Revealing the role of electron correlation in sequential double ionization

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The experimental observations of sequential double ionization (SDI) of Ar [A. N. Pfeiffer et al., Nat. Phys. 7, 428 (2011)], such as the four-peak momentum distribution and the ionization time of the first and second electrons, are investigated and reproduced with a quantum model by including and excluding the electron-electron correlation effect. Based on the comparison of experiment and simulation, the role of the electron-electron correlation in SDI is discussed. It is shown that the inclusion of electron-electron correlation is necessary to reproduce the momentum distribution of electrons.

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I. INTRODUCTION

Tunneling ionization of atoms, molecules, and semiconductors exposed to strong laser fields is one of the most fundamental quantum processes and has been of significant interest over the past several decades [1]. Theories [2,3] have been well established for single-electron ionization based on the Keldysh frame [4]. By numerically solving the time-dependent Schrödinger equation with a single-active-electron (SAE) approximation, Tong and Lin also proposed a more accurate Delone-Krainov ionization rate [2] to the numerical simulation theory [6]. In the latter mechanism, so-called sequential double ionization (SDI), the electron-electron correlation plays an essential role and the NSDI yield is remarkably enhanced compared with the independent-electron ionization process [1]. In the former mechanism, so-called nonsequential double ionization (NSDI), two electrons are released independently step by step, therefore the SDI yield can be simulated by using ionization theory based on the SAE approximation. Nevertheless, recent experiments [7–9] have doubted the validity of independent-electron approximation in the SDI regime. Specifically, Fleischer et al. [9] reported an angular correlation between the first and second ionization steps. In Ref. [7], Pfeiffer et al. measured the release time of SDI for Ar by using the attoclock technique [10]. The measured release time is in good agreement with the prediction of Tong and Lin’s formula for the first electron, but is much earlier than the theoretical prediction for the second electron. Zhou et al. [11] have reproduced the experimental release time with a classical model by including the electron-electron correlation. Later, Wang et al. [12] argued that the release time can be successfully explained with a similar classical model by properly adopting the soft-core parameter but excluding the electron-electron correlation, which implies that electron-electron correlation is not essential in SDI.

Even though the classical model is robust, which has been demonstrated extensively in previous investigations [13–16], and also provides a straightforward interpretation with electron trajectory analysis [11,12], the physical picture of SDI is still unclear. Actually, to produce the experimental data, a scaling factor was adopted in [11,12] to shift the simulation curve. Strictly speaking, the experiment has not been reproduced. Several questions remain: Does the electron-electron correlation play an essential role in SDI? What is its influence and how can its effect be observed? Can we find another indicator to identify the electron-electron correlation, except for ionization times and angular correlation [7,9]? To clarify these questions, we investigate SDI of Ar in an elliptically polarized field by using a quantum model. We show that the electron-electron correlation does affect SDI. It leads to a small part of the electrons ionizing in a correlated way. This effect influences the observed ionization time only slightly, but it affects the overall shape of the momentum distributions.

This work is organized as follows. In Sec. II the theoretical models are presented. Section III discusses the ionization time, momentum distribution of ions, and correlated radial momentum of electrons, respectively. A brief conclusion is given in Sec. IV.

II. THEORETICAL MODEL

A complete description of a two-electron system in an elliptically polarized field requires simulating the motion for each electron in at least two dimensions, which is almost impossible at present [17]. To overcome the enormous computational challenge, a quantum model [18] was introduced by Watson et al. that has excellently reproduced the knee structure of NSDI [6,19]. In Watson model, the total wave function is written

\[ \Psi(r_1, r_2, t) = \psi_1(r_1, t)\psi_2(r_2, t) + \psi_2(r_1, t)\psi_1(r_2, t). \] (1)

In the high-intensity elliptically polarized field, the electron is quickly removed from the core and the recollision is...
The inner and outer regions are smoothly divided by a time evolutions of the first [i.e., outer electron term] and can be negligible in comparison to the Coulomb significantly suppressed. Then we can assume that the overlap LAN, ZHOU, PFEIFFER, ZHANG, LU, AND MIDORIKAWA PHYSICAL REVIEW A 89

