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# Correlated electron emission in laser-induced nonsequence double ionization of helium

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In this paper, we have investigated the correlated electron emission of the nonsequence double ionization (NSDI) in an intense linearly polarized field. The theoretical model we employed is the semiclassical rescattering model, the model atom we used is the helium. We find a significant correlation between magnitude and direction of the momentum of two emission electrons, and give a good explanation for this striking phenomenon by observing the classical collisional trajectories. We argue that this correlation phenomenon is universal in NSDI process, as revealed by the recent experiment on the argon.

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The excessive double ionization observed in helium experiments [1–3] draws much attention to the multiple-electron dynamics in the laser-atom interaction. In these experiments the single ionization of He in a linearly polarized field is accurately predicted by the single active electron (SAE) approximation [2], well described by the Ammosov-Delone-Krainov tunneling theory [4]. However, the case of double ionization is more complicated. In the regime of very high intensities ($I > 10^{16}$ W/cm$^2$), where strong double ionization occurs, the double ionization keeps in good agreement with the sequential SAE models as that in the lower-intensities regime ($I < 10^{14}$ W/cm$^2$). The double ionization deviates seriously from the sequential SAE model and shows a great enhancement in a ''knee'' regime [$(0.8–3.0) \times 10^{15}$ W/cm$^2$]. This surprising large yields of the double ionization obviously indicates that the sequential ionization is no longer the dominating process in this regime and the electron-electron correlation has to be taken into account. Intense efforts to model the two-electron process of the double ionization in a laser field have reproduced the main feature of the knee structure in the double ionization yield as a function of laser peak intensity and, moreover, yielded quantitative agreement with the experiments in some cases [5,6].

The physical mechanism behind this nonsequential process is, however, still debatable. Both the ''shake-off'' model and the ''recollision'' model are suggested to describe the electron's correlation [1,3,7,8]. However, none of the two nonsequence double ionization (NSDI) mechanisms can completely explain the experimental observations. For the ''shake-off'' model, it cannot give the reason for the decrease in the double-ionization yields as the polarization of the laser field departs from linear [9–11]. In the ''recollision'' model, the returning electrons are known to have a maximum classical kinetic energy of $\sim 3.2U_p$ ($U_p = e^2F^2/4m_e\omega^2$), so one can determine a minimum intensity required for the rescattering electron to have enough energy to excite the inner electron. But the double-ionization yields observed in experiments has no such an intensity threshold. In fact, the double ionization process is rather complicated and subtle, both of the two NSDI processes and the sequential ionization have contributions to it and may dominate in the different regimes. In another aspect, Becker and Faisal proposed a ''correlated energy sharing'' model to describe the NSDI processes and nuclei recoil experiment [5,12,13]. The model is based on the so-called intense-field many-body *S*-matrix theory derived by a rearrangement of the usual *S*-matrix series and include time electron correlation and the rescattering mechanism.

We have employed the semiclassical model to study the double ionization of helium in intense linearly polarized field [6,14]. Our calculations reproduced the excessive double ionization and the photoelectron spectra observed experimentally both quantitatively and qualitively, and we argue that the classical collisional trajectories is the main source of the nonsequence double ionization of helium in the ''knee'' regime.

Recently, the observation of the correlated electron emission in laser-induced double ionization of argon [15] provided new insights into the NSDI process. These authors reported a strong correlation between the direction and the magnitude of the momenta of two electrons emitted from an argon atom: the momenta of the two emission electrons tend to have the same magnitude and sign in the polarization direction. On the theoretical side, by solving the time-dependent Schrödinger equation for two electrons in three dimensions, Taylor *et al.* [16] gave that the most of double-ionization probability flux tends to emerge to the same side of the ion. Similar conclusions have been drawn from one-dimensional (1D) model [17].

In this paper, based on the 3D semiclassical rescattering model developed recently [6,14], we investigate the dynamical behavior of the correlated electrons in the double-ionization process by analyzing their classical trajectories. This investigation, as shown later, is very helpful to understand the physical mechanism behind the momentum correlation. The model atom we use is the helium, however, we argue that our discussions are available to the other multi-electron atoms, like the argon in recent experiment [15].

First, we briefly present the semiclassical rescattering model adopted in our calculations. The ionization of the first electron from bound state to the continuous state is treated by the tunnelling ionization theory generalized by Delone and Krainov [18]. The subsequent evolution of the ionized electron and the bound electron in the combined Coulomb potential and the laser fields is described by a classical Newtonian equation. To emulate the evolution of the electron, a set of trajectories is launched with initial conditions taken into from the wave function of the tunneling electron.

