.3483    0.000          max(2)=        8 -> 14         0.40382
                max(1)=        9 -> 17         0.53366                 max(3)=        7 -> 15         0.03284
                max(2)=        9 -> 10         0.12544                 max(4)=        6 -> 11         0.01902
                max(3)=        9 -> 21         0.07785                 max(5)=        6 -> 18         0.01743
                max(4)=        9 -> 12         0.05906                 min(1)=        8 -> 11        -0.39084
```

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                min(2)=        7 -> 17        -0.07921                min(4)=        9 -> 27        -0.00431
                min(3)=        8 -> 22        -0.05494                min(5)=        9 -> 19        -0.00324
                min(4)=        7 -> 21        -0.05129                #CIs=29 #CIs>0=12 #CIs<0=17
                min(5)=        9 -> 25        -0.05098
                #CIs=39 #CIs>0=25 #CIs<0=14            14       Singlet-B2   12.1946        101.67    0.3237    0.000
                                                                       max(1)=        7 -> 10         0.60063
11       Singlet-B2   11.3469        109.27    0.0061    0.000         max(2)=        7 -> 12         0.06986
                max(1)=        8 -> 11         0.49666                max(3)=        7 -> 19         0.06028
                max(2)=        8 -> 14         0.42610                max(4)=        8 -> 16         0.02002
                max(3)=        7 -> 10         0.20797                max(5)=        6 -> 16         0.01014
                max(4)=        8 -> 18         0.11406                min(1)=        7 -> 17        -0.22526
                max(5)=        8 -> 16         0.03385                min(2)=        8 -> 14        -0.20293
                min(1)=        7 -> 17        -0.10755                min(3)=        8 -> 11        -0.13718
                min(2)=        7 -> 15        -0.02332                min(4)=        7 -> 15        -0.12838
                min(3)=        9 -> 25        -0.01713                min(5)=        6 -> 11        -0.02808
                min(4)=        7 -> 21        -0.01680                #CIs=39 #CIs>0=18 #CIs<0=21
                min(5)=        6 -> 11        -0.01480
                #CIs=39 #CIs>0=22 #CIs<0=16            15       Singlet-A1   12.5694         98.64    0.0002    0.000
                                                                       max(1)=        8 -> 17         0.46224
12       Singlet-A1   11.7640        105.39    0.0130    0.000         max(2)=        7 -> 11         0.11189
                max(1)=        8 -> 12         0.68494                max(3)=        8 -> 21         0.10632
                max(2)=        8 -> 10         0.09164                max(4)=        8 -> 10         0.09383
                max(3)=        8 -> 19         0.08109                max(5)=        7 -> 22         0.03225
                max(4)=        7 -> 16         0.07525                min(1)=        8 -> 15        -0.45146
                max(5)=        9 -> 13         0.04747                min(2)=        7 -> 16        -0.16463
                min(1)=        7 -> 11        -0.05168                min(3)=        7 -> 14        -0.14176
                min(2)=        9 -> 20        -0.05038                min(4)=        8 -> 23        -0.00950
                min(3)=        7 -> 18        -0.02623                min(5)=        9 -> 20        -0.00835
                min(4)=        7 -> 22        -0.02377                #CIs=47 #CIs>0=22 #CIs<0=25
                min(5)=        8 -> 21        -0.01438
                #CIs=47 #CIs>0=21 #CIs<0=26            16       Singlet-A1   13.3328         92.99    0.0645    0.000
                                                                       max(1)=        8 -> 15         0.46535
13       Singlet-B1   11.8629        104.51    0.0055    0.000         max(2)=        7 -> 11         0.32126
                max(1)=        8 -> 13         0.69367                max(3)=        8 -> 17         0.18482
                max(2)=        8 -> 20         0.11096                max(4)=        8 -> 10         0.17879
                max(3)=        9 -> 17         0.02142                max(5)=        8 -> 23         0.04971
                max(4)=        7 -> 25         0.01109                min(1)=        7 -> 16        -0.22566
                max(5)=        8 -> 26         0.00988                min(2)=        7 -> 14        -0.22559
                min(1)=        9 -> 15        -0.07336                min(3)=        9 -> 20        -0.05126
                min(2)=        9 -> 12        -0.01404                min(4)=        8 -> 19        -0.04961
                min(3)=        6 -> 26        -0.00436                min(5)=        6 -> 12        -0.01471
```

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                #CIs=48 #CIs>0=24 #CIs<0=23            20       Singlet-A2   14.5870         85.00    0.0000    0.000

                                                                       max(1)=        7 -> 13         0.69796
17       Singlet-B2   13.5681         91.38    0.0366    0.000          max(2)=        7 -> 20         0.11248
                max(1)=        8 -> 14         0.32443                 max(3)=        7 -> 26         0.00596
                max(2)=        8 -> 22         0.09473                 max(4)=        8 -> 25         0.00525
                max(3)=        7 -> 10         0.06733                 max(5)=        7 -> 31         0.00521
                max(4)=        7 -> 17         0.04674                 min(1)=        9 -> 16        -0.00456
                max(5)=        7 -> 21         0.03721                 min(2)=        9 -> 14        -0.00449
                min(1)=        8 -> 16        -0.54074                 min(3)=        6 -> 25        -0.00423
                min(2)=        8 -> 11        -0.26899                 min(4)=        9 -> 11        -0.00319
                min(3)=        7 -> 15        -0.09863                 min(5)=        9 -> 22        -0.00304
                min(4)=        7 -> 12        -0.02071                 #CIs=21 #CIs>0=10 #CIs<0=11
                min(5)=        7 -> 23        -0.01363
                #CIs=39 #CIs>0=15 #CIs<0=23            21       Singlet-B2   14.6146         84.84    0.0010    0.000

                                                                       max(1)=        7 -> 12         0.68744
18       Singlet-A1   13.6718         90.69    0.3968    0.000          max(2)=        7 -> 15         0.09721
                max(1)=        8 -> 17         0.40246                 max(3)=        7 -> 19         0.09134
                max(2)=        8 -> 15         0.23207                 max(4)=        6 -> 14         0.02397
                max(3)=        7 -> 16         0.20686                 max(5)=        8 -> 14         0.02176
                max(4)=        8 -> 10         0.19007                 min(1)=        7 -> 10        -0.07295
                max(5)=        9 -> 20         0.16130                 min(2)=        8 -> 16        -0.03018
                min(1)=        7 -> 11        -0.37018                 min(3)=        7 -> 21        -0.02906
                min(2)=        8 -> 12        -0.07097                 min(4)=        8 -> 11        -0.02359
                min(3)=        9 -> 13        -0.06362                 min(5)=        7 -> 17        -0.02095
                min(4)=        7 -> 18        -0.04802                 #CIs=39 #CIs>0=22 #CIs<0=17
                min(5)=        9 -> 26        -0.02918
                #CIs=48 #CIs>0=31 #CIs<0=16            22       Singlet-A2   14.7664         83.96    0.0000    0.000

                                                                       max(1)=        9 -> 18         0.69305
19       Singlet-A1   14.2641         86.92    0.1355    0.000          max(2)=        9 -> 22         0.04602
                max(1)=        7 -> 14         0.51662                 max(3)=        9 -> 28         0.04395
                max(2)=        7 -> 11         0.42603                 max(4)=        9 -> 16         0.01981
                max(3)=        7 -> 16         0.16742                 max(5)=        9 -> 34         0.01715
                max(4)=        8 -> 17         0.11107                 min(1)=        9 -> 11        -0.09065
                max(5)=        7 -> 18         0.08091                 min(2)=        9 -> 14        -0.08078
                min(1)=        9 -> 26        -0.04262                 min(3)=        9 -> 29        -0.01071
                min(2)=        6 -> 10        -0.02424                 min(4)=        6 -> 25        -0.00489
                min(3)=        6 -> 21        -0.01428                 min(5)=        8 -> 25        -0.00470
                min(4)=        7 -> 22        -0.01373                 #CIs=21 #CIs>0=11 #CIs<0=9
                min(5)=        8 -> 23        -0.00971
                #CIs=48 #CIs>0=21 #CIs<0=26            23       Singlet-B1   15.4506         80.25    0.0006    0.000

