

University of Groningen

## Nuclear anapole moment interaction in BaF from relativistic coupled-cluster theory

Hao, Yongliang; Ilias, Miroslav; Eliav, Ephraim; Schwerdtfeger, Peter; Flambaum, Victor V.; Borschevsky, Anastasia

*Published in:*  
Physical Review A

*DOI:*  
[10.1103/PhysRevA.98.032510](https://doi.org/10.1103/PhysRevA.98.032510)

**IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.**

*Document Version*  
Publisher's PDF, also known as Version of record

*Publication date:*  
2018

[Link to publication in University of Groningen/UMCG research database](#)

*Citation for published version (APA):*

Hao, Y., Ilias, M., Eliav, E., Schwerdtfeger, P., Flambaum, V. V., & Borschevsky, A. (2018). Nuclear anapole moment interaction in BaF from relativistic coupled-cluster theory. *Physical Review A*, *98*(3), [032510]. <https://doi.org/10.1103/PhysRevA.98.032510>

### Copyright

Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

The publication may also be distributed here under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license. More information can be found on the University of Groningen website: <https://www.rug.nl/library/open-access/self-archiving-pure/taverne-amendment>.

### Take-down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

*Downloaded from the University of Groningen/UMCG research database (Pure): <http://www.rug.nl/research/portal>. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.*

## Nuclear anapole moment interaction in BaF from relativistic coupled-cluster theory

Yongliang Hao,<sup>1</sup> Miroslav Iliáš,<sup>2</sup> Ephraim Eliav,<sup>3</sup> Peter Schwerdtfeger,<sup>4,5</sup> Victor V. Flambaum,<sup>6,7</sup> and Anastasia Borschevsky<sup>1,\*</sup>

<sup>1</sup>*Van Swinderen Institute for Particle Physics and Gravity, University of Groningen, Nijenborgh 4, 9747AG Groningen, The Netherlands*

<sup>2</sup>*Department of Chemistry, Faculty of Natural Sciences, Matej Bel University, Tajovského 40, 97401 Banská Bystrica, Slovakia*

<sup>3</sup>*School of Chemistry, Tel Aviv University, 6997801 Tel Aviv, Israel*

<sup>4</sup>*Centre for Theoretical Chemistry and Physics, New Zealand Institute for Advanced Study, Massey University Auckland, Private Bag 102904, 0745 Auckland, New Zealand*

<sup>5</sup>*Centre for Advanced Study (CAS) at the Norwegian Academy of Science and Letters, Drammensveien 78, NO-0271 Oslo, Norway*

<sup>6</sup>*Helmholtz Institute Mainz, Johannes Gutenberg University, 55099 Mainz, Germany*

<sup>7</sup>*School of Physics, University of New South Wales, Sydney, New South Wales 2052, Australia*



(Received 7 August 2018; published 27 September 2018)

We present high-accuracy relativistic coupled-cluster calculations of the  $P$ -odd interaction coefficient  $W_A$  describing the nuclear anapole moment effect on the molecular electronic structure. The molecule under study, BaF, is considered a promising candidate for the measurement of the nuclear anapole moment, and the preparation for the experiment is now underway [E. Altuntaş *et al.*, *Phys. Rev. Lett.* **120**, 142501 (2018)]. The influence of various computational parameters (size of the basis set, treatment of relativistic effects, and treatment of electron correlation) on the calculated  $W_A$  coefficient is investigated and a recommended value of 147.7 Hz with an estimated uncertainty of 1.5% is proposed.

DOI: [10.1103/PhysRevA.98.032510](https://doi.org/10.1103/PhysRevA.98.032510)

### I. INTRODUCTION

The standard model (SM) of particle physics, unifying the electromagnetic, weak, and strong forces by which the fundamental particles interact, has proven to be valid with unprecedented accuracy. However, the SM, as we know, is incomplete and many open questions remain that lie beyond its current formulation [1]; among the most important are the origin of dark matter and dark energy, neutrino mass and oscillations, matter-antimatter asymmetry, and the unification with the gravitational force. These open questions motivate both the formulation of new theories beyond the SM and the experimental search for new physical phenomena.

One prominent category of such experiments is the search for violation of parity ( $P$ ) and time ( $T$ ) reversal symmetries in atoms and molecules [2]. In particular, investigation of nuclear-spin-dependent parity-violating (NSD-PV) effects, which are the main focus of this work, can be used to test low-energy quantum chromodynamics and parity nonconservation in nuclei [3].

The NSD-PV term of the electronic Hamiltonian for a specific nucleus can be written as [2]

$$H_{\text{NSD}} = \frac{G_F}{\sqrt{2}I} \sum_i (\kappa_A + \kappa_{\text{ax}} + \kappa_{\text{hfs}}) (\boldsymbol{\alpha}_i \cdot \mathbf{I}) \rho(\mathbf{r}_i), \quad (1)$$

where  $G_F \approx 1.435850(1) \times 10^{-62} \text{ J m}^3 \approx 2.222516(1) \times 10^{-14} E_h a_B^3$  is the Fermi coupling constant [4],  $E_h$  is the Hartree energy,  $a_B$  is the Bohr radius,  $\boldsymbol{\alpha}_i$  are the Dirac matrices in the standard representation,  $\mathbf{I}$  is the nuclear spin,

$\mathbf{r}_i$  are the electronic coordinates for electron  $i$ , and  $\rho(\mathbf{r}_i)$  is the (normalized) nuclear density distribution. This contribution is only present for nuclei with  $I \neq 0$ , and for open-shell atoms or molecules because of Kramers symmetry. The three dimensionless nuclear  $\kappa$  parameters are associated with the different sources of the NSD-PV effects. The first term  $\kappa_A$  comes from the nuclear anapole moment interaction and will be discussed in more detail below. The second term  $\kappa_{\text{ax}}$  arises from the electroweak neutral coupling between the electron vector and nucleon axial-vector currents ( $\mathbf{V}_e \mathbf{A}_N$ ) [5]; theoretical prediction of  $\kappa_{\text{ax}}$  within the nuclear shell model can be found in Ref. [6]. The third contribution  $\kappa_{\text{hfs}}$  originates in the nuclear-spin-independent weak interaction combined with the hyperfine interaction [7]. The coefficients  $\kappa_{\text{hfs}}$  were derived using different models, for example in Refs. [8–10].

