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LETTER TO THE EDITOR

Electron correlation effects in the double ionization of HeDemetris G Lappas[†] and Robert van Leeuwen[‡][†] Department of Physics, Lund Institute of Technology, PO Box 118, S-221 00 Lund, Sweden[‡] Department of Theoretical Physics, University of Lund, Sölvegatan 14A, S-223 62 Lund, Sweden

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Abstract. The double-ionization yield of He is calculated with a one-dimensional fully correlated two-electron model for the low laser frequency of recent experiments. Results for a higher laser frequency also indicate a comparable very high double-ionization yield for sufficiently short pulses. It is shown that the Hartree–Fock approximation fails dramatically in describing the two-electron dynamics. Also, in a density functional theory approach, we demonstrate the need for an improved exchange correlation potential and for more accurate density functionals for the ionization probabilities.

The interaction of a strong laser pulse with an atom results in a variety of exciting new phenomena that have challenged our traditional understanding of light scattering and photoionization [1]. The strength of the laser–atom interaction, being of the order of the electric field that holds the electron in the ground state of the hydrogen atom, and the ultra-short duration of the pulse (tens of femtoseconds) have revealed a new regime for the multi-electron ionization of atoms.

Since the first short laser pulse experiments that showed evidence of direct double ionization in noble gases [2] there has been considerable progress in studying and understanding the process of correlated two-electron effects in ionization. Recently, the very accurate measurements of Fittinghof *et al* [3] and Walker *et al* [4], which showed a ‘knee’ structure in the ion yield curves of He, remain the best experimental evidence of direct double ionization for short-pulse (160 fs) irradiation. Theoretical models based on either a simplified dielectronic interaction [5] or more traditional perturbative methods [6] have reproduced most of the experimental data and illuminated much of the physical mechanism of the two-electron ejection.

In this letter we present an *ab initio* calculation of ionization yields of He in a one-dimensional model with two fully correlated electrons at the low laser frequency of recent experiments [4]. To the best of our knowledge this is the first ‘exact’ calculation for frequencies that are low enough to allow for tunnelling ionization [7]. For higher frequencies, where the ponderomotive energy of the electron is much lower, numerical calculations are less demanding with respect to space–time resolution. Results in this frequency regime were obtained recently by Bauer [8] with a model similar to ours. Furthermore, in this work, we can make a direct comparison with other approximate theoretical approaches, such as time-dependent Hartree–Fock (TDHF) and time-dependent density functional theory (TDDFT), and check their validity. Predictions can be made for different laser parameters, and we present calculations for a much shorter laser wavelength.

The detailed description of our two-dimensional model has been given elsewhere [9, 10]. Here, we only mention that the two electrons are allowed to move only along the x -axis in the neighbourhood of a nucleus that is fixed at the origin. In atomic units (au) the two-electron field-free Hamiltonian is

$$H = \frac{1}{2}p_1^2 + \frac{1}{2}p_2^2 + 2V(x_1) + 2V(x_2) - V(x_1 - x_2) \quad (1)$$

where both the electron–nucleus attraction and the electron–electron repulsion are described by the non-singular, soft-core and asymptotically Coulombic potential $V(x) = -1/\sqrt{x^2 + 1}$. The soft core of the interaction potential is physically reasonable in the sense that in a real atom the two electrons have enough available space to bypass each other [9]. The ground state of the system is found to be -2.238 au, by the method of imaginary-time evolution of the field-free Hamiltonian of equation (1). In our calculations the wavefunction $\psi(x_1, x_2)$ is taken to be symmetric with respect to exchange of the two electrons.