                                                                       max(1)=        9 -> 19         0.68763
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```
              max(2)=        9 -> 17         0.05786                  max(4)=        9 -> 13         0.05265
              max(3)=        9 -> 35         0.02229                  max(5)=        8 -> 19         0.05120
              max(4)=        9 -> 32         0.02053                  min(1)=        7 -> 16        -0.41110
              max(5)=        9 -> 15         0.01883                  min(2)=        9 -> 20        -0.40704
              min(1)=        9 -> 12        -0.11482                  min(3)=        7 -> 11        -0.22906
              min(2)=        9 -> 21        -0.09230                  min(4)=        8 -> 12        -0.01831
              min(3)=        9 -> 10        -0.01394                  min(5)=        9 -> 33        -0.01294
              min(4)=        6 -> 26        -0.00680                  #CIs=49 #CIs>0=25 #CIs<0=22
              min(5)=        9 -> 39        -0.00208
              #CIs=30 #CIs>0=14 #CIs<0=15            27       Singlet-A1   16.8777         73.46    0.0853    0.000
                                                                      max(1)=        9 -> 20         0.48422
24       Singlet-B2   15.5161         79.91    0.3631    0.000      max(2)=        7 -> 14         0.18679
              max(1)=        7 -> 17         0.45545                  max(3)=        8 -> 12         0.09887
              max(2)=        8 -> 16         0.13245                  max(4)=        8 -> 21         0.06091
              max(3)=        7 -> 12         0.10126                  max(5)=        7 -> 22         0.03031
              max(4)=        7 -> 21         0.07148                  min(1)=        7 -> 16        -0.37106
              max(5)=        7 -> 10         0.06620                  min(2)=        8 -> 19        -0.24936
              min(1)=        7 -> 15        -0.49512                  min(3)=        8 -> 17        -0.08455
              min(2)=        8 -> 18        -0.05767                  min(4)=        9 -> 13        -0.03978
              min(3)=        9 -> 25        -0.05562                  min(5)=        7 -> 18        -0.03846
              min(4)=        7 -> 24        -0.02477                  #CIs=48 #CIs>0=23 #CIs<0=24
              min(5)=        6 -> 16        -0.01968
              #CIs=39 #CIs>0=22 #CIs<0=17            28       Singlet-B2   17.9748         68.98    0.0145    0.000
                                                                      max(1)=        8 -> 18         0.68190
25       Singlet-B2   16.0080         77.45    0.1302    0.000      max(2)=        7 -> 17         0.09406
              max(1)=        7 -> 15         0.45665                  max(3)=        8 -> 22         0.07850
              max(2)=        7 -> 17         0.43819                  max(4)=        8 -> 16         0.05028
              max(3)=        7 -> 10         0.28284                  max(5)=        8 -> 28         0.03581
              max(4)=        7 -> 23         0.04826                  min(1)=        8 -> 11        -0.08703
              max(5)=        7 -> 21         0.03647                  min(2)=        8 -> 14        -0.07103
              min(1)=        7 -> 19        -0.08412                  min(3)=        9 -> 25        -0.03976
              min(2)=        8 -> 18        -0.07901                  min(4)=        8 -> 29        -0.00906
              min(3)=        6 -> 16        -0.01208                  min(5)=        7 -> 19        -0.00605
              min(4)=        7 -> 12        -0.01007                  #CIs=38 #CIs>0=23 #CIs<0=15
              min(5)=        7 -> 32        -0.00693
              #CIs=40 #CIs>0=19 #CIs<0=20            29       Singlet-B1   18.6697         66.41    0.0598    0.000
                                                                      max(1)=        9 -> 21         0.64725
26       Singlet-A1   16.1480         76.78    0.0166    0.000      max(2)=        9 -> 19         0.09579
              max(1)=        7 -> 14         0.30409                  max(3)=        8 -> 26         0.05518
              max(2)=        7 -> 22         0.07599                  max(4)=        9 -> 24         0.04728
              max(3)=        7 -> 18         0.06333                  max(5)=        7 -> 25         0.04344
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              min(1)=        8 -> 20        -0.23127                 min(3)=        8 -> 12        -0.05243
              min(2)=        9 -> 17        -0.07871                 min(4)=        7 -> 22        -0.05164
              min(3)=        9 -> 23        -0.03743                 min(5)=        8 -> 17        -0.04092
              min(4)=        6 -> 20        -0.01985                 #CIs=49 #CIs>0=24 #CIs<0=23
              min(5)=        9 -> 36        -0.01244
              #CIs=29 #CIs>0=12 #CIs<0=17            33       Singlet-A1   21.1122         58.73    0.0139    0.000
                                                                max(1)=        7 -> 18         0.54241
30       Singlet-A2   18.9977         65.26    0.0000    0.000 max(2)=        8 -> 21         0.36841
              max(1)=        9 -> 22         0.68842           max(3)=        8 -> 19         0.19227
              max(2)=        9 -> 16         0.14728           max(4)=        9 -> 20         0.09807
              max(3)=        9 -> 34         0.02703           max(5)=        8 -> 15         0.04426
              max(4)=        6 -> 30         0.00485           min(1)=        9 -> 26        -0.06394
              max(5)=        7 -> 13         0.00460           min(2)=        8 -> 23        -0.05659
              min(1)=        9 -> 18        -0.04861           min(3)=        8 -> 17        -0.05002
              min(2)=        9 -> 28        -0.03300           min(4)=        7 -> 11        -0.04866
              min(3)=        8 -> 25        -0.00660           min(5)=        7 -> 14        -0.04314
              min(4)=        9 -> 29        -0.00606           #CIs=49 #CIs>0=25 #CIs<0=22
              min(5)=        9 -> 37        -0.00514
              #CIs=21 #CIs>0=8 #CIs<0=13             34       Singlet-B2   21.5193         57.62    0.0041    0.000
                                                                max(1)=        7 -> 19         0.68470
31       Singlet-B1   19.0207         65.18    0.0035    0.000 max(2)=        7 -> 17         0.08298
              max(1)=        8 -> 20         0.65655           max(3)=        7 -> 15         0.05569
              max(2)=        9 -> 21         0.22658           max(4)=        6 -> 16         0.05139
              max(3)=        7 -> 25         0.03447           max(5)=        6 -> 14         0.04631
              max(4)=        8 -> 26         0.03269           min(1)=        7 -> 12        -0.10098
              max(5)=        9 -> 24         0.02881           min(2)=        9 -> 25        -0.04436
              min(1)=        8 -> 13        -0.10860           min(3)=        7 -> 21        -0.03604
              min(2)=        9 -> 17        -0.03139           min(4)=        7 -> 24        -0.02160
              min(3)=        6 -> 26        -0.01069           min(5)=        7 -> 10        -0.01446
              min(4)=        9 -> 15        -0.00857           #CIs=39 #CIs>0=23 #CIs<0=16
              min(5)=        9 -> 36        -0.00517
              #CIs=29 #CIs>0=13 #CIs<0=15            35       Singlet-A2   21.7805         56.92    0.0000    0.000
                                                                max(1)=        7 -> 20         0.69685
32       Singlet-A1   19.3606         64.04    0.0057    0.000 max(2)=        7 -> 33         0.02330
              max(1)=        8 -> 19         0.60591           max(3)=        7 -> 31         0.02098
              max(2)=        9 -> 20         0.17066           max(4)=        7 -> 26         0.01511
              max(3)=        7 -> 11         0.06404           max(5)=        8 -> 25         0.01491
              max(4)=        7 -> 14         0.05160           min(1)=        7 -> 13        -0.11293
              max(5)=        9 -> 26         0.03822           min(2)=        6 -> 25        -0.01006
              min(1)=        7 -> 18        -0.25028           min(3)=        8 -> 30        -0.00474
              min(2)=        7 -> 16        -0.13781           min(4)=        9 -> 18        -0.00455
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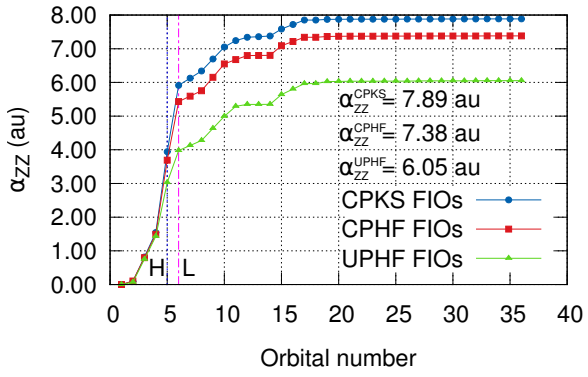
              min(5)=        9 -> 28        -0.00298                  max(5)=        6 -> 12         0.04296
              #CIs=21 #CIs>0=11 #CIs<0=10                              min(1)=        6 -> 17        -0.21465
                                                                       min(2)=        6 -> 15        -0.10396
36      Singlet-A1   21.9704         56.43    0.0001    0.000        min(3)=        9 -> 20        -0.06308
              max(1)=        8 -> 21         0.56708                  min(4)=        7 -> 18        -0.05537
              max(2)=        7 -> 22         0.06851                  min(5)=        8 -> 19        -0.04711
              max(3)=        6 -> 17         0.06815                  #CIs=49 #CIs>0=22 #CIs<0=25
              max(4)=        7 -> 16         0.06610
              max(5)=        8 -> 27         0.05054   39      Singlet-B2   23.1222         53.62    0.0003    0.000
              min(1)=        7 -> 18        -0.33552                  max(1)=        6 -> 16         0.46672
              min(2)=        6 -> 10        -0.12972                  max(2)=        6 -> 14         0.40758
              min(3)=        9 -> 20        -0.10719                  max(3)=        7 -> 23         0.01291
              min(4)=        8 -> 19        -0.08926                  max(4)=        7 -> 17         0.00920
              min(5)=        8 -> 17        -0.07249                  max(5)=        7 -> 27         0.00741
              #CIs=49 #CIs>0=20 #CIs<0=27                              min(1)=        6 -> 11        -0.30483
                                                                       min(2)=        6 -> 22        -0.08648
37      Singlet-B2   22.2530         55.72    0.0070    0.000        min(3)=        9 -> 25        -0.08632
              max(1)=        8 -> 22         0.67426                  min(4)=        7 -> 19        -0.07349
              max(2)=        8 -> 16         0.11639                  min(5)=        8 -> 22        -0.03450
              max(3)=        9 -> 25         0.11134                  #CIs=38 #CIs>0=16 #CIs<0=22
              max(4)=        7 -> 21         0.06162
              max(5)=        6 -> 14         0.04432   40      Singlet-B1   23.2765         53.27    0.0099    0.000
              min(1)=        8 -> 18        -0.07692                  max(1)=        9 -> 24         0.64169
              min(2)=        7 -> 17        -0.03774                  max(2)=        6 -> 13         0.01962
              min(3)=        7 -> 23        -0.03600                  max(3)=        9 -> 35         0.01888
              min(4)=        8 -> 28        -0.02761                  max(4)=        6 -> 20         0.01100
              min(5)=        6 -> 28        -0.01376                  max(5)=        9 -> 27         0.00959
              #CIs=39 #CIs>0=20 #CIs<0=19                              min(1)=        9 -> 23        -0.28361
                                                                       min(2)=        9 -> 21        -0.05587
38      Singlet-A1   23.1118         53.65    0.0167    0.000        min(3)=        8 -> 26        -0.03849
              max(1)=        6 -> 10         0.63434                  min(4)=        7 -> 25        -0.03128
              max(2)=        8 -> 21         0.10634                  min(5)=        9 -> 32        -0.02920
              max(3)=        7 -> 22         0.10216                  #CIs=30 #CIs>0=17 #CIs<0=12
              max(4)=        9 -> 26         0.05384
```

9 NH_3

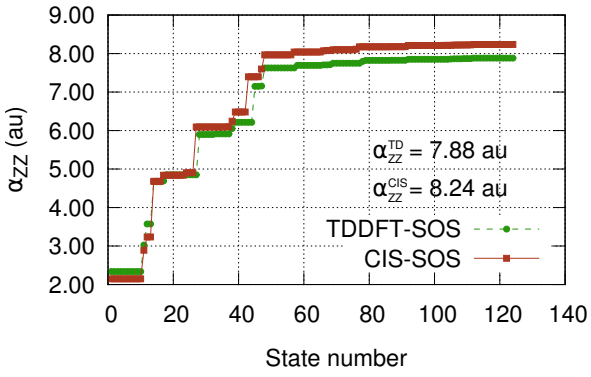
9.1 6-311G(d,p)

9.1.1 Plots

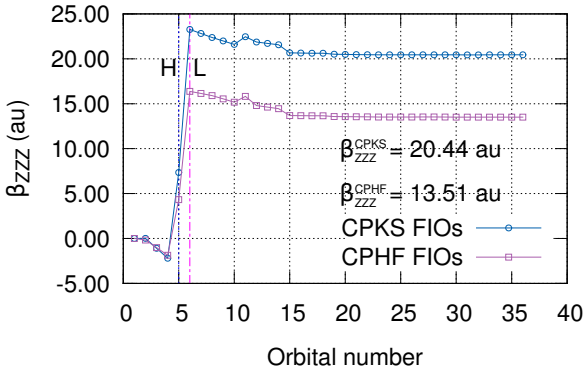
Figure S13: For NH_3 molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S13a and S13c) or states (SOS approaches, in Plots S13b and S13d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.02 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



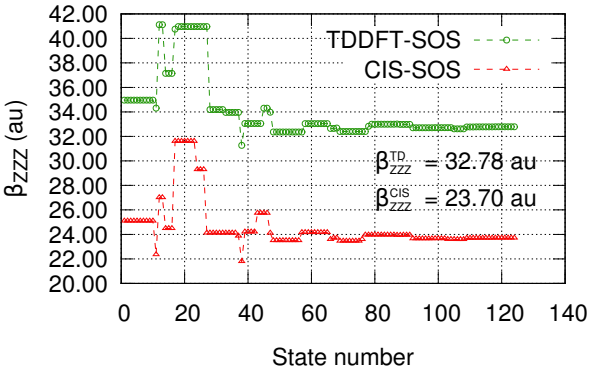
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

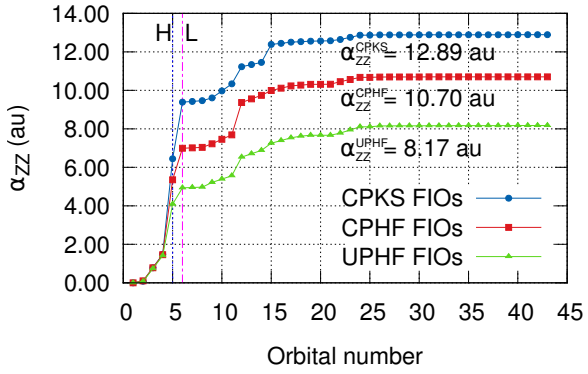


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

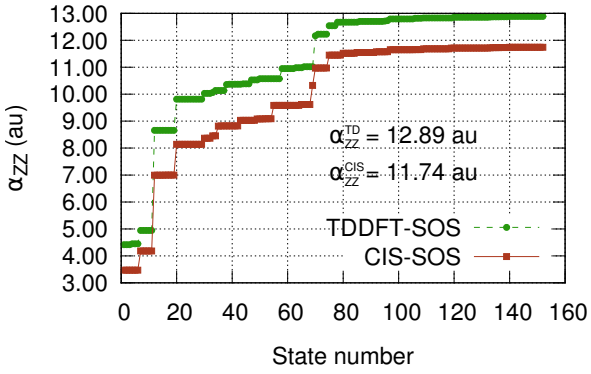
9.2 6-311++G(d,p)

9.2.1 Plots

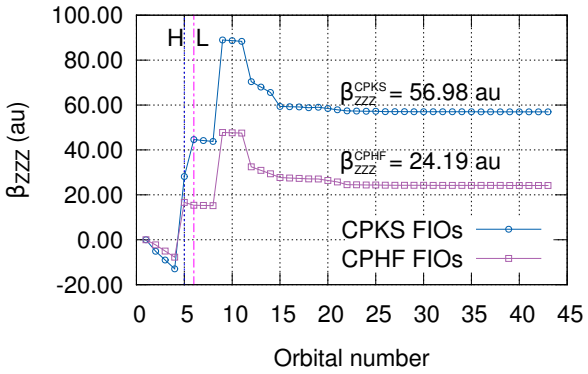
Figure S14: For NH_3 molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S14a and S14c) or states (SOS approaches, in Plots S14b and S14d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.12 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



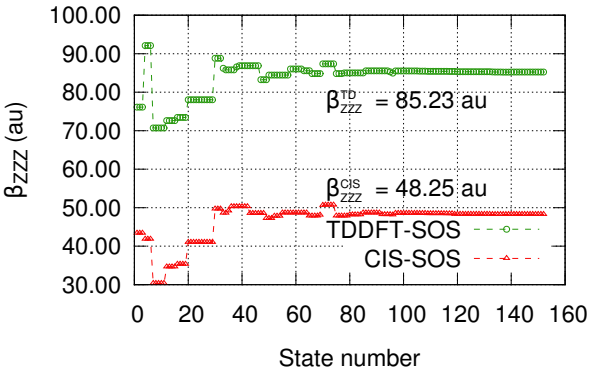
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



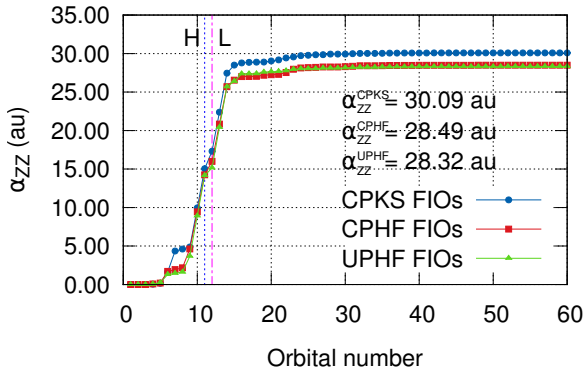
(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

10 F-CC-H

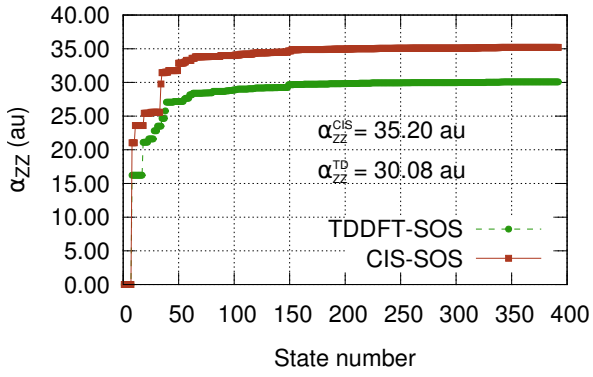
10.1 6-311G(d,p)

10.1.1 Plots

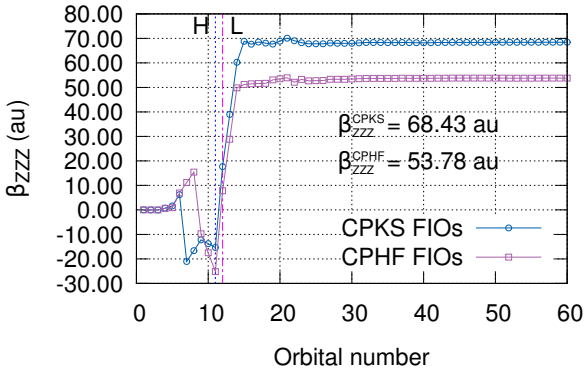
Figure S15: For F−C≡C−H molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S15a and S15c) or states (SOS approaches, in Plots S15b and S15d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.03 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



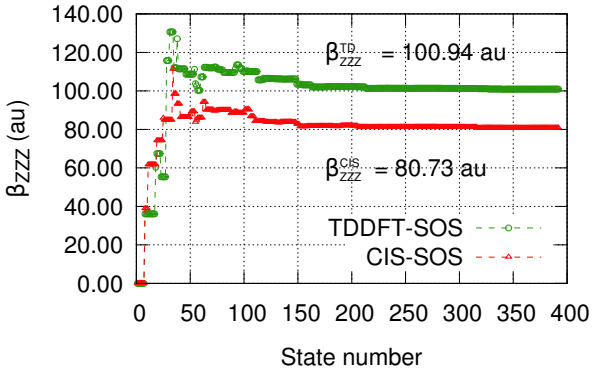
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

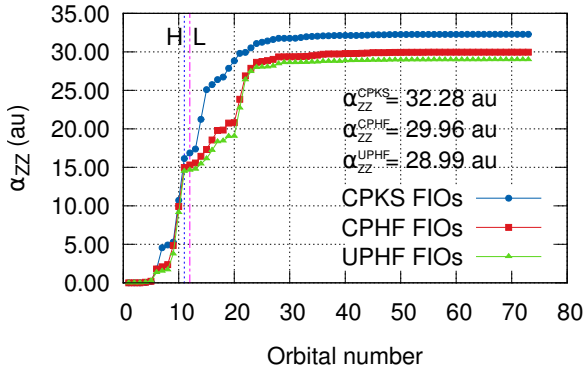


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

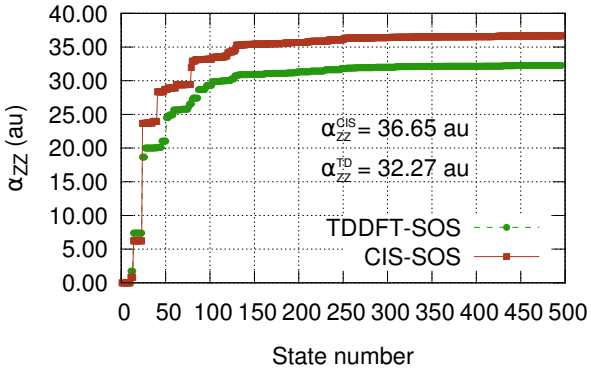
10.2 6-311++G(d,p)

10.2.1 Plots

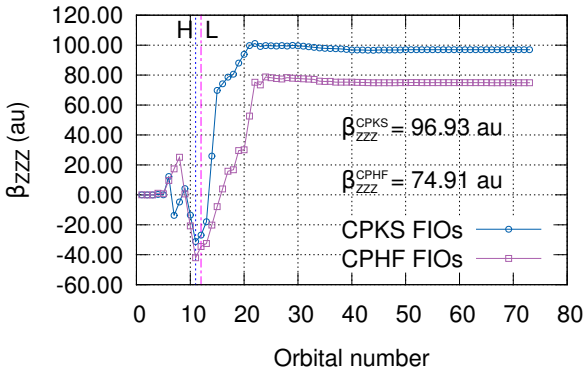
Figure S16: For F−C≡C−H molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (bottom) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S16a and S16c) or states (SOS approaches, in Plots S16b and S16d). For α FIOs, three approaches were considered: CPKS (CAM-B3LYP), CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. All elements of the β tensor were recomputed with an error less than 0.03 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



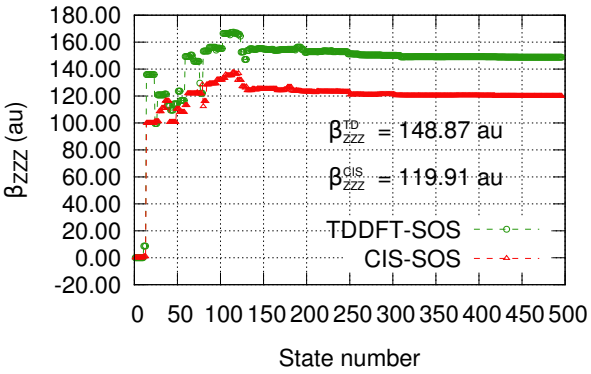
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.



(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.



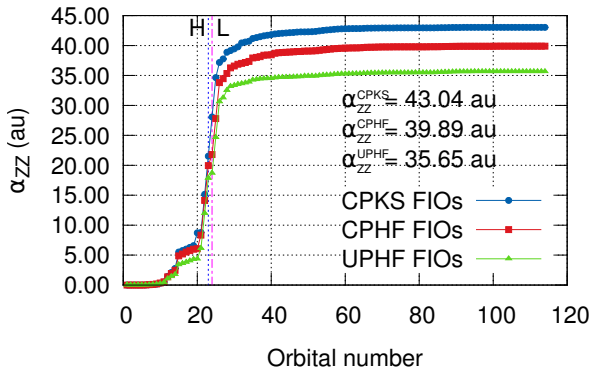
(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

11 $HCCCF_3$

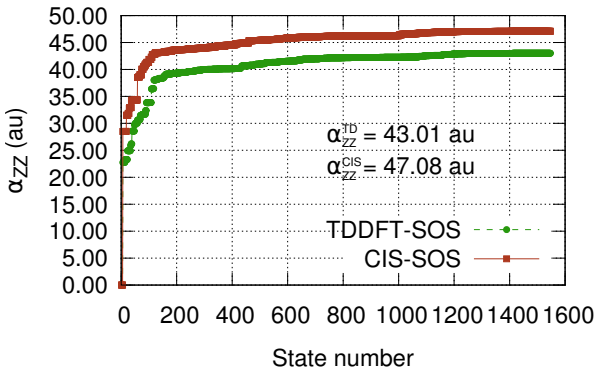
11.1 6-311G(d,p)

11.1.1 Plots

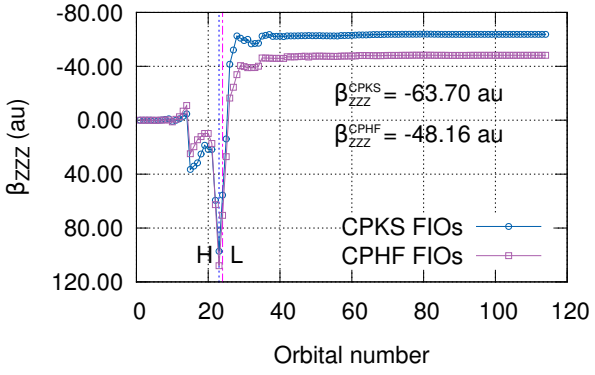
Figure S17: For H−C≡C−CF_3 molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S17a and S17b) or states (SOS approaches, in Plots S17c and S17d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S17e and S17f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.20 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



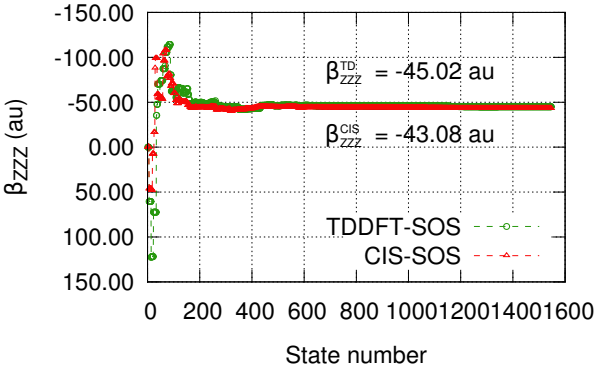
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

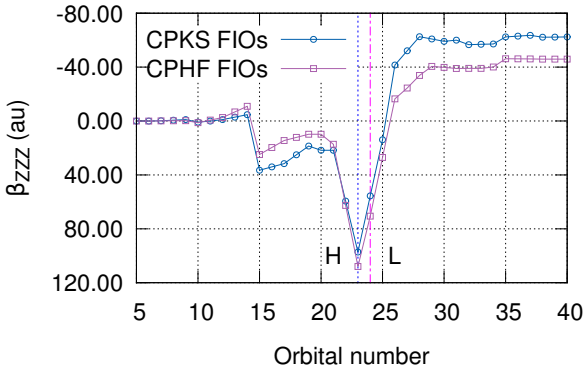


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

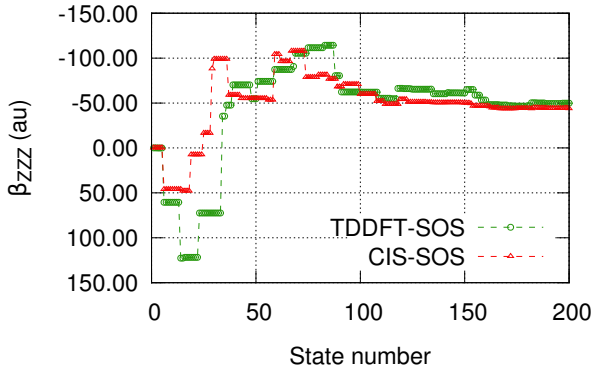


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S17: (continued) For H−C≡C−CF_3 molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S17a and S17b) or states (SOS approaches, in Plots S17c and S17d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S17e and S17f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.20 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 5 and 40.

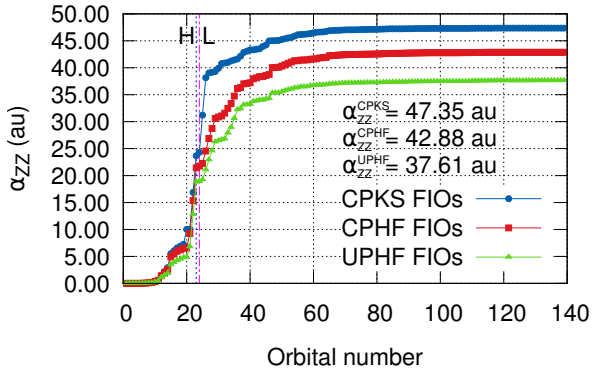


(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 200.

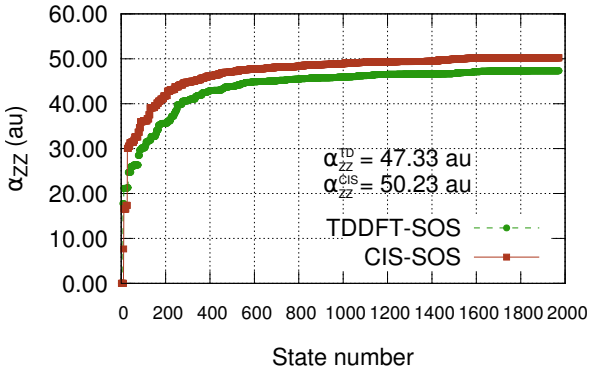
11.2 6-311++G(d,p)

11.2.1 Plots

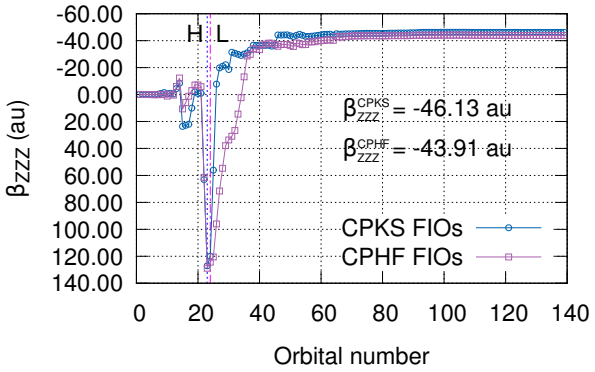
Figure S18: For H−C≡C−CF_3 molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S18a and S18b) or states (SOS approaches, in Plots S18c and S18d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S18e and S18f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.20 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



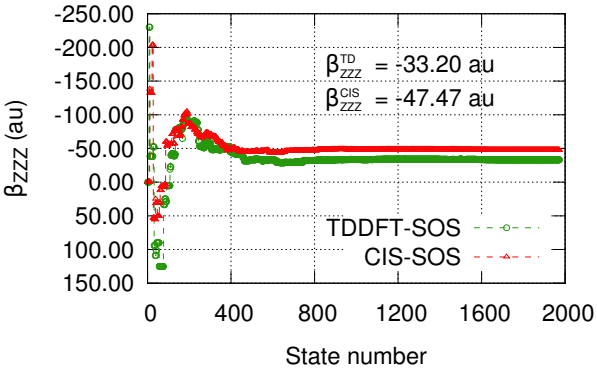
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

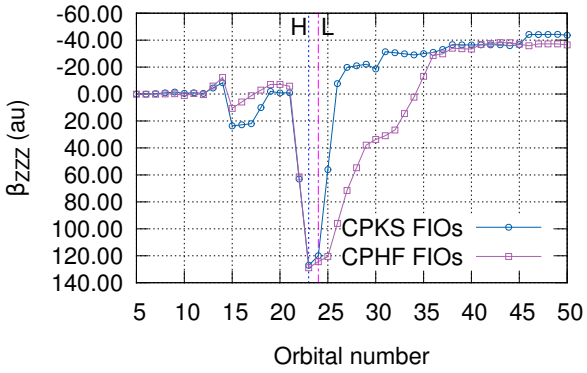


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

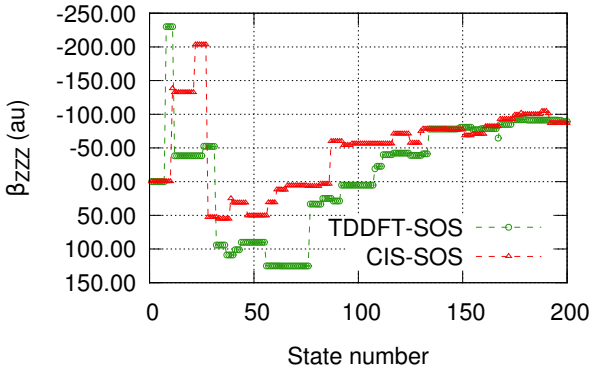


