

Correlation Effects in Two-Electron Model Atoms in Intense Laser Fields

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We report an efficient implementation of a spatial two-zone method for solving Schrödinger's equation numerically, using a canonical basis set decomposition, and also its application to two-electron wave functions in a study of double multiphoton ionization. We make the first calculation of the time-dependent degree of electron correlation for this poorly understood process. Our results show a particularly sensitive window of intensities coinciding exactly with the well-known "knee" regime in ion-count data.

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To date, almost all of the theoretical understanding of multiphoton processes of atoms in intense laser fields has been based on the single active electron picture. Even two-electron ionization can be so interpreted if the electrons are ejected sequentially via the Ammosov-Delone-Krainov (ADK) mechanism [1,2]. However, the excessive double ionization observed in helium experiments by Fittinghoff *et al.* [3], Walker *et al.* [4], and Sheehy *et al.* [5] is universally accepted as arising from correlated two-electron physics. Ion count data from these experiments show a departure from ADK theory with a characteristic "knee" structure in the ion yield curve of He^{++} under short-pulse (160 fs) irradiation in the intensity range 10^{14} – 10^{15} W/cm².

Partially *ad hoc* theoretical models based on either a simplified two-electron interaction [6–9] or many-body intense-field *S*-matrix theory [10] have reproduced features of the experimental data. Numerical techniques for integration of the two-electron Schrödinger equation are being developed but do not yet have the power to attack this problem directly [11]. The physical mechanism and the accompanying dynamics of non-ADK two-electron ionization remain unclear and are under active debate [5]. In particular, a successful semiclassical view of the two-electron process is proving more difficult to develop than anticipated, following the great success of the semiclassical recollision model [12] for high harmonic generation that takes place in essentially the same regime of laser intensity.

We believe that it will prove valuable to have a better command of the role of electron-electron interaction and correlation in the two-electron ionization process than is presently available. Fundamentally, one knows that all nontrivial effects of correlation are connected to the nonfactorable *e-e* coupling term in the two-electron Hamiltonian, but little more than this is known. It is not

known, for example, whether this term plays a persistent role during the ionization process, or if there is an intensity threshold where its effect "turns on" or "turns off," or even if there is a particularly sensitive window of intensities where the effect of correlation is most active. It is also not known how the effect of electron correlation is synchronized, if at all, with the time development of the ionizing field.

Electron correlation, *per se*, has not been isolated for study. However, as Grobe *et al.* have shown [13], there are several more or less equally sensible and closely compatible definitions of the degree of electron correlation that are, in principle, open to calculation. The difficulty in taking this route has been obtaining wave functions that are accurate enough over a sufficiently large spatial domain to permit the calculations to be made.

In this Letter we show that an expansion of the two-electron wave function into a sum of Slater determinants of one-electron orthonormal orbitals, as explained below, solves both of these problems. It provides a way to obtain two-electron wave functions efficiently over a much larger spatial domain than has been customary, and it provides the time-dependent degree of correlation during the ionization process. The penalty for this advance is that it can be implemented at the present time only within a one-dimensional (1D) model of a two-electron atom. The 1D model [14] is well known to provide a reliable first-cut view of one-electron ionization, and it has begun to be employed in the two-electron context [7–9,15] as well.

We will begin by confirming that our new implementation of this model is consistent with the most important characteristics of two-electron ionization data. Then we will report the first quantitative results on the time-dependent behavior of the degree of two-electron correlation, and relate them to experimentally observed features.

In the 1D two-electron model atom, each electron is allowed to move only along the x axis in relation to a fixed nucleus. In atomic units (a.u.) the two-electron field-free Hamiltonian is

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

where both the electron-nucleus attractions and the electron-electron repulsion are described by the soft-core Coulomb potential $V(x) = -1/\sqrt{x^2 + 1}$, and we will call C the “repulsion charge.” In the present note, $C = 1$ (helium analog) unless noted otherwise, and we will use the symbol He to refer to the model for convenience. The key structural facts about the model are as follows: the symmetric (electron exchange-invariant) ground state energy of the atom is -2.238 a.u., calculated by the method of imaginary-time propagation of the field-free Hamiltonian, and the ionization threshold is 0.754 a.u. (see also [16]).

Two-electron wave function calculations are time consuming because the two-electron probability distribution is needed for all space in order to account correctly for single and double ionization. The absence of an absorbing boundary creates the need for a very large computational box. With such difficulties, previous studies [7–9,15] could only treat the atom under very short pulses and with high frequency fields. To overcome this difficulty, and reach low enough frequencies to speak realistically of “multiphoton” ionization (in our case, both 5- and 13-photon ionization), we partition the wave function spatially into inner and outer parts [17,18]. The electron-electron interaction is fully accounted for in the inner part, but neglected in the outer part, where the electrons are typically far apart. The time propagation of the inner part is calculated exactly on a full numerical grid, and the time propagation of the outer part is accomplished using canonical basis states

$$\Psi(x_1, x_2, t) = \sum_a c_a(t) \psi_a(x_1, t) \psi_a(x_2, t). \quad (2)$$

This approach reduces the two-dimensional (2D) outer wave function (which extends to extremely large 2D space) into combinations of the 1D wave functions $\psi_a(x, t)$, which can be integrated on a very large grid. The decomposition is repeatedly updated during the integration process by the part of the wave function which reaches the inner region boundary and enters the outer zone. The transition from the inner to the outer zone is achieved by a smooth cutoff function [18].

