Role of the correlation charge in the double ionization of two-electron model atoms exposed to intense laser fields

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We investigate the influence of electron correlation in the double ionization of a two-electron model atom exposed to an intense laser field. Using the freedom of our model system, we vary in a systematic way the correlation strength between the two electrons. For the case of five-photon ionization, while adjusting the laser frequency according to the correlation charge, we investigate extreme situations of strong and weak two-electron interaction regimes. We monitor the single and double ionization yield and study the variation of the nonsequential ionization.

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It is commonly accepted that the influence of the correlation between the two electrons is relevant for an unexpectedly high yield of double ionization in helium. This nonsequential double ionization has been measured in experiments using intense femtosecond laser pulses [1] and compared to theoretical predictions according to the Ammosov-Delone-Krainov (ADK) mechanism [2]. Correlation also plays a prominent and universal role in the production of doubly or multiply charged ions in diverse multielectron systems including atoms [3], large molecules [4], and clusters [5]. However, only limited progress has been made with respect to a theoretical modeling and understanding of this effect, even for helium.

While time-independent analytical calculations are able to model well the final ionization yield [6], time-dependent calculations of the ionization of two-electron atoms in intense laser fields are still a prohibitive numerical task. Though a huge effort has been undertaken in Belfast for time-dependent simulations [7], these calculations on parallel computers are still carried out in a limited parameter range. In order to explore the physically relevant regime of intensities and laser frequencies, further approximations are necessary. Some results have been obtained from a simplified two-electron interaction [8]. In contrast, we use for our time-dependent calculations a fully-correlated atom [9] which is restricted in a different way, i.e., to a single space dimension. Despite this, it has previously provided valuable insight in strong field problems [10]. More recently, the time-dependent degree of electron correlation [11] has been studied using an efficient implementation [12] of this model. This approach to the nonsequential double ionization problem provides a complementary understanding of the physical mechanism of correlation and its impact.

In this Brief Report we exploit the flexibility of our model atom in a different way. We identify a ''correlation charge'' as the effective coupling constant for correlation physics, and present results for the single and double ionization as a function of intensity for different electron correlation charges. Our goal is to provide a series of simulations showing explicitly that depending on this coupling constant the double-ionization yield systematically exceeds the expected sequential prediction more or less strongly.

In the one-dimensional model atom, both electrons are allowed to move along the laser polarization x axis with respect to the fixed nucleus at the origin. In atomic units the two-electron field-free Hamiltonian reads

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

Both the electron-nucleus attraction and the electron-electron repulsion are described by the soft-core Coulomb potential $V(x)=-1/\sqrt{x^2+1}$. The parameter C is the ''correlation charge'' and is going to be varied in a large neighborhood of $C=1$ in this investigation. Since $C=1$ is the physical value, we will refer to our model system as ''helium.''

The time-dependent Schrödinger equation for the two-electron system is solved using a split-operator algorithm in a double-zone space in extension of grid-basis methods [13]. The time propagation of the inner part whose size is typically ± 250 a.u. is calculated exactly on a full numerical grid with spacing of 0.4 a.u., and the time propagation of the outer part uses a decomposition on canonical basis states. These one-dimensional wave functions can be integrated on a grid which is an order of magnitude larger that the standard two-dimensional one. In many ionization events the electron probability remains close to the nucleus at the initial stage of the ionization process, and we expect the missing contribution from electron-electron scattering in the outer region to be negligible.