The anapole moment was first predicted by Zel'dovich [11] in 1958. It appears in the second-order multipole expansion of the magnetic vector potential simultaneously with the  $P$ - and  $T$ -violating magnetic quadrupole moment [12]. In a simple valence nucleon model,  $\kappa_A$  has the following form [13]:

$$\kappa_A = 1.15 \times 10^{-3} \left( \frac{\mathcal{K}}{I+1} \right) A^{\frac{2}{3}} \mu_i g_i. \quad (2)$$

Here,  $\mathcal{K} = (-1)^{l+\frac{1}{2}-I} (I+1/2)$ ,  $l$  is the orbital angular momentum of the external unpaired nucleon  $i = n, p$ ;  $\mu_p = +2.8$ ,  $\mu_n = -1.9$ , and  $A$  is the atomic mass number. Theoretical estimates give the dimensionless strength constant for nucleon-nucleus weak potential  $|g_p| \approx 4.6$  for a proton [13], and  $|g_n| \sim 1$  for a neutron [14]. Due to the  $A^{2/3}$  scaling of this effect, the nuclear anapole moment provides the dominant NSD-PV contribution for systems containing heavy nuclei [3]. The determination of nuclear anapole effects can contribute

\*a.borschevsky@rug.nl

to the fundamental understanding of parity violation in the hadronic sector [3,15].

To date, only one observation of a nonzero nuclear anapole moment was achieved using a Stark-PV interference technique in an experiment on the  $^{133}\text{Cs}$  atom [16], where the main source of the anapole moment was due to the unpaired proton. The value of  $\kappa_A$  for  $^{133}\text{Cs}$  was determined as  $\kappa_A = 364(62) \times 10^{-3}$  [14]. Further measurements on Cs and other alkali atoms using the ground-state hyperfine splitting have been recently proposed [17]. Complementary measurements are also being performed on atoms with unpaired neutrons, such as  $^{171}\text{Yb}$  [18] and  $^{212}\text{Fr}$  [19].

It was shown early on [8,20,21] that NSD parity-violating effects are strongly enhanced in diatomic molecules with  $^2\Sigma_{1/2}$  and  $^2\Pi_{1/2}$  electronic states due to the mixing of close rotational states of opposite parity. Thus, these systems provide a different, advantageous route for the search for these phenomena. An experiment to measure NSD-PV effects using the Stark-PV interference technique in polar diatomic molecules was proposed in 2008 by DeMille *et al.* [22]. In this approach, the opposite parity rotational or hyperfine levels of ground-state molecules are tuned to near degeneracy by a magnetic field, causing dramatic amplification of the parity-violating effects [23]. Highly sensitive measurements of this type using the BaF molecule were demonstrated recently [24,25]. Another experiment based on optical rotation measurements in  $^{199}\text{HgH}$  was also proposed [26].

In diatomic molecules with nonzero nuclear spin and  $^2\Sigma_{1/2}$  and  $^2\Pi_{1/2}$  electronic states, the nuclear anapole moment interaction can be rewritten in a slightly simplified form,

$$H_A = \kappa_A \frac{G_F}{\sqrt{2}} \sum_i \rho(\mathbf{r}_i) \alpha_+, \quad (3)$$

with

$$\alpha_+ = \alpha_x + i\alpha_y = \begin{pmatrix} 0 & \sigma_x \\ \sigma_x & 0 \end{pmatrix} + i \begin{pmatrix} 0 & \sigma_y \\ \sigma_y & 0 \end{pmatrix}. \quad (4)$$

Here,  $\sigma_x$  and  $\sigma_y$  are the Pauli matrices. The  $^2\Sigma_{1/2}$  and  $^2\Pi_{1/2}$  open-shell electronic states are twofold degenerate, corresponding to the two possible projections of electronic angular momentum along  $\mathbf{n}$ , i.e.,  $|\Omega\rangle = |\pm\frac{1}{2}\rangle$ , where  $\mathbf{n}$  is the unit vector directed along the molecular axis from the heavier to the lighter nucleus. The interaction  $H_A$  removes the degeneracy and mixes  $|\Omega\rangle$  states with different signs (parities).

The  $P$ -odd interaction coefficient  $W_A$  is usually explored for the expression of the strength of coupling of the two different parity states. This coefficient depends on the electronic structure of the molecule and is defined for a given electronic state; it can be derived from the expression for  $H_A$  as the transition element between the two different  $|\Omega\rangle$  states [27],

$$W_A = \frac{G_F}{\sqrt{2}} \langle +\frac{1}{2} | \sum_i \rho(\mathbf{r}_i) \alpha_+ | -\frac{1}{2} \rangle. \quad (5)$$

Note that the matrix elements calculated between the same  $|\Omega\rangle$  states are zero. Thus, the coefficient  $W_A$  defines the amplitude of the expectation value of  $H_A$  in the mixed-parity state. Knowledge of  $W_A$  is required for extracting the nuclear anapole moment from experiment. It cannot be measured

directly and has to be provided by theory. Needless to say, the accuracy and reliability of the calculated  $W_A$  coefficients are important for the meaningful interpretation of any measurement, and it is thus most desirable to employ state-of-the-art relativistic quantum theoretical methods for such calculations.

Here we perform relativistic coupled-cluster calculations to obtain the  $W_A$  coefficient for BaF within the framework of a finite-field approach. We investigate the sensitivity of  $W_A$  to various computational parameters allowing us to estimate the uncertainty of our result, and finally propose a recommended value for interpretations of future experiments on this molecule.

There have been two previous studies that used coupled-cluster theory for the calculation of the  $W_A$  coefficient. Relativistic two-component Fock-space coupled-cluster theory was used to calculate  $W_A$  and other  $P$ - and  $(T, P)$ -odd parameters for RaF (the authors estimated the uncertainty of the results as 10%) [27]. More recently, the relativistic Fock-space coupled-cluster method was used to calculate the  $W_A$  coefficients of the  $^2\Sigma_{1/2}$  and the  $^2\Pi_{1/2}$  electronic states of HgH [26].

The majority of earlier investigations of NSD-PV effects in diatomic molecules such as BaF have relied on more approximate approaches such as semiempirical methods [22,28], where the  $W_A$  parameters were estimated using experimental spectroscopic data. Kozlov *et al.* performed relativistic effective core potential (RECP) calculations in the framework of a self-consistent-field (SCF) approach estimating core-polarization effects by an effective operator (EO) [29]. Nayak and Das [30] carried out Dirac-Hartree-Fock (DHF) calculation within restricted active space configuration interaction (RASCI). Isaev and Berger [31] used a quasirelativistic two-component zeroth-order regular approximation (ZORA) combined with Hartree-Fock (HF) and density functional theory (DFT), and scaled the results using a semiempirical model described in Ref. [32]. We have previously carried out both DHF and DFT calculations of this property for BaF and many other diatomic molecules [33,34]. In that work, the average of the DHF and the DFT results scaled by the effect of core polarization (CP) obtained from atomic calculations was taken as the recommended value; these results are designated here as DHF/DFT+CP.