Thus, \( V_2 \) includes a static effective potential due to the nucleus interaction with the first electron was taken into account. Where atomic units (a.u.) are adopted and \( r_n \) denotes two-dimensional coordinates \((x_n, y_n)\), \( n = 1, 2 \) refer to the first and second electrons, \( V_{\text{int}} = r_n \cdot E(t) \), and

\[
E(t) = E_0 \exp \left( -\frac{t^2}{\tau^2} / 2 \right) \frac{\epsilon}{\sqrt{\epsilon^2 + 1}} \cos (\omega t + \phi_0) \hat{x} + \frac{1}{\sqrt{\epsilon^2 + 1}} \sin (\omega t + \phi_0) \hat{y}
\]

is the driving laser field. Here, \( \epsilon, \tau, \) and \( \phi_0 \) refer to the ellipticity, duration, and carrier-envelope phase (CEP), respectively. In the Watson model, the first electron was assumed to move in a static effective potential due to the nucleus and inner electrons, i.e., \( V_1(r_1, t) = -1/\sqrt{r_1^2 + a_1} \). For the second electron, the interaction with the first electron was taken into account. Thus, \( V_2 \) includes a static effective potential due to the nucleus plus a time-dependent potential due to the electron

\[
V_2(r_2, t) = -\frac{2}{\sqrt{a_2 + r_2^2}} + \int d r_1 \frac{\alpha \psi_1^*(r_1, t) \psi_1(r_1, t)}{\beta + (r_1 - r_2)^2}.
\]

where \( a_1, a_2, \) and \( \beta \) are the soft-core parameters for the electron-ion and electron-electron interactions. The second electron is correlated with the first one through the time-dependent term of Eq. (3). We could include or exclude the electron-electron correlation by setting \( \alpha = 1 \) or 0. In the former case, we adjust the electron-electron correlation strength by changing \( \beta \) to identify its influence. Note that in the Watson model, the correlation of the first electron on the second is taken into account while its counteraction, i.e., the time-dependent potential of the second electron on the first electron, is not explicitly included. In order to remedy this seemingly unreasonable treatment, we performed other calculations where the time-dependent potential of the second electron on the first electron is also explicitly taken into account. Thus, the potential for the first electron is

\[
V_1(r_1, t) = -\frac{1}{\sqrt{a_1 + r_1^2}} + \int d r_2 \frac{\alpha \psi_2^*(r_2, t) \psi_2(r_2, t)}{\beta + (r_1 - r_2)^2}.
\]

We call this method the revised correlation model in this work.

We employ the split-operator method \([20]\) to numerically solve Eq. (2). Like in Ref. \([21]\), the two-dimensional space of each electron is partitioned into two regions: the outer region \( \{|r_n| > a\} \) and the inner region \( \{|r_n| < a\} \) with \( a = 100 \) a.u. In the inner region, the wave function is propagated exactly in the presence of combination of the Coulombic potential and the laser field. In the outer region, the wave function is propagated under the Volkov Hamiltonian analytically and the final momentum spectra of the first electron \( C_1(p_1) \) and the second \( C_2(p_2) \) are obtained from the wave function in this region \([21]\). The inner and the outer regions are smoothly divided by a splitting technique \([21]\). The ground states of the two electrons are obtained by imaginary-time method. In order to identify how electron-electron correlation affects SDI, calculations were performed by treating the time-dependent potential of electron-electron interaction in different ways as mentioned above, i.e., the revised correlation model, uncorrelated model, and Watson model.

III. RESULTS AND DISCUSSION

A. Ionization time

Figure 1 shows the simulated ionization times of the first and second electrons for the 7-fs and 33-fs pulses. The central wavelengths of the 7-fs and 33-fs pulses are 740 and 780 nm and the ellipticities are 0.78 and 0.77, respectively. For the 7-fs pulse, four CEP values, 0, \( \pi \), and \( \pm \pi/2 \), are adopted and the final simulation results are averaged over the CEP. For a comparison with experimental data, the intensity profile of the laser focus, the density distribution of the atoms in the gas jet, and the geometrical overlap of the focus and the gas jet need to be considered. Because of uncertainties of these parameters in experiment \([7]\), a three-dimensional (3D) focal volume averaging (assumption of constant gas density across the entire 3D Gaussian beam \([22]\)) and a 2D focal volume averaging (assumption of constant gas density only at the waist of the beam and zero gas density otherwise, indicating a gas jet small compared to the Rayleigh range of the laser focus) are tested here. We first analyze the result for the 3D focal volume averaging. As shown in Fig. 1(a), the simulated ionization time agrees well with the experimental data for both electrons, regardless of the electron-electron correlation being excluded.
agree better with the experimental data at low intensity. We also range. The calculations with the revised correlation model agree with the experimental data at the high-laser-light intensity range. For the second electron, the calculations with the uncorrelated model agree very well with the experimental data for the first electron. Figure 3 shows the simulated ionization times of the first and second electrons for the 7-fs and 33-fs pulses. We first analyze the result for the 3D focal volume averaging. As compared to the 3D result, the simulated electron-electron correlation indeed shifts the ionization time to an earlier time; however, the difference is less than 0.2 fs for the second electron and 0.1 fs for the first electron.

