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Electron correlation *versus* stabilization of atoms in intense laser pulses

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Abstract

We present a numerical study of the stabilization process for a fully correlated two-electron model atom in an intense laser pulse. A comparison with calculations for a "real" 3D He atom is also done. We concentrate on the very high frequency regime, where the photon energy exceeds the ionization energy of both electrons, outer *and* inner. Our results show that when correlation effects are included the ionization probability (IP) is enhanced. Nevertheless, we still observe a decreasing IP within a certain intensity domain. The results from the fully correlated simulations are compared with those from simpler, approximate models. The full numerical treatment for the He atom is not yet possible. We therefore present results obtained with "single active electron" approximation and time-dependent density functional theory. Our numerical simulations can be useful for the future understanding of the stabilization phenomenon for more-than-one electron atoms.

1. INTRODUCTION

The availability of high intensity lasers has stimulated a great interest in nonperturbative effects of atomic systems interacting with intense laser light (see Protopapas *et al.*, 1997). In particular, the decrease of the ionization rate with increasing intensity, known as *adiabatic stabilization*, has been, for the last fifteen years, one of the most discussed topics in the laser–atom interaction physics community. This kind of stabilization was already predicted in 1975 (Gersten & Mittleman, 1974). Experimentally, only stabilization of highly excited atoms has been reported (de Boer *et al.*, 1993; de Boer *et al.*, 1994; van Druten, 1997). Probing ground state stabilization is instead not yet possible because it would require high intensity X-ray lasers, that unfortunately are currently not available. Therefore most of the studies in this field are of analytical or numerical nature: "high-frequency theory" (Gavrila, 1992), Floquet calculations (Dörr *et al.*, 1991; Faisal *et al.*, 1995), the numerical treatment of 1D model atoms, quantum (Su *et al.*, 1990; Grobe & Eberly, 1992, 1993; Vivirito & Knight, 1995) and classical (Rosenberger *et al.*, 1997), as well as in two-color laser fields (Cheng *et al.*, 1999), 2D atoms in arbitrary polarized laser light (Patel *et al.*, 1998), and full 3D hydrogen (Kulander *et al.*, 1991). There is also some discussion in the literature about the possibility that the atom survives in a "real" laser pulse, where an up- and a down-ramping are present (Lambropoulos, 1985). In fact, when a "real" pulse is considered, the atom also experiences low intensity fields and therefore it must pass through the "death valley" of ionization before arriving at the "magic mountain" of stabilization.

The main task of this paper is to describe how in a two-electron model atom stabilization is affected when the electron correlation is included. The purpose of the fully-correlated calculations is to achieve a qualitative picture of the stabilization mechanism in a more-than-one electron atom. In that case, in fact the full *ab initio* studies would require calculation capabilities that are far above the standard at present available and therefore approximations like "single active electron" or time-dependent density functional theory are needed. To find out which approximation tails better, a comparison with the fully-correlated results will be done.

To our knowledge only a few other numerical studies of correlated two-electron systems in the stabilization regime are reported in the literature so far (Grobe & Eberly, 1993; Lewenstein *et al.*, 1993; Volkova *et al.*, 1998*a*,*b*; Bauer & Ceccherini, 1999).

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2. THE MODEL ATOM

The 1D model with the electron-electron correlation fully taken into account is described by a 2D time-dependent Schrödinger equation (TDSE)

$$i\,\frac{\partial}{\partial t}\,|\Psi(t)\rangle = \mathsf{H}(t)|\Psi(t)\rangle \tag{1}$$

with the Hamiltonian

$$\mathsf{H}(t) = \frac{1}{2}\,(\mathsf{p}_1 + A(t))^2 + \frac{1}{2}\,(\mathsf{p}_2 + A(t))^2 - \frac{2}{\sqrt{\mathsf{x}_1^2+\epsilon}} - \frac{2}{\sqrt{\mathsf{x}_2^2+\epsilon}} + \frac{1}{\sqrt{(\mathsf{x}_1-\mathsf{x}_2)^2+\epsilon}}. \tag{2}$$

where

x_i, p_i $(i = 1,2)$ = electron's coordinates and canonical momenta, respectively

$A(t)$ = vector potential

Atomic units (a.u.) are used throughout all the paper. We chose the parameter ϵ in such a way that the ionization potentials were similar to those of real helium (0.9 a.u. for the first electron, and 2 a.u. for the second one). We therefore used ϵ equal to 0.49.