The laser field is in velocity gauge and the photons have energy 0.0584 a.u. and 0.1837 a.u. We have used pulse durations of 6 and 16 optical cycles. The laser field is turned on and off with a linear profile over two cycles. The program was performed on the Cray C90 and J90, and Sun and PC workstations. The double-ionization

probability was defined as the total probability that both electrons are at least 5 a.u. away from the origin. This information is obtained directly from the fully correlated two-electron wave function as a two-dimensional integral of the joint probability density in the regime $|x_1|, |x_2| > 5$. The single-ionization probability is the probability that only one electron is within 5 a.u. of the origin.

As background, we show ionization signals from a 16-cycle laser pulse (wavelength $\lambda = 780$ nm) and a 6-cycle laser pulse ($\lambda = 248$ nm) in Fig. 1. The ionization signals from a one-dimensional He^+ ion, which represent the sequential ionization case in the condition that one electron is ionized very early, are also shown. Note the knee structure in the double-ionization signal at an intensity that is in the range familiar from double-ionization experiments [3–5]. Although the reduced dimensionality of our model does not allow for a quantitative comparison with experiment, the main experimental features are semiquantitatively clearly reproduced. For example, the double-ionization yield is correctly much 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} W/cm². In addition, the knee in the curve is located around the saturation intensity of single ionization. Figure 1 demonstrates good agreement with Lappas and Leeuwen’s results (with a 6-cycle laser pulse) [9] and shows that the knee structure appears very early in the ionization process.

Now let us turn to the main question of correlation. A degree of correlation K has been defined [13] by considering the two-electron wave function expanded as

$$\Psi(x_1, x_2) = \sum_a D_a \phi_a(x_1, x_2), \quad (3)$$

where ϕ_a represents a Slater determinant of single-electron orthonormal orbitals. The normalization of Ψ requires $\sum_a |D_a|^2 = 1$, and K is then defined via the average of the probabilities $|D_a|^2$:

$$K^{-1} = \sum_a |D_a|^4. \quad (4)$$

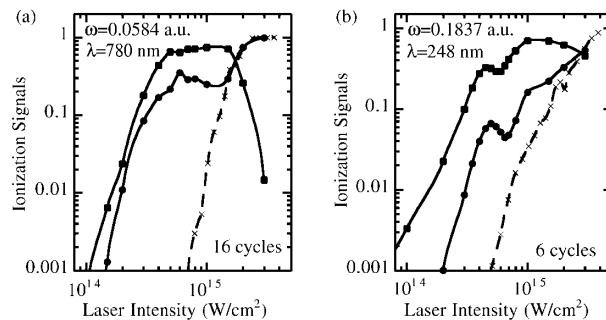


FIG. 1. Single- (squares) and double- (circles) ionization signals as functions of laser intensity. Laser wavelengths are 780 nm (a) and 248 nm (b), and the pulse duration is 16 and 6 optical cycles, respectively. The single-ionization signals (crosses) from the He^+ ion are also shown.

In practice, we can decompose $\Psi(x_1, x_2, t)$ into single-electron orthonormal orbitals via the single-particle density operator

$$\rho(x, x', t) = \int \Psi(x, x_2, t) \Psi^*(x', x_2, t) dx_2.$$

The operator $\rho(x, x', t)$ is Hermitian and its eigenvalues are the desired coefficients $|D_a|^2$.

The degree of correlation is not a static measure, and Fig. 2 shows that K is closely synchronized not only with the turn-on of the field intensity but even with the oscillations of its carrier wave. This is very classical-like electron behavior and suggests that a semiclassical view may ultimately be feasible. The short periods in which K rapidly increases occur during the laser field cycle when the field strength is reaching its peak, showing that the atom releases most of the ionized probability then. The plot also contains intensity information—we note that K is practically unresponsive to the field for $I = 3 \times 10^{14}$ W/cm², but reacts strongly at $I = 5 \times$ and 8×10^{14} W/cm². These latter intensities correlate with the onset of the knee region in Fig. 1.

It is nearly universally accepted that nontrivial electron correlation is responsible for the experimental features identified with non-ADK multiphoton double ionization (and reproduced in theoretical work as well), but this has not yet been tested. Here we use the flexibility of the numerical model to make a quantitative test. We have made small arbitrary changes in the value of the repulsion charge C in Eq. (1). Naturally this cannot be done experimentally, since fundamentally $C \equiv 1$ in helium, but it is a simple way to test the role of e - e repulsion numerically in regions of interest. Figure 3 shows several items of interest. First, K is sensitive to small modifications of the repulsion charge. Second, K

increases as C increases in the regime around $C = 1$. Third, it is notable how the sensitivity of K to small changes in C nearly disappears outside the intensity regime of the knee structure.