The evolution of the two electrons after the first electron tunneled are described by the classical equations (in atomic unit),

$$\frac{d^2\mathbf{r}_i}{dt^2} = -\nabla(V_n^i + V_{ee}) - \mathbf{F}(t), \quad i=1,2. \tag{1}$$

Here $\mathbf{F}(t) = F\cos(\omega t)\mathbf{e}_z$ is the laser field. The indices $i=1$ and 2 refer to the tunnel ionized and bound electron, respectively. The potentials are

$$V_n^i = -\frac{2}{|\mathbf{r}_i|}, \quad V_{ee} = \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}. \tag{2}$$

The initial condition of the tunneled electron, under the SAE approximation of $He^+$, is determined by a equation including the effective potential given in Ref. [19] and a generalized tunneling formula developed by Delone and Krainov [18]. In parabolic coordinates, the Schrödinger equation for a hydrogen-like atom in a uniform field $\epsilon$ is written (in atomic unit),

$$\frac{d^2\phi}{d\eta^2} + \left(\frac{I_{p1}}{2} + \frac{1}{2\eta} + \frac{1}{4\eta^2} + \frac{1}{4}\epsilon\eta\right)\phi = 0, \tag{3}$$

in which $I_{p1} = -0.9$ a.u. is the negative ionization potential of the outer electron.

The above equation has the form of the one-dimensional Schrödinger equation with the potential $U(\eta) = -1/4\eta - 1/8\eta^2 - \epsilon\eta/8$ and the energy $K = I_{p1}/4$.

The evolution of the outer electron is traced by launching a set of trajectories with different initial parameters $t_0$ and $v_{1x0}$, where $v_{1x0}$ is the initial velocity perpendicular to the polarization of the electric field. The initial position of the electron born at time $t_0$ is given by $x_{10} = y_{10} = 0$, $z_{10} = -\eta_0/2$ from the Eq. (3). The initial velocity is set to be $v_{1y0} = v_{1z0} = 0$, $v_{1x0} = v_{10}$. Thus, the weight of each trajectory is evaluated by Ref. [18],

$$w(t_0, v_{10}) = w(0)w(1), \tag{4}$$

$$w(1) = \frac{\sqrt{2I_{p1}}v_{10}}{\epsilon\pi}\exp(-\sqrt{2I_{p1}}v_{10}^2/\epsilon), \tag{5}$$

and where $w(0)$ is the tunneling rate in the quasistatic approximation [4,20].

The initial state of the bounded electron is described by assuming that the electron is in the ground state of $He^+$ with energy $E_2 = -2.0$ a.u. and its initial distribution is microcanonical distribution [6,21].

In our calculation, the Eqs. (1) are solved in a time interval between $t_0$ and $15T$ by employing the standard Runge-Kuta algorithm. During the first ten optical cycles the electric field amplitude is constant, and then the field is switched off using a $\cos^2$ envelope during three cycles, and during the last two optical cycles the electrons is free from the electric field. The wavelength is $\lambda = 780$ nm, which is so chosen to match the experiment [2,23], and the intensity of laser is $I = 1 \times 10^{15}$ W/cm$^2$ which is right at the ''knee'' region of the double ionization of helium. $10^6$ or more initial points are randomly distributed in the parameter plane $-\pi/2 < \omega t_0 < 3\pi/2$, $v_{1x0} > 0$ for the outer electron and in the microcanonical distribution for the inner electron. The distribution for the ionization electron can be obtained by making statistics on an ensemble of classical trajectories weighed by the Eq. (4). The results have been tested for numerical convergence by increasing the number of trajectories.

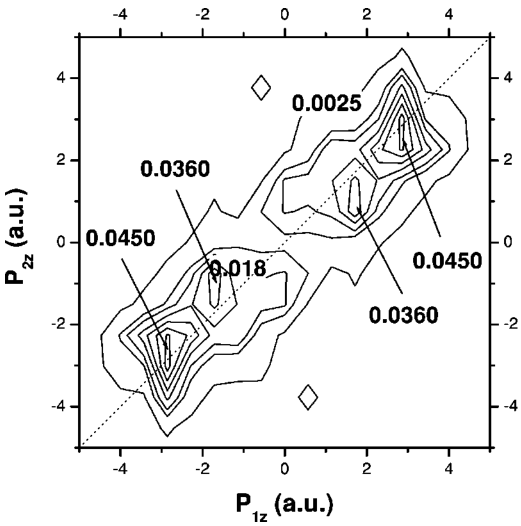


FIG. 1. Momentum correlation between the two emitted electrons given by present calculations.