## II. METHOD AND COMPUTATIONAL DETAILS

The calculations were carried out using the adapted version of the DIRAC program package [35] in the framework of the Dirac-Coulomb Hamiltonian,

$$H_0 = \sum_i [c\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i mc^2 + V(r_i)] + \sum_{i<j} \frac{1}{r_{ij}}, \quad (6)$$

where  $\boldsymbol{\alpha}_i$  and  $\beta$  are the Dirac matrices in standard representation. The Coulomb potential  $V(r_i)$  takes into account the finite size of the nuclei, modeled by Gaussian charge distributions [36].

The  $P$ -odd interaction constant is a property of a given nucleus within the molecular environment and for a diatomic molecule we have two  $W_A$  values. In this work, however,

we entirely focus on the  $W_A$  parameter at the metal nucleus relevant for future experiments.

In order to perform coupled-cluster calculations for the  $W_A$  parameter, we employ a finite-field (FF) approach [37,38]. Within this scheme, the entire Hamiltonian of the system  $H$  is regarded to be a function of some perturbation parameter  $\lambda$ ,

$$H(\lambda) = H_0 + \lambda \frac{G_F}{\sqrt{2}} \sum_i \rho(\mathbf{r}_i) \alpha_+. \quad (7)$$

The nuclear density  $\rho(\mathbf{r}_i)$  is of Gaussian shape, which is suitable for the fully relativistic framework of the present work. For small values of  $\lambda$ , the total energy can be expanded in Taylor series around  $\lambda = 0$ ,

$$E(\lambda) = E(0) + \lambda \left. \frac{dE(\lambda)}{d\lambda} \right|_{\lambda=0} + \dots \quad (8)$$

The calculations are performed at various perturbation strengths  $\lambda$ . If these are chosen to be small enough to remain in the linear regime, the higher-order terms can be ignored and  $W_A$  can be obtained numerically, according to the Hellmann-Feynman theorem, from the first derivative of the energy with respect to  $\lambda$ :

$$W_A = \left. \frac{dE(\lambda)}{d\lambda} \right|_{\lambda=0}. \quad (9)$$

The perturbation strength needs to be sufficiently large such that the change in total energy is not lost in the precision of the calculations. We have tested the linearity of the above expression with different perturbation strengths applied, i.e.,  $\lambda = 10^{-6}, 10^{-7}, 10^{-8}$ , and  $10^{-9}$ . Based on our results, we found that minimal error in the linear fit is obtained for perturbation strengths of the order of  $\sim 10^{-8}$ . Furthermore, the energy convergence requirement of the coupled-cluster iterations had to be set to  $10^{-12}$  a.u.

We have used and compared two variants of relativistic coupled-cluster theory: the standard single-reference coupled-cluster method with single, double, and perturbative triple contributions [CCSD(T)] [39] and the multireference Fock-space coupled-cluster (FSCC) approach [40]. Within the framework of the valence-universal FSCC approach, an effective Hamiltonian is defined and calculated in a low-dimensional model (or  $P$ ) space, constructed from zero-order wave functions (Slater determinants), with eigenvalues approximating some desirable eigenvalues of the Hamiltonian. According to Lindgren's formulation of the open-shell CC method [41], the effective Hamiltonian has the form

$$H_{\text{Eff}} = P H \Omega P, \quad \Omega = \exp S, \quad (10)$$

where  $\Omega$  is the normal-ordered wave operator and the excitation operator  $S$  is defined with respect to a closed-shell reference determinant (vacuum state) and partitioned according to the number of valence holes ( $m$ ) and valence particles ( $n$ ) with respect to this reference:

$$S = \sum_{m \geq 0} \sum_{n \geq 0} \left( \sum_{l \geq m+n} S_l^{(m,n)} \right). \quad (11)$$

Here,  $l$  is the number of excited electrons.

BaF has a single valence electron occupying the  $\sigma$  orbital and thus two different computational schemes are appropriate

for this system. In the first scheme, designated FSCC(0,1), we start with  $\text{BaF}^+$ . After solving the relativistic Dirac-Fock equations and correlating this closed-shell reference state, an electron is added to reach the neutral state. At each stage, the coupled-cluster equations are solved to obtain the correlated ground- and excited-state energies. The extra electron can be added to the lowest  $\sigma$  orbital or allowed to also occupy the higher states, thus yielding a number of energy levels and also improving the description of the ground-state energy and properties. We have tested the influence of the size of the model space  $P$  on the calculated  $W_A$  parameters. Within the second computational scheme [FSCC(1,0)], the calculation begins from the closed-shell negative ion  $\text{BaF}^-$ , and an electron is removed to obtain the neutral system. In principle, the two schemes should give very similar results for the ground state, with the main difference stemming from the different closed-shell reference states yielding different Hartree-Fock orbitals (i.e., relaxation effects). In addition to the coupled-cluster results, we also report  $W_A$  values from second-order Møller-Plesset perturbation theory (MP2) [42].

In order to further investigate the effects of electron correlation, we performed open-shell single-determinant average-of-configuration DHF [43] and relativistic DFT [44] calculations for  $W_A$  by evaluating the matrix elements of the  $\alpha\rho(\mathbf{r}_i)$  operator in the molecular spinor basis. To test the performance of various functionals for this property, the DFT calculations were carried out with the Perdew-Burke-Ernzerhof (PBE) functional [45,46], the Slater local exchange (SVWN5) functional [47], the Becke-Lee-Yang-Parr hybrid functional (B3LYP) [48–50], and its Coulomb-attenuated version (CAMB3LYP\*), adapted to accurately describe PV energy shifts in heavy atomic systems obtained from coupled-cluster theory [51,52].

Standard Dyall's basis sets of varying size [53,54] were employed to investigate the basis-set effects on the calculated  $W_A$  values. To further improve our results, we augmented the basis sets of the two atoms by additional large (tight) and small (diffuse) exponent functions (see below for details). Further investigated computational parameters were the active space in the electron-correlation procedure, i.e., the number of correlated electrons and the chosen virtual energy cutoff. In addition, we include the Gaunt term in our calculations [55] as part of the Breit interaction, which corrects the two-electron part of the Dirac-Coulomb Hamiltonian up to the order of  $(Z\alpha)^2$  [56]. The Gaunt interaction is included self-consistently at the DHF step. Alongside the detailed investigations of BaF, we also perform calculations for the  $W_A$  parameters of its lighter homologues BeF, MgF, CaF, and SrF in order to examine the dependence of  $W_A$  on the nuclear charge of the group 2 atom. The positions of the atoms were chosen according to the molecular experimental equilibrium bond lengths (1.361 Å for BeF, 1.750 Å for MgF, 1.967 Å for CaF [57], 2.076 Å for SrF [58], and 2.159 Å for BaF [59]).