The laser interaction is added in the minimal-coupling form, as a six-cycle duration pulse which is turned on and off linearly within two cycles, and has a two-cycle central part with constant amplitude. The time-dependent Schrödinger equation is solved numerically. The two-electron wavefunction is represented in space on a two-dimensional grid. We can employ an absorbing boundary at the ends of the spatial box of integration in order to avoid reflections, if this becomes necessary. Since at the frequency of $\omega = 0.0584$ au (780 nm laser) and intensities of up to 2×10^{15} W cm $^{-2}$ the ponderomotive energy of the electron is very high, a resolution of up to 2000 time steps per optical cycle is needed, with correspondingly high spatial resolution as well. These requirements make the calculations very time consuming and tedious, especially because in the present problem of correlated double ejection the total two-electron probability distribution in all space is needed in order to correctly account for single and double ionization. In this case the omission of absorbers at the box ends creates the need for rather long boxes. The two electrons can remain correlated at large distances from the atomic core and thus may not allow for an independent electron approximation away from the origin [11].

The time evolution of the two-electron wavefunction is performed with the split-operator method [12], which allows for much larger grid spacing Δx than that used in other non-spectral methods. The reason is that with our method the second space derivatives of the kinetic energy operator are expressed exactly in momentum space. The program runs were performed on a modern workstation.

The double-ionization probability was calculated as the total probability that both electrons are at least $R = 5$ au away from the origin at the end of the laser pulse \dagger . This information is obtained directly from the fully correlated two-electron wavefunction as a two-dimensional integral of the joint probability density in the four regions of the x_1 – x_2 plane, i.e.

$$P_2 = \int_{|x_1|>R} dx_1 \int_{|x_2|>R} dx_2 |\psi(x_1, x_2)|^2. \quad (2)$$

The single-ionization probability is the probability that only one electron is within a distance R from the origin,

$$P_1 = 2 \int_{|x_1|<R} dx_1 \int_{|x_2|>R} dx_2 |\psi(x_1, x_2)|^2 \quad (3)$$

where the factor of 2 is due to the symmetry under exchange. The method of projections to ionic bound states yields the same ionization results, as has been verified in a previous

\dagger The dominance of tunnelling in this low-frequency regime diminishes the probability of excitation to higher bound atomic states. A brief discussion on the validity of this definition of ionization can also be found in [13].

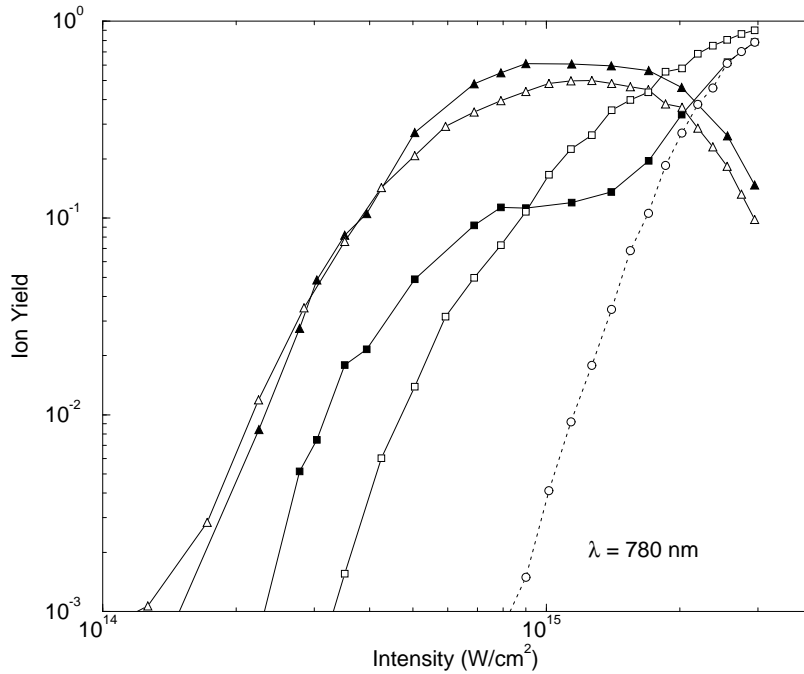


Figure 1. Single (triangles) and double (squares) ionization yields of He when irradiated by a 780 nm six-cycle laser pulse, as obtained from the ‘exact’ fully correlated two-electron model (full triangles and squares). The corresponding curves from a Hartree–Fock calculation (open triangles and squares) either overestimate or underestimate the actual yields, depending on the intensity regime. The (single) ionization yield from the He^+ ion is also shown (open circles connected with dotted lines).

