Strong-Field Double Ionization through Sequential Release from Double Excitation with Subsequent Coulomb Scattering

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We perform a triple coincidence study on differential momentum distributions of strong-field double ionization of Ar atoms in linearly polarized fields (795 nm, 45 fs, 7×10^{13} W/cm^2). Using a three-dimensional two-electron atomic-ensemble semiclassical model including the tunneling effect for both electrons, we retrieve differential momentum distributions and achieve a good agreement with the measurement. Ionization dynamics of the correlated electrons for the side-by-side and back-to-back emission is analyzed separately. According to the semiclassical model, we find that the doubly excited states are largely populated after the laser-assisted recollision and large amounts of double ionization dominantly takes place through sequential ionization of doubly excited states at such a low laser intensity. Compared with the Coulomb-free and Coulomb-corrected sequential tunneling models, we verify that electrons can obtain an energy as large as $\sim 6.5U_p$ through Coulomb scattering in the combined laser and doubly charged ionic fields.

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Completely understanding the dynamics of a three-body Coulomb system, i.e., nonsequential double ionization (NSDI), is a challenging task and represents one of the incompletely solved problems of theoretical and experimental physics, holding a variety of puzzles on both classical and quantum levels [1,2]. It is of paradigmatic importance since it reveals complicated and fundamental electron dynamics in atomic and molecular physics. Especially, the liberation of two or more electrons from an atom by strong laser fields is dominated by the electron correlation. Extensive studies of atoms and molecules in strong infrared light fields have revealed that electron correlation dominates the enhancement of double ionization yields at moderate laser intensity [2]. Much effort has been concentrated on exploring the physical mechanism of strong-field NSDI. Now, it has been widely accepted that the electron recollision-induced-direct-ionization mechanism [3,4] plays a key role for NSDI. The electrons were released nonsequentially into the same direction in the laser polarization plane (side-by-side emission) [5–9], and thus the momentum distribution of doubly charged ions exhibits the prominent "double hump" structure [10,11]. The pronounced "*V*" shaped structure was further observed in correlated electron momentum spectra of double ionization He atoms [8,9]. The findings imply that the dynamics of NSDI is more complicated than expected by the simple recollision-induced-direct-ionization picture. Moreover, the dominant back-to-back emission (so-called anticorrelation) for Ar atoms was found in the multiphoton double ionization regime [12]. Electron anticorrelation was basically explained within the mechanisms of multiple-recollision and recollision-induced-excitation tunneling [13].

Although the basic correlation dynamics for NSDI has been observed, the fundamental questions about the correlated emission of strong field double ionization are still hanging (see a recent review [14]). For example, the electron releasing time of sequential ionization in circularly polarized laser pulses was recently investigated [15]. The electron correlation is usually believed less important in the sequential double ionization process. Indeed, the question whether the electron correlation exists in strong-field double ionization by circularly polarized laser fields is still puzzling [16]. Nevertheless, the correlation dynamics in linearly polarized laser fields is more complicated, which demands the full differential measurement.

Previous measurement on electron correlation momentum spectra are usually based on double coincidence measurements (one electron and the parent doubly charged ion) [7–10,17–19]. In this case, the false coincidence events could be included because a large amount of electrons may come from other parent ions or the background; e.g., it could reach as much as ~25% of the total double ionization events [17]. In this Letter, we perform a triple coincidence study with the experimental measurement and model calculation on NSDI of Ar atoms in a linearly polarized field at intensity of 7×10^{13} W/cm^2 (795 nm, 45 fs), where

both the side-by-side and back-to-back emissions reveal important effects. Experimentally, the ionized two electrons are measured in coincidence and are recognized by their final drift momenta for each type of electron correlation; i.e., the side-by-side and back-to-back emission. As known, full-dimensional time-dependent quantum calculation on the three-body problem in infrared laser fields is extremely difficult. Successful classical and semiclassical models for strong-field double ionization [20–23] motivate this work. We use a dedicated 3D semiclassical two-electron atomic ensemble model including the tunneling effect for both electrons and retrieve the comprehensive collision and ionization dynamics for both types of electron correlation. We find that large amounts of doubly excited states are formed with laser-assisted hard recollision and sequential ionization from the doubly excited states is universal for strong-field double ionization at low laser intensity. Comparing with the Coulomb-free and Coulomb-corrected sequential tunneling models, we verify that the Coulomb scattering mediated by the laser fields in the doubly charged ions is responsible for the high energy photoelectrons of double ionization.