The electric field $E(t) = -\partial_t A(t)$ was a trapezoidal pulse with a rising edge over 5 optical cycles, 5 cycles of constant amplitude $\hat{E}$, and a down-ramp over, again, 5 cycles. The pulse was chosen quite short in order to avoid all the complications due to the presence of low-frequency Rabi floppings between the eigenstates of the averaged potential (Protopapas *et al.*, 1997).

All our simulations were initialized with the field-free ground state $|\Psi(0)\rangle$. The propagation in time of the wavefunction $\Psi(x_1, x_2, t) = \langle x_1 x_2|\Psi(t)\rangle$ was realized with an unconditionally stable, explicit "grid hopping" algorithm (DeRaedt, 1987). The probability amplitude $\Psi(x_1, x_2)$ approaching the grid boundary was continuously removed by an imaginary potential. The numerical grid has always been kept much larger (at least 10 times) than the excursion length

$$\hat{\alpha} = |\hat{E}/\omega^2| = |\hat{A}/\omega| \tag{3}$$

of a classical electron oscillating in the laser field. As usual, ω and $\hat{E}$ are the frequency and the amplitude of the electric field, $\hat{A}$ is the vector potential amplitude.

In the "single active electron" (SAE) approximation one assumes that an *inner* and an *outer* electron respond independently to the laser field. The inner electron "feels" the bare nuclear potential ($Z_i = 2$, hydrogen-like), whereas the outer one sees an effective nuclear charge $Z_o = 1.1$. In this way the correct ionization potentials are kept.

Thus, in the SAE approximation, we solved two independent TDSEs with no dynamic correlation at all,

$$i\partial_t \Psi_i(x,t) = \left(\frac{1}{2}\,(-i\partial_x + A(t))^2 - \frac{Z_i}{\sqrt{x^2+\epsilon}}\right)\Psi_i(x,t), \tag{4}$$

$$i\partial_t \Psi_o(x,t) = \left(\frac{1}{2}\,(-i\partial_x + A(t))^2 - \frac{Z_o}{\sqrt{x^2+\epsilon}}\right)\Psi_o(x,t). \tag{5}$$

Our model system will then be also studied applying time-dependent density functional theory (TDDFT) (see Gross *et al.*, 1996 for an overview) in local density and exchange-only approximation. This method will lead to solve the (non-linear) TDSE for *one* Kohn–Sham orbital $\Phi(x,t)$,

$$i\partial_t \Phi(x,t) = \left(\frac{1}{2}\,(-i\partial_x + A(t))^2 - \frac{2}{\sqrt{x^2+\epsilon}} + \int \frac{|\Phi(x',t)|^2}{\sqrt{(x-x')^2+\epsilon}}\,dx'\right)\Phi(x,t). \tag{6}$$

The wavefunction $\Phi(x,t)$ cannot be interpreted in terms of a single electron orbital. Therefore it is usually not straightforward to obtain one-electron quantities (like single ionization for instance).

3. HIGH-FREQUENCY RESULTS

In our simulations we will observe the amount of probability density $|\Psi|^2$ inside a box $x_1, x_2 \in [-5,+5]$ (PDIB). Before the pulse is switched on the whole wavefunction is well contained in the box and therefore PDIB is very near to unity. After the pulse is over only a fraction of probability density is still inside the box. The missing fraction has been removed by the absorbing boundaries of the grid. The survived amount of PDIB can be intrerpreted as the probability of the atom to remain neutral, that is, to survive the pulse. Within this meaning we can say that stabilization is the increase of remaining PDIB with increasing laser intensity. In our discussion we will refer to an electron as "stabilized" when its PDIB *versus* intensity shows the characteristic stabilization behavior. The remaining PDIB (or the product of the two PDIB in the two-electron case) will be referred as "stabilization probability."