### III. RESULTS AND DISCUSSION

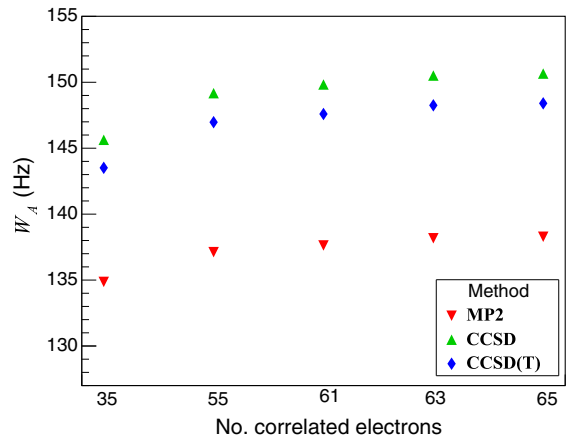
The first important step in our investigation was a detailed study of the influence of the basis set size on the  $W_A$  parameters; we also use this study to determine the best basis set which is still computationally affordable. These tests were

TABLE I. Basis-set dependence of the calculated  $W_A$  coefficient (Hz) of BaF.

Basis (Ba)	Basis (F)	DHF	CCSD	CCSD(T)
v2z	v2z	99.12	131.04	129.22
v3z	v3z	110.70	143.51	141.42
v4z	v4z	112.27	144.02	141.84
Diffuse functions				
s-aug-v4z	dyall.v4z	112.25	143.95	141.76
d-aug-v4z	dyall.v4z	112.25	143.95	141.76
dyall.v4z	s-aug-v4z	112.09	143.36	141.16
dyall.v4z	d-aug-v4z	112.04	143.27	141.05
Tight functions				
v4z+ts	v4z	112.30	144.08	141.89
v4z+tp	v4z	112.39	144.15	141.97
v4z+td	v4z	112.27	144.00	141.82
v4z+tf	v4z	112.32	145.17	143.02
v4z+2tf	v4z	112.33	145.28	143.08
v4z+tg	v4z	112.27	144.42	142.31
v4z	v4z+ts	112.27	144.02	141.84
v4z	v4z+tp	112.27	144.02	141.84
v4z	v4z+td	112.27	144.00	141.82
v4z	v4z+tf	112.27	144.00	141.82

performed within the DHF, CCSD, and CCSD(T) framework. In the coupled-cluster calculations, 35 electrons were correlated and virtual orbitals with energies above 30.0 a.u. were excluded. We used the standard Dyall's relativistic basis sets of double-, triple-, and quadruple- $\zeta$  quality [53,54]. To check the influence of diffuse functions, we have augmented the dyall.v4z basis with a single diffuse function for each symmetry (s-aug-dyall.v4z) and with two diffuse functions (d-aug-dyall.v4z). While diffuse functions are usually more important for chemical properties, a good description of the electronic wave function in the nuclear region is essential for obtaining reliable results for parity-violating effects [60]. In particular, it was demonstrated in our earlier work [33,34,61] that tight  $s$  and  $p$  functions have a considerable influence on the  $W_A$  parameter at the DHF level, especially for the lighter elements. Thus, we also tested the effect of adding different types of tight functions to the basis sets (designated as  $ts$  for high exponent  $s$  function,  $tp$  for high exponent  $p$ , etc.). The augmentations (both with the diffuse and the tight functions) were carried out separately for each of the atoms, and the results are summarized in Table I.

Going from a double- to triple- $\zeta$  quality basis set increases the calculated  $W_A$  value by  $\sim 10\%$ ; moving to quadruple- $\zeta$  quality leads to a further increase of less than a single percent on the coupled-cluster level. The correlation part of the  $W_A$  coefficient does not scale smoothly with the size of the basis set, and hence we did not perform extrapolation to the complete basis-set limit. Adding diffuse functions for barium has negligible effects on the results, while augmenting the basis of fluorine reduces the  $W_A$  value by 0.5% as the fluorine orbitals extend into the domain of the Ba atom. Out of the large exponent functions, only the tight  $f$ -type function has a discernible influence on the calculated  $W_A$ , raising its value by  $\sim 1\%$  on the CCSD and CCSD(T) level (but having no impact

FIG. 1. Calculated  $W_A$  coefficients of BaF using different number of correlated electrons.

on the DHF results). Adding a second tight  $f$  orbital leaves the calculated  $W_A$  almost unchanged. We thus assume that the results are converged (close to the basis-set limit) and perform the rest of our calculations using the optimized dyall.v4z+ $tf$  basis set.

Next we explored the effect of the number of correlated electrons and the size of the virtual space on  $W_A$ . In the first set of calculations, we keep the energy cutoff for the virtual space at a rather high value (500 a.u.) and vary the number of correlated electrons. Figure 1 presents the calculated MP2, CCSD, and CCSD(T)  $W_A$  values. Overall, the difference between including 35 electrons in the calculation (corresponding to the commonly used cutoff of  $-20.0$  a.u. in the space of the occupied orbitals) and correlating all 65 electrons is  $\sim 3\%$  for the three methods. We note that the major part of this difference does not come from the  $1s$  orbital alone, despite its proximity to the nucleus, but rather comes from all the core shells. In order to achieve  $<1\%$  accuracy, all the electrons should be included in the electron-correlation procedure.

In the next step, we perform calculations where all electrons are correlated, and vary the energy cutoff in the virtual space (Fig. 2). Unlike many other atomic or molecular properties, the calculated  $W_A$  value does not saturate at the energy cutoff of about 30 a.u., but continues to increase. The difference in the value corresponding to cutoff of 500 a.u. compared to 30 a.u. is  $\sim 3\%$ , and the  $W_A$  value continues to increase further beyond this point, albeit at a much lower rate ( $W_A = 148.91$  Hz for cutoff of 1000 a.u. vs 148.40 Hz for 500 a.u.). The importance of the inclusion of high-lying virtual orbitals for the correlation of the core electrons was also observed by Skripnikov *et al.* [62] for the scalar-pseudoscalar interaction constant  $R_s$  in the francium atom. We selected a final cutoff of 500 a.u. for the following calculations, which is a compromise between optimal accuracy and computational feasibility.