The experimental setup included a high power femtosecond laser oscillator operating with a repetition of 6 MHz [24] and a dedicated reaction microscopy (REMI) ([25]). In REMI, the electrons and doubly charged ions generated from atoms of a supersonic jet in the tightly focused laser field were guided towards two position-sensitive delay-line equipped multi-channel plate detectors by applying weak homogenous electric (2 V/cm) and magnetic (4.5 Gauss) fields along the laser polarization direction. The laser polarization is in parallel to the time-of-flight direction (longitudinal) of the spectrometer. All of charged particles (two electrons and the parent doubly charged ions) were coincidently measured for a double ionization event. As seen in Fig. 1(a) (the triple coincidence conserved momentum spectrum), the space charge effect and false coincidence events can be completely avoided in the measurement. In order to achieve enough triple coincidence counts, it took several weeks to collect the experimental data.

We concentrate on the differential information for each type of electron correlation. Since there is no "color" between electrons, the electron detector cannot distinguish the outer electron (tunneled electron) or the inner electron (struck electron) from strong-field double ionization. But, the detector can tell which is the "fast" electron and which is the "slow" electron in a correlated electron pair. Thus, we can determine the final drift momentum as the color. We define the larger longitudinal momentum electron as the fast electron and the smaller as the slow electron for the side-by-side emission. For the back-to-back emission, we define the electron moving with the positive momentum as the fast electron and the electron moving with the negative momentum as the slow electron. For the side-by-side emission, both electrons could be released into "left"

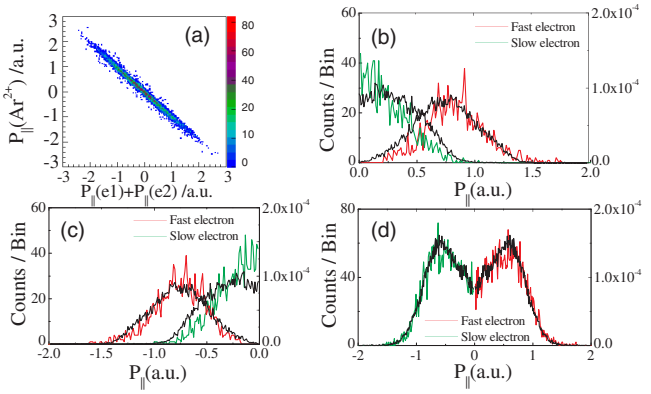

FIG. 1 (color online). (a) The momentum conservation spectrum measured by the triple coincidence. The experimental longitudinal momentum distributions of the fast electron and slow electron of the side-by-side emission for positive momenta (b) and for negative momenta (c), and of the back-to-back emission (d). The black curves show the corresponding momentum distribution by the semiclassical simulation, with the scale of the y axis.

(negative momentum) or "right" (positive momentum) along the field polarization. Those two cases are symmetrical for a long laser pulse. The integrated experimental momentum distributions of the fast electron or slow electron for the side-by-side and back-to-back emission are shown in Figs. 1(b),1(c), and 1(d).

Several interesting features can be quantitatively observed from the triple coincidence analysis. For the side-by-side emission, the integrated momentum distributions of the fast electron and slow electron show pronounced Gaussian-like and half-Gaussian-like distribution, respectively. The fast electrons can obtain longitudinal momentum as high as $p^{\mathrm{fast}}_{z,\mathrm{max}} = 1.4$ a.u. [$E \sim 6.5Up$, $U_p = \varepsilon^2/4\omega^2$, the ponderomotive potential, ε is field amplitude, ω is the field frequency, and atomic units (a.u.) are used unless otherwise specified]. While for the slow electrons, the momentum distribution approximately decreases from the origin to $p^{\mathrm{slow}}_{z,\mathrm{max}} = 0.75$ a.u. ($E \sim 2\ Up$), which behave much like that of direct ionization in the single ionization process. For the back-to-back emission, the distribution shape shows the edge-cut Gaussian distribution with the maximum yield at ~ 0.75 a.u. and the cutoff longitudinal momentum $p^{\mathrm{fast}}_{z,\mathrm{max}} = 1.4$ a.u.