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S18: (continued) For H−C≡C−CF_3 molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S18a and S18b) or states (SOS approaches, in Plots S18c and S18d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S18e and S18f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.20 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 5 and 50.



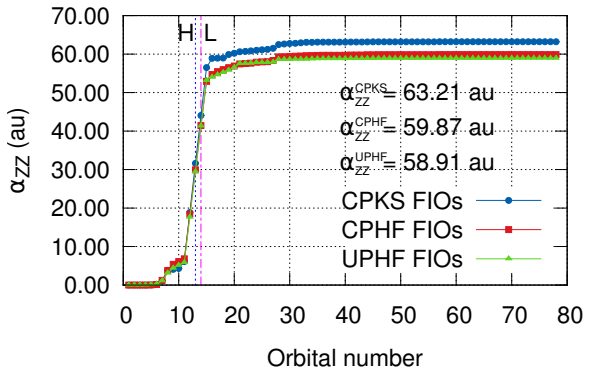
(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 200.

12 CN-CC-H

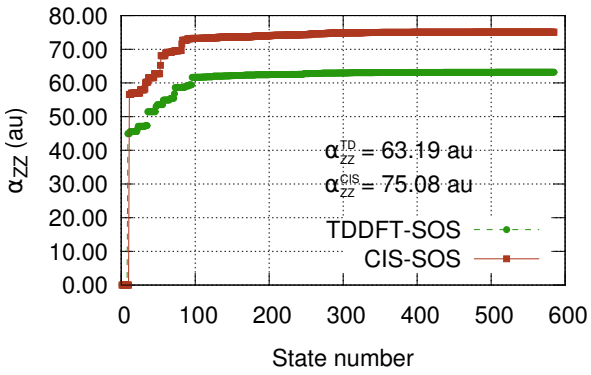
12.1 6-311G(d,p)

12.1.1 Plots

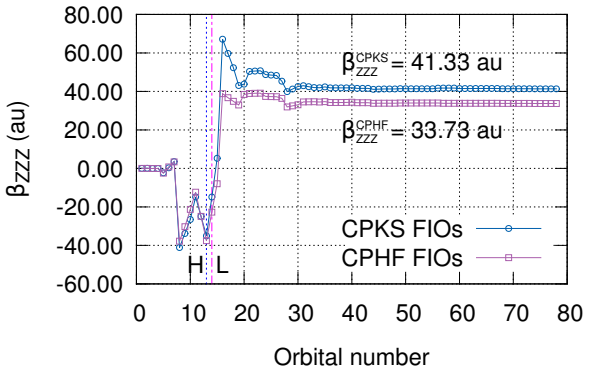
Figure S19: For CN−C≡C−H molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S19a and S19b) or states (SOS approaches, in Plots S19c and S19d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S19e and S19f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.13 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



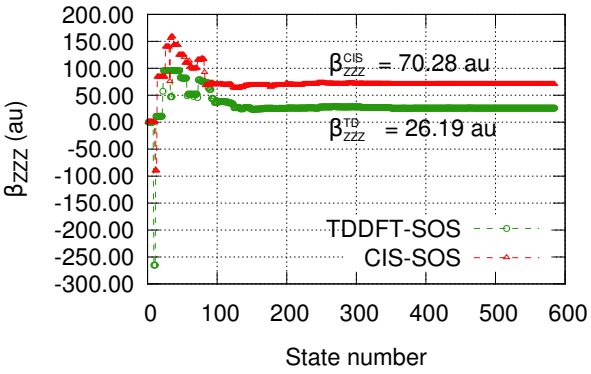
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

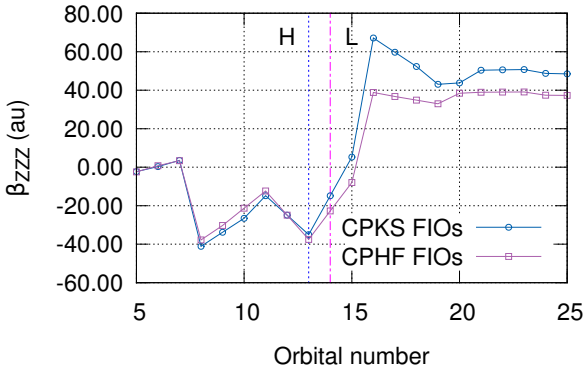


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

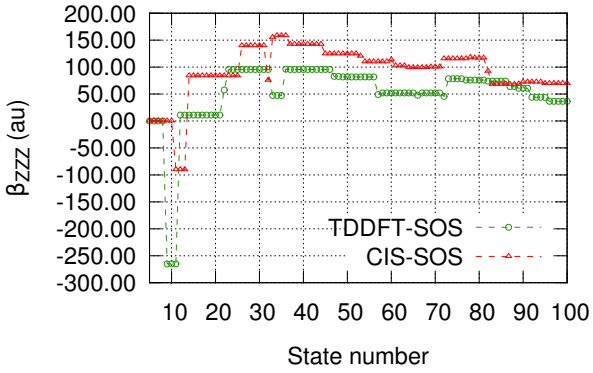


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S19: (continued) For CN−C≡C−H molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S19a and S19b) or states (SOS approaches, in Plots S19c and S19d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S19e and S19f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.13 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 5 and 25.

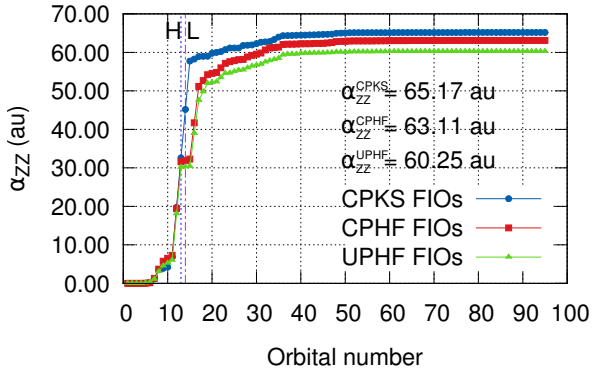


(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 5 and 100.

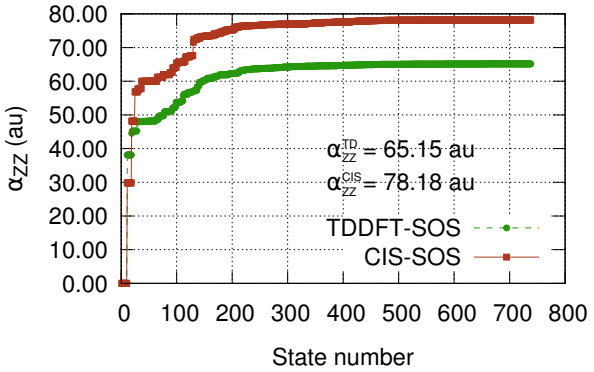
12.2 6-311++G(d,p)

12.2.1 Plots

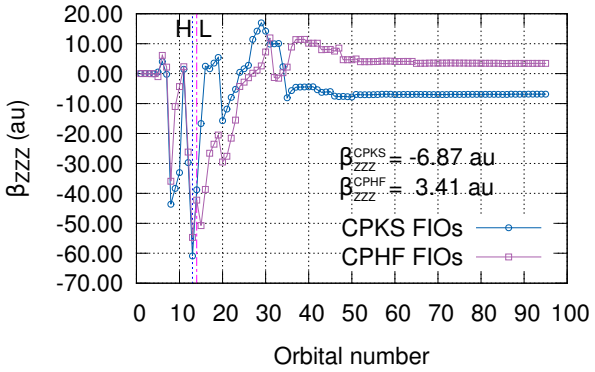
Figure S20: For CN−C≡C−H molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S20a and S20b) or states (SOS approaches, in Plots S20c and S20d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S20e and S20f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.13 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



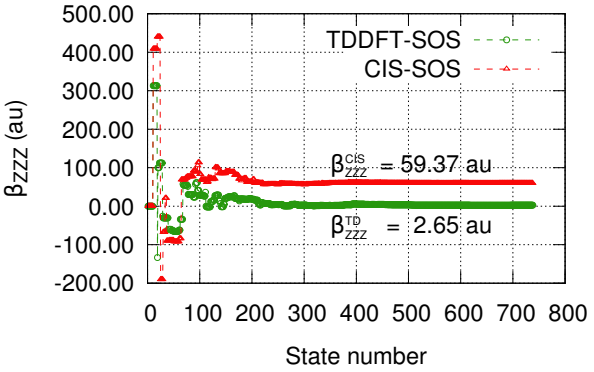
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

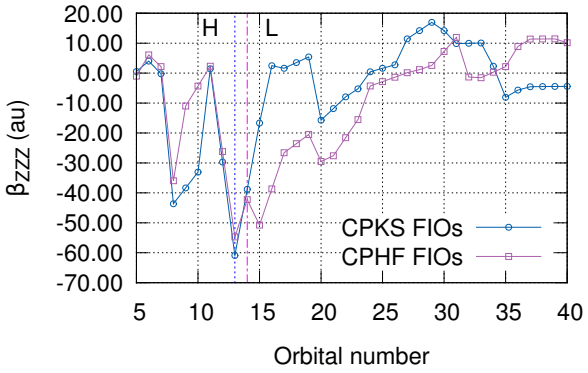


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

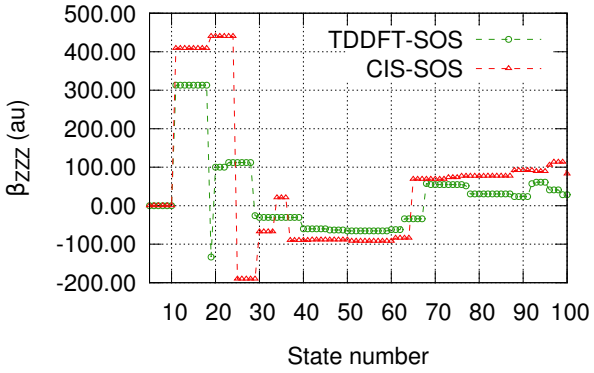


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S20: (continued) For CN−C≡C−H molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S20a and S20b) or states (SOS approaches, in Plots S20c and S20d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S20e and S20f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.13 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 5 and 40.



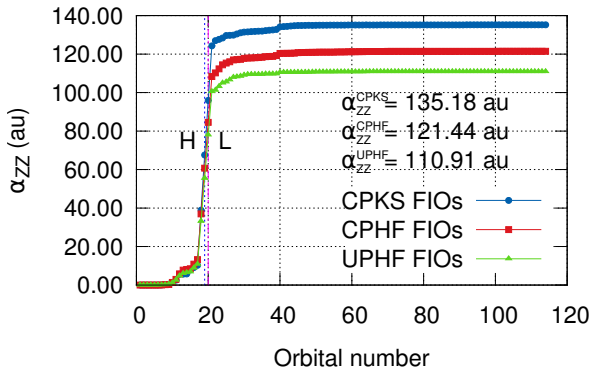
(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 5 and 100.

13 CN-CC-CC-H

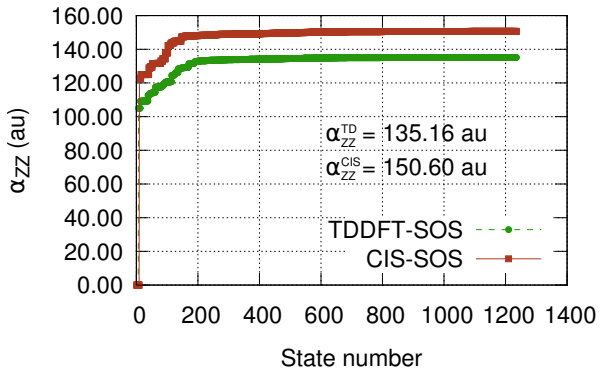
13.1 6-311G(d,p)

13.1.1 Plots

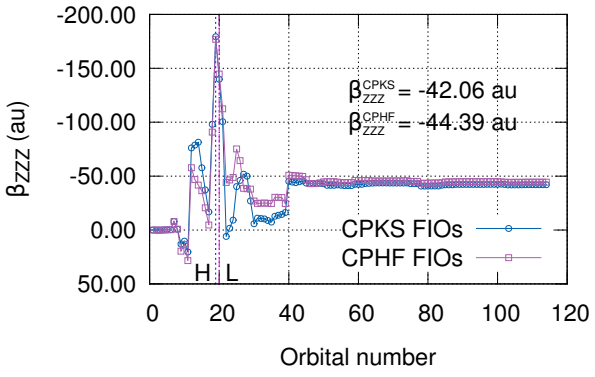
Figure S21: For CN−C≡C−C≡C−H molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S21a and S21b) or states (SOS approaches, in Plots S21c and S21d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S21e and S21f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.12 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



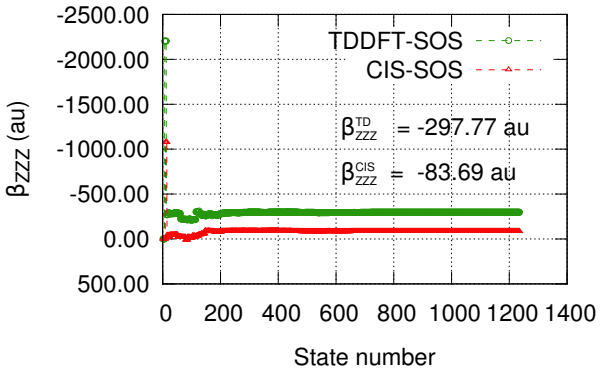
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

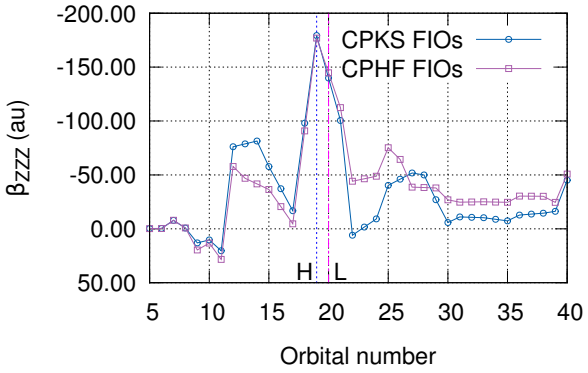


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

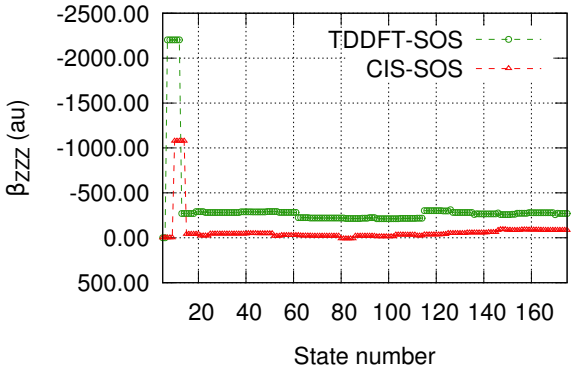


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S21: (continued) For CN−C≡C−C≡C−H molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S21a and S21b) or states (SOS approaches, in Plots S21c and S21d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S21e and S21f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.12 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 5 and 40.

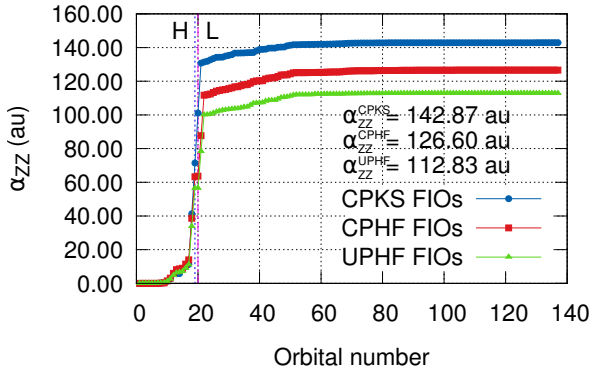


(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 5 and 175.

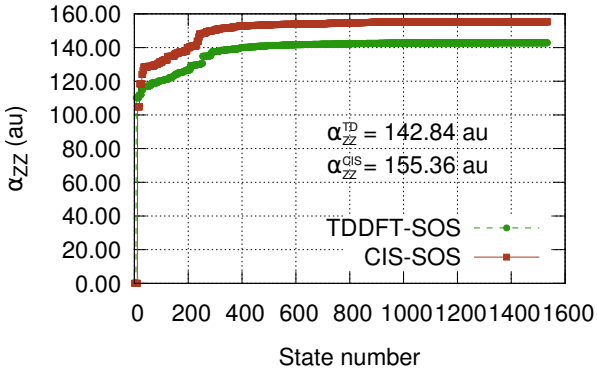
13.2 6-311++G(d,p)

13.2.1 Plots

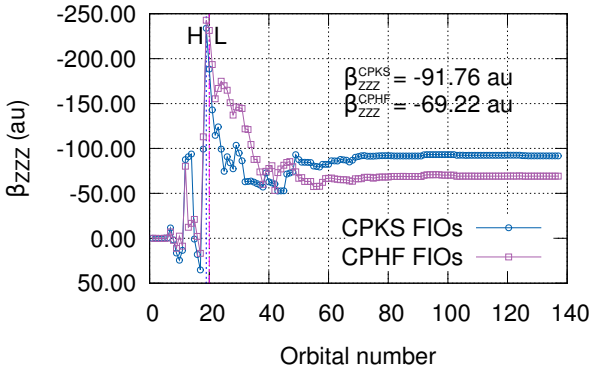
Figure S22: For CN−C≡C−C≡C−H molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S22a and S22b) or states (SOS approaches, in Plots S22c and S22d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S22e and S22f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.24 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



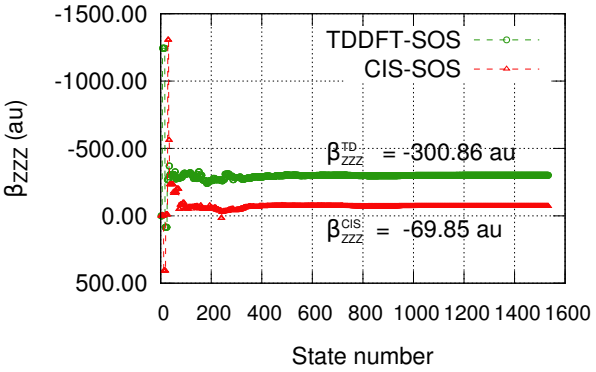
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

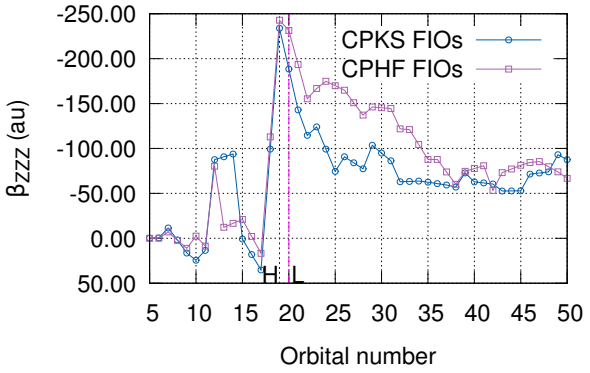


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

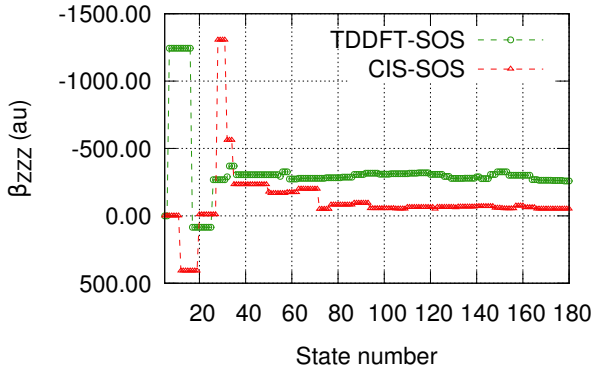


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S22: (continued) For CN−C≡C−C≡C−H molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S22a and S22b) or states (SOS approaches, in Plots S22c and S22d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S22e and S22f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.24 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 5 and 40.



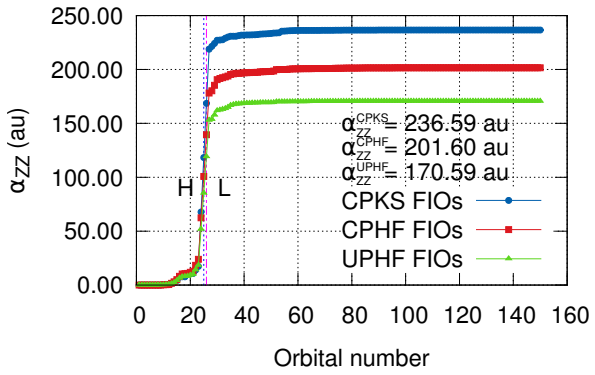
(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 5 and 175.

14 CN-CC-CC-CC-H

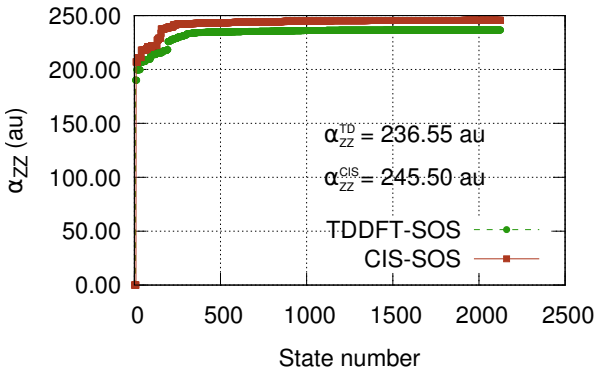
14.1 6-311G(d,p)

14.1.1 Plots

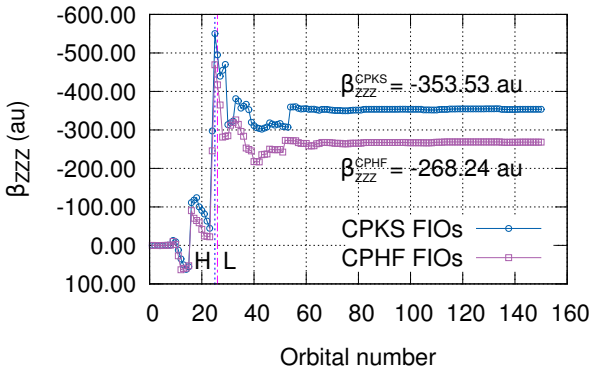
Figure S23: For CN−C≡C−C≡C−C≡C−H molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S23a and S23b) or states (SOS approaches, in Plots S23c and S23d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S23e and S23f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.41 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



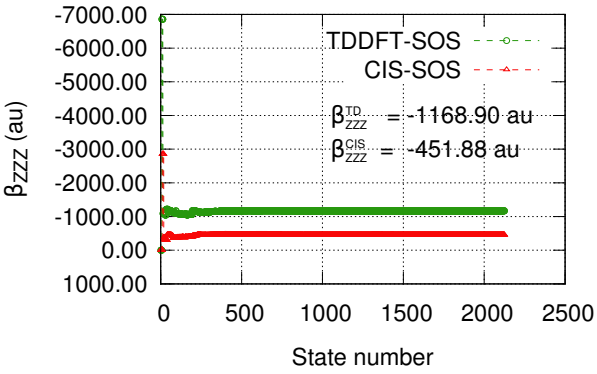
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

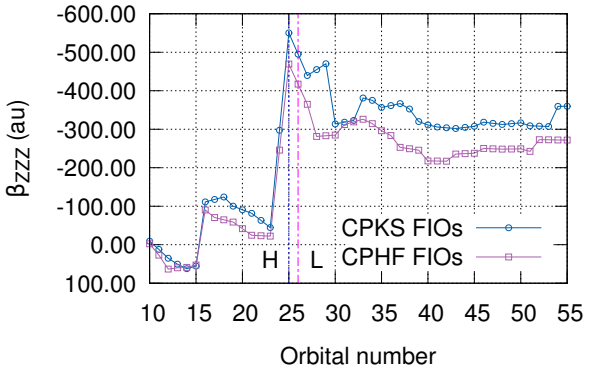


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

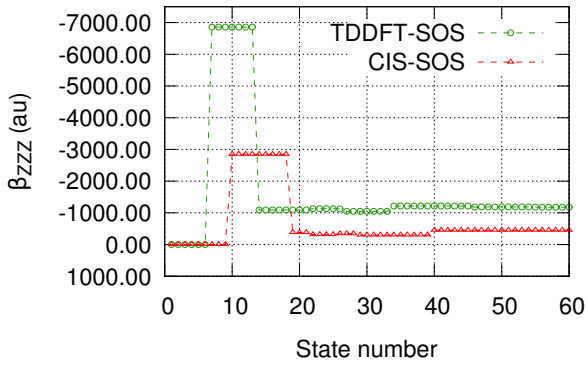


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S23: (continued) For CN−C≡C−C≡C−C≡C−H molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S23a and S23b) or states (SOS approaches, in Plots S23c and S23d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S23e and S23f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.41 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 10 and 55.

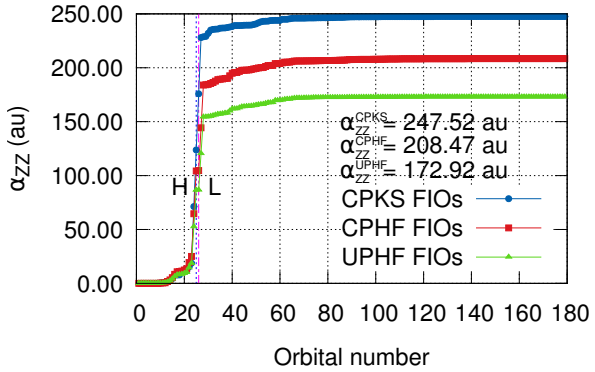


(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 60.

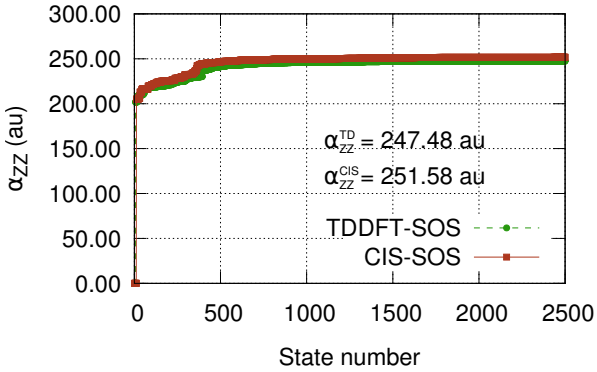
14.2 6-311++G(d,p)

14.2.1 Plots

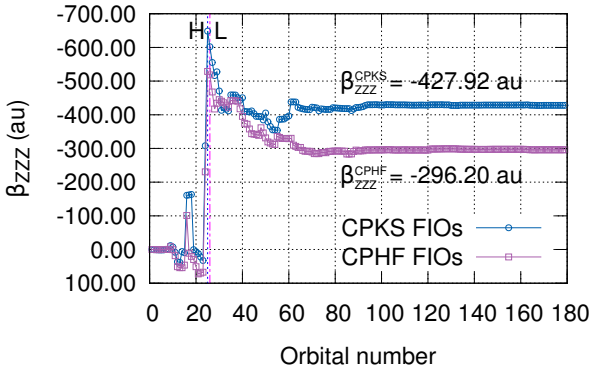
Figure S24: For CN−C≡C−C≡C−C≡C−H molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S24a and S24b) or states (SOS approaches, in Plots S24c and S24d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S24e and S24f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.21 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



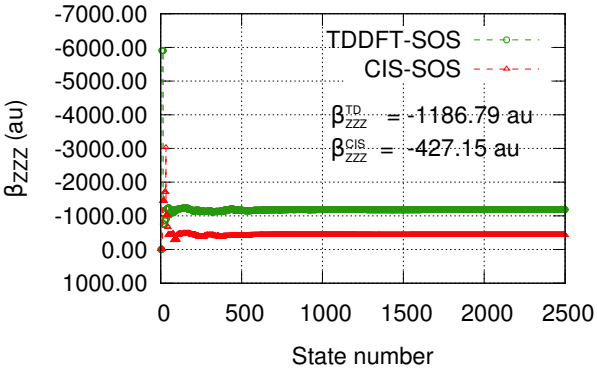
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

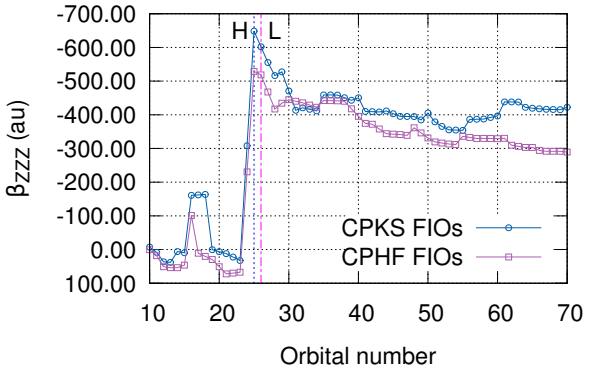


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

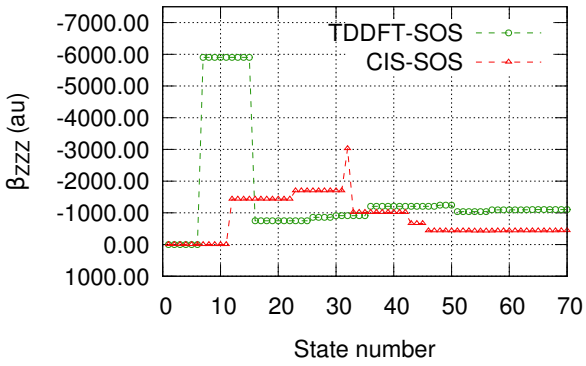


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S24: (continued) For CN−C≡C−C≡C−C≡C−H molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S24a and S24b) or states (SOS approaches, in Plots S24c and S24d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S24e and S24f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.21 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 10 and 70.



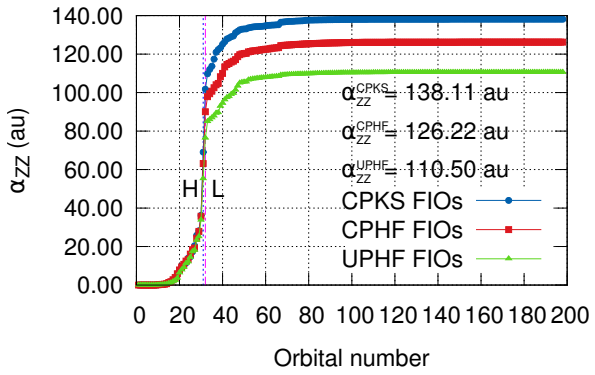
(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 70.

15 *p*-cyanoaniline

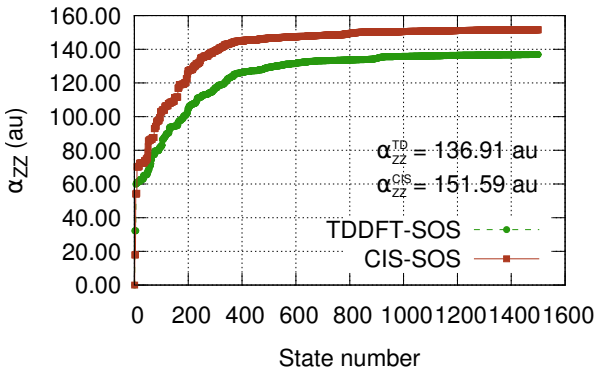
15.1 6-311G(d,p)

15.1.1 Plots

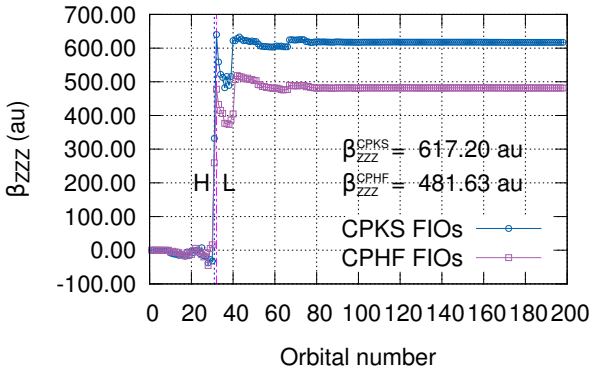
Figure S25: For *p*-cyanoaniline molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S25a and S25b) or states (SOS approaches, in Plots S25c and S25d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S25d and S25f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.11 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



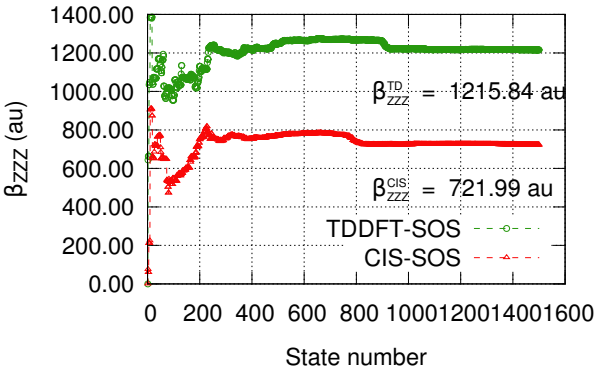
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

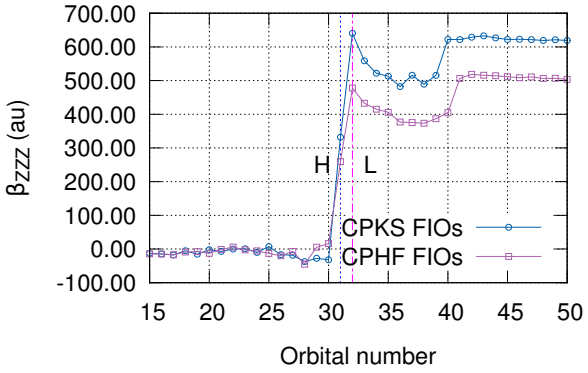


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

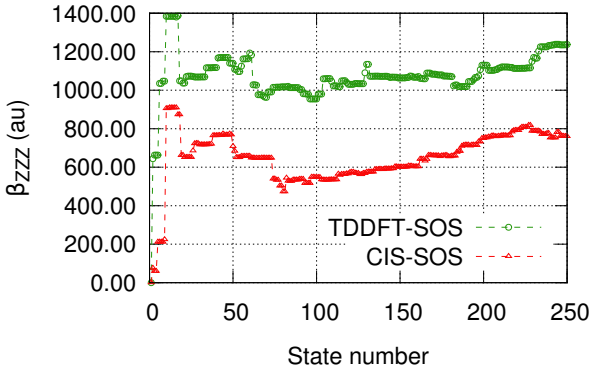


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S25: (continued) For *p*-cyanoaniline molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S25a and S25b) or states (SOS approaches, in Plots S25c and S25d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S25d and S25f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.11 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 15 and 50.

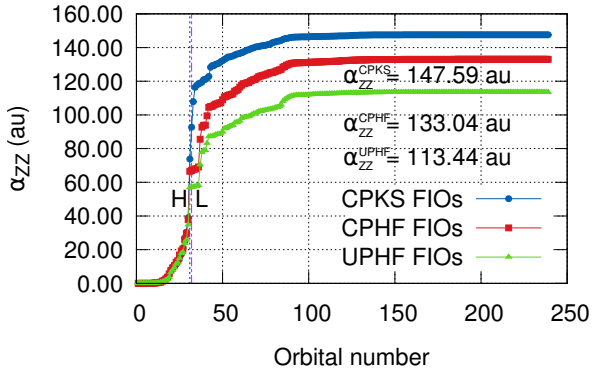


(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 250.

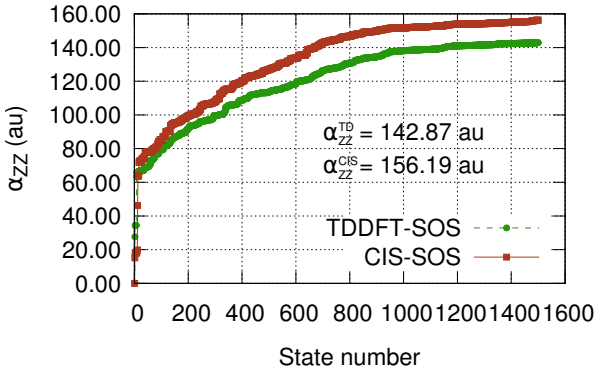
15.2 6-311++G(d,p)

15.2.1 Plots

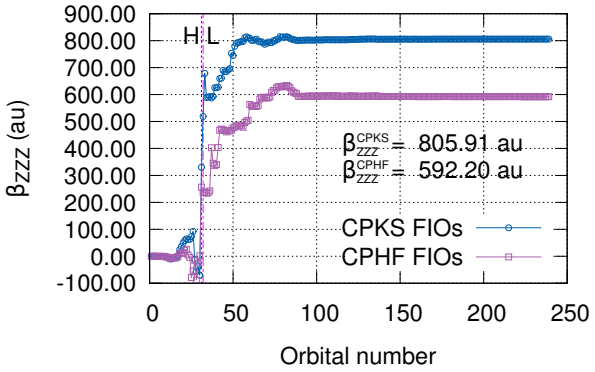
Figure S26: For *p*-cyanoaniline molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S26a and S26b) or states (SOS approaches, in Plots S26c and S26d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S26d and S26f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.11 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



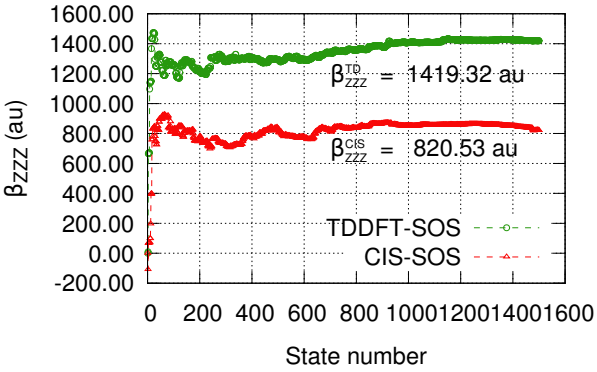
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

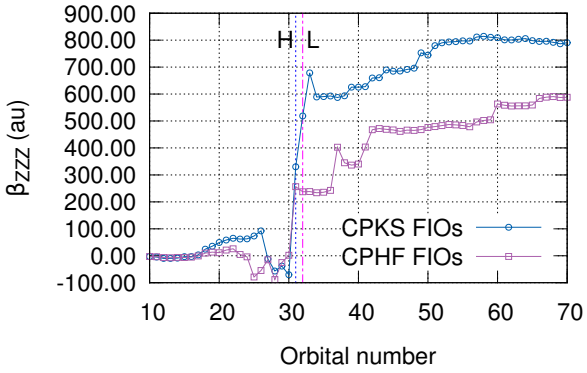


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

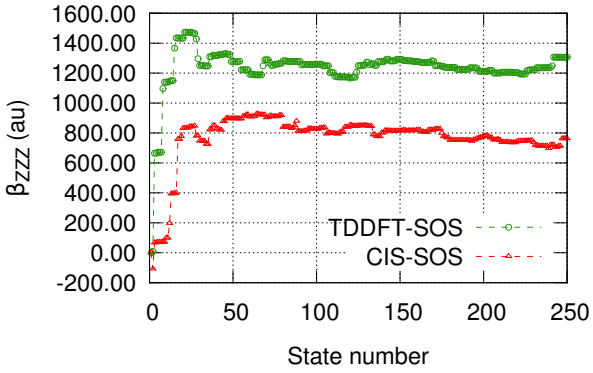


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S26: (continued) For *p*-cyanoaniline molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S26a and S26b) or states (SOS approaches, in Plots S26c and S26d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S26d and S26f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.11 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 15 and 50.



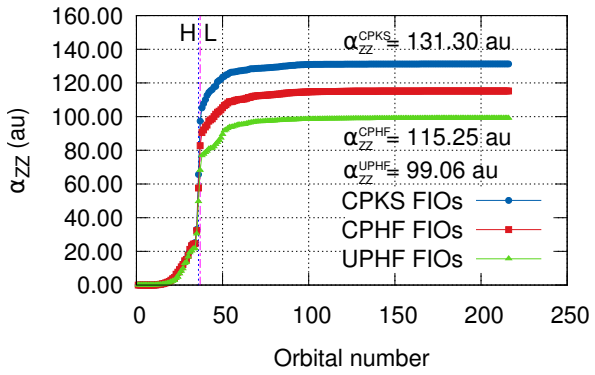
(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 250.

16 *p*-nitroaniline

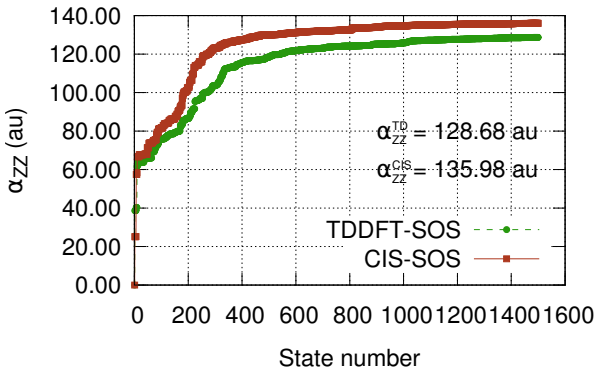
16.1 6-311G(d,p)

16.1.1 Plots

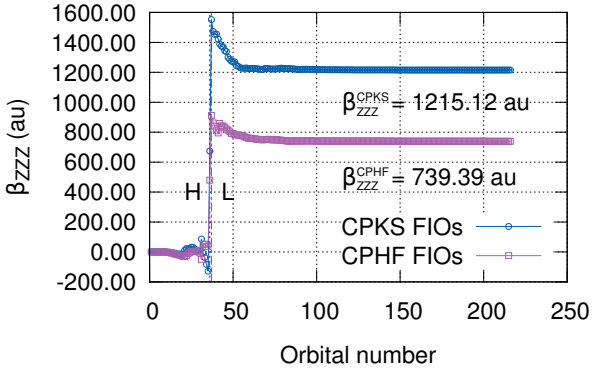
Figure S27: For *p*-nitroaniline molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S27a and S27b) or states (SOS approaches, in Plots S27c and S27d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S27e and S27f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.37 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



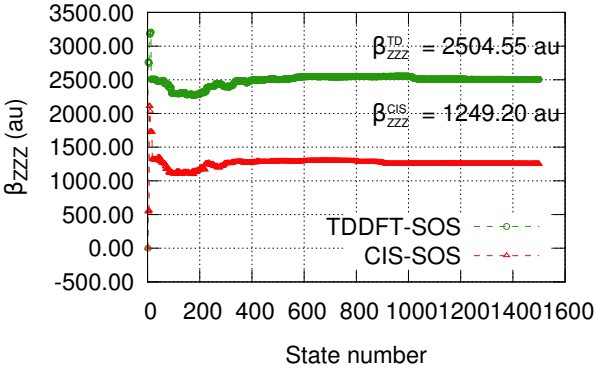
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

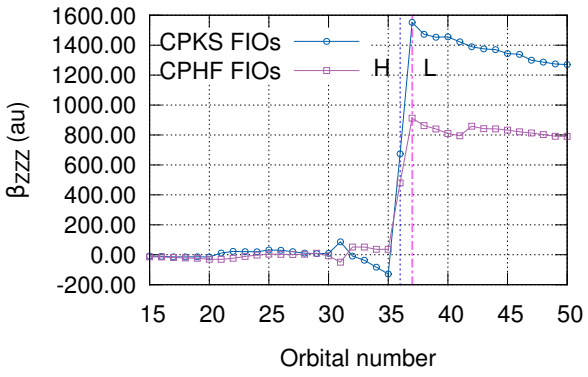


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

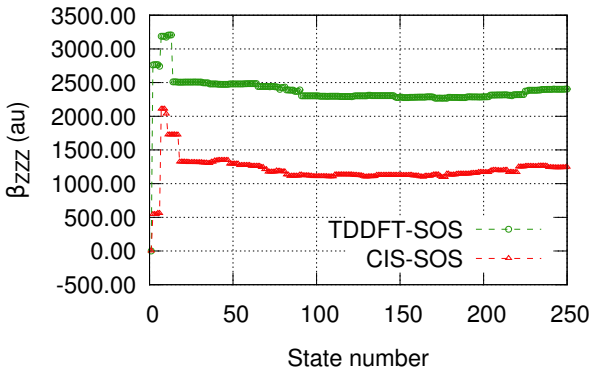


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S27: (continued) For *p*-nitroaniline molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S27a and S27b) or states (SOS approaches, in Plots S27c and S27d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S27e and S27f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.37 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 15 and 50.



(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 250.

16.1.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_
                                                    2        Singlet-A'    4.5920        270.00    0.3670    0.000
1        Singlet-A"    4.1147        301.32    0.0000    0.000          max(1)=       36 -> 37         0.68965
                max(1)=       34 -> 37         0.67431                  max(2)=       35 -> 38         0.10043
                max(2)=       27 -> 40         0.01118                  max(3)=       31 -> 40         0.04187
                max(3)=       26 -> 37         0.01054                  max(4)=       31 -> 39         0.03005
                max(4)=       30 -> 37         0.00937                  max(5)=       29 -> 37         0.01343
                max(5)=       27 -> 39         0.00637                  min(1)=       31 -> 37        -0.09692
                min(1)=       34 -> 40        -0.17798                  min(2)=       30 -> 50        -0.02180
                min(2)=       34 -> 39        -0.10948                  min(3)=       32 -> 47        -0.01837
                min(3)=       34 -> 46        -0.02774                  min(4)=       32 -> 37        -0.01730
                min(4)=       27 -> 37        -0.01731                  min(5)=       34 -> 45        -0.01695
                min(5)=       34 -> 42        -0.01418                  #CIs=2384 #CIs>0=1051 #CIs<0=1061
                #CIs=2195 #CIs>0=781 #CIs<0=803
```

```
3       Singlet-A'    4.6429        267.04    0.0004    0.000          max(2)=       31 -> 39         0.11595
              max(1)=       32 -> 37         0.67546                  max(3)=       35 -> 38         0.05525
              max(2)=       25 -> 37         0.02320                  max(4)=       31 -> 37         0.03750
              max(3)=       28 -> 37         0.01979                  max(5)=       29 -> 39         0.01269
              max(4)=       36 -> 37         0.01789                  min(1)=       36 -> 40        -0.16744
              max(5)=       29 -> 37         0.01470                  min(2)=       31 -> 40        -0.06675
              min(1)=       32 -> 40        -0.17158                  min(3)=       36 -> 42        -0.04391
              min(2)=       32 -> 39        -0.10632                  min(4)=       31 -> 42        -0.04329
              min(3)=       32 -> 46        -0.02523                  min(5)=       35 -> 41        -0.04238
              min(4)=       25 -> 40        -0.01736                  #CIs=2380 #CIs>0=1048 #CIs<0=994
              min(5)=       32 -> 42        -0.01392
              #CIs=2370 #CIs>0=920 #CIs<0=932         7       Singlet-A'    6.6277        187.07    0.0327    0.000
                                                                     max(1)=       36 -> 40         0.53002
4       Singlet-A"    4.9397        251.00    0.0078    0.000          max(2)=       35 -> 38         0.42128
              max(1)=       36 -> 38         0.56495                  max(3)=       31 -> 37         0.11707
              max(2)=       33 -> 37         0.06008                  max(4)=       36 -> 39         0.09771
              max(3)=       22 -> 38         0.01137                  max(5)=       32 -> 47         0.03359
              max(4)=       33 -> 40         0.01077                  min(1)=       31 -> 39        -0.05435
              max(5)=       35 -> 68         0.00757                  min(2)=       33 -> 38        -0.05033
              min(1)=       35 -> 37        -0.39877                  min(3)=       36 -> 37        -0.04930
              min(2)=       35 -> 40        -0.11204                  min(4)=       29 -> 37        -0.04028
              min(3)=       35 -> 39        -0.05993                  min(5)=       31 -> 40        -0.02849
              min(4)=       31 -> 38        -0.04252                  #CIs=2386 #CIs>0=1091 #CIs<0=1029
              min(5)=       35 -> 42        -0.01043
              #CIs=2284 #CIs>0=938 #CIs<0=1006        8       Singlet-A"    6.7048        184.92    0.1439    0.000
                                                                     max(1)=       33 -> 37         0.69091
5       Singlet-A"    5.8000        213.77    0.0805    0.000          max(2)=       32 -> 54         0.04678
              max(1)=       35 -> 37         0.57965                  max(3)=       32 -> 50         0.04168
              max(2)=       36 -> 38         0.39505                  max(4)=       34 -> 53         0.03616
              max(3)=       33 -> 37         0.06330                  max(5)=       34 -> 55         0.03615
              max(4)=       31 -> 38         0.04339                  min(1)=       36 -> 38        -0.08037
              max(5)=       35 -> 56         0.01607                  min(2)=       31 -> 38        -0.06296
              min(1)=       35 -> 40        -0.04377                  min(3)=       34 -> 51        -0.05866
              min(2)=       35 -> 39        -0.02591                  min(4)=       33 -> 40        -0.02648
              min(3)=       36 -> 41        -0.01366                  min(5)=       33 -> 39        -0.02329
              min(4)=       26 -> 48        -0.01303                  #CIs=2286 #CIs>0=998 #CIs<0=977
              min(5)=       27 -> 48        -0.01253
              #CIs=2288 #CIs>0=941 #CIs<0=1005        9       Singlet-A"    7.1817        172.64    0.0007    0.000
                                                                     max(1)=       36 -> 41         0.66631
6       Singlet-A'    6.2642        197.92    0.0016    0.000          max(2)=       31 -> 41         0.10009
              max(1)=       36 -> 39         0.66406                  max(3)=       35 -> 40         0.02612
```