If not stated otherwise, the frequency used in our calculations is $\omega = \pi$. The time-steps and spatial grid-steps are 0.1 and 0.2, respectively.

3.1. Single active electron-approximation

In the SAE approximation both electrons should stabilize, in particular the outer one since the frequency is 3.5 times larger than the ionization potential $\mathcal{E}_o = 0.9$. In Figure 1, the PDIB *versus* time for $\hat{\alpha} = 0.5$, 1.0, 1.5, and 2.0 is shown for both electrons. As expected we observe that the outer electron is more stabilized than the inner one: the amount of PDIB after

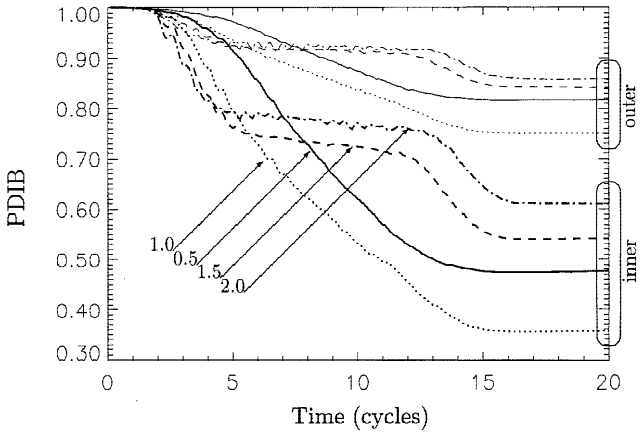

Fig. 1. The amount of probability density inside the box $x_1, x_2 \in [-5,+5]$ *versus* time for $\hat{\alpha} = 0.5$, 1.0, 1.5, and 2.0 (drawn solid, dotted, dashed, and dashed-dotted, respectively) for the inner and the outer electron in "single active electron"-approximation. The laser pulse of frequency $\omega = \pi$ was ramped-up linearly (in field) over 5 cycles, held constant for another 5 cycles before the linear down-ramp between $t = 10$ and $t = 15$ cyc. As expected, the outer electron is more stabilized. For the two higher $\hat{\alpha}$-values we clearly see that ionization is weak during the *intense, constant* part of the pulse.

the pulse has gone is greater for the outer electron. When the field intensity is higher ($\hat{\alpha} = 1.5$, drawn dashed, and $\hat{\alpha} = 1.5$, 2.0, drawn dashed-dotted) it is possible to observe that the electron ionization occurs mainly during the two rampings, in particular after the up-ramping the curves bend sharply.

Finally, after the pulse is over (at $t = 15$ cycles) the amount of PDIB is stationary. Moreover we observe that there is no monotonous increase of remaining PDIB with increasing intensity and the so-called death valley in our model seems to be located around $\hat{\alpha} = 1$ for *both* electrons.

In Figure 2 the PDIB for both electrons is shown *versus* the excursion $\hat{\alpha}$. The quiver amplitude $\hat{\alpha}$ scales to the laser intensity as $\hat{\alpha} = I^{1/2}\omega^{-2}$. The PDIB of the inner electron exhibits an oscillatory behavior. A maximum is obtained for $\hat{\alpha} \approx 4$, if the field intensity is increased ionization increases again up to a PDIB minimum around $\hat{\alpha} \approx 7$.

An oscillatory character of the ionization rate in 1D systems has been observed by other authors as well (Yao & Chu, 1992; Millack, 1993; Su *et al.*, 1996). The overall behavior observed is summarized by a decreasing of the atomic population with increasing intensity. In contrast to our results, an overall increase of stabilization with increasing intensity was also found (Yao & Chu, 1992; Millack, 1993). We think that this is mainly due to the fact that we are looking at the ionization *probability* after the pulse is over while in analytical papers often the ionization *rate* is discussed and moreover in this latter case the ionization during the up- and down-ramps is not usually taken into account.