Following the distinguishable spirit, we perform a 3D semiclassical two-electron atomic ensemble model calculation. Briefly, in the model, one electron is released at the outer edge of the field-suppressed Coulomb barrier through tunneling with a rate given by the ADK theory [26]. The bound electron is sampled from a microcanonical distribution. The subsequent evolution of the two electrons with the above initial conditions is governed by Newton's equations of motion: $(d^2r_i/dt^2) = -\varepsilon(t) - \nabla_{r_i}(V^i_{ne} + V_{ee})$. Here, index i denotes the two electrons. $V^i_{ne} = -(2/|r_i|)$ and $V_{ee} = (1/|r_1 - r_2|)$ are Coulomb interactions between

the nucleus and electrons and between two electrons, respectively. The laser field $\varepsilon(t)$ with a cosine waveform has constant amplitude for the first 10 cycles and is turned off with a 3-cycle ramp. We consider the recollision-induced excitation tunneling effect in the model, which is done by allowing the inner electron to tunnel through the potential barrier whenever it reaches the outer turning point, where $P_{i,z} = 0$ and $z_i\varepsilon(t) < 0$, with a tunneling probability P_i^{tul} given by the Wentzel-Kramers-Brillouin (WKB) approximation $P_i^{\mathrm{tul}} = \exp[-2\sqrt{2}\int_{z_i^{\mathrm{in}}}^{z_i^{\mathrm{out}}} \sqrt{V(z_i) - V(z_z^{\mathrm{in}})}dz_i]$, where z_i^{in} and z_i^{out} are the two roots ($|z_i^{\mathrm{in}}| < |z_i^{\mathrm{out}}|$) of the equation for the z_i, $V(z_i) = -2/r_i + z_i\varepsilon(t) = -2/z_i^{\mathrm{in}} + z_i^{\mathrm{in}}\varepsilon(t)$. In the calculation, the first and second ionization potentials of the two-electron atom are chosen as $I_{p1} = -0.58$ and $I_{p2} = -1.02$ a.u., respectively, to match the argon atom. The calculated integrated momentum distributions of the fast electron and slow electron in the side-by-side and back-to-back emission are shown in Fig. 1 (black curves). The model calculation agrees perfectly with the experimental measurement.

Using our model, we first coincidently trace back the ionization time of the fast electron and slow electron for each type of electron correlation with respect to the field phase to investigate the temporal correlation. The ionization time is defined as the instant when the electron energy $E_e = -2/r + (p_x^2 + p_y^2 + p_z^2)/2$ turns from negative to positive and no longer returns to be negative. Note that the ionization time is different with the tunneling time. The zero time starts at the first maximum for the laser field. Here, for the side-by-side emission we only study the case when both electrons move with the positive momentum. As shown in Figs. 2(a) and 2(b), the temporal correlation spectra of two released electrons show the complicated ionization signature. Besides the instantaneous escape, strong-field double ionization does mainly occur at the regular time delay for several laser periods. From the simulation, one can observe that the "simultaneous" ionization (so-called nonsequential) events, i.e., $t_{\mathrm{ionization}}^{\mathrm{slow}} \approx t_{\mathrm{ionization}}^{\mathrm{fast}}$ [events indicated as the main diagonal lines in Figs. 2(a) and 2(b)], just contributes to a very small fraction (~10%) of the total double ionization.

For the side-by-side emission, the fast electrons usually leave the nucleus earlier than the slow electron, as seen in Fig. 2c. The slow electrons could move around the nucleus for several laser periods after the fast electrons leave the nucleus. The emission time of the fast electron appears mainly at the instant of t at $\sim 1.4, 2.4, 3.4, \ldots$, optical cycles (o.c.). The slow electrons prefer to be released each half-laser cycle more closely to the field maximum, dominating the emergence of each integer laser cycle with the same direction of the fast electrons, and thus obtaining little drift momentum. The ionization fluxes persist as long as the field is still on, becoming weaker and weaker as the time goes on. Instead, the ionization sequence is much different for the back-to-back emission pair. The ionization