max(4)= 35 -> 44 0.02377
max(5)= 36 -> 52 0.01737
min(1)= 35 -> 39 -0.13953
min(2)= 36 -> 43 -0.10302
min(3)= 35 -> 42 -0.07181
min(4)= 31 -> 43 -0.06226
min(5)= 36 -> 45 -0.02544
#CIs=2279 #CIs>0=896 #CIs<0=941

10 Singlet-A’ 7.1897 172.45 0.4941 0.000
max(1)= 35 -> 38 0.53271
max(2)= 36 -> 42 0.08728
max(3)= 36 -> 46 0.03948
max(4)= 28 -> 49 0.02864
max(5)= 30 -> 50 0.02776
min(1)= 36 -> 40 -0.36762
min(2)= 31 -> 37 -0.19607
min(3)= 36 -> 39 -0.12175
min(4)= 36 -> 37 -0.09733
min(5)= 31 -> 40 -0.04234
#CIs=2383 #CIs>0=1034 #CIs<0=1070

11 Singlet-A’ 7.4228 167.03 0.0220 0.000
max(1)= 36 -> 42 0.66257
max(2)= 34 -> 38 0.15575
max(3)= 31 -> 37 0.06325
max(4)= 36 -> 39 0.05626
max(5)= 36 -> 49 0.04302
min(1)= 35 -> 38 -0.07269
min(2)= 35 -> 41 -0.06998
min(3)= 31 -> 39 -0.05663
min(4)= 36 -> 44 -0.05501
min(5)= 31 -> 44 -0.04814
#CIs=2382 #CIs>0=1044 #CIs<0=1007

12 Singlet-A’ 7.4434 166.57 0.0007 0.000
max(1)= 34 -> 38 0.68458
max(2)= 27 -> 38 0.04068
max(3)= 35 -> 41 0.01889
max(4)= 36 -> 44 0.01465
max(5)= 31 -> 39 0.01255
min(1)= 36 -> 42 -0.15049
min(2)= 28 -> 37 -0.02735
min(3)= 31 -> 37 -0.02660
min(4)= 26 -> 38 -0.02507
min(5)= 30 -> 38 -0.02437
#CIs=2377 #CIs>0=924 #CIs<0=958

13 Singlet-A" 7.5448 164.33 0.0000 0.000
max(1)= 30 -> 37 0.66268
max(2)= 32 -> 38 0.16383
max(3)= 30 -> 40 0.11088
max(4)= 30 -> 39 0.05602
max(5)= 26 -> 40 0.03096
min(1)= 26 -> 37 -0.08087
min(2)= 27 -> 37 -0.05126
min(3)= 25 -> 38 -0.04847
min(4)= 26 -> 46 -0.02996
min(5)= 18 -> 37 -0.02679
#CIs=2246 #CIs>0=914 #CIs<0=880

14 Singlet-A’ 7.6230 162.64 0.0391 0.000
max(1)= 31 -> 37 0.63006
max(2)= 35 -> 38 0.09946
max(3)= 28 -> 37 0.08293
max(4)= 36 -> 37 0.07994
max(5)= 31 -> 40 0.03923
min(1)= 36 -> 40 -0.20456
min(2)= 29 -> 37 -0.10439
min(3)= 36 -> 39 -0.10260
min(4)= 33 -> 38 -0.08697
min(5)= 36 -> 42 -0.04849
#CIs=2384 #CIs>0=984 #CIs<0=1104

15 Singlet-A" 7.7354 160.28 0.0001 0.000
max(1)= 32 -> 38 0.67753
max(2)= 19 -> 38 0.02657
max(3)= 27 -> 37 0.02113
max(4)= 27 -> 46 0.01497
max(5)= 17 -> 38 0.01420
min(1)= 30 -> 37 -0.16407
min(2)= 28 -> 38 -0.05006

```
               min(3)=       34 -> 40        -0.04518                  max(4)=       29 -> 41         0.05276
               min(4)=       29 -> 38        -0.03759                  max(5)=       35 -> 47         0.03375
               min(5)=       25 -> 38        -0.03612                  min(1)=       35 -> 40        -0.36951
               #CIs=2267 #CIs>0=826 #CIs<0=884                         min(2)=       36 -> 43        -0.13542
                                                                       min(3)=       31 -> 38        -0.09207
16      Singlet-A"    7.8651        157.64    0.0993    0.000         min(4)=       36 -> 45        -0.06702
               max(1)=       35 -> 40         0.47058                  min(5)=       31 -> 43        -0.05483
               max(2)=       35 -> 39         0.40130                  #CIs=2281 #CIs>0=932 #CIs<0=910
               max(3)=       31 -> 38         0.29149
               max(4)=       36 -> 38         0.12151  19      Singlet-A’    8.2584        150.13    0.0072    0.000
               max(5)=       36 -> 41         0.08271                  max(1)=       30 -> 38         0.69695
               min(1)=       35 -> 37        -0.05165                  max(2)=       34 -> 38         0.02548
               min(2)=       33 -> 40        -0.02993                  max(3)=       29 -> 37         0.02274
               min(3)=       33 -> 39        -0.02822                  max(4)=       28 -> 37         0.02218
               min(4)=       30 -> 49        -0.01664                  max(5)=       30 -> 67         0.02019
               min(5)=       27 -> 48        -0.01563                  min(1)=       25 -> 37        -0.05918
               #CIs=2287 #CIs>0=979 #CIs<0=975                         min(2)=       25 -> 46        -0.05514
                                                                       min(3)=       35 -> 41        -0.04625
17      Singlet-A’    7.9323        156.30    0.0001    0.000         min(4)=       18 -> 38        -0.02335
               max(1)=       33 -> 38         0.69822                  min(5)=       36 -> 44        -0.02257
               max(2)=       31 -> 37         0.09378                  #CIs=2365 #CIs>0=883 #CIs<0=926
               max(3)=       35 -> 38         0.03286
               max(4)=       36 -> 40         0.01651  20      Singlet-A"    8.2595        150.11    0.0000    0.000
               max(5)=       32 -> 42         0.01537                  max(1)=       34 -> 40         0.55705
               min(1)=       22 -> 37        -0.01804                  max(2)=       34 -> 39         0.34114
               min(2)=       34 -> 43        -0.01095                  max(3)=       34 -> 37         0.21020
               min(3)=       22 -> 46        -0.00934                  max(4)=       34 -> 46         0.11978
               min(4)=       33 -> 43        -0.00677                  max(5)=       32 -> 38         0.05640
               min(5)=       34 -> 41        -0.00641                  min(1)=       26 -> 37        -0.04796
               #CIs=2377 #CIs>0=976 #CIs<0=971                         min(2)=       28 -> 38        -0.02493
                                                                       min(3)=       21 -> 37        -0.02372
18      Singlet-A"    8.1246        152.60    0.0000    0.000         min(4)=       29 -> 38        -0.01843
               max(1)=       35 -> 39         0.45817                  min(5)=       26 -> 40        -0.01467
               max(2)=       35 -> 42         0.30658                  #CIs=2255 #CIs>0=846 #CIs<0=885
               max(3)=       36 -> 41         0.11483
```

16.1.3 Main contributions from different excited states at CIS approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_   Singlet-A"    5.5098        225.03    0.0000    0.000
                                                                       max(1)=       33 -> 37         0.58784
```

```
              max(2)=       33 -> 41         0.05815
              max(3)=       33 -> 47         0.05713
              max(4)=       27 -> 42         0.03547
              max(5)=       33 -> 44         0.02378
              min(1)=       33 -> 42        -0.36190
              min(2)=       33 -> 39        -0.05465
              min(3)=       33 -> 46        -0.05187
              min(4)=       33 -> 92        -0.04773
              min(5)=       27 -> 37        -0.03918
              #states=936 #states>0=500 #states<0=436 5       Singlet-A'    7.3223        169.33    0.0023    0.000
                                                                    max(1)=       36 -> 39         0.64341
2       Singlet-A'    5.7079        217.22    0.3688    0.000         max(2)=       32 -> 39         0.19934
              max(1)=       36 -> 37         0.63296                max(3)=       31 -> 39         0.06652
              max(2)=       36 -> 42         0.09732                max(4)=       29 -> 39         0.05574
              max(3)=       32 -> 42         0.05243                max(5)=       32 -> 37         0.04943
              max(4)=       32 -> 47         0.02898                min(1)=       35 -> 40        -0.09036
              max(5)=       32 -> 39         0.02407                min(2)=       36 -> 41        -0.08159
              min(1)=       35 -> 38        -0.24506                min(3)=       32 -> 41        -0.07216
              min(2)=       32 -> 37        -0.09776                min(4)=       29 -> 41        -0.04635
              min(3)=       31 -> 37        -0.03827                min(5)=       32 -> 42        -0.02954
              min(4)=       29 -> 42        -0.03238                #states=1237 #states>0=606 #states<0=631
              min(5)=       32 -> 46        -0.03001
              #states=1569 #states>0=810 #states<0=759 6       Singlet-A"    7.4245        166.99    0.1003    0.000
                                                                    max(1)=       35 -> 37         0.47927
3       Singlet-A'    5.8463        212.07    0.0001    0.000         max(2)=       34 -> 37         0.31672
              max(1)=       31 -> 37         0.56145                max(3)=       33 -> 53         0.03411
              max(2)=       32 -> 42         0.10947                max(4)=       34 -> 41         0.02654
              max(3)=       31 -> 41         0.05369                max(5)=       35 -> 42         0.02202
              max(4)=       31 -> 47         0.05100                min(1)=       36 -> 38        -0.34403
              max(5)=       28 -> 37         0.04833                min(2)=       34 -> 42        -0.16032
              min(1)=       31 -> 42        -0.33298                min(3)=       36 -> 40        -0.03537
              min(2)=       32 -> 37        -0.18679                min(4)=       34 -> 39        -0.02745
              min(3)=       31 -> 39        -0.05136                min(5)=       30 -> 49        -0.01941
              min(4)=       25 -> 42        -0.04556                #states=1511 #states>0=734 #states<0=777
              min(5)=       31 -> 46        -0.04288
              #states=1129 #states>0=554 #states<0=575 7       Singlet-A'    7.6171        162.77    0.8431    0.000
                                                                    max(1)=       35 -> 38         0.57447
4       Singlet-A"    5.8607        211.55    0.0093    0.000         max(2)=       36 -> 37         0.24933
              max(1)=       36 -> 38         0.55319                max(3)=       32 -> 42         0.08185
              max(2)=       35 -> 37         0.40760                max(4)=       36 -> 41         0.04127
              max(3)=       35 -> 42         0.13750                max(5)=       29 -> 37         0.03859
```