work [10]. Our results are presented in figure 1 (full triangles and squares). We see a clear knee structure in the double-ionization curve at an intensity that is reasonably close to the experimental one. Although the reduced dimensionality of our model does not allow for a direct quantitative comparison with experiment, most of the experimental features are reproduced. For example, the double-ionization yield is two to three orders of magnitude higher than what one would expect from a ‘sequential’ process in which the second electron comes only from the ionization of He^+ , for intensities up to $10^{15} \text{ W cm}^{-2}$. We observe that the knee structure is located around the saturation intensity of single ionization. Similar curves were obtained by Watson *et al* [5] for partially correlated electrons in one and three space dimensions. Becker and Faisal [6] also calculated the double-ionization yield, but only up to the saturation intensity of single ionization. The origin of this enormous direct signal is traced to the strong energy transfer between the two electrons in their mutual interactions while being driven by the laser [5, 6].

Density functional theory and its time-dependent version [14] has the potential of reducing a many-body problem into a set of one-body problems that are computationally tractable. In this work, we test the applicability of such an approach in our case of the strongly correlated system of the He atom. This is the best testing ground for TDDFT, in view of the discovery of one of the most important manifestations of electron correlation in atomic physics, namely the very high probability of direct double ionization in a strong and short-pulsed laser field.

In figure 1 we show the results from a one-dimensional TDHF calculation, that has the exchange interaction only. Within the framework of a TDDFT approach [15, 16], this is equivalent to solving the Kohn–Sham (KS) equation for a single orbital in a system of two equivalent electrons, with exchange, but without correlation interaction:

$$i\frac{\partial}{\partial t}\phi(x; t) = \left[-\frac{1}{2}\frac{\partial^2}{\partial x^2} + V_{\text{KS}}(x; t) \right] \phi(x; t) \quad (4)$$

where the KS time-dependent potential V_{KS} is a functional of the total electron density $n(x; t) = 2|\phi(x; t)|^2$, i.e.

$$V_{\text{KS}}[n](x; t) = V_{\text{ext}}(x; t) + \int dy \frac{n(y; t)}{\sqrt{(x-y)^2 + 1}} + V_{\text{XC}}[n](x; t). \quad (5)$$

Here $V_{\text{ext}}(x; t)$ includes the potential of the nucleus and the external laser potential in the dipole approximation. The exchange-correlation potential V_{XC} is a functional of the density in such a way that the density $n(x; t)$ of the fully interacting two-electron system is reproduced at all times. We use only the exchange part of the exchange-correlation potential, which reads

$$V_{\text{X}}(x; t) = -\frac{1}{2} \int dy \frac{n(y; t)}{\sqrt{(x-y)^2 + 1}}. \quad (6)$$

It is evident from figure 1 that the agreement between the two methods—the fully correlated and the TDHF—is very poor and, most importantly, no knee structure similar to that in the experimental and model curves is observed in the TDHF curves. Earlier TDHF calculations have also explicitly shown the inadequacy of the method in cases of describing correlated electronic wavefunctions [17].

In order to show the onset of a ‘sequential’ process, we also show in figure 1 the ionization yield from a one-dimensional He^+ model ion (open circles connected with dotted lines). In this case the electron only experiences the potential of the He nucleus and the laser, a process that can dominate in the ionization of the He atom only if one electron is ejected very early, almost at the beginning of the laser pulse. In the latter case, the full ionization process could be described as ‘sequential’. No clear distinction between ‘nonsequential’ (or ‘direct’) and ‘sequential’ ionization exists, because both electrons are always active at the same time. It is only the short duration of the laser pulse that allows for the observation of a high double-ionization signal before the depletion of the neutral atoms.