The probability to have a neutral He atom after the pulse is over is, in SAE approximation, simply the product of the probabilities for each electron to remain bound. In Figure 2 the corresponding curve (indicated by $\mathrm{i}\cdot\mathrm{o}$) and the fully correlated results are shown. Because it seems that SAE treatment overestimates the probability of the atom to remain neutral, we could argue that *if* the system stabilizes it will probably stabilize in such a way that the correlation energy is minimized. Therefore it might be more adequate to deal with two inner electrons (the correlation term is crossed out). We will refer to this model as the "independent electron" (IE) model. The result is included in Figure 2, labeled $\mathrm{i}\cdot\mathrm{i}$; apart from oscillations in the IE result the agreement with the fully correlated one is good. Electron correlation obviously washes out these oscillations in the stabilization probability.

It is useful to study the stabilization dynamics in the time-averaged Kramers–Henneberger potential (TAKHP), that is, the frame of reference where the quivering electron is at rest but the nuclear potential oscillates, and averages over one cycle,

$$V_{KH}^{corr}(x_1, x_2) = \sum_{i=1}^{2} \frac{\omega}{2\pi} \int_0^{2\pi/\omega} \frac{-2}{\sqrt{(x_i + \alpha(t))^2 + 0.49}}\, dt + \frac{1}{\sqrt{(x_1 - x_2)^2 + 0.49}}. \quad (7)$$

When the frequency is sufficiently high this is the leading term in a perturbation series in ω^{-1} (Gavrila, 1992). This potential is compared in Figure 3 with that one where the correlation is neglected. With correlation, there are two minima near $x_1 = \hat{\alpha}$, $x_2 = -\hat{\alpha}$, and $x_1 = -\hat{\alpha}$, $x_2 = \hat{\alpha}$ whereas without correlation there are two more, energetically equivalent minima at $x_1 = x_2 = \hat{\alpha}$ and $x_1 = x_2 = -\hat{\alpha}$. Since the

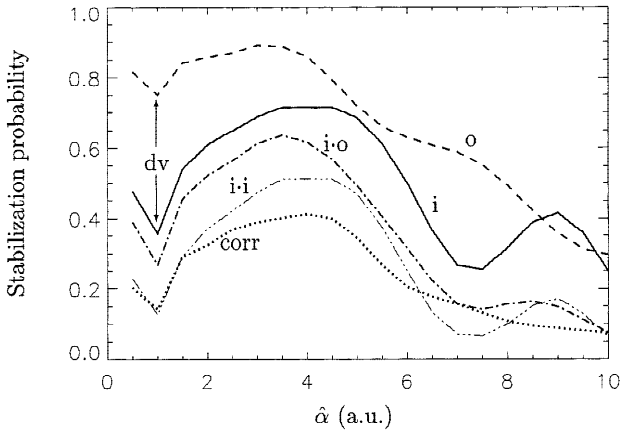

Fig. 2. Stabilization probability for inner (i) and outer (o) electron in SAE-approximation for $\omega = \pi$. The fully correlated treatment leads to the stabilization probability (that is, the probability for the He model atom to survive as *neutral* He) drawn dotted and indicated with "corr." From a SAE viewpoint one would expect the product of the curves i and o, indicated with "$\mathrm{i}\cdot\mathrm{o}$". Obviously, in SAE approximation the stabilization probability is overestimated. The so-called death valley (dv) for small $\hat{\alpha}$ is located around $\hat{\alpha} \approx 1$ for both, SAE and fully correlated results. The $\mathrm{i}\cdot\mathrm{i}$-curve results from an independent electron model with two inner electrons. This curve seems to oscillate around the correct curve "corr."