```
                min(1)=       36 -> 42        -0.26216                min(3)=       32 -> 37        -0.10099
                min(2)=       32 -> 37        -0.04104                min(4)=       35 -> 38        -0.09470
                min(3)=       29 -> 46        -0.03809                min(5)=       36 -> 44        -0.08983
                min(4)=       32 -> 47        -0.03222                #states=1381 #states>0=695 #states<0=686
                min(5)=       35 -> 40        -0.02944
                #states=1497 #states>0=746 #states<0=751 11      Singlet-A'    8.6676        143.04    0.2843    0.000
                                                                      max(1)=       36 -> 42         0.53589
8       Singlet-A"    8.0297        154.41    0.5747    0.000      max(2)=       32 -> 37         0.28404
                max(1)=       34 -> 37         0.49417              max(3)=       35 -> 38         0.22981
                max(2)=       36 -> 40         0.21626              max(4)=       36 -> 41         0.14066
                max(3)=       36 -> 38         0.21187              max(5)=       31 -> 37         0.09234
                max(4)=       32 -> 38         0.05888              min(1)=       29 -> 37        -0.07116
                max(5)=       32 -> 40         0.05388              min(2)=       35 -> 40        -0.06031
                min(1)=       35 -> 37        -0.24062              min(3)=       36 -> 46        -0.06008
                min(2)=       34 -> 42        -0.21080              min(4)=       32 -> 39        -0.05625
                min(3)=       35 -> 39        -0.08995              min(5)=       36 -> 44        -0.05621
                min(4)=       36 -> 43        -0.06050              #states=1589 #states>0=808 #states<0=781
                min(5)=       35 -> 41        -0.04964
                #states=1565 #states>0=837 #states<0=728 12      Singlet-A"    9.0484        137.02    0.0097    0.000
                                                                      max(1)=       35 -> 41         0.42280
9       Singlet-A"    8.1873        151.43    0.1561    0.000      max(2)=       35 -> 39         0.37819
                max(1)=       36 -> 40         0.53804              max(3)=       36 -> 40         0.21023
                max(2)=       35 -> 37         0.13588              max(4)=       29 -> 40         0.10646
                max(3)=       32 -> 40         0.12651              max(5)=       32 -> 40         0.03901
                max(4)=       34 -> 42         0.07477              min(1)=       36 -> 43        -0.23315
                max(5)=       35 -> 44         0.05043              min(2)=       36 -> 45        -0.15818
                min(1)=       35 -> 39        -0.23817              min(3)=       32 -> 43        -0.12828
                min(2)=       34 -> 37        -0.17936              min(4)=       32 -> 45        -0.06225
                min(3)=       35 -> 41        -0.17678              min(5)=       31 -> 43        -0.04259
                min(4)=       36 -> 43        -0.10495              #states=1112 #states>0=559 #states<0=553
                min(5)=       36 -> 38        -0.09972
                #states=1439 #states>0=694 #states<0=745 13      Singlet-A"    9.4584        131.08    0.0001    0.000
                                                                      max(1)=       30 -> 37         0.62707
10      Singlet-A'    8.4423        146.86    0.0443    0.000      max(2)=       30 -> 42         0.20653
                max(1)=       36 -> 41         0.60649              max(3)=       25 -> 38         0.08646
                max(2)=       36 -> 39         0.09661              max(4)=       26 -> 47         0.07326
                max(3)=       36 -> 49         0.07681              max(5)=       26 -> 42         0.06247
                max(4)=       32 -> 41         0.04884              min(1)=       31 -> 38        -0.08043
                max(5)=       36 -> 57         0.03479              min(2)=       26 -> 37        -0.07234
                min(1)=       35 -> 40        -0.18959              min(3)=       26 -> 46        -0.06514
                min(2)=       36 -> 42        -0.10571              min(4)=       18 -> 37        -0.06292
```

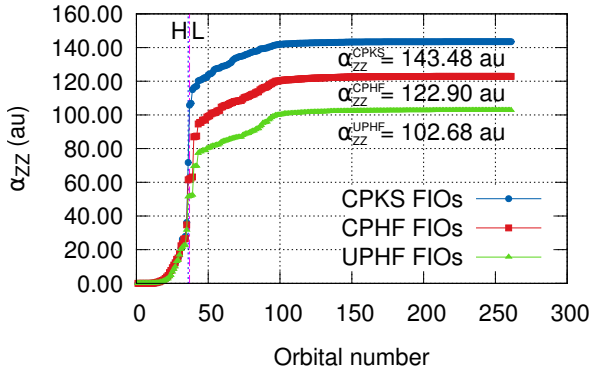
```
                min(5)=       27 -> 37        -0.05227
                #states=1177 #states>0=581 #states<0=596 17      Singlet-A"    9.9934        124.07    0.0538    0.000
                                                                        max(1)=       35 -> 42         0.31484
14      Singlet-A'    9.5700        129.56    0.0321    0.000          max(2)=       32 -> 40         0.12946
                max(1)=       35 -> 40         0.53968                max(3)=       35 -> 39         0.07920
                max(2)=       36 -> 41         0.21176                max(4)=       35 -> 47         0.07183
                max(3)=       36 -> 44         0.19202                max(5)=       29 -> 38         0.04884
                max(4)=       30 -> 38         0.15759                min(1)=       36 -> 43        -0.40473
                max(5)=       35 -> 43         0.15757                min(2)=       35 -> 41        -0.28927
                min(1)=       32 -> 41        -0.13994                min(3)=       32 -> 38        -0.17211
                min(2)=       36 -> 47        -0.06309                min(4)=       36 -> 40        -0.16871
                min(3)=       36 -> 46        -0.06235                min(5)=       36 -> 45        -0.10903
                min(4)=       32 -> 39        -0.05651                #states=1321 #states>0=647 #states<0=674
                min(5)=       31 -> 41        -0.04790
                #states=1053 #states>0=529 #states<0=524 18      Singlet-A'   10.0448        123.43    0.0418    0.000
                                                                        max(1)=       32 -> 37         0.53182
15      Singlet-A"    9.8449        125.94    0.0088    0.000          max(2)=       32 -> 42         0.18720
                max(1)=       36 -> 43         0.35496                max(3)=       31 -> 37         0.16067
                max(2)=       35 -> 42         0.31945                max(4)=       28 -> 37         0.08075
                max(3)=       35 -> 39         0.31872                max(5)=       36 -> 37         0.07715
                max(4)=       36 -> 40         0.23643                min(1)=       36 -> 42        -0.28445
                max(5)=       36 -> 45         0.09520                min(2)=       35 -> 38        -0.14260
                min(1)=       32 -> 38        -0.22183                min(3)=       29 -> 37        -0.06647
                min(2)=       35 -> 41        -0.11590                min(4)=       36 -> 46        -0.04413
                min(3)=       32 -> 40        -0.07422                min(5)=       22 -> 42        -0.04243
                min(4)=       35 -> 37        -0.07377                #states=1556 #states>0=769 #states<0=787
                min(5)=       31 -> 38        -0.06564
                #states=1277 #states>0=634 #states<0=643 19      Singlet-A"   10.1433        122.23    0.0055    0.000
                                                                        max(1)=       35 -> 41         0.37503
16      Singlet-A'    9.8472        125.91    0.0037    0.000          max(2)=       35 -> 42         0.33782
                max(1)=       30 -> 38         0.64910                max(3)=       35 -> 44         0.11629
                max(2)=       25 -> 37         0.09071                max(4)=       35 -> 48         0.06013
                max(3)=       25 -> 46         0.06155                max(5)=       29 -> 43         0.05686
                max(4)=       32 -> 41         0.02758                min(1)=       35 -> 39        -0.38699
                max(5)=       19 -> 37         0.02354                min(2)=       32 -> 38        -0.20323
                min(1)=       35 -> 40        -0.15095                min(3)=       31 -> 38        -0.07251
                min(2)=       25 -> 47        -0.06939                min(4)=       35 -> 37        -0.06022
                min(3)=       33 -> 38        -0.06715                min(5)=       35 -> 46        -0.04214
                min(4)=       28 -> 37        -0.06247                #states=1218 #states>0=618 #states<0=600
                min(5)=       27 -> 38        -0.05452
                #states=1242 #states>0=622 #states<0=620 20      Singlet-A"   10.4551        118.59    0.0429    0.000
```

```
max(1)=       28 -> 38         0.41404
max(2)=       32 -> 38         0.30771
max(3)=       35 -> 42         0.15807
max(4)=       27 -> 37         0.14279
max(5)=       29 -> 38         0.13131
min(1)=       31 -> 38        -0.30491
min(2)=       26 -> 37        -0.11425
min(3)=       27 -> 47        -0.07235
min(4)=       30 -> 37        -0.06792
min(5)=       21 -> 37        -0.04397
#states=1360 #states>0=681 #states<0=679
```

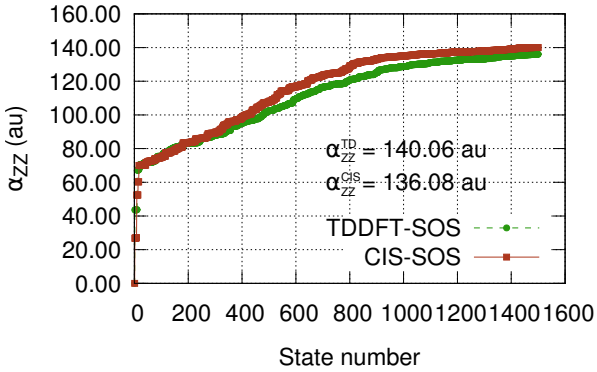
16.2 6-311++G(d,p)

16.2.1 Plots

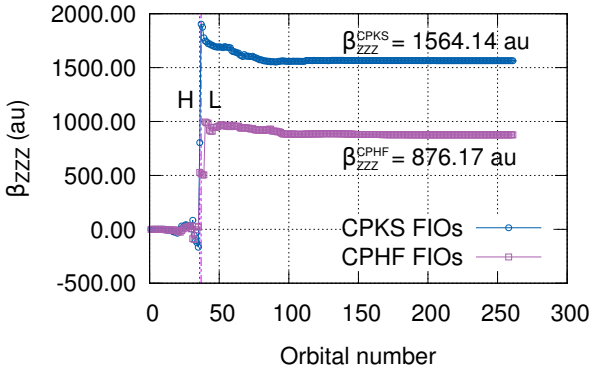
Figure S28: For *p*-nitroaniline molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S28a and S28b) or states (SOS approaches, in Plots S28c and S28d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S28e and S28f as well (bottom). All elements of the β tensor were recomputed with an error less than 1.00 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



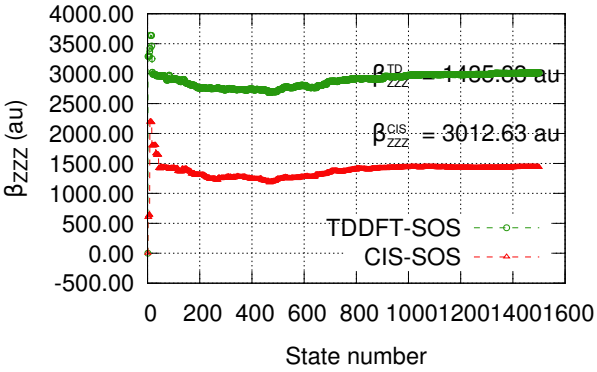
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

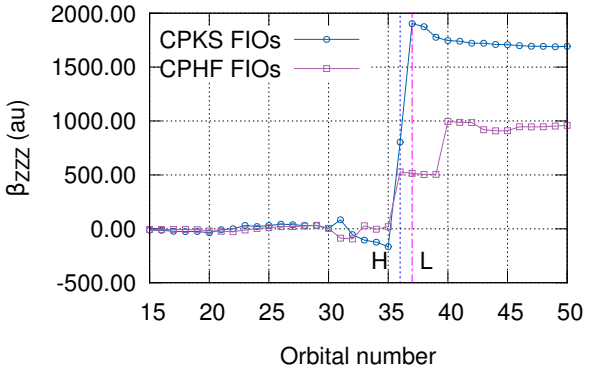


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

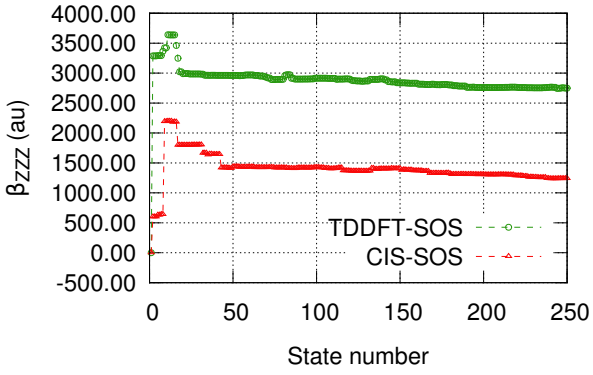


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S28: (continued) For *p*-nitroaniline molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S28a and S28b) or states (SOS approaches, in Plots S28c and S28d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S28e and S28f as well (bottom). All elements of the β tensor were recomputed with an error less than 1.00 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 15 and 50.



(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 250.

16.2.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_
                                                    2        Singlet-A'    4.4033        281.57    0.3807    0.000
1        Singlet-A"    4.0380        307.04    0.0000    0.000          max(1)=       36 -> 37         0.68961
                max(1)=       34 -> 37         0.67514                 max(2)=       35 -> 39         0.09295
                max(2)=       34 -> 51         0.04697                 max(3)=       31 -> 43         0.05405
                max(3)=       34 -> 47         0.03502                 max(4)=       36 -> 43         0.02201
                max(4)=       34 -> 55         0.03023                 max(5)=       29 -> 37         0.01632
                max(5)=       34 -> 38         0.02310                 min(1)=       31 -> 37        -0.09702
                min(1)=       34 -> 43        -0.19017                 min(2)=       31 -> 55        -0.01284
                min(2)=       34 -> 44        -0.02750                 min(3)=       30 -> 64        -0.01111
                min(3)=       34 -> 88        -0.01996                 min(4)=       30 -> 90        -0.01042
                min(4)=       34 -> 41        -0.01990                 min(5)=       29 -> 43        -0.01023
                min(5)=       34 -> 70        -0.01800                 #CIs=2983 #CIs>0=1335 #CIs<0=1343
                #CIs=2762 #CIs>0=994 #CIs<0=1022
```

```
3       Singlet-A'    4.6012        269.46    0.0005    0.000
              max(1)=       32 -> 37         0.67632
              max(2)=       32 -> 51         0.04506
              max(3)=       32 -> 47         0.03300
              max(4)=       32 -> 55         0.02868
              max(5)=       25 -> 37         0.02423
              min(1)=       32 -> 43        -0.18398
              min(2)=       32 -> 44        -0.02653
              min(3)=       32 -> 88        -0.02030
              min(4)=       32 -> 41        -0.01939
              min(5)=       25 -> 43        -0.01768
              #CIs=2927 #CIs>0=1152 #CIs<0=1123

4       Singlet-A"    4.8452        255.89    0.0038    0.000
              max(1)=       36 -> 39         0.54751
              max(2)=       33 -> 37         0.05185
              max(3)=       35 -> 47         0.05100
              max(4)=       35 -> 55         0.02119
              max(5)=       35 -> 63         0.01279
              min(1)=       35 -> 37        -0.42106
              min(2)=       35 -> 43        -0.10818
              min(3)=       36 -> 40        -0.04074
              min(4)=       31 -> 39        -0.03949
              min(5)=       36 -> 58        -0.03053
              #CIs=2854 #CIs>0=1217 #CIs<0=1280

5       Singlet-A'    5.3620        231.23    0.0020    0.000
              max(1)=       36 -> 38         0.67834
              max(2)=       31 -> 38         0.09982
              max(3)=       36 -> 50         0.08541
              max(4)=       36 -> 45         0.08150
              max(5)=       31 -> 50         0.02506
              min(1)=       36 -> 48        -0.06907
              min(2)=       36 -> 41        -0.04091
              min(3)=       31 -> 41        -0.03789
              min(4)=       36 -> 52        -0.03591
              min(5)=       35 -> 40        -0.03430
              #CIs=2974 #CIs>0=1235 #CIs<0=1213

6       Singlet-A"    5.5921        221.71    0.0721    0.000
              max(1)=       35 -> 37         0.56127
              max(2)=       36 -> 39         0.41501
              max(3)=       33 -> 37         0.08803
              max(4)=       35 -> 47         0.04140
              max(5)=       31 -> 39         0.03590
              min(1)=       35 -> 43        -0.03629
              min(2)=       33 -> 43        -0.01788
              min(3)=       29 -> 49        -0.01043
              min(4)=       35 -> 63        -0.00994
              min(5)=       30 -> 76        -0.00968
              #CIs=2854 #CIs>0=1277 #CIs<0=1244

7       Singlet-A"    6.1467        201.71    0.0000    0.000
              max(1)=       36 -> 40         0.66365
              max(2)=       36 -> 53         0.12893
              max(3)=       31 -> 40         0.05851
              max(4)=       36 -> 46         0.05195
              max(5)=       36 -> 39         0.03825
              min(1)=       36 -> 42        -0.11042
              min(2)=       35 -> 38        -0.10476
              min(3)=       31 -> 42        -0.04446
              min(4)=       35 -> 41        -0.04142
              min(5)=       33 -> 37        -0.02922
              #CIs=2853 #CIs>0=1172 #CIs<0=1186

8       Singlet-A'    6.2617        198.00    0.0013    0.000
              max(1)=       36 -> 43         0.50496
              max(2)=       36 -> 41         0.38669
              max(3)=       35 -> 39         0.23111
              max(4)=       31 -> 37         0.12674
              max(5)=       36 -> 44         0.05491
              min(1)=       36 -> 52        -0.06324
              min(2)=       35 -> 40        -0.04274
              min(3)=       33 -> 39        -0.03313
              min(4)=       36 -> 37        -0.03080
              min(5)=       36 -> 55        -0.02845
              #CIs=2983 #CIs>0=1331 #CIs<0=1372

9       Singlet-A'    6.3706        194.62    0.0086    0.000
              max(1)=       36 -> 41         0.55650
              max(2)=       36 -> 45         0.08250
              max(3)=       36 -> 59         0.05380
```

```
                max(4)=       36 -> 38         0.02285                 min(1)=       36 -> 50        -0.13576
                max(5)=       33 -> 39         0.02265                 min(2)=       36 -> 38        -0.04860
                min(1)=       36 -> 43        -0.34025                 min(3)=       36 -> 41        -0.04205
                min(2)=       35 -> 39        -0.18111                 min(4)=       31 -> 48        -0.03759
                min(3)=       36 -> 52        -0.08849                 min(5)=       36 -> 48        -0.03572
                min(4)=       31 -> 37        -0.07197                 #CIs=2971 #CIs>0=1234 #CIs<0=1235
                min(5)=       31 -> 38        -0.05881
                #CIs=2984 #CIs>0=1376 #CIs<0=1313       13      Singlet-A"    7.0142        176.76    0.0019    0.000
                                                                       max(1)=       35 -> 38         0.62903
10      Singlet-A"    6.4572        192.01    0.1724    0.000          max(2)=       35 -> 41         0.22546
                max(1)=       33 -> 37         0.68122                 max(3)=       36 -> 40         0.09845
                max(2)=       34 -> 96         0.05025                 max(4)=       35 -> 45         0.09103
                max(3)=       34 -> 44         0.03938                 max(5)=       36 -> 46         0.04799
                max(4)=       34 -> 48         0.03795                 min(1)=       36 -> 42        -0.10323
                max(5)=       32 -> 90         0.03702                 min(2)=       35 -> 52        -0.08787
                min(1)=       36 -> 39        -0.08917                 min(3)=       35 -> 48        -0.06013
                min(2)=       33 -> 43        -0.06393                 min(4)=       35 -> 43        -0.04093
                min(3)=       31 -> 39        -0.05969                 min(5)=       33 -> 38        -0.04007
                min(4)=       35 -> 37        -0.04034                 #CIs=2845 #CIs>0=1093 #CIs<0=1103
                min(5)=       34 -> 54        -0.03209
                #CIs=2856 #CIs>0=1256 #CIs<0=1264       14      Singlet-A"    7.1298        173.89    0.0001    0.000
                                                                       max(1)=       36 -> 42         0.63959
11      Singlet-A’    6.9434        178.56    0.5070    0.000          max(2)=       36 -> 40         0.14774
                max(1)=       35 -> 39         0.61556                 max(3)=       35 -> 38         0.11679
                max(2)=       31 -> 55         0.02354                 max(4)=       36 -> 56         0.08620
                max(3)=       36 -> 70         0.02034                 max(5)=       35 -> 45         0.03191
                max(4)=       27 -> 86         0.01854                 min(1)=       36 -> 46        -0.14068
                max(5)=       36 -> 61         0.01844                 min(2)=       35 -> 41        -0.07388
                min(1)=       36 -> 43        -0.26399                 min(3)=       31 -> 40        -0.06385
                min(2)=       31 -> 37        -0.13989                 min(4)=       35 -> 43        -0.05714
                min(3)=       36 -> 37        -0.08901                 min(5)=       36 -> 67        -0.04713
                min(4)=       36 -> 51        -0.07876                 #CIs=2837 #CIs>0=1150 #CIs<0=1154
                min(5)=       31 -> 43        -0.07366
                #CIs=2984 #CIs>0=1274 #CIs<0=1375       15      Singlet-A"    7.4100        167.32    0.0000    0.000
                                                                       max(1)=       30 -> 37         0.67688
12      Singlet-A’    6.9779        177.68    0.0040    0.000          max(2)=       30 -> 43         0.10821
                max(1)=       36 -> 44         0.51090                 max(3)=       32 -> 39         0.08552
                max(2)=       36 -> 45         0.42854                 max(4)=       26 -> 37         0.08246
                max(3)=       35 -> 40         0.11198                 max(5)=       27 -> 37         0.05401
                max(4)=       36 -> 65         0.05676                 min(1)=       30 -> 47        -0.05384
                max(5)=       35 -> 53         0.03004                 min(2)=       25 -> 39        -0.04243
```

```
               min(3)=       26 -> 43        -0.03745                  max(4)=       31 -> 41         0.08042
               min(4)=       30 -> 63        -0.02272                  max(5)=       36 -> 62         0.07406
               min(5)=       30 -> 55        -0.02160                  min(1)=       36 -> 44        -0.34822
               #CIs=2810 #CIs>0=1147 #CIs<0=1170                       min(2)=       35 -> 40        -0.17109
                                                                       min(3)=       36 -> 48        -0.09933
16      Singlet-A'    7.4219        167.05    0.0052    0.000         min(4)=       36 -> 41        -0.07089
               max(1)=       34 -> 39         0.51139                  min(5)=       36 -> 38        -0.06655
               max(2)=       36 -> 43         0.13467                  #CIs=2983 #CIs>0=1352 #CIs<0=1276
               max(3)=       36 -> 45         0.10919
               max(4)=       29 -> 37         0.05526   19      Singlet-A"    7.5125        165.04    0.1146    0.000
               max(5)=       33 -> 39         0.04262                  max(1)=       35 -> 43         0.57148
               min(1)=       31 -> 37        -0.41041                  max(2)=       35 -> 41         0.27311
               min(2)=       36 -> 44        -0.07501                  max(3)=       31 -> 39         0.17692
               min(3)=       28 -> 37        -0.07433                  max(4)=       36 -> 49         0.12499
               min(4)=       35 -> 40        -0.06690                  max(5)=       36 -> 42         0.11060
               min(5)=       36 -> 37        -0.05811                  min(1)=       35 -> 38        -0.05084
               #CIs=2984 #CIs>0=1365 #CIs<0=1343                       min(2)=       35 -> 37        -0.04955
                                                                       min(3)=       35 -> 52        -0.04762
17      Singlet-A'    7.4309        166.85    0.0069    0.000         min(4)=       31 -> 40        -0.04005
               max(1)=       34 -> 39         0.47294                  min(5)=       33 -> 43        -0.03767
               max(2)=       31 -> 37         0.46124                  #CIs=2856 #CIs>0=1268 #CIs<0=1254
               max(3)=       36 -> 37         0.06585
               max(4)=       36 -> 44         0.05242   20      Singlet-A'    7.5199        164.87    0.0472    0.000
               max(5)=       35 -> 39         0.05097                  max(1)=       36 -> 47         0.64074
               min(1)=       36 -> 43        -0.14697                  max(2)=       35 -> 40         0.21820
               min(2)=       29 -> 37        -0.09327                  max(3)=       36 -> 44         0.07051
               min(3)=       36 -> 45        -0.08373                  max(4)=       35 -> 39         0.05217
               min(4)=       33 -> 39        -0.04826                  max(5)=       36 -> 51         0.04827
               min(5)=       34 -> 40        -0.03174                  min(1)=       36 -> 45        -0.10077
               #CIs=2985 #CIs>0=1388 #CIs<0=1324                       min(2)=       36 -> 50        -0.04312
                                                                       min(3)=       31 -> 41        -0.03790
18      Singlet-A'    7.4595        166.21    0.0389    0.000         min(4)=       31 -> 37        -0.03734
               max(1)=       36 -> 45         0.47267                  min(5)=       36 -> 52        -0.02799
               max(2)=       36 -> 47         0.19990                  #CIs=2980 #CIs>0=1355 #CIs<0=1283
               max(3)=       31 -> 37         0.18109
```