Table II contains the  $W_A$  constant of BaF (for the Ba atom), calculated at the DHF, MP2, and DFT levels of theory, using different functionals, and within various coupled-cluster schemes. These results were obtained using the optimized basis set (dyall.v4z augmented by a single tight  $f$  function), and

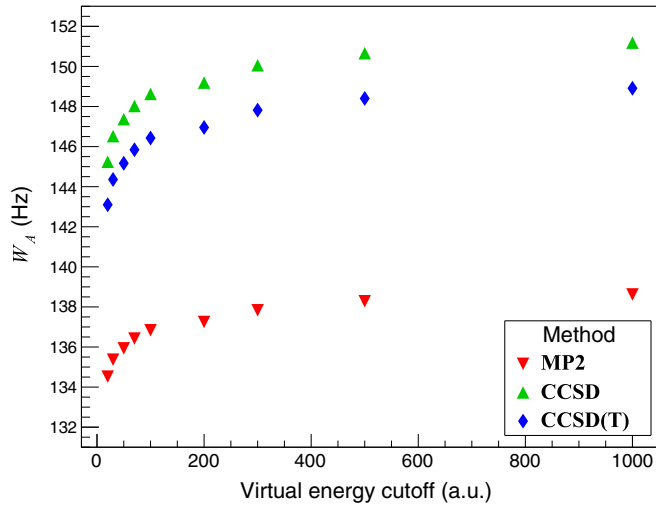


FIG. 2. Calculated  $W_A$  coefficients of BaF using different sizes of the virtual space.

in the MP2 and CC calculations all electrons were correlated with the energy cutoff for the virtual space set to 500 a.u.

Electron correlation clearly plays an important role for this property, and the CCSD results are 25% higher than the corresponding DHF values. MP2, however, performs remarkably well and captures the majority of electron correlation, differing from the CCSD value by only 8%.

DFT results tend to be very close to the DHF value; in particular, the result obtained with the CAMB3LYP\* functional, which is generally expected to perform well for parity-violating properties [52], is almost identical to the DHF result, which is somewhat disappointing. Note that similarly disappointing results are obtained for electric-field gradients of molecules containing transition metals [63], and arguments about the failure of DFT can be found in Ref. [64]. As we expect that most density functionals lie in-between the DHF and the local density approximation (SVWN5) results, here this deficiency cannot be so easily fixed by adjusting the Hartree-Fock contribution in the hybrid functional, as was done for electric-field gradients [63].

Moving to the coupled-cluster results, the triple excitations contribute very little and lower the  $W_A$  value by  $\sim 1.5\%$  only. We thus expect that the higher-order excitations in the CC procedure will not play an important role for this property.

As already mentioned, we have tested two variants of FSCC. In the first one, FSCC(0,1), the calculation starts from  $\text{BaF}^+$  and an electron is added into the virtual orbitals. Here we test two sizes of the model space: the minimal one, designated model space I, where the additional electron is allowed to occupy only the lowest  $\sigma$  orbital (yielding the ground  $X^2\Sigma_{1/2}$  state), and model space II, which contains 2  $\sigma$ , 2  $\pi$ , and 2  $\delta$  orbitals.

The second FSCC scheme, FSCC(1,0), starts with  $\text{BaF}^-$  as a reference state, and an electron is removed to reach the neutral system. Usually, one expects the FSCC results to be situated between the CCSD and the CCSD(T) values (for recent reviews of the relativistic FSCC approach, see Ref. [65]). The sector (0,1) results are extremely close to the CCSD(T) values; superior performance of FSCC in particle sectors compared to single-reference CCSD has been observed in the

TABLE II. Calculated  $W_A$  coefficient (Hz) of BaF within different correlation approaches and compared to previous predictions. The present recommended value [CCSD(T)+Gaunt] is given in bold font.

$W_A$ (Hz)	Method	Reference
112.32	DHF	This work
138.28	MP2	This work
150.66	CCSD	This work
148.40	CCSD(T)	This work
147.89	CCSD+T	This work
148.59	CCSD-T	This work
148.84	FSCC(0,1)-Model space I	This work
148.25	FSCC(0,1)-Model space II	This work
151.98	FSCC(1,0)	This work
<b>147.71</b>	<b>CCSD(T)+Gaunt</b>	<b>This work</b>
123.50	DFT(SVWN5)	This work
116.18	DFT(B3LYP)	This work
116.08	DFT(PBE)	This work
112.92	DFT(CAMB3LYP*)	This work
210–240	Semiempirical	[28]
111	RECP-SCF	[29]
181	RECP-SCF+EO <sup>a</sup>	[29]
164	Semiempirical	[22]
135	DHF	[30]
160	4c-RASCI	[30]
111	ZORA-HF	[31]
119	ZORA-DFT(B3LYP)	[31]
190	Scaled ZORA-HF <sup>b</sup>	[31]
112.9	DHF	[34]
111.6	DFT(CAMB3LYP*)	[34]
146.0	DHF/DFT+CP <sup>c</sup>	[34]

<sup>a</sup>RECP-SCF+EO: RECP SCF calculation with an effective operator describing valence-core correlations.

<sup>b</sup>ZORA-HF results with semiempirical scaling.

<sup>c</sup>Average of DHF and DFT values, scaled by a core-polarization parameter.

past [65]. It should be noted that in this case, increasing the size of the model space has a negligible influence on the results. The sector (1,0) values are slightly higher than the CCSD ones, rather than lower as one would expect. This is probably due to the fact that the (1,0) calculation starts from a negative closed-shell reference state, and the basis which was optimized for the neutral system does not provide sufficient description of the more diffuse orbitals in  $\text{BaF}^-$ .

The Gaunt interaction lowers the  $W_A$  (on the DHF level) by 0.7 Hz. We add the Gaunt contribution from the DHF calculation to the CCSD(T) result to provide the final (recommended) value for the  $W_A$  constant of BaF. This value is designated as CCSD(T)+Gaunt in Table II.

In order to put an error bar on this value, we need to examine the remaining sources of uncertainty within our computational approach. These include basis-set deficiencies, the unaccounted full triple and higher-order contributions in the coupled-cluster procedure, the choice of the virtual space cutoff, and neglect of the full Breit and higher-order QED effects. From the investigation of the basis-set effects (Table I), we see that the contribution from the diffuse functions on the F atom, which we neglect here, is around  $-0.6$  Hz, while the