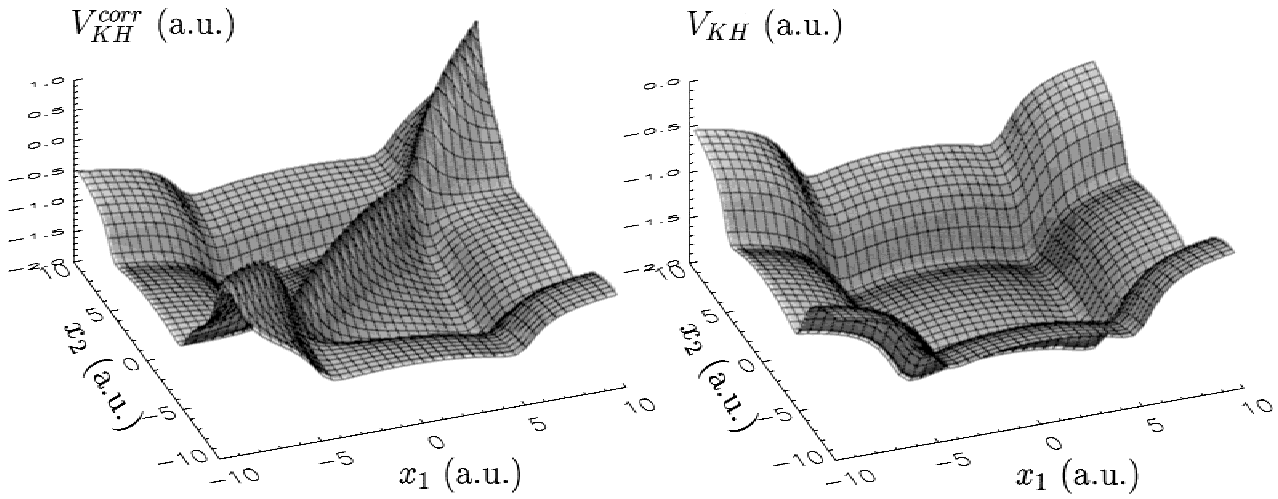

Fig. 3. The time-averaged Kramers–Henneberger potential with (V_{KH}^{corr}) and without (V_{KH}) electron correlation included for $\hat{\alpha} = 5$. With correlation, there are two minima near $x_1 = \hat{\alpha}$, $x_2 = -\hat{\alpha}$ and $x_1 = -\hat{\alpha}$, $x_2 = \hat{\alpha}$ whereas without correlation there are two more, energetically equivalent minima at $x_1 = x_2 = \hat{\alpha}$ and $x_1 = x_2 = -\hat{\alpha}$. However, once the fully correlated system is in the ground state corresponding to V_{KH}^{corr}, the correlation energy is small (for not too small $\hat{\alpha}$) since the interparticle distance is $2\hat{\alpha}$.

interparticle distance is $2\hat{\alpha}$, the higher $\hat{\alpha}$ the lower the correlation energy. We therefore believe that this is the physical reason that, for increasing $\hat{\alpha}$, the agreement of the IE results with the fully correlated becomes better. Moreover, this viewpoint is further supported by examining the probability density of the fully correlated system during the pulse. In Figure 4, $|\Psi(x_1, x_2)|^2$ is shown for $\omega = \pi$, $\hat{\alpha} = 4.0$ at $t = 7.5$ cycles, that is, in the middle of the constant part of the trapezoidal pulse. Clear *dichotomy* is observable, that is, two probability density peaks at the classical turning points, the other two peaks at $x_1 = x_2 = \pm 4$ are suppressed by correlation. The correlation energy is rather small since the distance between the two peaks in the x_1x_2-plane is $\sqrt{8}\hat{\alpha}$. In the work by Mittleman (Mittleman, 1990) such multielectron "dichotomized" bound states are calculated.

3.2. Time-dependent density functional theory

In Figure 5 we present the results obtained from the TDDFT calculations. We take the Kohn–Sham orbital $\Phi(x, t)$ as an approximation to a single electron orbital. Therefore we can look at $|\Phi(x, t)|^2 \cdot |\Phi(x, t)|^2$, integrated over the region $-5 < x < 5$ after the pulse is over, as our TDDFT stabilization probability. We see that for $\hat{\alpha} < 1.5$ the agreement between TDDFT and the correct result is very good. For low intensities ($\hat{\alpha}$ up to about 5.5) electron correlation suppresses sta-