16.2.3 Main contributions from different excited states at CIS approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_   Singlet-A"    5.4601        227.07    0.0000    0.000
                                                                       max(1)=       32 -> 40         0.56057
```

```
              max(2)=       32 -> 46         0.21314                  max(4)=       31 -> 51         0.15770
              max(3)=       32 -> 59         0.20856                  max(5)=       31 -> 49         0.10335
              max(4)=       32 -> 51         0.16403                  min(1)=       31 -> 50        -0.13834
              max(5)=       32 -> 49         0.10395                  min(2)=       31 -> 85        -0.08107
              min(1)=       32 -> 50        -0.14314                  min(3)=       33 -> 40        -0.07846
              min(2)=       32 -> 85        -0.08570                  min(4)=       31 -> 39        -0.06999
              min(3)=       32 -> 39        -0.06996                  min(5)=       31 -> 61        -0.03180
              min(4)=       32 -> 61        -0.03493                  #CIs=2677 #CIs>0=1235 #CIs<0=1213
              min(5)=       32 -> 52        -0.02717
              #CIs=2523 #CIs>0=1143 #CIs<0=1128       5       Singlet-A'    6.0323        205.53    0.0036    0.000
                                                                     max(1)=       36 -> 37         0.62829
2       Singlet-A'    5.5320        224.12    0.3691    0.000      max(2)=       36 -> 45         0.15624
              max(1)=       36 -> 40         0.61814                  max(3)=       33 -> 37         0.11191
              max(2)=       36 -> 37         0.07568                  max(4)=       36 -> 64         0.03261
              max(3)=       35 -> 44         0.06593                  max(5)=       33 -> 50         0.02337
              max(4)=       35 -> 58         0.05880                  min(1)=       36 -> 52        -0.12608
              max(5)=       33 -> 77         0.02798                  min(2)=       36 -> 47        -0.12334
              min(1)=       35 -> 43        -0.21217                  min(3)=       36 -> 40        -0.08206
              min(2)=       36 -> 39        -0.10936                  min(4)=       35 -> 38        -0.06464
              min(3)=       36 -> 49        -0.09839                  min(5)=       33 -> 39        -0.05356
              min(4)=       33 -> 40        -0.09761                  #CIs=2840 #CIs>0=1304 #CIs<0=1317
              min(5)=       36 -> 59        -0.07120
              #CIs=2897 #CIs>0=1389 #CIs<0=1383       6       Singlet-A"    6.7357        184.07    0.0030    0.000
                                                                     max(1)=       36 -> 38         0.60907
3       Singlet-A"    5.7395        216.02    0.0062    0.000      max(2)=       36 -> 53         0.17128
              max(1)=       36 -> 43         0.52167                  max(3)=       33 -> 38         0.06875
              max(2)=       35 -> 40         0.38920                  max(4)=       35 -> 52         0.06733
              max(3)=       36 -> 38         0.03241                  max(5)=       36 -> 44         0.04821
              max(4)=       36 -> 57         0.03108                  min(1)=       35 -> 37        -0.20201
              max(5)=       35 -> 65         0.02952                  min(2)=       36 -> 41        -0.10169
              min(1)=       36 -> 44        -0.14860                  min(3)=       35 -> 40        -0.08146
              min(2)=       36 -> 58        -0.11972                  min(4)=       35 -> 39        -0.07873
              min(3)=       35 -> 49        -0.09802                  min(5)=       33 -> 41        -0.04583
              min(4)=       35 -> 59        -0.08874                  #CIs=2705 #CIs>0=1281 #CIs<0=1231
              min(5)=       35 -> 39        -0.05974
              #CIs=2735 #CIs>0=1259 #CIs<0=1311       7       Singlet-A'    6.9208        179.15    0.0115    0.000
                                                                     max(1)=       36 -> 39         0.62081
4       Singlet-A'    5.8059        213.55    0.0005    0.000      max(2)=       36 -> 45         0.16666
              max(1)=       31 -> 40         0.55976                  max(3)=       36 -> 40         0.11158
              max(2)=       31 -> 46         0.20766                  max(4)=       36 -> 61         0.08302
              max(3)=       31 -> 59         0.19698                  max(5)=       36 -> 60         0.06382
```

```
              min(1)=       36 -> 50        -0.14421                min(3)=       35 -> 52        -0.09551
              min(2)=       36 -> 51        -0.08616                min(4)=       35 -> 47        -0.08585
              min(3)=       33 -> 37        -0.07892                min(5)=       35 -> 51        -0.07931
              min(4)=       35 -> 38        -0.06887                #CIs=2664 #CIs>0=1220 #CIs<0=1235
              min(5)=       29 -> 37        -0.03420
              #CIs=2844 #CIs>0=1325 #CIs<0=1322       11      Singlet-A'    7.5006        165.30    0.0092    0.000
                                                                    max(1)=       36 -> 45         0.45034
8       Singlet-A"    7.0942        174.77    0.1002    0.000   max(2)=       35 -> 38         0.26614
              max(1)=       35 -> 40         0.47223                max(3)=       36 -> 52         0.12928
              max(2)=       34 -> 40         0.28945                max(4)=       35 -> 53         0.08970
              max(3)=       34 -> 46         0.09456                max(5)=       36 -> 64         0.08536
              max(4)=       36 -> 38         0.07751                min(1)=       36 -> 42        -0.34821
              max(5)=       34 -> 59         0.07741                min(2)=       36 -> 39        -0.10921
              min(1)=       36 -> 43        -0.33466                min(3)=       36 -> 37        -0.09211
              min(2)=       35 -> 39        -0.10578                min(4)=       36 -> 46        -0.09183
              min(3)=       34 -> 50        -0.05909                min(5)=       36 -> 50        -0.07922
              min(4)=       35 -> 46        -0.04391                #CIs=2824 #CIs>0=1295 #CIs<0=1307
              min(5)=       34 -> 39        -0.03449
              #CIs=2764 #CIs>0=1334 #CIs<0=1311       12      Singlet-A"    7.6771        161.50    0.4758    0.000
                                                                    max(1)=       34 -> 40         0.44373
9       Singlet-A'    7.3490        168.71    0.6163    0.000   max(2)=       36 -> 43         0.17733
              max(1)=       35 -> 43         0.46680                max(3)=       34 -> 46         0.12478
              max(2)=       36 -> 46         0.32288                max(4)=       34 -> 59         0.10269
              max(3)=       36 -> 40         0.18878                max(5)=       34 -> 49         0.09601
              max(4)=       36 -> 49         0.18059                min(1)=       36 -> 41        -0.29941
              max(5)=       36 -> 59         0.13681                min(2)=       35 -> 40        -0.18282
              min(1)=       35 -> 44        -0.13310                min(3)=       36 -> 38        -0.11498
              min(2)=       35 -> 58        -0.09137                min(4)=       35 -> 37        -0.10557
              min(3)=       33 -> 40        -0.08952                min(5)=       34 -> 50        -0.08083
              min(4)=       36 -> 39        -0.05736                #CIs=2771 #CIs>0=1351 #CIs<0=1304
              min(5)=       36 -> 50        -0.05735
              #CIs=2892 #CIs>0=1373 #CIs<0=1399       13      Singlet-A"    7.7719        159.53    0.2152    0.000
                                                                    max(1)=       36 -> 41         0.52931
10      Singlet-A"    7.4772        165.82    0.0000    0.000   max(2)=       34 -> 40         0.24608
              max(1)=       35 -> 37         0.50125                max(3)=       36 -> 38         0.14201
              max(2)=       35 -> 39         0.31202                max(4)=       36 -> 43         0.12393
              max(3)=       36 -> 38         0.20028                max(5)=       35 -> 37         0.10787
              max(4)=       35 -> 45         0.15582                min(1)=       35 -> 40        -0.16573
              max(5)=       36 -> 44         0.08418                min(2)=       36 -> 44        -0.14958
              min(1)=       36 -> 41        -0.13539                min(3)=       33 -> 38        -0.06021
              min(2)=       35 -> 50        -0.11241                min(4)=       33 -> 44        -0.05106
```

```
                min(5)=       36 -> 68        -0.05081
                #CIs=2772 #CIs>0=1318 #CIs<0=1328       17      Singlet-A'    8.2644        150.02    0.2966    0.000
                                                                      max(1)=       35 -> 43         0.20136
14      Singlet-A'    7.7816        159.33    0.2110    0.000          max(2)=       33 -> 40         0.19315
                max(1)=       36 -> 46         0.50483                max(3)=       36 -> 46         0.16526
                max(2)=       36 -> 45         0.20155                max(4)=       36 -> 50         0.10622
                max(3)=       36 -> 42         0.12923                max(5)=       36 -> 45         0.05643
                max(4)=       35 -> 44         0.09358                min(1)=       36 -> 49        -0.54999
                max(5)=       35 -> 48         0.08210                min(2)=       36 -> 59        -0.16457
                min(1)=       35 -> 43        -0.29792                min(3)=       36 -> 51        -0.13190
                min(2)=       36 -> 40        -0.14992                min(4)=       35 -> 44        -0.05889
                min(3)=       33 -> 40        -0.13218                min(5)=       29 -> 40        -0.04521
                min(4)=       36 -> 51        -0.06724                #CIs=2885 #CIs>0=1381 #CIs<0=1352
                min(5)=       36 -> 47        -0.05133
                #CIs=2891 #CIs>0=1357 #CIs<0=1385       18      Singlet-A"    8.2945        149.48    0.0011    0.000
                                                                      max(1)=       35 -> 39         0.51715
15      Singlet-A'    7.9885        155.20    0.0398    0.000          max(2)=       35 -> 40         0.13513
                max(1)=       35 -> 38         0.53063                max(3)=       36 -> 41         0.13071
                max(2)=       36 -> 37         0.16237                max(4)=       35 -> 61         0.06443
                max(3)=       35 -> 53         0.15578                max(5)=       35 -> 47         0.06388
                max(4)=       36 -> 39         0.14934                min(1)=       35 -> 37        -0.34594
                max(5)=       36 -> 46         0.11778                min(2)=       35 -> 42        -0.14199
                min(1)=       36 -> 45        -0.28426                min(3)=       35 -> 50        -0.13977
                min(2)=       33 -> 39        -0.06547                min(4)=       35 -> 51        -0.10355
                min(3)=       36 -> 42        -0.06364                min(5)=       33 -> 38        -0.03193
                min(4)=       36 -> 62        -0.05141                #CIs=2716 #CIs>0=1269 #CIs<0=1260
                min(5)=       33 -> 37        -0.03875
                #CIs=2783 #CIs>0=1290 #CIs<0=1260       19      Singlet-A"    8.5058        145.76    0.0033    0.000
                                                                      max(1)=       35 -> 45         0.49950
16      Singlet-A'    8.1699        151.76    0.0072    0.000          max(2)=       35 -> 52         0.14012
                max(1)=       36 -> 42         0.51288                max(3)=       35 -> 56         0.10254
                max(2)=       35 -> 38         0.24333                max(4)=       35 -> 64         0.07537
                max(3)=       36 -> 45         0.19098                max(5)=       35 -> 65         0.06999
                max(4)=       35 -> 41         0.08936                min(1)=       35 -> 42        -0.27621
                max(5)=       36 -> 62         0.07853                min(2)=       36 -> 44        -0.19296
                min(1)=       36 -> 46        -0.20932                min(3)=       35 -> 46        -0.13266
                min(2)=       36 -> 47        -0.19989                min(4)=       35 -> 39        -0.12101
                min(3)=       36 -> 37        -0.05265                min(5)=       36 -> 41        -0.08612
                min(4)=       36 -> 67        -0.04304                #CIs=2733 #CIs>0=1252 #CIs<0=1299
                min(5)=       36 -> 65        -0.03742
                #CIs=2826 #CIs>0=1303 #CIs<0=1307       20      Singlet-A"    8.7087        142.37    0.0164    0.000
```

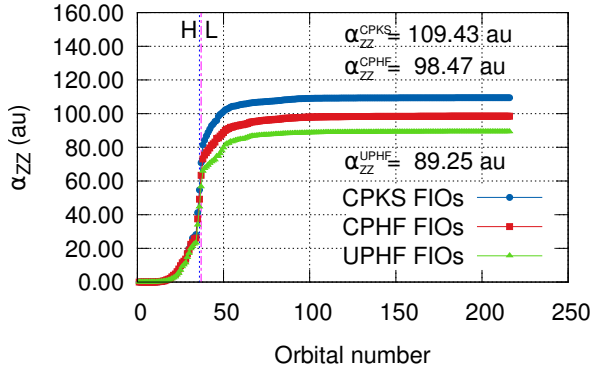
```
max(1)=       36 -> 44         0.39478          min(2)=       34 -> 40        -0.05794
max(2)=       36 -> 48         0.37400          min(3)=       36 -> 38        -0.05773
max(3)=       35 -> 45         0.25623          min(4)=       36 -> 75        -0.04844
max(4)=       35 -> 46         0.22651          min(5)=       35 -> 69        -0.03570
max(5)=       36 -> 41         0.10705          #CIs=2770 #CIs>0=1307 #CIs<0=1315
min(1)=       35 -> 39        -0.10682
```

17 *m*-nitroaniline

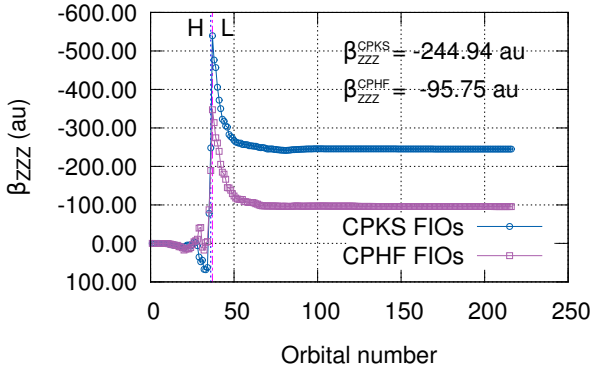
17.1 6-311G(d,p)

17.1.1 Plots

Figure S29: For *m*-nitroaniline molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S29a and **??**) or states (SOS approaches, in Plots S29b and **??**). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S29c and **??** as well (bottom). All elements of the β tensor were recomputed with an error less than 0.10 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.

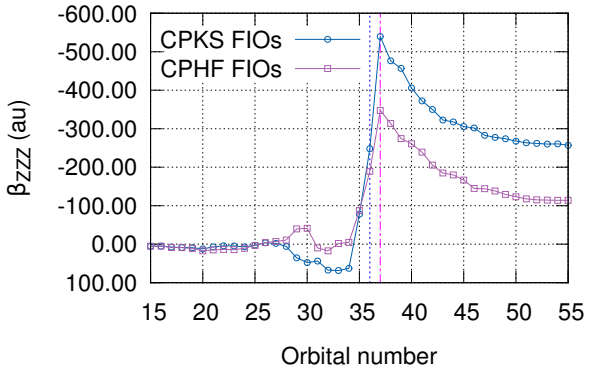


(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

Figure S29: (continued) For *m*-nitroaniline molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S29a and **??**) or states (SOS approaches, in Plots S29b and **??**). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S29c and **??** as well (bottom). All elements of the β tensor were recomputed with an error less than 0.37 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



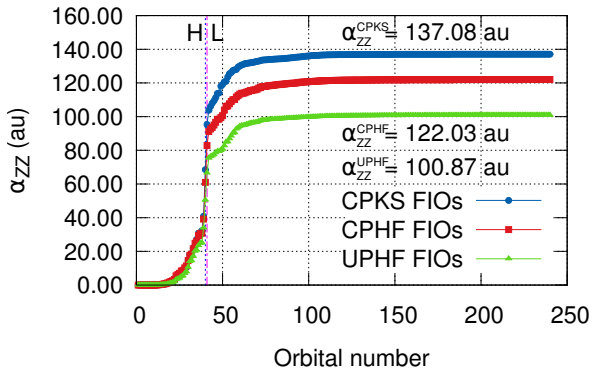
(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 15 and 55.

18 *p*-methoxy-nitrobenzene

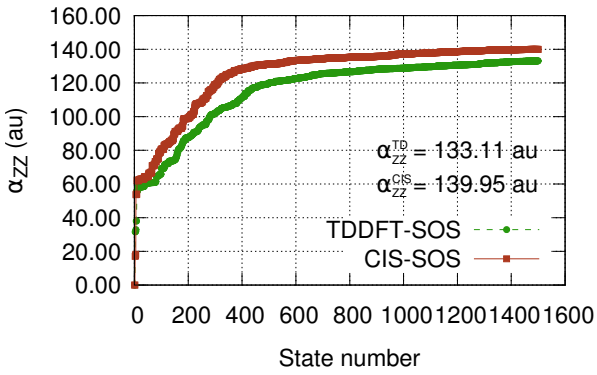
18.1 6-311G(d,p)

18.1.1 Plots

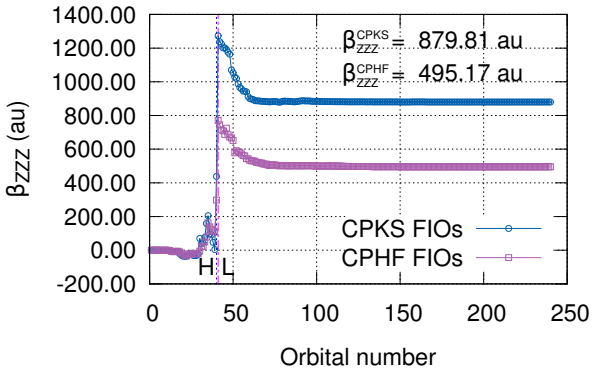
Figure S30: For *p*-methoxy-nitrobenzene molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S30a and S30b) or states (SOS approaches, in Plots S30c and S30d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S30e and S30f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.51 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



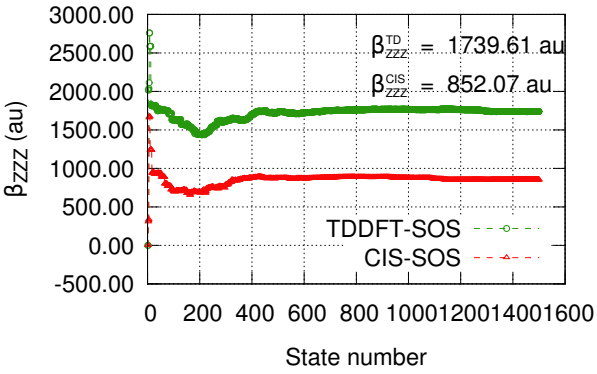
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

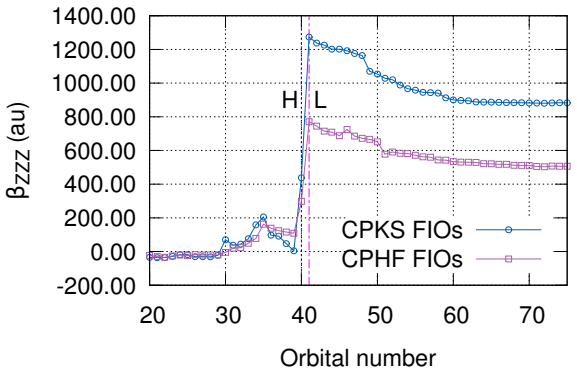


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

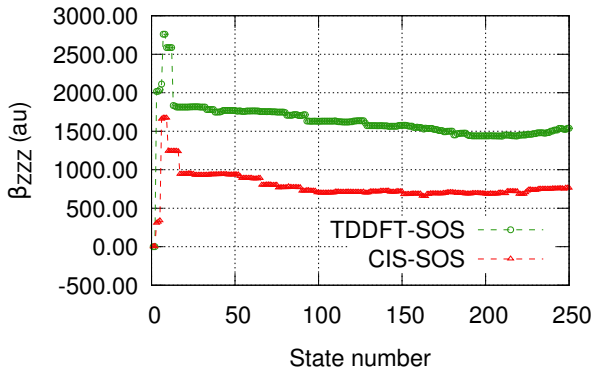


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S30: (continued) For *p*-methoxy-nitrobenzene molecule and 6-311G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S30a and S30b) or states (SOS approaches, in Plots S30c and S30d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S30e and S30f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.51 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 20 and 70.



(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 250.

18.1.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

```
#_exc.st ___symm___     Exc.E Osc._Strength   ___f___ _<S**2>_          #CIs=2265 #CIs>0=849 #CIs<0=819

1        Singlet-A"    4.1023        302.23    0.0000    20.000    Singlet-A"    4.6149        268.66    0.0001    0.000
                max(1)=       38 -> 41         0.67368                   max(1)=       36 -> 41         0.67564
                max(2)=       38 -> 42         0.02078                   max(2)=       36 -> 42         0.01993
                max(3)=       30 -> 41         0.01671                   max(3)=       31 -> 43         0.01869
                max(4)=       35 -> 43         0.01198                   max(4)=       28 -> 43         0.01613
                max(5)=       29 -> 41         0.00843                   max(5)=       29 -> 41         0.01013
                min(1)=       38 -> 43        -0.20915                   min(1)=       36 -> 43        -0.20131
                min(2)=       38 -> 52        -0.02528                   min(2)=       31 -> 41        -0.02534
                min(3)=       35 -> 41        -0.02455                   min(3)=       36 -> 52        -0.02263
                min(4)=       30 -> 43        -0.01284                   min(4)=       28 -> 41        -0.01770
                min(5)=       38 ->101        -0.01049                   min(5)=       30 -> 41        -0.01316
```

```
                #CIs=2276 #CIs>0=921 #CIs<0=901          6        Singlet-A'    6.7020        185.00    0.1597    0.000

                                                                        max(1)=       37 -> 41         0.63153
3        Singlet-A'    4.8366        256.35    0.3324    0.000          max(2)=       40 -> 43         0.15679
                max(1)=       40 -> 41         0.68850                 max(3)=       34 -> 41         0.05029
                max(2)=       34 -> 43         0.04889                 max(4)=       39 -> 41         0.03565
                max(3)=       32 -> 41         0.02924                 max(5)=       38 -> 62         0.02688
                max(4)=       36 -> 53         0.01721                 min(1)=       39 -> 42        -0.20550
                max(5)=       38 -> 51         0.01583                 min(2)=       40 -> 42        -0.11877
                min(1)=       39 -> 42        -0.10676                 min(3)=       39 -> 43        -0.05829
                min(2)=       34 -> 41        -0.09180                 min(4)=       38 -> 57        -0.05788
                min(3)=       32 -> 43        -0.02268                 min(5)=       34 -> 42        -0.04164
                min(4)=       36 -> 45        -0.01367                 #CIs=3513 #CIs>0=1592 #CIs<0=1538
                min(5)=       35 -> 56        -0.01241
                #CIs=3510 #CIs>0=1483 #CIs<0=1566          7        Singlet-A'    6.7415        183.91    0.1247    0.000

                                                                        max(1)=       39 -> 42         0.50139
4        Singlet-A'    5.0828        243.93    0.0011    0.000          max(2)=       37 -> 41         0.26366
                max(1)=       39 -> 41         0.52452                 max(3)=       40 -> 41         0.06912
                max(2)=       40 -> 42         0.45028                 max(4)=       32 -> 41         0.05114
                max(3)=       39 -> 43         0.12534                 max(5)=       34 -> 43         0.04716
                max(4)=       37 -> 41         0.05425                 min(1)=       40 -> 43        -0.38820
                max(5)=       40 -> 43         0.01601                 min(2)=       34 -> 41        -0.09085
                min(1)=       34 -> 42        -0.04868                 min(3)=       39 -> 43        -0.03049
                min(2)=       39 -> 42        -0.01687                 min(4)=       38 -> 57        -0.02533
                min(3)=       39 -> 48        -0.01053                 min(5)=       40 -> 42        -0.01811
                min(4)=       40 -> 41        -0.00987                 #CIs=3515 #CIs>0=1561 #CIs<0=1594
                min(5)=       39 -> 76        -0.00756
                #CIs=3497 #CIs>0=1444 #CIs<0=1530          8        Singlet-A"    7.2821        170.26    0.0000    0.000

                                                                        max(1)=       35 -> 41         0.61418
5        Singlet-A'    5.9952        206.81    0.0892    0.000          max(2)=       35 -> 43         0.17491
                max(1)=       40 -> 42         0.49975                 max(3)=       38 -> 43         0.04769
                max(2)=       37 -> 41         0.12410                 max(4)=       38 -> 41         0.03530
                max(3)=       39 -> 43         0.11675                 max(5)=       29 -> 43         0.03053
                max(4)=       32 -> 41         0.01387                 min(1)=       33 -> 41        -0.26577
                max(5)=       38 -> 45         0.01105                 min(2)=       29 -> 41        -0.05835
                min(1)=       39 -> 41        -0.46933                 min(3)=       36 -> 42        -0.05006
                min(2)=       39 -> 42        -0.02469                 min(4)=       31 -> 41        -0.04116
                min(3)=       34 -> 41        -0.01961                 min(5)=       38 -> 42        -0.03996
                min(4)=       39 -> 64        -0.01757                 #CIs=2281 #CIs>0=1004 #CIs<0=997
                min(5)=       38 -> 57        -0.01691
                #CIs=3508 #CIs>0=1542 #CIs<0=1489          9        Singlet-A'    7.3195        169.39    0.4348    0.000