Figure 1 shows the momentum correlation between the two emission electrons in the double ionization of the present calculations. The horizontal axis shows the momentum component of the first electron in the direction of polarization ($P_{1z}$) and the vertical axis the same momentum component of the second electron ($P_{2z}$). This figure shows a strong correlation between the momenta of the two electrons. There is a clear maximum for both electrons being emitted with the same momentum component in the direction of polarization axis of about 2.7 a.u., and emission to opposite half planes is strongly suppressed, i.e., both two electrons tend to fly to same side of ion in the direction of polarization. This phenomena has been observed in the ''knee'' region for argon [15]. On the other hand, from Fig. 1, we see that the maximum momentum of both electrons is about 4.5 a.u., which is consistent with the electron-ion coincidence experiment observation of helium [23], in which the maximum energy of emission electron in NSDI process is $4U_p$, since the perpendicular component of momentum is small, the maximum momentum component in the polarization direction can be approximate obtained as $P_{z\max} = \sqrt{8U_p} \simeq 4$ a.u.

An useful alternative perspective on Fig. 2 is obtained by rotating the distribution by 45°. Then we can get two new distributions. In Fig. 2(a), we show the distribution of the sum momentum $P^+ = (P_{1z} + P_{2z})$; in Fig. 2(b), we show the distribution of the difference momentum $P^- = (P_{1z} - P_{2z})$. Owing to momentum conservation, the sum momentum of emission electrons is equal and opposite to the $He^{2+}$ recoil

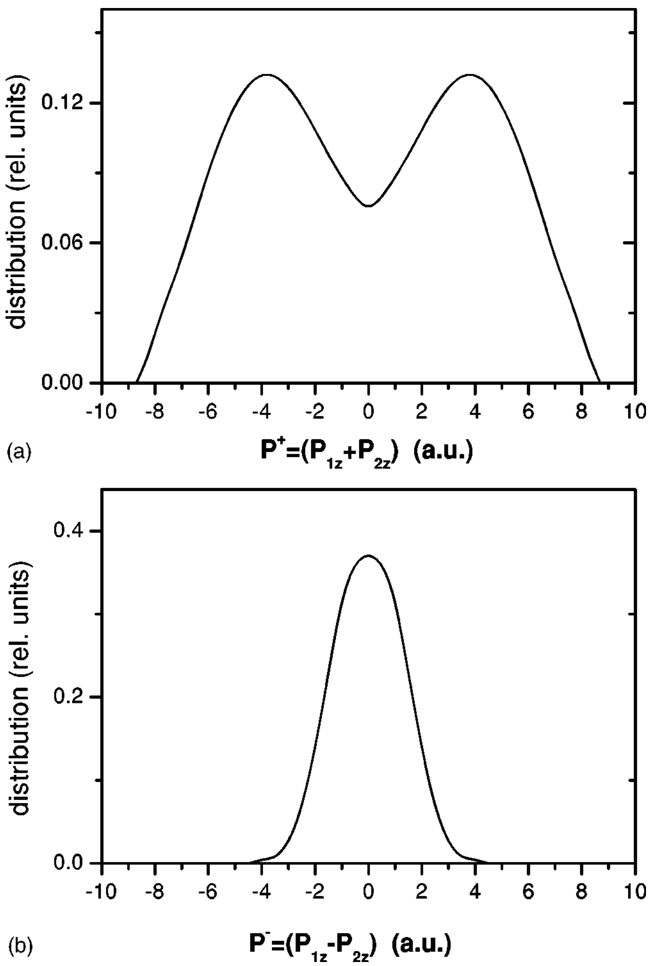


FIG. 2. The distribution of momentum parallel to the polarization axis: (a) the sum momentum of the two emission electrons. (b) the difference momentum of the two emission electrons.

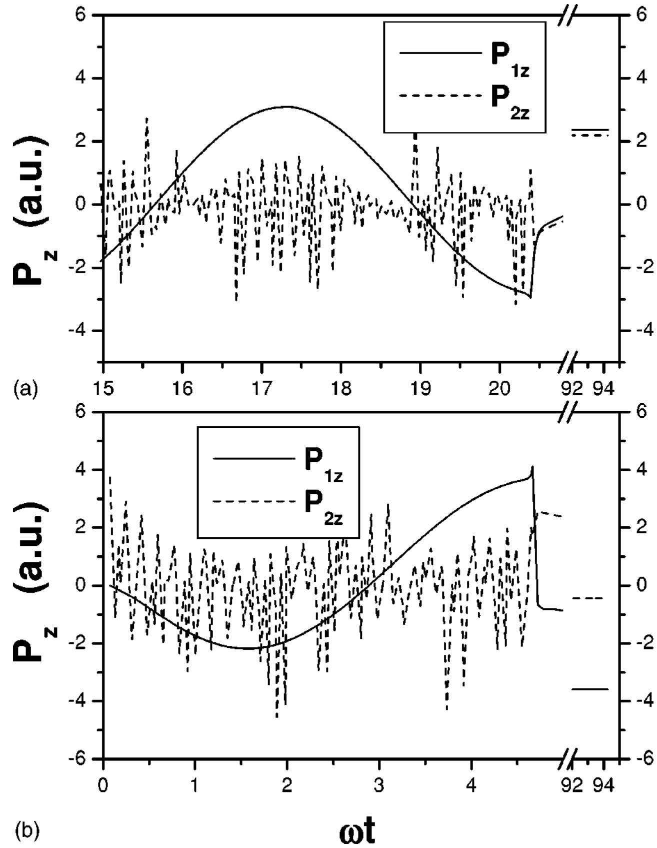


FIG. 3. Two typical trajectories of the “ recollision” mechanism: (a) the case that the difference momentum is small. (b) the case that the difference momentum is nearly the maximum.