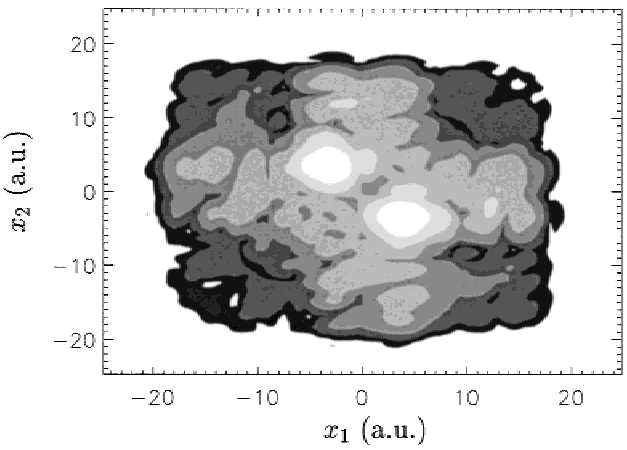

Fig. 4. Probability density $|\Psi(x_1, x_2)|^2$ for $\omega = \pi$, $\hat{\alpha} = 4.0$ at $t = 7.5$ cycles, that is, in the middle of the constant part of the trapezoidal pulse. We clearly observe *dichotomy*, that is, two probability density peaks around $x_1 = \mp 4, x_2 = \pm 4$. Therefore, the correlation energy is rather small since the distance between the two peaks in the x_1x_2-plane is $\sqrt{8}\hat{\alpha}$.

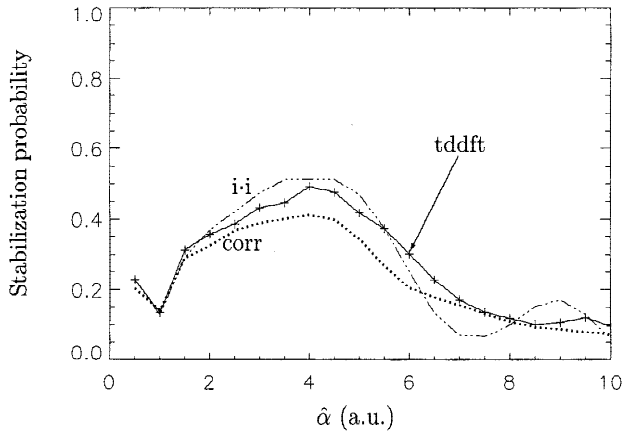

Fig. 5. Stabilization probability in the high-frequency case ($\omega = \pi$) calculated from time-dependent density functional theory (TDDFT). The fully correlated results (labelled 'corr') and the independent electron curve (indicated by i·i) are included for comparison. The agreement of TDDFT with the exact result is good: "death valley" and stabilization maximum are at the same $\hat{\alpha}$ position, respectively. For higher $\hat{\alpha}$ the agreement is even better.

bilization compared to the IE approximation. In that region TDDFT agrees better with the full result. The TDDFT result agrees very well with the fully correlated curve also for $\hat{\alpha} \geq 7$. In summary, we can conclude that the TDDFT result is in good agreement with the exact, fully correlated stabilization probability. Both approximation (IE and TDDFT) have their maximum around $\hat{\alpha} \approx 4$ and the "death valley" is also at the same position. For higher intensities the agreement is even better.

4. 3D HE ATOM

We also performed some simulations for a 3D He atom. Using a linear polarized laser we have to deal with two numerical dimensions for each electron. A fully correlated treatment is therefore not possible. We used again "single active electron" approximation and TDDFT. The results for the frequency $\omega = 3.4$ are shown in Figure 6. The qualitative behavior does not differ too much compared to the 1D model: the "death valley" structure is present, followed by a stabilization maximum. It seems quite evident that, compared to the 1D model, there is a general increase of ionization probability and stabilization is more difficult to achieve.

5. DISCUSSION AND SUMMARY

In this paper we studied the probability for stabilization of a two-electron atom when correlation effects are included. In spite of a general increase of the ionization probability stabilization is still present. We observed clear stabilization for frequencies that exceed *both* ionization potentials. The necessity to fulfill this frequency condition was also stressed by Dörr *et al.* (1995). When this high-frequency regime is considered the two electrons behave more like two independent *inner* electrons. Similar results were obtained for a H^- model-ion (Grobe *et al.*, 1993).