                                                                        max(1)=       40 -> 43         0.47634
```

```
              max(2)=       39 -> 42         0.41656
              max(3)=       34 -> 41         0.27191
              max(4)=       40 -> 41         0.09293
              max(5)=       34 -> 43         0.04124
              min(1)=       32 -> 41        -0.04631
              min(2)=       37 -> 42        -0.03911
              min(3)=       40 -> 52        -0.03158
              min(4)=       34 -> 52        -0.02720
              min(5)=       36 -> 53        -0.02650
              #CIs=3505 #CIs>0=1528 #CIs<0=1521       13       Singlet-A'    7.8309        158.33    0.0612    0.000
                                                                    max(1)=       34 -> 41         0.58382
10       Singlet-A"    7.4686        166.01    0.0000    0.000    max(2)=       34 -> 43         0.07720
              max(1)=       38 -> 42         0.69198              max(3)=       40 -> 41         0.07164
              max(2)=       35 -> 42         0.07191              max(4)=       40 -> 42         0.02889
              max(3)=       35 -> 41         0.03536              max(5)=       40 -> 52         0.01755
              max(4)=       31 -> 41         0.02301              min(1)=       40 -> 43        -0.29090
              max(5)=       40 -> 45         0.02240              min(2)=       32 -> 41        -0.16931
              min(1)=       40 -> 44        -0.06014              min(3)=       37 -> 42        -0.13082
              min(2)=       30 -> 42        -0.04222              min(4)=       39 -> 42        -0.10273
              min(3)=       33 -> 41        -0.03507              min(5)=       39 -> 43        -0.04538
              min(4)=       38 -> 43        -0.03244              #CIs=3510 #CIs>0=1517 #CIs<0=1498
              min(5)=       38 -> 41        -0.03056
              #CIs=2278 #CIs>0=960 #CIs<0=902          14       Singlet-A'    7.9597        155.77    0.1890    0.000
                                                                    max(1)=       39 -> 43         0.63604
11       Singlet-A"    7.5127        165.03    0.0155    0.000    max(2)=       37 -> 42         0.08477
              max(1)=       40 -> 44         0.66539              max(3)=       34 -> 41         0.05477
              max(2)=       34 -> 44         0.06977              max(4)=       37 -> 43         0.03654
              max(3)=       38 -> 42         0.06432              max(5)=       37 -> 41         0.01815
              max(4)=       34 -> 45         0.04359              min(1)=       34 -> 42        -0.21352
              max(5)=       40 -> 46         0.03712              min(2)=       40 -> 42        -0.17111
              min(1)=       40 -> 45        -0.16458              min(3)=       40 -> 43        -0.04024
              min(2)=       39 -> 44        -0.09140              min(4)=       39 -> 48        -0.02797
              min(3)=       39 -> 46        -0.05065              min(5)=       39 -> 41        -0.02716
              min(4)=       40 -> 66        -0.01977              #CIs=3509 #CIs>0=1525 #CIs<0=1477
              min(5)=       40 -> 51        -0.01947
              #CIs=2278 #CIs>0=953 #CIs<0=953          15       Singlet-A'    7.9704        155.56    0.0015    0.000
                                                                    max(1)=       37 -> 42         0.68495
12       Singlet-A"    7.7384        160.22    0.0000    0.000    max(2)=       34 -> 41         0.13032
              max(1)=       36 -> 42         0.69305              max(3)=       40 -> 42         0.02240
              max(2)=       31 -> 42         0.06138              max(4)=       34 -> 43         0.02117
              max(3)=       35 -> 41         0.05891              max(5)=       37 -> 48         0.01892
```

```
               min(1)=       39 -> 43        -0.08931                max(3)=       38 -> 41         0.19536
               min(2)=       39 -> 42        -0.03473                max(4)=       38 -> 52         0.10676
               min(3)=       37 -> 43        -0.02219                max(5)=       38 -> 42         0.05201
               min(4)=       32 -> 41        -0.02016                min(1)=       30 -> 41        -0.09128
               min(5)=       24 -> 41        -0.01735                min(2)=       26 -> 41        -0.03753
               #CIs=3512 #CIs>0=1454 #CIs<0=1436                     min(3)=       25 -> 41        -0.03567
                                                                     min(4)=       40 -> 45        -0.03295
16      Singlet-A"    8.0487        154.04    0.0029    0.000       min(5)=       35 -> 52        -0.02818
               max(1)=       35 -> 42         0.63529                #CIs=2283 #CIs>0=989 #CIs<0=983
               max(2)=       38 -> 43         0.03758
               max(3)=       36 -> 42         0.02602   19      Singlet-A"    8.3200        149.02    0.0020    0.000
               max(4)=       35 -> 43         0.02493                max(1)=       39 -> 44         0.45209
               max(5)=       38 -> 41         0.01536                max(2)=       40 -> 45         0.23296
               min(1)=       33 -> 42        -0.28015                max(3)=       40 -> 44         0.14028
               min(2)=       38 -> 42        -0.07496                max(4)=       40 -> 49         0.12169
               min(3)=       28 -> 52        -0.04642                max(5)=       39 -> 46         0.05302
               min(4)=       28 -> 41        -0.04135                min(1)=       39 -> 45        -0.32054
               min(5)=       33 -> 41        -0.02624                min(2)=       40 -> 46        -0.27199
               #CIs=2277 #CIs>0=974 #CIs<0=940                       min(3)=       34 -> 46        -0.06695
                                                                     min(4)=       40 -> 50        -0.05688
17      Singlet-A"    8.1010        153.05    0.0049    0.000       min(5)=       34 -> 50        -0.03130
               max(1)=       40 -> 45         0.60894                #CIs=2281 #CIs>0=995 #CIs<0=962
               max(2)=       40 -> 46         0.14342
               max(3)=       39 -> 45         0.14131   20      Singlet-A"    8.4095        147.43    0.0001    0.000
               max(4)=       40 -> 44         0.11644                max(1)=       33 -> 41         0.53185
               max(5)=       34 -> 44         0.09018                max(2)=       35 -> 41         0.20826
               min(1)=       39 -> 44        -0.20381                max(3)=       35 -> 43         0.17436
               min(2)=       40 -> 50        -0.06878                max(4)=       29 -> 41         0.12321
               min(3)=       40 -> 53        -0.04256                max(5)=       33 -> 43         0.10802
               min(4)=       34 -> 50        -0.04134                min(1)=       38 -> 43        -0.21945
               min(5)=       32 -> 46        -0.03640                min(2)=       36 -> 43        -0.13569
               #CIs=2282 #CIs>0=974 #CIs<0=1001                      min(3)=       30 -> 41        -0.10369
                                                                     min(4)=       31 -> 41        -0.07894
18      Singlet-A"    8.1969        151.26    0.0000    0.000       min(5)=       38 -> 41        -0.06440
               max(1)=       38 -> 43         0.61220                #CIs=2278 #CIs>0=1075 #CIs<0=1011
               max(2)=       33 -> 41         0.21891
```

18.1.3 Main contributions from different excited states at CIS approach

#_exc.st	___symm___	Exc.E	Osc._Strength	___f___	_<S**2>_
1	Singlet-A"	5.4890	225.88	0.0000	0.000
	max(1)=	37 -> 41	0.58235		
	max(2)=	37 -> 46	0.36535		
	max(3)=	37 ->101	0.04189		
	max(4)=	37 -> 42	0.03832		
	max(5)=	30 -> 41	0.03673		
	min(1)=	34 -> 41	-0.07416		
	min(2)=	37 -> 53	-0.07198		
	min(3)=	34 -> 46	-0.05147		
	min(4)=	36 -> 41	-0.03624		
	min(5)=	36 -> 98	-0.02687		
	#states=840 #states>0=400 #states<0=440				
2	Singlet-A"	5.8145	213.23	0.0001	0.000
	max(1)=	36 -> 41	0.59144		
	max(2)=	36 -> 46	0.35599		
	max(3)=	28 -> 46	0.03825		
	max(4)=	36 ->101	0.03774		
	max(5)=	36 -> 42	0.03751		
	min(1)=	36 -> 53	-0.06488		
	min(2)=	31 -> 41	-0.04925		
	min(3)=	31 -> 46	-0.04465		
	min(4)=	37 -> 98	-0.02842		
	min(5)=	30 -> 41	-0.02690		
	#states=1029 #states>0=512 #states<0=517				
3	Singlet-A'	5.8919	210.43	0.2659	0.000
	max(1)=	40 -> 41	0.61052		
	max(2)=	39 -> 46	0.03767		
	max(3)=	40 -> 48	0.02787		
	max(4)=	39 -> 48	0.01468		
	max(5)=	32 -> 48	0.01378		
	min(1)=	39 -> 42	-0.27165		
	min(2)=	35 -> 41	-0.09381		
	min(3)=	39 -> 41	-0.09099		
	min(4)=	40 -> 42	-0.09079		
	min(5)=	40 -> 46	-0.08810		
	#states=2172 #states>0=1083 #states<0=1089				
	Singlet-A'	6.0167	206.07	0.0085	0.000
	max(1)=	40 -> 42	0.48970		
	max(2)=	39 -> 41	0.46178		
	max(3)=	40 -> 41	0.11220		
	max(4)=	39 -> 48	0.03575		
	max(5)=	32 -> 42	0.02833		
	min(1)=	39 -> 46	-0.14255		
	min(2)=	39 -> 42	-0.06232		
	min(3)=	40 -> 46	-0.03830		
	min(4)=	35 -> 42	-0.03114		
	min(5)=	40 -> 48	-0.02249		
	#states=1870 #states>0=949 #states<0=921				
5	Singlet-A'	7.5232	164.80	0.0663	0.000
	max(1)=	38 -> 41	0.39897		
	max(2)=	40 -> 42	0.35179		
	max(3)=	38 -> 46	0.19531		
	max(4)=	36 -> 58	0.02875		
	max(5)=	37 -> 87	0.02740		
	min(1)=	39 -> 41	-0.38865		
	min(2)=	37 -> 59	-0.04054		
	min(3)=	35 -> 42	-0.03109		
	min(4)=	37 -> 73	-0.02368		
	min(5)=	37 -> 66	-0.02301		
	#states=2181 #states>0=1077 #states<0=1104				
6	Singlet-A'	7.6657	161.74	0.9391	0.000
	max(1)=	39 -> 42	0.56871		
	max(2)=	40 -> 41	0.27636		
	max(3)=	40 -> 46	0.22247		
	max(4)=	40 -> 42	0.06229		
	max(5)=	32 -> 53	0.04479		
	min(1)=	38 -> 41	-0.08592		
	min(2)=	35 -> 46	-0.07240		
	min(3)=	35 -> 41	-0.05075		
	min(4)=	39 -> 41	-0.04565		
	min(5)=	32 -> 41	-0.03974		
	#states=2143 #states>0=1087 #states<0=1056				
7	Singlet-A'	8.1377	152.36	0.8699	0.000
	max(1)=	38 -> 41	0.46251		

```
                max(2)=       39 -> 41         0.31719
                max(3)=       38 -> 46         0.19474
                max(4)=       39 -> 42         0.10510
                max(5)=       40 -> 41         0.06137
                min(1)=       40 -> 42        -0.31141
                min(2)=       37 -> 59        -0.04261
                min(3)=       37 -> 66        -0.02771
                min(4)=       37 -> 65        -0.02510
                min(5)=       37 -> 73        -0.02424
                #states=2171 #states>0=1072 #states<0=1099

8       Singlet-A"    8.6473        143.38    0.0227    0.000
                max(1)=       40 -> 43         0.54002
                max(2)=       35 -> 43         0.08322
                max(3)=       39 -> 50         0.07128
                max(4)=       40 -> 52         0.06277
                max(5)=       40 -> 45         0.04883
                min(1)=       40 -> 44        -0.29367
                min(2)=       39 -> 43        -0.22191
                min(3)=       39 -> 45        -0.16523
                min(4)=       40 -> 50        -0.06307
                min(5)=       40 -> 55        -0.03921
                #states=1095 #states>0=593 #states<0=502

9       Singlet-A"    8.9099        139.15    0.0001    0.000
                max(1)=       39 -> 43         0.38454
                max(2)=       39 -> 52         0.05471
                max(3)=       39 -> 49         0.04791
                max(4)=       40 -> 49         0.04709
                max(5)=       40 -> 50         0.04512
                min(1)=       39 -> 44        -0.40047
                min(2)=       40 -> 44        -0.29040
                min(3)=       40 -> 45        -0.24405
                min(4)=       35 -> 45        -0.08245
                min(5)=       32 -> 45        -0.07836
                #states=1023 #states>0=505 #states<0=518

10      Singlet-A'    8.9479        138.56    0.2917    0.000
                max(1)=       40 -> 46         0.56553
                max(2)=       40 -> 53         0.05479
                max(3)=       35 -> 53         0.03614
                max(4)=       39 -> 41         0.02131
                max(5)=       34 -> 43         0.02061
                min(1)=       35 -> 41        -0.29035
                min(2)=       39 -> 42        -0.21885
                min(3)=       32 -> 41        -0.11058
                min(4)=       40 -> 41        -0.05291
                min(5)=       40 -> 48        -0.04139
                #states=2135 #states>0=1117 #states<0=1018

11      Singlet-A"    9.2454        134.10    0.0001    0.000
                max(1)=       34 -> 41         0.56076
                max(2)=       37 -> 41         0.08002
                max(3)=       33 -> 46         0.06566
                max(4)=       28 -> 42         0.06094
                max(5)=       34 -> 48         0.05118
                min(1)=       33 -> 41        -0.26131
                min(2)=       34 -> 46        -0.22610
                min(3)=       29 -> 53        -0.08083
                min(4)=       29 -> 41        -0.05940
                min(5)=       36 -> 42        -0.05801
                #states=1266 #states>0=644 #states<0=622

12      Singlet-A"    9.5991        129.16    0.0005    0.000
                max(1)=       40 -> 43         0.36726
                max(2)=       40 -> 44         0.30890
                max(3)=       39 -> 45         0.26705
                max(4)=       39 -> 43         0.22991
                max(5)=       40 -> 45         0.21866
                min(1)=       39 -> 44        -0.13174
                min(2)=       39 -> 47        -0.10809
                min(3)=       32 -> 43        -0.07972
                min(4)=       33 -> 42        -0.06650
                min(5)=       35 -> 45        -0.04637
                #states=1162 #states>0=593 #states<0=569

13      Singlet-A"    9.6814        128.06    0.0502    0.000
                max(1)=       34 -> 42         0.33701
                max(2)=       39 -> 45         0.21689
                max(3)=       39 -> 44         0.17619
                max(4)=       40 -> 45         0.16607
                max(5)=       40 -> 52         0.08363
```

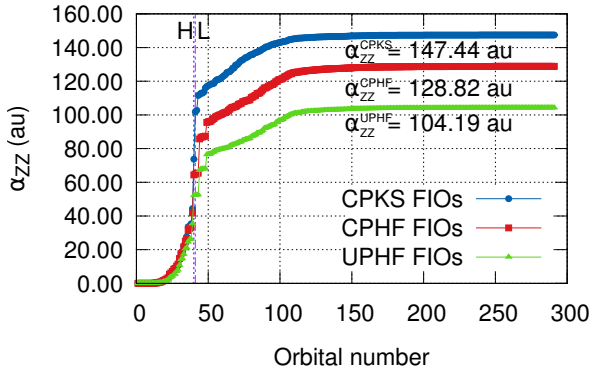
```
              min(1)=       40 -> 44        -0.34549
              min(2)=       33 -> 42        -0.20276
              min(3)=       40 -> 47        -0.14885
              min(4)=       35 -> 43        -0.12033
              min(5)=       40 -> 43        -0.10712
              #states=1286 #states>0=615 #states<0=671
14      Singlet-A"    9.7815        126.75    0.0046    0.000
              max(1)=       34 -> 42         0.44076
              max(2)=       40 -> 44         0.17112
              max(3)=       40 -> 47         0.11986
              max(4)=       37 -> 42         0.09907
              max(5)=       35 -> 43         0.08598
              min(1)=       33 -> 42        -0.26005
              min(2)=       40 -> 45        -0.24336
              min(3)=       39 -> 45        -0.20851
              min(4)=       39 -> 43        -0.13004
              min(5)=       39 -> 44        -0.07889
              #states=1345 #states>0=688 #states<0=657
15      Singlet-A"   10.0305        123.61    0.0001    0.000
              max(1)=       40 -> 45         0.45114
              max(2)=       34 -> 42         0.06004
              max(3)=       32 -> 43         0.05436
              max(4)=       39 -> 52         0.05320
              max(5)=       40 -> 44         0.03495
              min(1)=       39 -> 45        -0.29857
              min(2)=       39 -> 44        -0.28953
              min(3)=       40 -> 51        -0.15475
              min(4)=       39 -> 49        -0.13526
              min(5)=       40 -> 43        -0.12113
              #states=1234 #states>0=603 #states<0=631
16      Singlet-A'   10.0659        123.17    0.1781    0.000
              max(1)=       39 -> 46         0.62218
              max(2)=       35 -> 42         0.21246
              max(3)=       39 -> 41         0.10398
              max(4)=       32 -> 42         0.09787
              max(5)=       40 -> 42         0.09364
              min(1)=       39 -> 48        -0.08816
              min(2)=       34 -> 43        -0.02394
              min(3)=       29 -> 54        -0.02023
              min(4)=       35 -> 46        -0.02008
              min(5)=       30 -> 43        -0.01738
              #states=1891 #states>0=968 #states<0=923
17      Singlet-A'   10.3444        119.86    0.0320    0.000
              max(1)=       35 -> 41         0.54493
              max(2)=       40 -> 46         0.28334
              max(3)=       32 -> 41         0.17849
              max(4)=       40 -> 41         0.07830
              max(5)=       24 -> 46         0.04531
              min(1)=       35 -> 46        -0.19056
              min(2)=       39 -> 42        -0.10950
              min(3)=       40 -> 53        -0.08082
              min(4)=       35 -> 42        -0.06437
              min(5)=       39 -> 46        -0.04269
              #states=2101 #states>0=1061 #states<0=1040
18      Singlet-A"   10.4528        118.61    0.0000    0.000
              max(1)=       36 -> 42         0.34733
              max(2)=       31 -> 42         0.23736
              max(3)=       33 -> 41         0.23233
              max(4)=       30 -> 42         0.16546
              max(5)=       34 -> 41         0.10761
              min(1)=       30 -> 41        -0.22885
              min(2)=       40 -> 51        -0.17485
              min(3)=       26 -> 41        -0.13565
              min(4)=       34 -> 46        -0.09912
              min(5)=       33 -> 42        -0.08241
              #states=1546 #states>0=775 #states<0=771
19      Singlet-A"   10.5039        118.04    0.0000    0.000
              max(1)=       40 -> 51         0.44010
              max(2)=       35 -> 51         0.21498
              max(3)=       36 -> 42         0.17584
              max(4)=       40 -> 50         0.15036
              max(5)=       31 -> 42         0.14499
              min(1)=       40 -> 47        -0.13417
              min(2)=       39 -> 43        -0.11072
              min(3)=       39 -> 44        -0.10335
              min(4)=       40 -> 44        -0.09769
```

```
                min(5)=       35 -> 43        -0.07781                max(5)=       30 -> 41         0.14460
                #states=1558 #states>0=761 #states<0=797              min(1)=       33 -> 41        -0.21356
                                                                      min(2)=       29 -> 42        -0.17358
20      Singlet-A"   10.6873        116.01    0.0008    0.000         min(3)=       30 -> 42        -0.14985
                max(1)=       31 -> 42         0.28813                min(4)=       26 -> 42        -0.13181
                max(2)=       31 -> 41         0.25985                min(5)=       40 -> 51        -0.10522
                max(3)=       37 -> 42         0.25573                #states=1429 #states>0=729 #states<0=700
                max(4)=       36 -> 42         0.16339
```

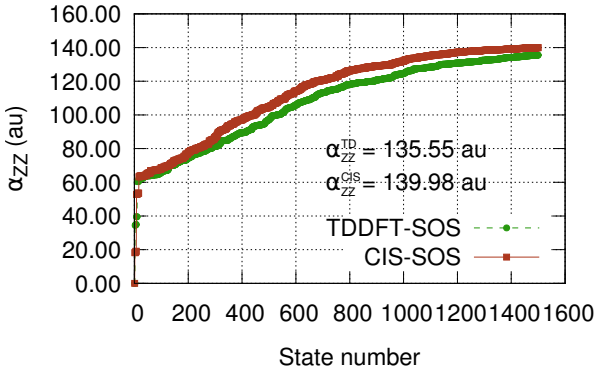
18.2 6-311++G(d,p)

18.2.1 Plots

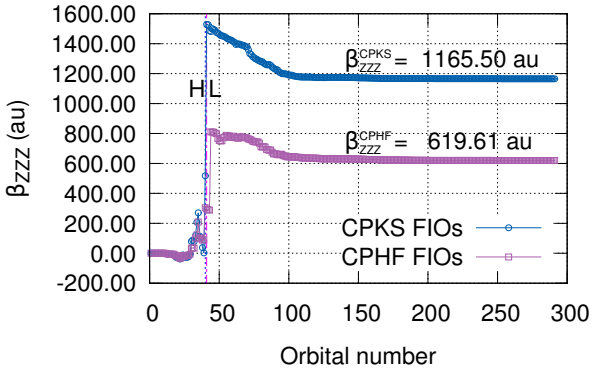
Figure S31: For *p*-methoxy-nitrobenzene molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S31a and S31b) or states (SOS approaches, in Plots S31c and S31d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S31e and S31f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.51 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



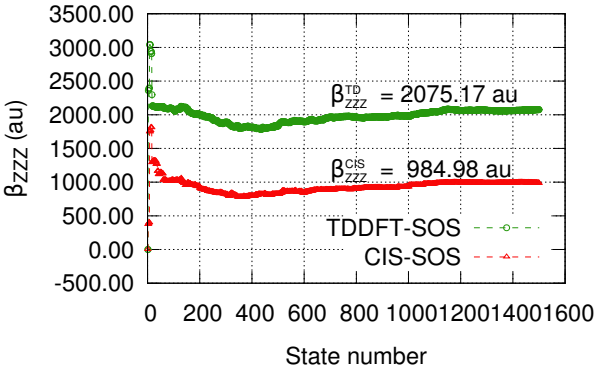
(a) CPKS-, CPHF- and UPHF-FIOs decomposition of α_{ZZ} into MOs.



(b) TDDFT- and CIS-SOS decomposition of α_{ZZ} into states.

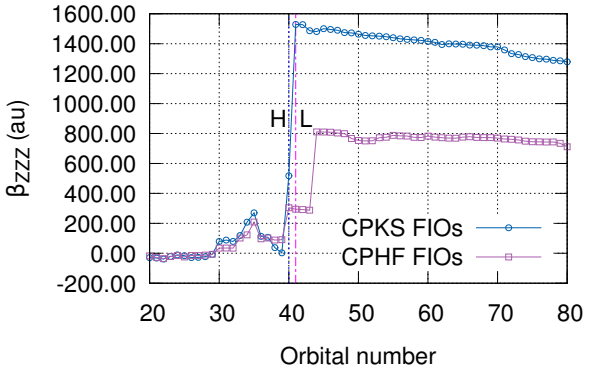


(c) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} into MOs.

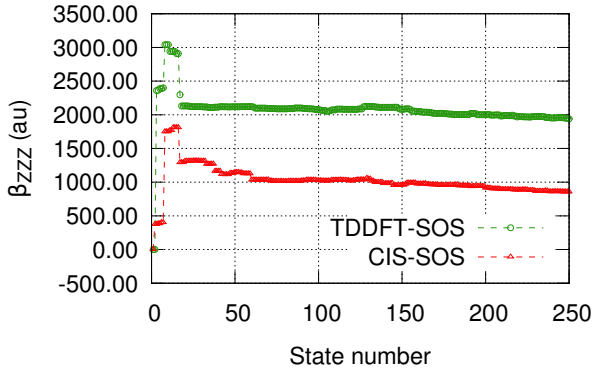


(d) TDDFT- and CIS-SOS decomposition of β_{ZZZ} into states.

Figure S31: (continued) For *p*-methoxy-nitrobenzene molecule and 6-311++G(d,p) basis set, variation of α_{ZZ} (top) and β_{ZZZ} (middle) with respect to the number of orbitals (FIOs decomposition presented in this work, in Plots S31a and S31b) or states (SOS approaches, in Plots S31c and S31d). For α FIOs, three approaches were considered: CPKS, CPHF and UPHF. For β FIOs, the results of CPKS and CPHF approaches are provided. Besides, a shorten range of number of orbitals/states can be seen in Plots S31e and S31f as well (bottom). All elements of the β tensor were recomputed with an error less than 0.51 au in the case of the FIOs. For SOS approach, TDDFT (CAM-B3LYP) and CIS methods were employed. Recomputed values of α_{ZZ} and β_{ZZZ} for the different approaches are included in each plot. HOMO (H) and LUMO (L) are represented by blue and pink dotted lines, respectively.



(e) CPKS- and CPHF-FIOs decomposition of β_{ZZZ} between MOs 20 and 70.



(f) TDDFT- and CIS-SOS decomposition of β_{ZZZ} between states 1 and 250.

18.2.2 Main contributions from different excited states at TDDFT (CAM-B3LYP) approach

#_exc.st	___symm___	Exc.E	Osc._Strength	___f___	_<S**2>_	#CIs=2799 #CIs>0=1065 #CIs<0=1120				
1	Singlet-A"	4.0301	307.65	0.0000	0.000	Singlet-A"	4.5769	270.89	0.0004	0.000
	max(1)=	38 -> 41	0.67612			max(1)=	36 -> 41	0.67784		
	max(2)=	38 -> 46	0.07207			max(2)=	36 -> 46	0.06939		
	max(3)=	38 -> 58	0.03313			max(3)=	36 -> 58	0.03143		
	max(4)=	38 -> 43	0.02194			max(4)=	36 -> 43	0.02109		
	max(5)=	38 -> 97	0.01694			max(5)=	28 -> 41	0.01886		
	min(1)=	38 -> 48	-0.18123			min(1)=	36 -> 48	-0.17439		
	min(2)=	35 -> 41	-0.02622			min(2)=	31 -> 41	-0.02686		
	min(3)=	38 -> 57	-0.02058			min(3)=	36 -> 57	-0.01969		
	min(4)=	30 -> 41	-0.01747			min(4)=	28 -> 48	-0.01439		
	min(5)=	38 -> 54	-0.01287			min(5)=	36 -> 54	-0.01247		

#CIs=2815 #CIs>0=1204 #CIs<0=1114

3 Singlet-A’ 4.6655 265.75 0.3377 0.000

max(1)= 40 -> 41 0.68869

max(2)= 34 -> 48 0.04650

max(3)= 32 -> 41 0.03304

max(4)= 36 -> 75 0.01424

max(5)= 37 -> 43 0.01117

min(1)= 39 -> 43 -0.09737

min(2)= 34 -> 41 -0.09645

min(3)= 32 -> 48 -0.01966

min(4)= 34 -> 43 -0.01226

min(5)= 37 -> 86 -0.01198

#CIs=4451 #CIs>0=2002 #CIs<0=1980

4 Singlet-A’ 4.9848 248.72 0.0013 0.000

max(1)= 39 -> 41 0.55676

max(2)= 40 -> 43 0.41162

max(3)= 39 -> 48 0.10910

max(4)= 37 -> 41 0.04036

max(5)= 40 -> 48 0.02276

min(1)= 34 -> 43 -0.04580

min(2)= 39 -> 46 -0.04157

min(3)= 39 -> 65 -0.02292

min(4)= 39 -> 53 -0.01856

min(5)= 39 -> 43 -0.01781

#CIs=4453 #CIs>0=1940 #CIs<0=1950

5 Singlet-A’ 5.8296 212.68 0.0727 0.000

max(1)= 40 -> 43 0.52325

max(2)= 37 -> 41 0.16026

max(3)= 39 -> 48 0.11622

max(4)= 40 -> 46 0.02045

max(5)= 32 -> 41 0.01426

min(1)= 39 -> 41 -0.42632

min(2)= 39 -> 53 -0.03907

min(3)= 39 -> 43 -0.02866

min(4)= 39 -> 46 -0.02284

min(5)= 34 -> 41 -0.02225

#CIs=4451 #CIs>0=1933 #CIs<0=2027

6 Singlet-A" 6.3456 195.39 0.0153 0.000

max(1)= 40 -> 42 0.64733

max(2)= 40 -> 45 0.13022

max(3)= 40 -> 44 0.12554

max(4)= 40 -> 56 0.10508

max(5)= 40 -> 50 0.10036

min(1)= 39 -> 42 -0.04343

min(2)= 40 -> 51 -0.03984

min(3)= 34 -> 45 -0.03592

min(4)= 40 -> 60 -0.02224

min(5)= 40 -> 52 -0.01912

#CIs=2811 #CIs>0=1190 #CIs<0=1090

7 Singlet-A’ 6.4800 191.33 0.1925 0.000

max(1)= 37 -> 41 0.66115

max(2)= 40 -> 48 0.06831

max(3)= 39 -> 41 0.06545

max(4)= 38 ->109 0.04083

max(5)= 34 -> 41 0.03239

min(1)= 40 -> 43 -0.14305

min(2)= 39 -> 43 -0.08166

min(3)= 39 -> 48 -0.05865

min(4)= 37 -> 48 -0.05592

min(5)= 34 -> 43 -0.04068

#CIs=4451 #CIs>0=1950 #CIs<0=2013

8 Singlet-A’ 6.5823 188.36 0.0921 0.000

max(1)= 39 -> 43 0.49637

max(2)= 40 -> 46 0.22150

max(3)= 37 -> 41 0.11606

max(4)= 40 -> 41 0.05619

max(5)= 32 -> 41 0.05603

min(1)= 40 -> 48 -0.39922

min(2)= 34 -> 41 -0.11344

min(3)= 39 -> 48 -0.01776

min(4)= 40 -> 58 -0.01445

min(5)= 34 -> 74 -0.01399

#CIs=4454 #CIs>0=2010 #CIs<0=2032

9 Singlet-A" 6.8634 180.64 0.0001 0.000

max(1)= 40 -> 44 0.60394