ion momentum [14,22], so its distribution, as we know, shows a characteristic double-hump structure with a central minima. A vast amount of literature has devoted to discuss the double-hump structure of momenta of recoil ions [14,22], here we only pay attention to the correlation between the two emission electrons.

Comparing the Fig. 2(a) with Fig. 2(b), one finds that the sum momentum at the peak is about 4.3 a.u., almost above the maximum momentum of each electron, and the maximum sum momentum is almost 8.7 a.u., about twice of the maximum momentum of one electron; furthermore the distribution width of difference moment is much smaller than the one of the sum momentum. These features indicate the momenta parallel to polarization axis of the two emission electrons likely have the same direction. On the other hand, the peak of the distribution of difference momentum is at zero, so the momenta of the two emission electron tend to have the equal magnitude.

To study the origin of the correlation of electrons emission, we show two trajectories of the electrons in Fig. 3. Figure 3(a) shows a typical trajectory of which the difference momentum in polarization direction is small and Fig. 3(b) shows a typical trajectory of which the difference momentum in polarization direction is nearly the maximum. From these trajectories, it can be concluded that after the second electron ionized the momentum transfer received from the laser field is almost identical for both electrons. Because the electron can obtain very high energy from the laser, the momenta of the two, thus, be accelerated to the same direction. On the other hand, this part of the acceleration only adds to the sum of the momenta of the two electrons, but does not change the difference of the momenta. Therefore, the difference of the momenta is only determined by the ionization process of the second electron.

As we have discussed before, the second electron is mostly ionized by the “recollision” mechanism: the second electron is ionized by a collision with the tunneled electron [6]. Because the collision between the two electrons is almost instantaneous so that the energy is conserved approximately when the collision happens. The total energy of the two electrons can be expressed by

$$E=E_T+\left(\frac{P_{1z}+P_{2z}}{2}\right)^2+\left(\frac{P_{1z}-P_{2z}}{2}\right)^2, \qquad (6)$$

where $E_T$ includes the potentials and the kinetic energy of the perpendicular parts. To the best of our knowledge, so far the energy distribution of the returning electron has not been reported, but we believe that it exhibits a peak at zero energy and decreases as the energy increases, on the other hand, only the returning electron energy of which is above the ionization potential of the bound electron $I_{p2}$, can cause the second electron ionized, so the total energy of the two electrons soon after the collision occurred is small and its distribution peaks at zero. Therefore the $P^-$ is more likely zero, i.e., the momentum components in the polarization direction of the two emission electrons are likely equal to each other. The returning electrons are known to have a maximum classical kinetic energy of $\sim 3.2U_p$, therefore the maximum difference momentum must satisfy $(P_{1z}-P_{2z}/2)^2 \simeq 3.2U_p - I_{p2}$, so $|P^-|_{\max}=2\sqrt{3.2U_p-I_{p2}} \simeq 4.2$ a.u.

On the other hand, since the total energy of the two electrons soon after the collision occurring is small, the total final energy of the two emission electrons mostly reflect the energy transfer received from the field. The field acceleration make the sum momentum increase. The maximum energy for an electron in the double-ionization process is about $4U_p$ [23], from Eq. (6), assuming the two emission electron have the same energy, we can obtain the maximum sum momentum $|P^+|_{\max} \simeq 8$ a.u.. This indicates that the joint acceleration of the electrons in the laser field clearly dominates over the influence of the electron repulsion, and both electrons ionized are driven by the laser to the same side.

In conclusion, we have investigated the momentum correlation between magnitude and direction of the two emission electrons in nonsequence double ionization. The numerical results on the helium show a significant correlation on the momentum of the two electrons: the emission electrons tend to have the same momentum component in the polarization direction. The phenomena can be directly comprehended from the classical collisional trajectories. These discussions suggest that the correlated electrons emission in double-ionization process is a semiclassical process. We also evaluated the width of distribution of the sum and difference momentum. Because the difference momentum is only determined by the ionization process, so it is important to verify the dominating process in the ‘‘knee’’ regime. Based on the rescattering model, we argue that the maximum difference momentum of the two emission electrons is $|P^-|_{\max}=2\sqrt{3.2U_p-I_{p2}}$. The predictions coincide with the argon experiment [15]. We hope our discussions will stimulate the experimental works on the helium in the direction.

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