Figures 1(c) and 1(d) show the simulation results for the 33-fs pulse by assuming a 3D and a 2D focal configuration, respectively. One can see that the electron-electron correlation indeed influences the ionization time, but the influence is slight. Additionally, the absolute values of the ionization times depend also on the details of the focal geometry, which is unknown in experiment. Thus, it is difficult to judge the role of electron-electron correlation in the SDI-based ionization time.

For comparison, we also simulated the ionization time with the Watson model. Figure 3 shows the simulated ionization times of the first and second electrons for the 7-fs and 33-fs pulses. We first analyze the result for the 3D focal volume averaging. The dashed magenta line with triangles in Fig. 3(a) shows the simulated ionization times for the second electron with 2D focal volume averaging plays a less significant for the 33-fs pulse than that for the 7-fs pulse. This is because the electric field changes more slowly for the long pulse. For instance, at 3 PW/cm², the first ionization time t₁ occurs at ~4.7 fs, which is at the very beginning of the laser pulse. When the laser intensity is 1.25 PW/cm², the first ionization time is almost the same as that for the 33-fs pulse with the Watson model by assuming a 3D and a 2D focal configuration, respectively. Similar to Figs. 1(c) and 1(d), the ionization times simulated by excluding and including the electron-electron correlation shifts the second ionization step to earlier ionization times, resulting in a good agreement of experimental data and simulation for β = 2.2.

On the other hand, one can see from Figs. 1 and 3 that the focal volume averaging plays a less significant for the 33-fs pulse than that for the 7-fs pulse. This is because the electric field changes more slowly for the long pulse. For instance, at 3 PW/cm², the first ionization time t₁ occurs at ~4.7 fs, which is at the very beginning of the laser pulse. When the laser intensity is 1.25 PW/cm², the first ionization time t₁ occurs at ~3.0 fs, which is closer to the pulse peak. When the laser intensity is further reduced, the
ionization will gradually shift to the pulse peak or may occur at the descending part of the pulse when the laser intensity is lower than 0.6 PW/cm². In other words, for the 7-fs pulse, the ionization time is shifted in a very large region when the laser intensity is changed. The focal volume effect forces us to average these results and therefore it plays a significant role. In contrast, for the 33-fs pulse, because the pulse duration is longer, the ionization time for the first electron is still in the ascending part of the pulse even if the laser intensity is reduced to 0.6 PW/cm². Moreover, the electric field of the long pulse changes slowly. The ionization time changes in a smaller region as compared with that in the 7-fs pulse and thus the focal volume averaging plays a less significant role too.

The above results indicate that for both the revised correlation model and the Watson model the electron-electron correlation indeed influences the ionization time; however, the influence is slight. By accounting for the uncertainties of the focal geometry, the experiment can be explained by the uncorrelated model, the revised correlation model, or the Watson model. Thus, it is difficult to judge the role of electron-electron correlation in SDI-based ionization time. These ionization times read in the experiments and our calculations are integral signals, which have erased the details in SDI, obstructing our way to revealing the role of electron-electron correlation in SDI. Therefore, another indicator that keeps more details of the ionization process and is robust against uncertainties in the focal volume averaging is needed to identify the role of electron-electron correlation.