effect of tight functions beyond tight  $f$  (mostly stemming from the tight  $g$ ) is  $+0.5$  Hz. These two effects cancel out, but we take a conservative estimate of basis-set uncertainty of about 0.5 Hz (of the order of magnitude of these effects) to account for any further shortfalls of the basis set. The difference in the values of  $W_A$  calculated with a virtual space cutoff of 500 and 1000 a.u. is about 0.5 Hz and it seems that saturation is reached; we thus take 1 Hz as the corresponding uncertainty. The contribution of perturbative triple excitations in the calculation via the CCSD(T) scheme is  $-2.26$  Hz. In CCSD(T), the triple excitations are fully included in the fourth order in perturbation theory, and part of the fifth-order terms is also included [66]. To test the stability of this scheme, we present the results of the CCSD-T calculation, where further fifth-order terms are included [67], as well as the CCSD+T approach [68], where the triple corrections are treated only at the fourth-order level. These values are also shown in Table II. While CCSD+T has the strongest effect on the calculated  $W_A$  ( $-2.77$  Hz), the addition of the fifth-order terms moderates the contribution of the triple excitations, and the difference between CCSD(T) and CCSD-T is negligible. Currently there is no possibility to evaluate the contribution of quadruple and higher excitation, but as the triple-excitation contribution is already quite small, we may safely neglect the higher-order ones. We take twice the difference between CCSD+T and CCSD-T (1.5 Hz) as the uncertainty due to incomplete treatment of the correlation. We assume that the effect of replacing the Breit term by the Gaunt interaction and neglecting QED effects is not more than the contribution of the Gaunt term itself ( $\sim 0.7$  Hz). The final source of uncertainty is in the numerical nature of the finite-field approach, where a slight dependence on the size of the perturbation can emerge, and for small fields numerical noise might be a factor. Test calculations that we carried out show that these effects are small, up to 0.5 Hz. Summing up all of the above effects, we get an uncertainty estimate of 2 Hz, or 1.5%.

Table II also contains the results of the previous investigations of the  $W_A$  parameter of BaF. The majority of these studies used approximate methods, such as DHF and DFT, or semiempirical approaches. Our investigation of this property in BaF is within a relativistic coupled-cluster approach, and thus direct comparison with earlier values is perhaps difficult. Our present DHF value is in excellent agreement with the RECP-SCF result of Kozlov *et al.* [29]. However, when these authors include an effective operator (EO) to account for core-polarization effects, their final value overshoots the result obtained here. The DHF and DFT results of Ref. [31] are close to the corresponding present values, but the scaling scheme seems to overcompensate for the spin-polarization effects, similar to that employed in Ref. [29]. Our earlier DHF and DFT calculations are in good agreement with the present results, as expected, and the final value in that publication, corrected for core polarization, is in fact very close to our CCSD(T) result, supporting the use of this scaling scheme. The DHF result of Ref. [30] is larger than our value and other uncorrelated calculations [29,34,69], but their RASCI value is again close to our present CCSD(T) result.

TABLE III.  $W_A$  coefficients (Hz) for alkaline-earth fluorides.

Molecule	DHF	B3LYP	MP2	CCSD(T)
BeF	0.38	0.40	0.44	0.46
MgF	3.67	4.34	4.41	4.91
CaF	7.74	8.39	9.55	10.75
SrF	37.29	41.50	45.79	50.87
BaF	112.27	116.02	138.23	147.16

It is expected that the magnitude of  $\frac{|W_A|}{R_W}$  of the  $^2\Sigma_{1/2}$  electronic state scales as  $Z^2$  [7], where the relativistic enhancement parameter  $R_W$  ( $R_W \geq 1$ ) is defined as follows [8]:

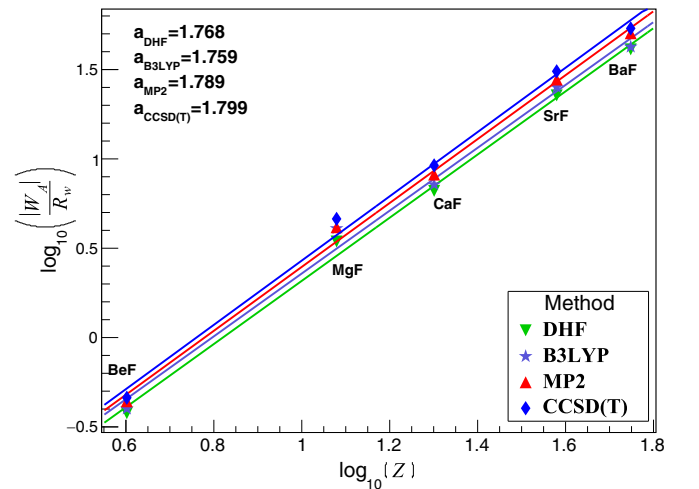
$$R_W = \frac{2\gamma + 1}{3} \left( \frac{a_B}{2Zr_0A^{1/3}} \right)^{2-2\gamma} \frac{4}{[\Gamma(2\gamma + 1)]^2}, \quad (12)$$

$$\gamma = \sqrt{1 - (Z\alpha)^2}. \quad (13)$$

Here,  $\alpha$  is the fine-structure constant,  $r_0$  is the nuclear radius, taken here as  $r_0 = 1.2 \times 10^{-15}$  m [3],  $\Gamma(x)$  is the gamma function, and  $a_B$  is the Bohr radius. To test this dependence, we have calculated the  $W_A$  parameters of the other alkaline-earth metal fluorides. These calculations were performed with the standard Dyall's v4z basis set; all the electrons were correlated and the energy cutoff for the virtual space was again set at 500 a.u. Table III contains the calculated  $W_A$  parameters at the DHF, DFT, MP2, and CCSD(T) levels of theory. In Fig. 3, we show  $\log_{10}(\frac{|W_A|}{R_W})$  as a function of  $\log_{10}(Z)$  for these systems. The results are fitted by a linear function,

$$\log_{10} \left( \frac{|W_A|}{R_W} \right) = a \log_{10}(Z) + b. \quad (14)$$

For the four computational methods, the scaling factors  $a$  are more or less identical [1.77 for DHF, 1.76 for B3LYP, 1.79 for MP2, and 1.80 for CCSD(T)], in spite of very different  $W_A$  values, implying that the trend is not sensitive to the treatment of electron correlation. Gaul *et al.* report a similar finding concerning the scaling of the  $P$ - and  $T$ -violating parameters

FIG. 3. Scaling of  $\log_{10}(\frac{W_A}{R_W})$  with  $\log_{10}(Z)$  for the selected alkaline-earth fluorides.

$W_d$  and  $W_s$  in this group of molecules [70]. It should be mentioned that for other sets of molecules investigated in Ref. [70] (i.e., group 4 oxides and group 12 hydrides), this is not the case and the Hartree-Fock and DFT scaling differ significantly. The scaling we obtain here is close (if slightly lower) to the expected  $Z^2$  dependence and in good agreement with the scaling derived from the earlier DHF+DFT results [34] and that of Ref. [31]. In this group of molecules, no additional enhancement due to electronic structure effects is observed (unlike in group 12 fluorides, for example, where the scaling is predicted to be 2.4 [34]).