These results show that the time development of K appears to be closely related with two-electron dynamics. But, since K is not a familiar measure in photoionization studies, its role should be regarded as tentative until more experience is acquired with it. Nonetheless, it clearly already has the power to raise questions and make suggestions that may be useful. For example, the results presented here suggest another test of the effect of correlation that can only be accomplished numerically, as follows.

Not only the degree of correlation K but also the double-ionization signals themselves are sensitive to the repulsion charge, as Fig. 4 shows. The double-ionization signal increases significantly with increasing repulsion charge, but again solely in the knee regime. The double-ionization signals below and above this domain change little with small adjustments of C . Several studies have made clear that double ionization can be described by ADK sequential ionization [2,4,6] in the high-intensity regime beyond the knee structure. One can infer that the knee can be regarded as a “transition zone” of intensities. Above the transition zone (higher intensities), the double-ionization signal is mainly generated via the ADK sequential ionization channel, and below the transition zone (lower intensities), the double-ionization signal is mainly generated via the non-ADK ionization channel. We conclude that the effect of the increasing repulsion charge (i.e., the stronger electron-electron correlation) is to move the transition zone to higher intensities. This conclusion satisfactorily explains that the degree of

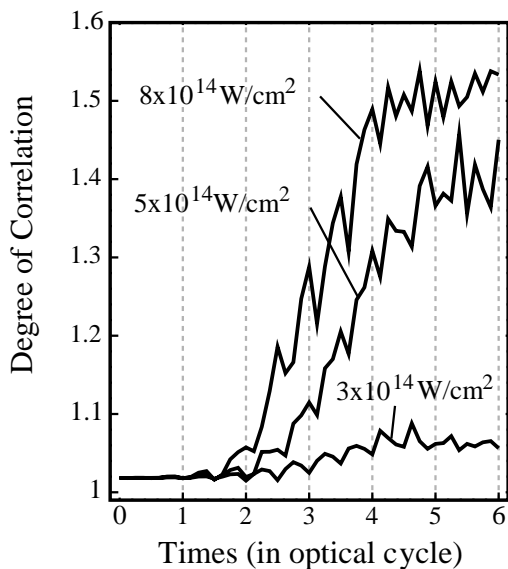


FIG. 2. Degree of correlation as a function of time in a 248 nm, 6-cycle laser pulse at different laser intensities.

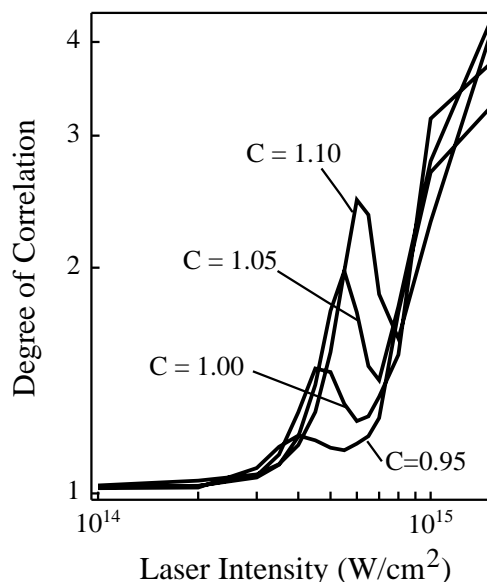


FIG. 3. Degree of correlation with different repulsion charge C , as a function of laser intensity in a 248 nm, 6-cycle laser pulse.

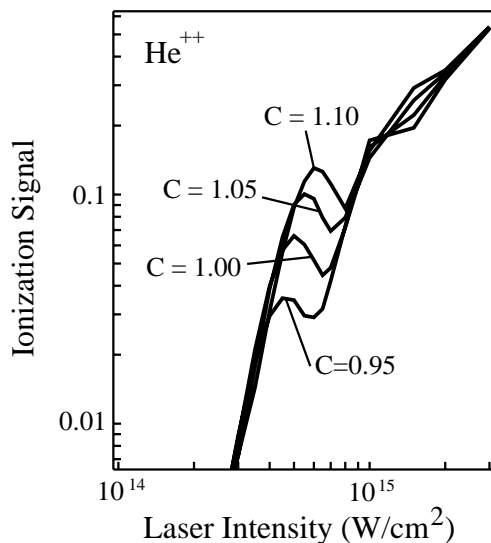


FIG. 4. Double-ionization signals with different repulsion charges C in a 248 nm, 6-cycle laser pulse.

correlation and the double-ionization signal change significantly only around the transition zone.

In summary, we have developed an efficient 1D implementation of a two-zone two-electron wave function integration method, using a canonical basis set decomposition. Using two-electron wave functions obtained in this way we have first reproduced the main experimental features of double ionization of helium in the short-pulse low frequency multiphoton regime. We then used these wave functions to calculate $K(t)$, a quantitative time-dependent measure of electron correlation during the ionization process. We demonstrated that the repulsion charge C surprisingly plays a nearly vanishing role during the ionization process, except in a particularly sensitive window of intensities. We also showed that this window coincided exactly with the well-known knee regime. Finally, while an adjustable C is only a useful theoretical trick, we want to emphasize that calculations of K are much more than that.

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