The one and double ionization are determined at the end of the pulse by calculating wave function probability as a function of position. This is indicated in Fig. 1. The actual box size has only an overall quantitative influence on the resulting ionization yields. This efficient one-dimensional implementation [12] of the two-zone two-electron wave function integration method has been able to reproduce the

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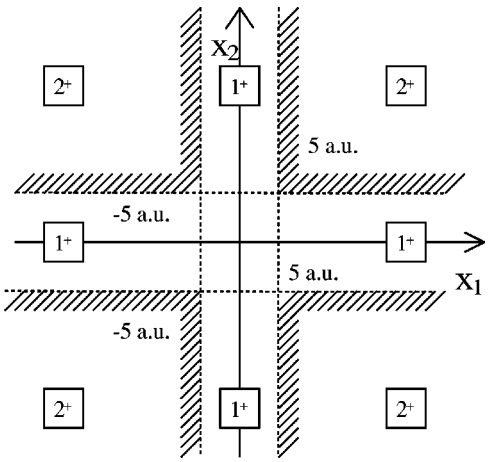

FIG. 1. Population within a box of ± 5 a.u. around the nucleus is counted as bound, population within 5 a.u. in one direction only is counted as single ionization, and population outside the box in both directions corresponds to double ionization.

main experimental features [1] of double ionization of helium in the short-pulse regime.

The key structural features of the model are well known for the case of repulsion charge $C=1$ [14]. The symmetric (electron-exchange-invariant) ground state energy is -2.2304 a.u. and the ionization threshold to the He^+ ground state at -1.4836 a.u. is 0.7468 a.u. A typical laser angular frequency for simulations is $\omega=0.1837$ a.u. However, the structure of the one-dimensional model atom depends on the adjustable correlation charge C. In order to obtain a systematic series of simulations the laser angular frequency needs to be changed for maintaining the five-photon ionization condition.

TABLE I. Energy values for helium ground states and laser angular frequencies ω for different correlation charges C.

Correl. charge	He ground state	ω laser
$C=0.1$	-2.8914 a.u.	0.3450 a.u.
$C=0.5$	-2.5944 a.u.	0.2761 a.u.
$C=0.7$	-2.4497 a.u.	0.2373 a.u.
$C=0.9$	-2.3080 a.u.	0.2025 a.u.
$C=1.0$	-2.2304 a.u.	0.1837 a.u.
$C=1.1$	-2.1697 a.u.	0.1677 a.u.
$C=1.3$	-2.0356 a.u.	0.1356 a.u.
$C=1.5$	-1.9060 a.u.	0.1038 a.u.

Now we are going to present the results of our simulations of the ionization yield from the model atom exposed to strong six-cycle pulses. The pulse is ramped linearly in two cycles to the maximal field strength value and switched off again linearly in the two final cycles. The yield for single- and double-ionization is determined at the end of the pulse. A typical run for one particular intensity takes about six to eight hours on a Pentium II, Pentium III, or Macintosh G3 personal computer.

For different correlation charges C in Table I the computed values for the helium ground states and laser angular frequencies ω chosen are shown. Eight values in the range from 0.1 to 1.5 were chosen in order to cover a broad variety of different interaction regimes. The single- and double-ionization yields as a function of laser intensity are shown in Figs. 2 and 3.

The series of graphs in Fig. 2 shows the effects of systematically reduced correlation charge: $C=0.9$ through $C=0.1$. The already familiar picture of the ionization yield for