#### IV. CONCLUSIONS

In this work, we presented high-accuracy relativistic coupled-cluster calculations for the nuclear-spin-dependent  $P$ -odd interaction constant  $W_A$  of BaF. The effect of various computational parameters on the obtained result was explored; these include the choice of the basis set, treatment of electron correlation, number of correlated electrons, size of the virtual space, and inclusion of the Gaunt term. We find that inclusion of electron correlation raises the calculated  $W_A$  value by about 25%; the rest of the parameters have a much weaker effect on the results, of the order of a single percent. Furthermore, the performance of various DFT functionals for

this property was investigated and found lacking. We propose a final recommended value of  $W_A = 147.7$  Hz for BaF, obtained from the CCSD(T) calculation using the optimized basis set and corrected for the Gaunt contribution. This result, with its estimated uncertainty of 1.5%, will be useful for the interpretation of future experiments on this system. We have also investigated the scaling of the  $W_A$  parameter in group 2 fluorides and found it to be close to the expected  $Z^2$  behavior.

#### ACKNOWLEDGMENTS

The authors would like to thank the Center for Information Technology of the University of Groningen for providing access to the Peregrine high performance computing cluster and for their technical support. M.I. acknowledges the support of the Slovak Research and Development Agency and the Scientific Grant Agency, Grants No. APVV-15-0105 and No. VEGA 1/0737/17, respectively. This research used the resources of a High Performance Computing Center of the Matej Bel University in Banská Bystrica using the HPC infrastructure acquired in Projects No. ITMS 26230120002 and No. 26210120002 (Slovak infrastructure for high performance computing) supported by the Research and Development Operational Programme funded by the ERDF. P.S. acknowledges support from the Centre for Advanced Study at the Norwegian Academy of Science and Letters.

- 
- [1] C. Patrignani *et al.* (Particle Data Group), *Chin. Phys. C* **40**, 100001 (2016).
- [2] M. S. Safronova, D. Budker, D. DeMille, D. F. J. Kimball, A. Derevianko, and C. W. Clark, *Rev. Mod. Phys.* **90**, 025008 (2018).
- [3] J. Ginges and V. V. Flambaum, *Phys. Rep.* **397**, 63 (2004).
- [4] P. J. Mohr, D. B. Newell, and B. N. Taylor, *J. Phys. Chem. Ref. Data* **45**, 043102 (2016).
- [5] V. N. Novikov, O. P. Sushkov, V. V. Flambaum, and I. B. Khriplovich, *Zh. Eksp. Teor. Fiz* **73**, 802 (1977) [*Sov. Phys. JETP* **46**, 420 (1977)].
- [6] V. V. Flambaum and I. B. Khriplovich, *Zh. Eksp. Teor. Fiz* **79**, 1656 (1980) [*Sov. Phys. JETP* **52**, 835 (1980)].
- [7] V. V. Flambaum and I. B. Khriplovich, *Zh. Eksp. Teor. Fiz* **89**, 1505 (1985) [*Sov. Phys. JETP* **62**, 872 (1985)].
- [8] V. V. Flambaum and I. B. Khriplovich, *Phys. Lett. A* **110**, 121 (1985).
- [9] C. Bouchiat and C. A. Pickett, *Z. Phys. C* **49**, 91 (1991).
- [10] W. R. Johnson, M. S. Safronova, and U. I. Safronova, *Phys. Rev. A* **67**, 062106 (2003).
- [11] Y. B. Zel'dovich, *Zh. Eksp. Teor. Fiz* **33**, 1531 (1957) [*Sov. Phys. JETP* **6**, 1184 (1958)].
- [12] O. Sushkov, V. Flambaum, and I. Khriplovich, *Zh. Eksp. Teor. Fiz* **87**, 1521 (1984) [*Sov. Phys. JETP* **60**, 873 (1984)].
- [13] V. V. Flambaum, I. B. Khriplovich, and O. P. Sushkov, *Phys. Lett. B* **146**, 367 (1984).
- [14] V. V. Flambaum and D. W. Murray, *Phys. Rev. C* **56**, 1641 (1997).
- [15] W. C. Haxton and C. E. Wieman, *Annu. Rev. Nucl. Part. Sci.* **51**, 261 (2001).
- [16] C. S. Wood, S. C. Bennett, D. Cho, B. P. Masterson, J. L. Roberts, C. E. Tanner, and C. E. Wieman, *Science* **275**, 1759 (1997).
- [17] J. Choi and D. S. Elliott, *Phys. Rev. A* **93**, 023432 (2016).
- [18] D. Antypas, A. Fabricant, L. Bougas, K. Tsigutkin, and D. Budker, *Hyperfine Interact.* **238**, 21 (2017).
- [19] S. Aubin, J. A. Behr, R. Collister, V. V. Flambaum, E. Gomez, G. Gwinner, K. P. Jackson, D. Melconian, L. A. Orozco, M. R. Pearson, D. Sheng, G. D. Sprouse, M. Tandecki, J. Zhang, and Y. Zhao, *Hyperfine Interact.* **214**, 163 (2013).
- [20] L. N. Labzovsky, *Zh. Eksp. Teor. Fiz* **48**, 434 (1978) [*Sov. Phys. JETP* **48**, 434 (1978)].
- [21] O. P. Sushkov and V. V. Flambaum, *Zh. Eksp. Teor. Fiz* **75**, 1208 (1978) [*Sov. Phys. JETP* **48**, 608 (1978)].
- [22] D. DeMille, S. B. Cahn, D. Murphree, D. A. Rahmlow, and M. G. Kozlov, *Phys. Rev. Lett.* **100**, 023003 (2008).
- [23] M. G. Kozlov, L. N. Labzovskii, and A. O. Mitrushchenkov, *Zh. Eksp. Teor. Fiz* **100**, 761 (1991) [*Sov. Phys. JETP* **73**, 415 (1991)].
- [24] E. Altıntaş, J. Ammon, S. B. Cahn, and D. DeMille, *Phys. Rev. A* **97**, 042101 (2018).
- [25] E. Altıntaş, J. Ammon, S. B. Cahn, and D. DeMille, *Phys. Rev. Lett.* **120**, 142501 (2018).
- [26] A. J. Geddes, L. V. Skripnikov, A. Borschevsky, J. C. Berengut, V. V. Flambaum, and T. P. Rakitzis, *Phys. Rev. A* **98**, 022508 (2018).
- [27] A. D. Kudashov, A. N. Petrov, L. V. Skripnikov, N. S. Mosyagin, T. A. Isaev, R. Berger, and A. V. Titov, *Phys. Rev. A* **90**, 052513 (2014).