In figure 1, the convergence of the two curves at high intensities is due to the fact that when the single ionization saturates early during the interaction with the laser pulse—for such high intensities—only the He^+ ions remain and can produce ion yields for the most part of the pulse. We observe that the high signal of double ionization extends only up to the saturation intensity of single ionization. Since for higher intensities the direct curve has to merge with the sequential one, the famous knee structure is produced when the curve bends to the right, simply due to a reduced number of available neutral He atoms that can yield a direct signal. It is therefore reasonable to expect that one can move the position of the knee, and consequently increase or reduce the direct signal, by simply changing the saturation intensity. For example, a lower saturation intensity appears when the laser pulse is longer, because it can then deplete the neutral He atoms and still have sufficient time to interact with the remaining ions, yielding a relatively much higher sequential than direct signal. In this case the knee is moved to such low intensities that make its observation difficult due to the high noise-to-signal ratio in the experimental detection. This explains

why the knee structure of the double-ionization yield had not been discovered before the advance of short-pulse lasers [2].

An important issue that has to be addressed at this point is that in calculations where the electronic density rather than the orbitals have a physical interpretation, the definition of single and double ionization is less clear, as we lack information on the joint two-electron detection probability (which, of course, is a functional of the density). However, a single-orbital definition of ionization can be given approximately as follows: let $\phi(x)$ be the orbital that describes each of the two equivalent electrons of He. At some time t after the interaction with the laser, the probability $p = p(t)$ that an electron is within a distance $R = 5$ au from the origin is simply

$$p(t) = \int_{-R}^{+R} dx |\phi(x; t)|^2. \quad (7)$$

The probability P_0 that both electrons are within the distance R is $P_0 = p^2$. The single-ionization probability P_1 is the probability that only one of the two electrons is within R , i.e.

$$P_1 = 2p(1 - p) \quad (8)$$

and the double-ionization probability P_2 is then

$$P_2 = (1 - p)^2. \quad (9)$$

Note that the above equations follow simply by putting a product wavefunction into equations (2) and (3). We can check the accuracy of the above approximate formulae of

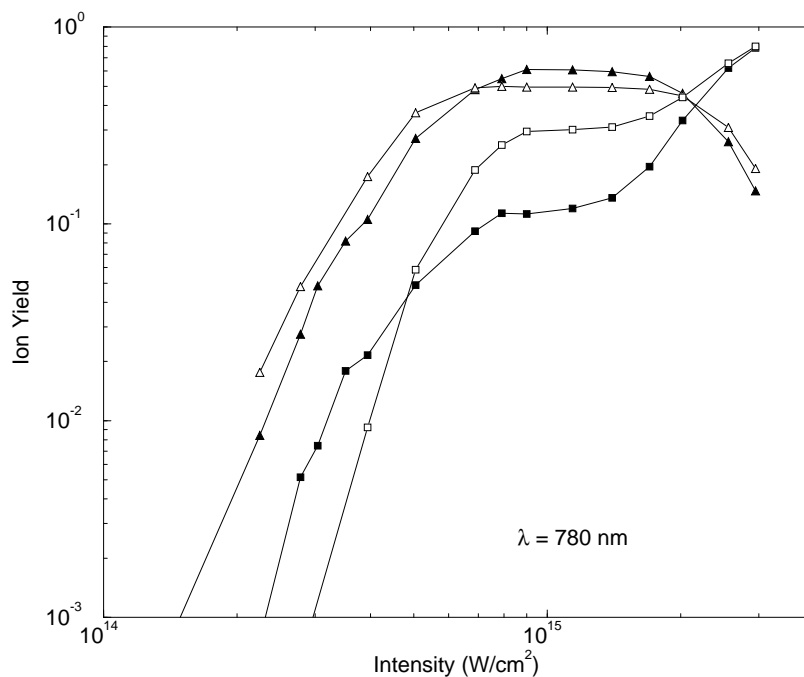


Figure 2. Single- and double-ionization yields of He from the fully correlated exact model (full triangles and squares), and the estimate based on equations (8) and (9) in the text with the same 'exact' electron densities (open triangles and squares).