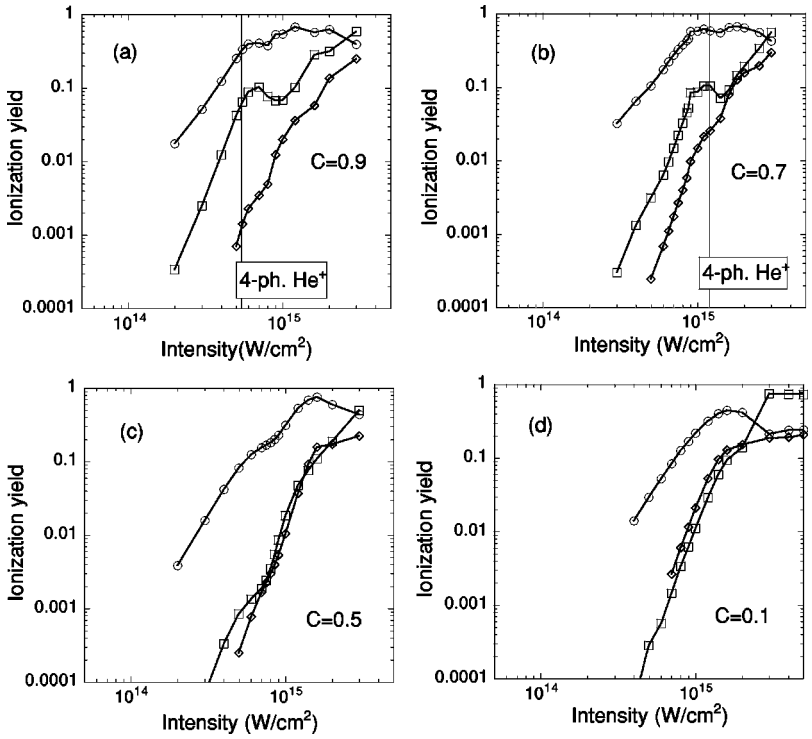

FIG. 2. Ionization yield as a function of peak laser intensity for reduced correlation charge $C<1$. The dotted curves correspond to the single-ionization signal, the boxed curves denote the double-ionization signal, and the diamond curves are the expected sequential double-ionization signal.

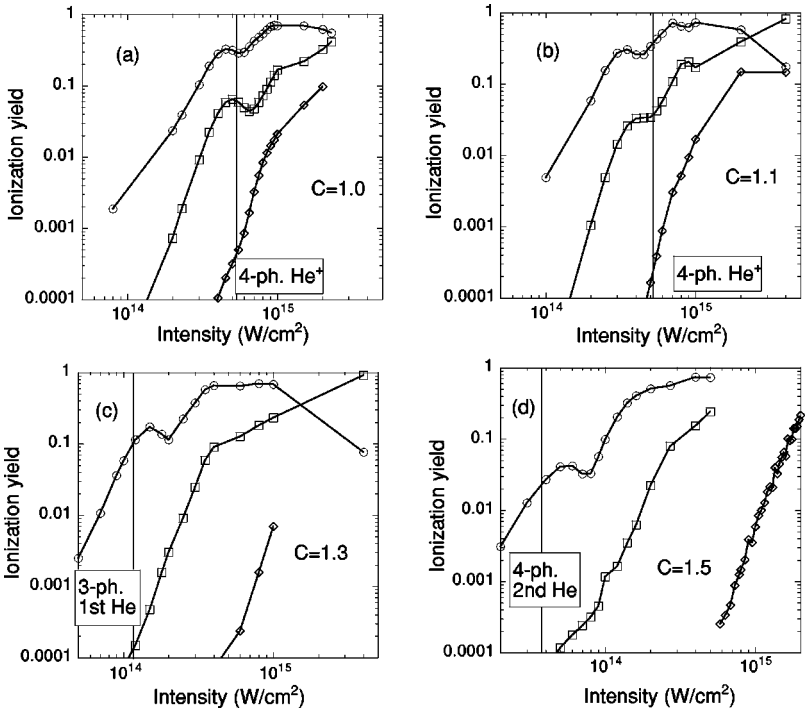

FIG. 3. Ionization yield as a function of peak laser intensity for increased correlation charge $C>1$. The dotted curves correspond to the single-ionization signal, the boxed curves denote the double-ionization signal, and the diamond curves are the expected sequential double-ionization signal.

correlation charge $C=1$, together with examples of the cases of increasing correlation charge, is shown in Fig. 3: $C=1$ through $C=1.5$.

We begin our discussion with a comparison of general qualitative features of the data. The single-ionization signal (circles) is in general one or two orders of magnitude larger than the double-ionization one (boxes). The third curve (diamonds) denotes the simple expectation of double ionization in a sequential process. The ''knee structure'' meaning the excess yield due to nonsequential ionization is located near the saturation intensity of the single-ionization signal. Along with the decrease in laser frequency with increasing correlation charge the onset of the ionization signal shifts to lower intensities.