- [28] M. G. Kozlov and L. N. Labzowsky, *J. Phys. B: At., Mol. Opt. Phys.* **28**, 1933 (1995).
- [29] M. G. Kozlov, A. V. Titov, N. S. Mosyagin, and P. V. Souchko, *Phys. Rev. A* **56**, R3326 (1997).
- [30] M. K. Nayak and B. P. Das, *Phys. Rev. A* **79**, 060502 (2009).
- [31] T. A. Isaev and R. Berger, *Phys. Rev. A* **86**, 062515 (2012).
- [32] M. G. Kozlov, *Zh. Eksp. Teor. Fiz.* **89**, 1933 (1985) [*Sov. Phys. JETP* **62**, 1114 (1985)].
- [33] A. Borschevsky, M. Iliaš, V. A. Dzuba, K. Beloy, V. V. Flambaum, and P. Schwerdtfeger, *Phys. Rev. A* **85**, 052509 (2012).
- [34] A. Borschevsky, M. Iliaš, V. A. Dzuba, V. V. Flambaum, and P. Schwerdtfeger, *Phys. Rev. A* **88**, 022125 (2013).
- [35] Computer code DIRAC, a relativistic *ab initio* electronic structure program, Release DIRAC17 (2017), written by L. Visscher, H. J. Aa. Jensen, R. Bast, and T. Saue, with contributions from V. Bakken, K. G. Dyall, S. Dubillard, U. Ekström, E. Eliav, T. Enevoldsen, E. Faßhauer, T. Fleig, O. Fossgaard, A. S. P. Gomes, E. D. Hedegård, T. Helgaker, J. Henriksson, M. Iliaš, Ch. R. Jacob, S. Knecht, S. Komorovský, O. Kullie, J. K. Lærdahl, C. V. Larsen, Y. S. Lee, H. S. Nataraj, M. K. Nayak, P. Norman, G. Olejniczak, J. Olsen, J. M. H. Olsen, Y. C. Park, J. K. Pedersen, M. Pernpointner, R. di Remigio, K. Ruud, P. Sałek, B. Schimmelpfennig, A. Shee, J. Sikkema, A. J. Thorvaldsen, J. Thyssen, J. van Stralen, S. Villaume, O. Visser, T. Winther, and S. Yamamoto, see <http://www.diracprogram.org>.
- [36] L. Visscher and K. G. Dyall, *At. Data Nucl. Data Tabl.* **67**, 207 (1997).
- [37] H. J. Monkhorst, *Int. J. Quantum Chem.* **12**, 421 (1977).
- [38] J. Thyssen, J. K. Laerdahl, and P. Schwerdtfeger, *Phys. Rev. Lett.* **85**, 3105 (2000).
- [39] L. Visscher, T. Lee, and K. G. Dyall, *J. Chem. Phys.* **105**, 8769 (1996).
- [40] L. Visscher, E. Eliav, and U. Kaldor, *J. Chem. Phys.* **115**, 9720 (2001).
- [41] I. Lindgren, in *Many-Body Methods in Quantum Chemistry*, edited by U. Kaldor (Springer-Verlag, Berlin, Heidelberg, 1989), p. 293.
- [42] C. Møller and M. S. Plesset, *Phys. Rev.* **46**, 618 (1934).
- [43] J. Thyssen, Ph.D. dissertation, Department of Chemistry, University of Southern Denmark, Odense, Denmark, 2001.
- [44] T. Saue and T. Helgaker, *J. Comput. Chem.* **23**, 814 (2002).
- [45] J. P. Perdew, K. Burke, and M. Ernzerhof, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [46] K. Burke, J. P. Perdew, and M. Ernzerhof, *Phys. Rev. Lett.* **78**, 1396 (1997).
- [47] S. H. Vosko, L. Wilk, and M. Nusair, *Canad. J. Phys.* **58**, 1200 (1980).
- [48] C. Lee, W. Yang, and R. G. Parr, *Phys. Rev. B* **37**, 785 (1988).
- [49] A. D. Becke, *J. Chem. Phys.* **98**, 5648 (1993).
- [50] A. D. Becke, *J. Comput. Chem.* **20**, 63 (1999).
- [51] T. Yanai and D. P. Tew, *Chem. Phys. Lett.* **393**, 51 (2004).
- [52] C. Thierfelder, G. Rauhut, and P. Schwerdtfeger, *Phys. Rev. A* **81**, 032513 (2010).
- [53] K. G. Dyall, *J. Phys. Chem. A* **113**, 12638 (2009).
- [54] K. G. Dyall, *Theoret. Chem. Acc.* **135**, 128 (2016).
- [55] J. A. Gaunt, *Proc. Royal Soc. Lond. A: Math. Phys. Eng. Sci.* **122**, 513 (1929).
- [56] G. Breit, *Phys. Rev.* **34**, 375 (1929).
- [57] *NIST Chemistry Webbook, NIST Standard Reference Database No. 69*, edited by P. J. Linstrom and W. G. Mallard (National Institute of Standards and Technology, Gaithersburg, MD, 20899), <http://webbook.nist.gov>.
- [58] R. F. Barrow and J. R. Beale, *Chem. Commun. (London)* **12**, 606a (1967).
- [59] C. Ryzlewicz and T. Törring, *Chem. Phys.* **51**, 329 (1980).
- [60] J. K. Laerdahl and P. Schwerdtfeger, *Phys. Rev. A* **60**, 4439 (1999).
- [61] A. Borschevsky, M. Iliaš, V. A. Dzuba, K. Beloy, V. V. Flambaum, and P. Schwerdtfeger, *Phys. Rev. A* **86**, 050501 (2012).
- [62] L. V. Skripnikov, D. E. Maison, and N. S. Mosyagin, *Phys. Rev. A* **95**, 022507 (2017).
- [63] R. Bast and P. Schwerdtfeger, *J. Chem. Phys.* **119**, 5988 (2003).
- [64] P. Schwerdtfeger, M. Pernpointner, and J. K. Laerdahl, *J. Chem. Phys.* **111**, 3357 (1999).
- [65] E. Eliav, A. Borschevsky, and U. Kaldor, High-accuracy relativistic coupled-cluster calculations for the heaviest elements, in *Handbook of Relativistic Quantum Chemistry*, edited by W. Liu (Springer-Verlag, Berlin, 2017), pp. 819–849.
- [66] K. Raghavachari, G. W. Trucks, J. A. Pople, and M. Head-Gordon, *Chem. Phys. Lett.* **157**, 479 (1989).
- [67] M. J. O. Deegan and P. J. Knowles, *Chem. Phys. Lett.* **227**, 321 (1994).
- [68] M. Urban, J. Noga, S. J. Cole, and R. J. Bartlett, *J. Chem. Phys.* **83**, 4041 (1985).
- [69] T. A. Isaev and R. Berger, [arXiv:1302.5682](https://arxiv.org/abs/1302.5682).
- [70] K. Gaul, S. Marquardt, T. Isaev, and R. Berger, [arXiv:1805.05494](https://arxiv.org/abs/1805.05494).