equations (8) and (9) by using the exact density of the two-electron model calculation in the above formulae and comparing the resulting ion yields. In the TDDFT terminology this is equivalent to solving the KS equation (4) with the *exact* exchange-correlation potential in equation (5) and using the KS product wavefunction as an approximation to the full two-electron wavefunction. From figure 2 (open squares) we see that a knee structure in the double-ionization curve is now reproduced, but still does not agree with the fully correlated results (full squares). This indicates that the ionization probabilities must not be given accurately by the TDDFT calculation. In the case of the TDHF calculation the knee structure is not reproduced at all, as is evident in figure 1 (open squares). Therefore, an improved single-particle KS potential with a more accurate exchange-correlation part, probably involving memory effects [18, 19], should be included in a TDDFT approach, rather than TDHF, in order to produce the correct density. The fact that the TDDFT curves, calculated with the exact density (i.e. with an exact exchange-correlation potential, but approximate expressions for the probabilities) still do not agree, indicates that the definition of single and double ionization in TDDFT also has to be modified. Therefore, we need better density functionals for the probabilities than those in equations (8) and (9). One way of doing that could be by introducing a pair correlation function [16].

We have found that the physics of double direct ionization is strongly dependent on the laser frequency. In figure 3 we show the same ion yields for He, but for a 248 nm ($\omega = 0.183$ au) laser pulse. The structure of the curves is rather rich compared to the previous low-frequency case. This is an indication of the presence of multi-photon resonances, as we are now away from the tunnelling ionization regime. Similar structures

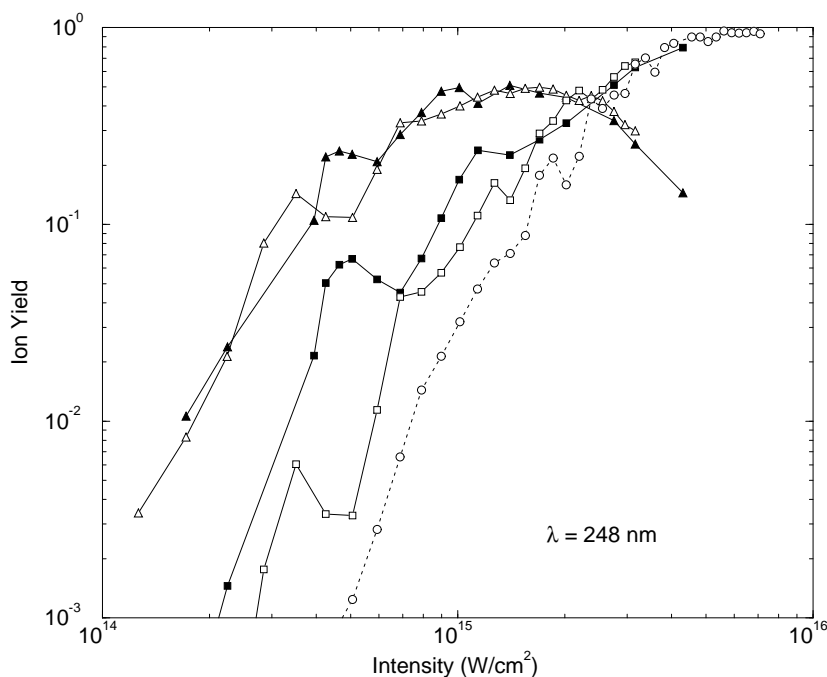


Figure 3. Single- and double-ionization yields of He when irradiated by a 248 nm six-cycle laser pulse, as obtained from the ‘exact’ fully correlated two-electron model. The (single) ionization yield from the He^+ ion is also shown (curves as in figure 1).

have been observed already in experiments and other Hartree–Fock calculations [20]. We observe that the Hartree–Fock yields are now much closer to the exact yields than they were before, in the low-frequency case. The prominent curve structures are now reproduced by both calculations, indicating that the Hartree–Fock method is better suited to the high-frequency multi-photon regime, at least for the problem of ionization. Other calculations [8], with even higher frequencies ($\omega = 0.4$ au), reached similar conclusions regarding the insufficiency of the Hartree–Fock approximation.