Figure 2 shows that with decreasing correlation charge the difference between expected and actual double-ionization yield gradually decreases. While in Fig. 2(a) for $C=0.9$ the difference is well pronouced and in part (b) for $C=0.7$ is still well visible, the difference is only marginal in the case of $C=0.5$ shown in part (c). Finally, in the case of $C=0.1$ in part (d) there seems to be virtually no difference any more.

In the simulations with increased correlation charge shown in Fig. 3 the tendency of increased difference between expected and actual double-ionization yield is again clear: Starting from the case $C=1$ in part (a), the difference increases from $C=1.1$ to $C=1.5$ in part (d).

In addition to these features, in several pictures local maxima are visible in both the single- and the double-ionization yield. Though we do not want to give here precise and complete spectroscopic data on the model atom, a simple inspection of low-lying states and of their corresponding Stark shifts due to the electric field E_0 reveals the essential behavior of the system and gives a fair estimate of the intensities at which resonances are expected to occur.

The Stark shifts of the ground states can be estimated by calculating the static polarizability of the helium atom and the He^+ ion ($\alpha_{He}^+=0.92$ a.u.) for various C. The Stark shifts of the helium excited states are approximated by the ponderomotive energy $E_0^2/4\omega^2$ in the strong laser field. The intensity which will bring a Stark-shifted ground state E_g into n-photon resonance with a shifted higher lying state with energy E_e can then be determined using the equations

$$E_g-\frac{\alpha_{He}}{2}E_0^2+n\omega=E_e+\frac{E_0^2}{4\omega^2},$$

$$E_g-\frac{\alpha_{He}}{2}E_0^2+n\omega=E_e-\frac{\alpha_{He}^+}{2}E_0^2. \qquad (2)$$

For more accurate results, we should of course use the frequency-dependent dynamic polarizability instead. We expect the dynamic one to rise slightly with increasing frequency. However, the frequencies being considered here are far from any single-photon resonances and the deviation from the static case will not be very significant. Strictly speaking, since ionization is time dependent in the applied pulse, it is clear that the use of the full ponderomotive po-

TABLE II. Estimated Stark resonances at intensity I which lead to additional structure in the ionization yield for the model atom.

Correlation	α_{He}	photons	resonant state	I [W/cm^2]
$C=0.7$	2.37 a.u.	4	-1.6861 a.u.	1.2×10^{15}
$C=0.9$	2.63 a.u.	4	-1.6108 a.u.	5.3×10^{14}
$C=1.0$	2.80 a.u.	4	-1.6104 a.u.	5.3×10^{14}
$C=1.1$	3.01 a.u.	4	-1.4836 a.u.	5.1×10^{14}
$C=1.3$	3.60 a.u.	3	-1.5810 a.u.	1.1×10^{14}
$C=1.5$	4.52 a.u.	4	-1.4630 a.u.	3.9×10^{13}

tential is only a fair description for pulses with fast turn-on.This procedure should slightly underestimate the intensity actually necessary for achieving resonance.

Estimations of resonances are compiled in Table II and are in fair agreement with the local maxima occurring in the simulated ionization yields. It turns out that for the four cases $C=0.9$ [Fig. 2(a)], $C=0.7$ [Fig. 2(b)], $C=1.0$ [Fig. 3(a)], and $C=1.5$ [Fig. 3(d)] the four-photon transition between the helium ground state and the second excited state become resonant due to ac-Stark shifts. For the resonance in the case $C=1.1$ [Fig. 3(b)] the four-photon transition between the helium ground state and the He^+ ground state is responsible, and for the case $C=1.3$ [Fig. 3(c)] it is the three-photon transition between the helium ground state and the first excited state.

Our series of calculations shows the flexibility of the one-dimensional two-electron model atom for a huge variety of different simulations of parameter variations not easily accessible in experiments. On the one hand, in the case of reduced correlation $C<1$ we showed that the nonsequential double ionization knee disappears gradually with decreasing C as expected. On the other hand, in the case of increased correlation $C>1$ the excess of the double ionization becomes gradually more pronounced with increasing C. We have explicitly proven the dependence of the excess nonsequential ionization on the strength of the electron correlation.

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