The agreement of the exact numerical result with TDDFT in the high-frequency case was quite good.

Finally, we would like to compare the results of the 1D model with those obtained for the "real" 3D helium atom. It is quite clear that in the 3D case stabilization is less pronounced. Moreover, while in the 1D model TDDFT approaches the IE approximation for the real atom it seems that SAE is a better model. To understand the physical reason further studies must be undertaken. In 3D the oscillatory character is less visible, that is, we observe a single stabilization maximum followed by a rather monotonous increase of the ionization probability. The difference of 1D models

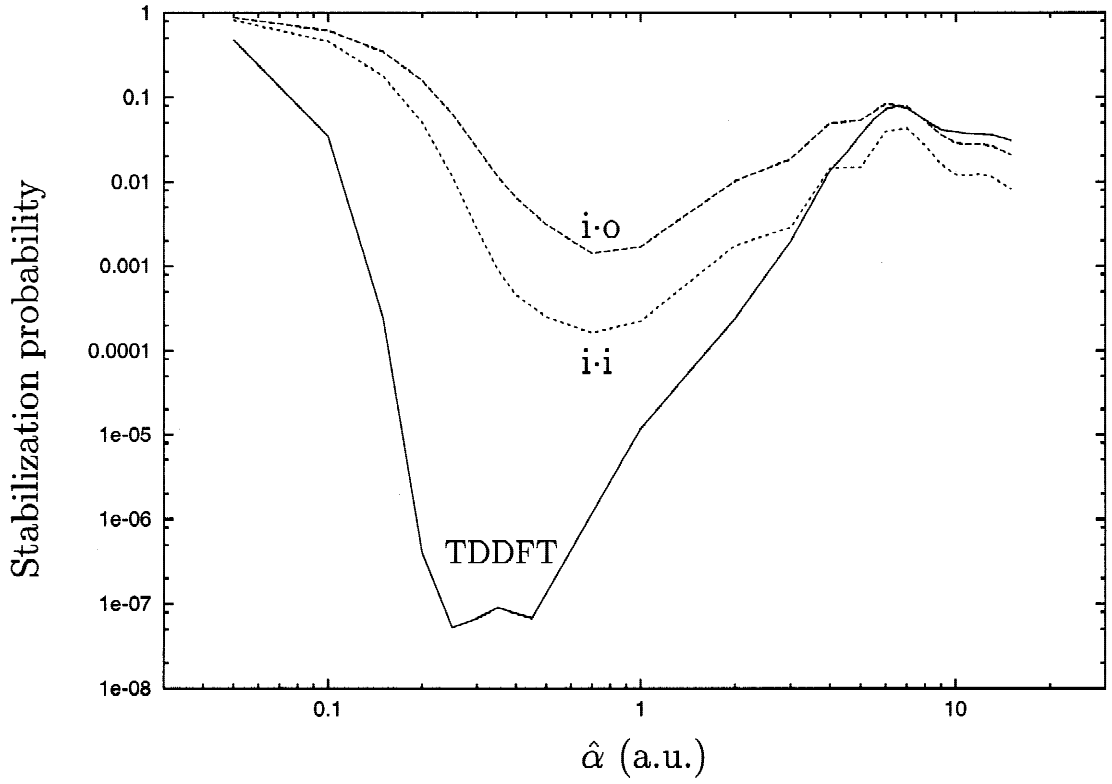

Fig. 6. Stabilization probability for a 3D He atom. The results from "single active electron" (SAE) and "independent electron" (IE) approximations (labeled i · o and i · i, respectively) are compared with that one from time-dependent density functional theory (TDDFT). While in the 1D model TDDFT approaches the IE approximation for the real atom it seems to be the other way around.

and 3D hydrogen was also studied (Millack, 1993). The effect of electron correlation in 3D stabilization will be the subject of a future paper (Ceccherini, *et al.*).

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