From figure 3, we observe that the double-ionization probability is one to two orders of magnitude larger than one would expect from a purely sequential process (see the open circles connected with dotted lines). At the laser wavelength of 248 nm, where tunnelling and ‘recollisions’ [21] cannot be a valid picture of understanding the two-electron effects, the direct double-electron ejection occurs no less dramatically than for the long wavelength of 780 nm. We are currently investigating the importance of the ultra-short duration of the laser pulses in probing the initial correlated wavefunction, which appears to be the origin of the enormous direct double-ionization effect.

Finally, in figure 4, we repeat the same calculations as for figure 2, but at the new wavelength of 248 nm. In this case, we observe that the approximate probability formulae (8) and (9), with the exact densities, are much closer to the exact curves, than in the previous case of the long wavelength. Also, since formulae (8) and (9), with the exact densities, are remarkably closer to the exact curves than the Hartree–Fock curves of figure 3, we conclude that the error is mainly due to the densities rather than to the formulae that define the ionization. Hence, in this case, the main error is due to the ‘exchange-only’ approximation for the exchange–correlation potential in equation (6).

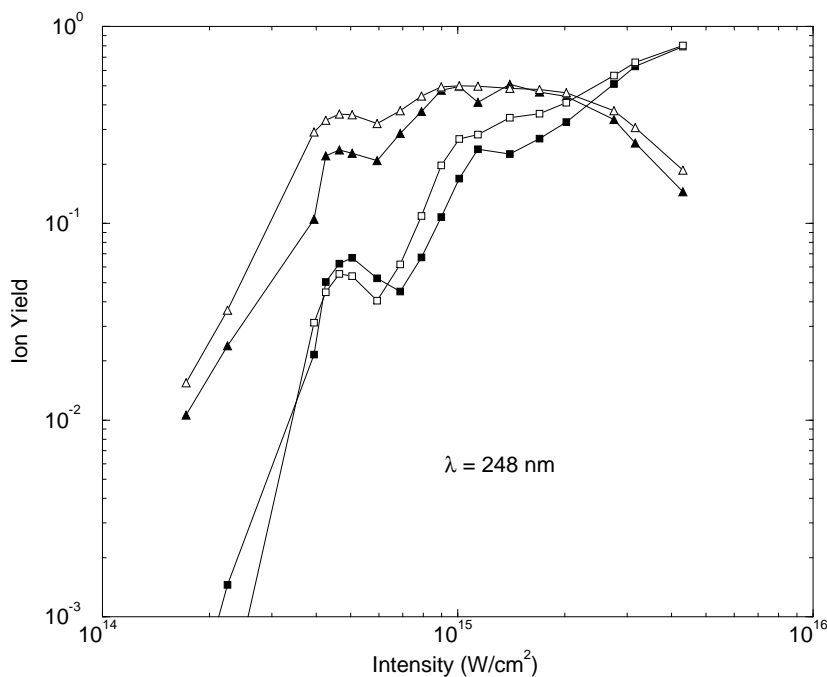


Figure 4. Single- and double-ionization yields of He, as in figure 3, from the fully correlated ‘exact’ model (full triangles and squares), and the estimate based on equations (8) and (9) in the text with the same exact electron densities (open triangles and squares).

In summary, we have reproduced the experimental features for the direct double-ionization yield of He in the short-pulse low-frequency regime using a one-dimensional two-electron model with full electron correlation. Comparison with TDHF/TDDFT calculations has shown very poor agreement. We conclude that more accurate density functionals for the exchange-correlation potential and the ionization probabilities are needed for a TDDFT approach, so that one can correctly account for the essential aspects of electron correlation in He. From our fully correlated calculations we also demonstrate that a very high signal of direct double ionization can be detected for much shorter wavelengths, if the laser pulse duration is sufficiently short.

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