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**Fig. 4.** (Color online) Calculated $p_x$ and $p_y$ momentum final distributions for a 33-fs pulse at (a) 3 PW/cm² and (b) 1 PW/cm² using the revised correlation model ($\alpha = 1$, $\beta = 4$, $\alpha_1 = 0.18$, and $\alpha_2 = 1.04$). Projections are shown for $p_x$ (solid line, integration is over all $p_x$) and $p_y$ (dashed line, integration is over all $p_y$). The calculated $\text{Ar}^{2+}$ $p_x$ momentum distribution as a function of laser intensity for (c) a 33-fs pulse and (d) a 7-fs pulse is also shown for the revised correlation model ($\alpha = 1$, $\beta = 4$, $\alpha_1 = 0.18$, and $\alpha_2 = 1.04$). All results are spatially averaged over the 3D laser focus.

**Fig. 5.** (Color online) Calculated $p_x$ and $p_y$ momentum final distributions for a 33-fs pulse at (a) 3 PW/cm² and (b) 1 PW/cm² using the Watson model ($\alpha = 1$, $\beta = 4$, $\alpha_1 = 0.4$, and $\alpha_2 = 1.0$). Projections are shown for $p_x$ (solid line, integration over all $p_x$) and $p_y$ (dashed line, integration over all $p_y$). The calculated $\text{Ar}^{2+}$ $p_y$ momentum distribution as a function of laser intensity for (c) a 33-fs pulse and (d) a 7-fs pulse is also shown for the Watson model ($\alpha = 1$, $\beta = 2.2$, $\alpha_1 = 0.4$, and $\alpha_2 = 1.0$). All results are spatially averaged over the 3D laser focus.

### B. Momentum distributions of ions

Figures 4(a) and 4(b) show the two-dimensional momentum distribution of $\text{Ar}^{2+}$ for the 33-fs pulse at 3 and 1 PW/cm², respectively. Here the electron-electron correlation is taken into account by using the revised correlation model and the 3D focal volume configuration is considered. By projecting onto the major axis of the ellipse (the $y$ axis), the momentum distribution is close to Gaussian, whereas along the minor axis (the $x$ axis), it displays three peaks at low intensity and four peaks at high intensity. The outer two peaks correspond to electrons that are emitted into the parallel direction and the inner peaks correspond to antiparallel electron emission [7,23]. Figure 4(c) shows the momentum distribution along the minor axis as a function of laser intensity. As shown in this figure, the inner band gradually expands with increasing intensity and bifurcates into two branches from 1.5 PW/cm². In contrast, for the 7-fs pulse, as shown in Fig. 4(d), the inner band also expands with increasing intensity but does not bifurcate over the intensity range considered in this work. All these features agree well with the experiment [7]. Simulations are also performed by using the Watson model and the uncorrelated model. The results are shown in Figs. 5 and 6, respectively. Comparing with Fig. 4, one could see that the overall features are almost the same for the cases of excluding and including electron correlation. The only difference between these three models is the depth of the valley between the four peaks. Compared to the uncorrelated simulation, the valley between the inner two peaks is shallower in the Watson model and the valley between the inner and outer peaks is shallower in the revised correlation model.
and py for the 7-fs pulse. Figure 7(c) presents the correlation spectrum using the uncorrelated model and the revised correlation model of the first electron which are respectively obtained from the momentum spectra. This feature can be more clearly seen in the reduced spectra, electron correlation is fatter than that excluding electron-electron correlation are narrower than the experimental data. These comparisons indicate that SDI indeed is influenced by electron-electron correlation and this influence leaves an imprint on the electron momentum spectra.

The correlation spectrum of radial momentum is also simulated with the Watson model and the results are shown in Fig. 9(a). In comparison with the uncorrelated simulation, the correlation spectrum is broader and the global shape becomes a half ball when electron-electron correlation is included in the Watson model. The detailed feature can be more clearly seen from the reduced radial momentum spectrum. As shown in Fig. 9(b), the reduced spectra of both electrons are Gaussian for the case of no electron-electron correlation. The widths of the radial momentum spectra are narrower than the experimental data. Note that the Watson model explicitly includes the electron-electron correlation only for the second electron. Therefore, the radial momentum of the first electron from the Watson model is also narrower than that in the experiments. However, for the second electron, an additional peak (denoted by $M_2$) appears at low momentum beside the main peak (denoted by $M_1$). Then the width of the radial momentum spectrum becomes broader than the experimental data. These comparisons indicate that SDI indeed is influenced by electron-electron correlation and this influence leaves an imprint on the electron momentum spectra.