```
              max(2)=       40 -> 50         0.17378                 max(4)=       38 -> 41         0.03411
              max(3)=       39 -> 42         0.15203                 max(5)=       28 -> 43         0.02925
              max(4)=       40 -> 47         0.12851                 min(1)=       33 -> 41        -0.27420
              max(5)=       40 -> 49         0.08067                 min(2)=       29 -> 41        -0.06219
              min(1)=       40 -> 42        -0.14861                 min(3)=       35 -> 46        -0.06137
              min(2)=       40 -> 60        -0.09906                 min(4)=       31 -> 41        -0.04126
              min(3)=       40 -> 45        -0.05847                 min(5)=       36 -> 43        -0.03636
              min(4)=       40 -> 71        -0.03000                 #CIs=2818 #CIs>0=1239 #CIs<0=1272
              min(5)=       40 -> 56        -0.02663
              #CIs=2819 #CIs>0=1195 #CIs<0=1172       13       Singlet-A"    7.2331        171.41    0.0004    0.000
                                                                     max(1)=       39 -> 42         0.49158
10       Singlet-A"    7.0788        175.15    0.0021    0.000     max(2)=       39 -> 45         0.25476
              max(1)=       40 -> 45         0.61872                 max(3)=       39 -> 44         0.14370
              max(2)=       40 -> 44         0.10334                 max(4)=       40 -> 42         0.13192
              max(3)=       39 -> 44         0.05688                 max(5)=       39 -> 56         0.09789
              max(4)=       40 -> 61         0.05206                 min(1)=       40 -> 50        -0.25576
              max(5)=       40 -> 56         0.04704                 min(2)=       40 -> 47        -0.17130
              min(1)=       40 -> 49        -0.14969                 min(3)=       40 -> 45        -0.12935
              min(2)=       40 -> 50        -0.13099                 min(4)=       40 -> 49        -0.05762
              min(3)=       40 -> 47        -0.11137                 min(5)=       40 -> 55        -0.04840
              min(4)=       40 -> 42        -0.09398                 #CIs=2820 #CIs>0=1230 #CIs<0=1195
              min(5)=       40 -> 59        -0.09289
              #CIs=2814 #CIs>0=1193 #CIs<0=1195       14       Singlet-A"    7.3079        169.66    0.0004    0.000
                                                                     max(1)=       40 -> 50         0.41409
11       Singlet-A'    7.1549        173.29    0.4820    0.000     max(2)=       39 -> 42         0.30174
              max(1)=       39 -> 43         0.45938                 max(3)=       40 -> 47         0.25805
              max(2)=       40 -> 48         0.42136                 max(4)=       40 -> 45         0.16131
              max(3)=       34 -> 41         0.24928                 max(5)=       39 -> 45         0.13968
              max(4)=       40 -> 41         0.09324                 min(1)=       40 -> 44        -0.27780
              max(5)=       34 -> 48         0.04544                 min(2)=       40 -> 52        -0.06599
              min(1)=       40 -> 46        -0.13900                 min(3)=       39 -> 44        -0.05174
              min(2)=       40 -> 54        -0.04478                 min(4)=       40 -> 42        -0.05164
              min(3)=       32 -> 41        -0.04055                 min(5)=       34 -> 42        -0.04168
              min(4)=       40 -> 53        -0.04017                 #CIs=2818 #CIs>0=1275 #CIs<0=1211
              min(5)=       34 -> 43        -0.02576
              #CIs=4448 #CIs>0=1978 #CIs<0=1989       15       Singlet-A'    7.4243        167.00    0.0118    0.000
                                                                     max(1)=       40 -> 46         0.61644
12       Singlet-A"    7.1634        173.08    0.0001    0.000     max(2)=       40 -> 48         0.25621
              max(1)=       35 -> 41         0.61247                 max(3)=       34 -> 46         0.05892
              max(2)=       35 -> 48         0.15640                 max(4)=       34 -> 54         0.02950
              max(3)=       38 -> 48         0.03829                 max(5)=       40 -> 65         0.02038
```

```
              min(1)=       40 -> 53        -0.15894                max(3)=       40 -> 53         0.09348
              min(2)=       40 -> 58        -0.09569                max(4)=       39 -> 53         0.04460
              min(3)=       39 -> 43        -0.06345                max(5)=       40 -> 41         0.03375
              min(4)=       39 -> 46        -0.05092                min(1)=       39 -> 46        -0.32247
              min(5)=       40 -> 70        -0.03678                min(2)=       40 -> 48        -0.15014
              #CIs=4431 #CIs>0=1770 #CIs<0=1825                     min(3)=       40 -> 43        -0.13385
                                                                    min(4)=       34 -> 43        -0.13256
16      Singlet-A"    7.4766        165.83    0.0000    0.000      min(5)=       32 -> 41        -0.07470
              max(1)=       38 -> 43         0.69260              #CIs=4443 #CIs>0=1926 #CIs<0=1995
              max(2)=       35 -> 43         0.07815
              max(3)=       30 -> 43         0.04095   19     Singlet-A"    7.7200        160.60    0.0130    0.000
              max(4)=       38 -> 46         0.03919              max(1)=       39 -> 44         0.45546
              max(5)=       31 -> 41         0.02549              max(2)=       40 -> 47         0.30224
              min(1)=       33 -> 41        -0.03325              max(3)=       40 -> 49         0.28181
              min(2)=       38 -> 41        -0.03220              max(4)=       39 -> 50         0.10045
              min(3)=       38 -> 54        -0.02708              max(5)=       40 -> 52         0.09736
              min(4)=       36 -> 48        -0.02334              min(1)=       40 -> 50        -0.19210
              min(5)=       33 -> 43        -0.02161              min(2)=       40 -> 51        -0.08793
              #CIs=2812 #CIs>0=1204 #CIs<0=1163                   min(3)=       39 -> 60        -0.07783
                                                                  min(4)=       40 -> 42        -0.07371
17      Singlet-A'    7.6496        162.08    0.0893    0.000      min(5)=       40 -> 44        -0.07293
              max(1)=       34 -> 41         0.53837              #CIs=2815 #CIs>0=1227 #CIs<0=1255
              max(2)=       39 -> 46         0.18040
              max(3)=       40 -> 46         0.10485   20     Singlet-A"    7.7763        159.44    0.0171    0.000
              max(4)=       40 -> 43         0.08529              max(1)=       39 -> 44         0.39781
              max(5)=       40 -> 41         0.07290              max(2)=       40 -> 50         0.33177
              min(1)=       39 -> 48        -0.23496              max(3)=       40 -> 51         0.08126
              min(2)=       40 -> 48        -0.20826              max(4)=       39 -> 50         0.07246
              min(3)=       32 -> 41        -0.16141              max(5)=       40 -> 55         0.07031
              min(4)=       39 -> 43        -0.08881              min(1)=       40 -> 47        -0.38169
              min(5)=       37 -> 43        -0.06543              min(2)=       40 -> 52        -0.12976
              #CIs=4451 #CIs>0=2034 #CIs<0=1954                   min(3)=       39 -> 42        -0.11391
                                                                  min(4)=       39 -> 60        -0.06045
18      Singlet-A'    7.6903        161.22    0.1834    0.000      min(5)=       40 -> 45        -0.05422
              max(1)=       39 -> 48         0.48646              #CIs=2818 #CIs>0=1243 #CIs<0=1216
              max(2)=       34 -> 41         0.25947
```

18.2.3 Main contributions from different excited states at CIS approach

#_exc.st	___symm___	Exc.E	Osc._Strength	___f___	_<S**2>_
1	Singlet-A"	5.4417	227.84	0.0000	0.000
	max(1)=	37 -> 44	0.56394		
	max(2)=	37 -> 55	0.21093		
	max(3)=	37 -> 53	0.16581		
	max(4)=	37 -> 77	0.08136		
	max(5)=	37 -> 52	0.08087		
	min(1)=	37 -> 60	-0.17457		
	min(2)=	37 -> 65	-0.12725		
	min(3)=	37 -> 45	-0.07900		
	min(4)=	34 -> 44	-0.07854		
	min(5)=	37 -> 95	-0.07442		
	#CIs=2612 #CIs>0=1158 #CIs<0=1156				
2	Singlet-A'	5.7590	215.29	0.2717	0.000
	max(1)=	40 -> 44	0.61076		
	max(2)=	39 -> 49	0.24810		
	max(3)=	40 -> 49	0.07286		
	max(4)=	40 -> 65	0.07161		
	max(5)=	39 -> 52	0.04367		
	min(1)=	35 -> 44	-0.09621		
	min(2)=	39 -> 44	-0.07625		
	min(3)=	40 -> 55	-0.07272		
	min(4)=	40 -> 45	-0.07159		
	min(5)=	35 -> 55	-0.03788		
	#CIs=4323 #CIs>0=2038 #CIs<0=2064				
3	Singlet-A"	5.7760	214.66	0.0004	0.000
	max(1)=	36 -> 44	0.57446		
	max(2)=	36 -> 55	0.20997		
	max(3)=	36 -> 53	0.16263		
	max(4)=	36 -> 52	0.07995		
	max(5)=	36 -> 77	0.07250		
	min(1)=	36 -> 60	-0.16891		
	min(2)=	36 -> 65	-0.12044		
	min(3)=	36 -> 45	-0.07973		
	min(4)=	36 -> 95	-0.07176		
	min(5)=	36 -> 49	-0.04839		
	#CIs=2667 #CIs>0=1210 #CIs<0=1215				
	Singlet-A'	5.9273	209.18	0.0051	0.000
	max(1)=	40 -> 49	0.46402		
	max(2)=	39 -> 55	0.11611		
	max(3)=	40 -> 57	0.07390		
	max(4)=	40 -> 52	0.06926		
	max(5)=	40 -> 69	0.06310		
	min(1)=	39 -> 44	-0.46429		
	min(2)=	40 -> 44	-0.09742		
	min(3)=	39 -> 65	-0.09139		
	min(4)=	40 -> 68	-0.06034		
	min(5)=	39 -> 49	-0.04954		
	#CIs=4237 #CIs>0=1921 #CIs<0=2079				
5	Singlet-A"	6.9182	179.21	0.0204	0.000
	max(1)=	40 -> 41	0.53951		
	max(2)=	40 -> 43	0.29808		
	max(3)=	40 -> 48	0.15344		
	max(4)=	40 -> 54	0.12210		
	max(5)=	40 -> 61	0.05593		
	min(1)=	40 -> 50	-0.16048		
	min(2)=	40 -> 56	-0.13192		
	min(3)=	39 -> 42	-0.07691		
	min(4)=	40 -> 42	-0.06092		
	min(5)=	39 -> 41	-0.05089		
	#CIs=2666 #CIs>0=1213 #CIs<0=1210				
6	Singlet-A'	7.2310	171.46	0.0443	0.000
	max(1)=	38 -> 44	0.40097		
	max(2)=	38 -> 55	0.15162		
	max(3)=	38 -> 53	0.08027		
	max(4)=	38 -> 68	0.04275		
	max(5)=	38 -> 52	0.04169		
	min(1)=	39 -> 44	-0.37360		
	min(2)=	40 -> 49	-0.34379		
	min(3)=	38 -> 60	-0.07751		
	min(4)=	38 -> 65	-0.06061		
	min(5)=	38 -> 45	-0.05084		
	#CIs=4276 #CIs>0=2020 #CIs<0=2056				
7	Singlet-A"	7.3532	168.61	0.0003	0.000
	max(1)=	39 -> 41	0.38704		

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              max(2)=       39 -> 43         0.28650                  max(4)=       40 -> 54         0.12104
              max(3)=       40 -> 43         0.12592                  max(5)=       40 -> 48         0.11890
              max(4)=       39 -> 54         0.09573                  min(1)=       40 -> 41        -0.29797
              max(5)=       40 -> 48         0.07344                  min(2)=       40 -> 51        -0.18154
              min(1)=       40 -> 42        -0.40149                  min(3)=       39 -> 43        -0.09425
              min(2)=       39 -> 56        -0.13192                  min(4)=       40 -> 58        -0.09237
              min(3)=       40 -> 59        -0.11795                  min(5)=       35 -> 41        -0.08849
              min(4)=       40 -> 41        -0.11630                  #CIs=2677 #CIs>0=1239 #CIs<0=1223
              min(5)=       39 -> 50        -0.07058
              #CIs=2655 #CIs>0=1188 #CIs<0=1184       11       Singlet-A'    7.8317        158.31    0.8939    0.000
                                                                     max(1)=       38 -> 44         0.43510
8       Singlet-A'    7.4607        166.18    0.8529    0.000    max(2)=       39 -> 44         0.31852
              max(1)=       39 -> 49         0.52800                  max(3)=       40 -> 49         0.29890
              max(2)=       40 -> 45         0.12052                  max(4)=       38 -> 55         0.14301
              max(3)=       40 -> 49         0.10690                  max(5)=       38 -> 53         0.07932
              max(4)=       40 -> 65         0.10093                  min(1)=       39 -> 49        -0.10908
              max(5)=       38 -> 44         0.08678                  min(2)=       38 -> 60        -0.08179
              min(1)=       40 -> 44        -0.23406                  min(3)=       40 -> 45        -0.07872
              min(2)=       40 -> 55        -0.19049                  min(4)=       39 -> 52        -0.07098
              min(3)=       40 -> 53        -0.10875                  min(5)=       39 -> 53        -0.05942
              min(4)=       39 -> 45        -0.08302                  #CIs=4294 #CIs>0=2031 #CIs<0=2056
              min(5)=       40 -> 52        -0.07993
              #CIs=4316 #CIs>0=2025 #CIs<0=2083       12       Singlet-A"    7.8498        157.94    0.0028    0.000
                                                                     max(1)=       39 -> 42         0.36451
9       Singlet-A"    7.6934        161.16    0.0023    0.000    max(2)=       40 -> 48         0.35346
              max(1)=       40 -> 42         0.47577                  max(3)=       39 -> 59         0.11836
              max(2)=       39 -> 41         0.31673                  max(4)=       39 -> 41         0.11813
              max(3)=       39 -> 43         0.23904                  max(5)=       40 -> 54         0.09701
              max(4)=       40 -> 41         0.14332                  min(1)=       40 -> 43        -0.26168
              max(5)=       39 -> 54         0.11718                  min(2)=       40 -> 50        -0.25083
              min(1)=       39 -> 50        -0.11336                  min(3)=       39 -> 43        -0.07811
              min(2)=       40 -> 48        -0.09826                  min(4)=       39 -> 48        -0.05394
              min(3)=       39 -> 56        -0.08577                  min(5)=       40 -> 82        -0.05046
              min(4)=       39 -> 51        -0.04678                  #CIs=2692 #CIs>0=1264 #CIs<0=1220
              min(5)=       35 -> 46        -0.04517
              #CIs=2694 #CIs>0=1211 #CIs<0=1231       13       Singlet-A'    8.0037        154.91    0.0064    0.000
                                                                     max(1)=       40 -> 45         0.58291
10      Singlet-A"    7.8093        158.77    0.0137    0.000    max(2)=       40 -> 44         0.14178
              max(1)=       40 -> 43         0.44909                  max(3)=       40 -> 55         0.08463
              max(2)=       40 -> 47         0.24365                  max(4)=       40 -> 49         0.07134
              max(3)=       40 -> 42         0.14855                  max(5)=       35 -> 45         0.04660
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                min(1)=       40 -> 52        -0.19725                min(3)=       39 -> 51        -0.13080
                min(2)=       40 -> 53        -0.19655                min(4)=       40 -> 42        -0.11140
                min(3)=       39 -> 49        -0.11713                min(5)=       39 -> 62        -0.07575
                min(4)=       40 -> 60        -0.10028                #CIs=2681 #CIs>0=1234 #CIs<0=1224
                min(5)=       40 -> 57        -0.08786
                #CIs=4179 #CIs>0=1931 #CIs<0=1928       17      Singlet-A'    8.5830        144.45    0.3249    0.000
                                                                      max(1)=       40 -> 55         0.52281
14      Singlet-A"    8.1909        151.37    0.0290    0.000       max(2)=       39 -> 49         0.22491
                max(1)=       39 -> 42         0.43307              max(3)=       40 -> 52         0.10998
                max(2)=       40 -> 50         0.21093              max(4)=       39 -> 52         0.09261
                max(3)=       40 -> 41         0.17001              max(5)=       40 -> 57         0.06617
                max(4)=       40 -> 43         0.13250              min(1)=       35 -> 44        -0.23292
                max(5)=       39 -> 59         0.11765              min(2)=       40 -> 65        -0.16409
                min(1)=       40 -> 48        -0.26679              min(3)=       40 -> 60        -0.10900
                min(2)=       39 -> 43        -0.14822              min(4)=       39 -> 45        -0.09423
                min(3)=       39 -> 48        -0.12208              min(5)=       32 -> 44        -0.09161
                min(4)=       40 -> 42        -0.11948              #CIs=4300 #CIs>0=2034 #CIs<0=2073
                min(5)=       40 -> 54        -0.10381
                #CIs=2713 #CIs>0=1270 #CIs<0=1241       18      Singlet-A"    8.6446        143.42    0.0123    0.000
                                                                      max(1)=       40 -> 47         0.49541
15      Singlet-A"    8.4294        147.09    0.0003    0.000       max(2)=       40 -> 46         0.32563
                max(1)=       40 -> 46         0.55741              max(3)=       40 -> 41         0.12348
                max(2)=       40 -> 51         0.13526              max(4)=       40 -> 62         0.09108
                max(3)=       40 -> 64         0.11410              max(5)=       40 -> 63         0.06750
                max(4)=       40 -> 43         0.11244              min(1)=       40 -> 43        -0.18706
                max(5)=       39 -> 41         0.09334              min(2)=       40 -> 51        -0.11391
                min(1)=       40 -> 47        -0.26919              min(3)=       39 -> 42        -0.11307
                min(2)=       39 -> 43        -0.11028              min(4)=       40 -> 42        -0.10609
                min(3)=       39 -> 47        -0.08316              min(5)=       40 -> 54        -0.09694
                min(4)=       40 -> 41        -0.07283              #CIs=2727 #CIs>0=1223 #CIs<0=1264
                min(5)=       35 -> 50        -0.05311
                #CIs=2711 #CIs>0=1249 #CIs<0=1257       19      Singlet-A"    8.6824        142.80    0.0009    0.000
                                                                      max(1)=       39 -> 43         0.34062
16      Singlet-A"    8.5288        145.37    0.0049    0.000       max(2)=       39 -> 47         0.30103
                max(1)=       39 -> 48         0.46032              max(3)=       39 -> 42         0.22707
                max(2)=       39 -> 54         0.14575              max(4)=       40 -> 46         0.16421
                max(3)=       39 -> 56         0.08654              max(5)=       39 -> 54         0.09044
                max(4)=       39 -> 75         0.08463              min(1)=       39 -> 41        -0.35398
                max(5)=       39 -> 59         0.07813              min(2)=       39 -> 51        -0.15009
                min(1)=       39 -> 50        -0.30107              min(3)=       39 -> 58        -0.07170
                min(2)=       39 -> 43        -0.25453              min(4)=       40 -> 42        -0.06026
```

		min(5)=	39 -> 50	-0.05533			max(5)=	39 -> 69	0.03783
		#CIs=2699 #CIs>0=1227 #CIs<0=1238					min(1)=	39 -> 53	-0.27774
							min(2)=	40 -> 52	-0.20099
20	Singlet-A'	8.6992	142.52	0.0135	0.000		min(3)=	39 -> 52	-0.18626
		max(1)=	39 -> 45	0.52645			min(4)=	39 -> 57	-0.08587
		max(2)=	39 -> 49	0.13107			min(5)=	40 -> 49	-0.06516
		max(3)=	40 -> 55	0.09209			#CIs=4215 #CIs>0=1984 #CIs<0=1977		
		max(4)=	40 -> 57	0.04490					