However, as shown in Figs. 7–9, the peaks of the momentum spectra do not change dramatically when electron-electron correlation is excluded or included. Therefore, the mean value of radial momentum is not dramatically influenced by the
respective, the reduced spectra of $3P_W$ simulated with the uncorrelated model ($\alpha_1, \beta_1 = 0.15, 0.95$). All results are 3D focal volume integrated.

The origin of the broadening of the reduced momentum spectra is related to the time intervals when the first and the second ionizations occur. Figure 10(a) shows the ionization rate as a function of time at 3 PW/cm$^2$ for the 7-fs pulse. The bold and thin solid lines correspond to the first and second electrons without electron-electron correlation. Because the laser intensity is much higher than the saturation intensity, the ionization is confined in a single cycle around $t_1$ and $t_2$ at the ascending part of the pulse, respectively. After ionization, the electrons are accelerated by the laser field and finally released with a momentum of $P_{r1}$ and $P_{r2}$, as indicated in Fig. 10(a); however, because of no correlation, the second electron is not freed when $t < t_1$ and the first ionization process is already finished when $t \geq t_2$. In contrast, when the electron-electron correlation is accounted for [Fig. 10(b)], the centers of the ionization time ($t_1$ and $t_2$) for both electrons remain virtually unchanged. This is consistent with the above results that the ionization time is not effectively influenced by the electron-electron correlation. However, as shown in Fig. 10(b), when $t < t_1$, a small percentage of the second electron starts to be released and also a small percentage of the first electron is released at $t \geq t_2$. Consequently, the ionization windows of the first and second ionization processes become broader than those excluding the electron-electron correlation. The long ionization window broadens the width of the momentum distribution. Moreover, because the first electron quickly moves away after ionization, its correlation with the second electron drops quickly. Only a small percentage of the first and second electrons can be ionized in a correlated way. By increasing the electron-electron correlation, a larger percentage of the electrons can be ionized in a correlated way and the momentum spectra will be further broadened as shown in Fig. 8. On the other hand, Fig. 10(c) shows the ionization rate simulated with the Watson model. In comparison with Fig. 10(b), the first electron is already completely released before $t_2$ but a larger percentage of the second electron is ionized before $t_1$. In other words, the ionization window in the Watson model is narrower for the first electron but broader for the second electron as compared with those in the revised correlation model. Accordingly, the radial momentum of the

FIG. 8. (Color online) (a) Calculated correlation spectrum of radial momenta $P_{r1}$ and $P_{r2}$ for the 7-fs pulse with a peak intensity of 3 PW/cm$^2$ using the Watson model ($\alpha = 1, \beta = 2.2, a_1 = 0.4$, and $a_2 = 1.0$). The bold and thin lines in (b) show, respectively, the reduced spectra of $P_{r1}$ and $P_{r2}$ of experimental data. The thin line with squares shows the reduced spectra of $P_{r2}$ simulated with the Watson model ($\alpha = 1, \beta = 2.2, a_1 = 0.4$, and $a_2 = 1.0$). The dashed bold line and dashed thin line show, respectively, the reduced spectra of $P_{r1}$ and $P_{r2}$ simulated with the uncorrelated model ($\alpha = 0, a_1 = 0.4$, and $a_2 = 1.0$). All results are 3D focal volume integrated.
the uncorrelated model (field and the radial momentum, respectively. Simulation results using dots and the dashed lines represent the vector potential of the laser show in (c).

The subtle features of momentum distribution require further experimental measurements with low noise and high accuracy.

IV. CONCLUSION

Sequential double ionization of Ar subjected to elliptically polarized pulses was investigated using quantum models. It was shown that the electron-electron correlation indeed slightly shifts the ionization time, but its influence is not crucial to explain the experiment [7]. Moreover, uncertainties in the focal volume averaging also influence the apparent ionization times. It is difficult to decisively reveal the role of electron-electron correlations based on the ionization timing data alone. However, our simulations show that the electron-electron correlation broadens the time windows for the first and second ionizations. Though this effect could not be distinguished in the ionization time of the second electron, it affects the momentum distribution. The signature of electron-electron correlation can be identified in the width of the electron momentum distribution. In comparison with the experiment [7], the width of the electron momentum distribution for the 7-fs pulse agrees well with the simulation including a fair electron-electron correlation, which indeed indicates the exist of electron-electron correlation in SDI.

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