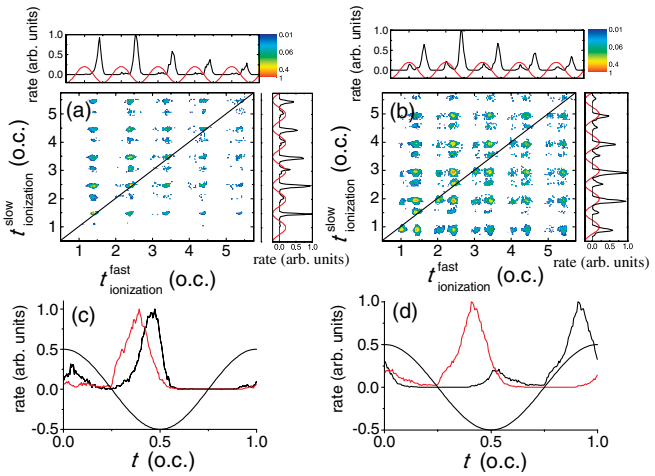

FIG. 2 (color online). Temporal correlation of the side-by-side emission (a) and back-to-back (b) emission. The main diagonal lines in (a) and (b) indicate the simultaneous double ionization events. The ionization rates of the fast electron and slow electron for the side-by-side and back-to-back pairs in the laser field are indicated as the side plots. (c) and (d) show the statistical ionization time of fast electrons (red curves) and slow electrons (black curves) of the side-by-side and back-to-back emission, respectively.

fluxes of the fast electrons mainly appear at $\sim 1.4, 2.4, 3.4\ldots$ o. c. and the slow electrons mainly appear at $t \sim 0.9, 1.9, 2.9, 3.9, \ldots$ o. c. Generally, the ionization of the fast electron and slow electron of the side-by-side emission pair is "in phase", i.e., with the same field direction. And the fast electron and slow electron of the back-to-back emission are dominantly released "out of phase", i.e., with the opposite field direction, as shown in Figs. 2(c) and 2(d).

Those regular delayed double ionization with more than one o.c. indeed arises from sequential ionization of doubly excited states that are formed at the instant of a laser-assisted electron hard recollision. Usually, the later ionized electrons can be released through the overbarrier ionization or tunneling ionization (see the Supplemental Material [27] for the details). We have analyzed the statistical binding energies of the tunneled ones and struck electrons in the doubly excited states, as shown in Fig. 3(a). One can find that two electrons usually have very similar binding energy of ~ 0.35 a.u. Because the maximal energy of struck electrons in a classical laser field is about $3.17Up$, one can estimate the energies of the doubly excited states are about $3.17Up - I_p$ (I_p is the first ionization potential of Ar). The dashed line in Fig. 3(a) indicates the events which satisfy the energy relation of $E_{\mathrm{struck}} + E_{\mathrm{tunneled}} = 3.17Up - I_p$. The simulation reveals that the tunneled and struck electrons usually share the energy in the doubly excitation states.

To understand the ionization dynamics from the double excitation states, we have further analyzed the binding

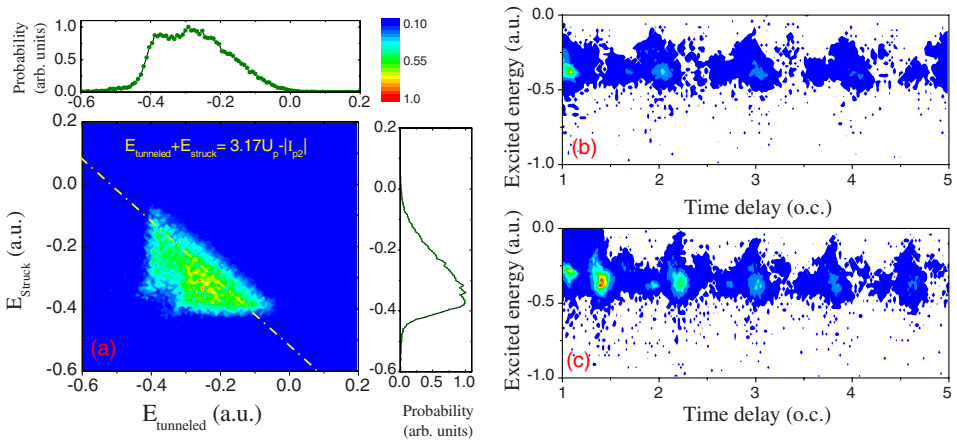

FIG. 3 (color online). (a) The binding energy of tunneled and struck electrons in the doubly excited states. The doubly excited energies are calculated at an instant slightly (5 a.u. in time) after the recollision. The side plots show the statistic binding energy of tunneled electrons and struck electrons. The white dashed line indicates the double ionization events that satisfy the relation of $E_{\text{struck}} + E_{\text{tunneled}} = 3.17\,Up - I_p$. (b) and (c) show the binding energy of later ionized electrons versus the ionization time delay for the side-by-side and back-to-back emission, respectively.

energy of the later ionized electrons for the side-by-side and back-to-back emission, respectively, as shown in Fig. 3(b) and Fig. 3(c). The binding energy of the later ionized electrons is about $I_p = 0.3$–0.4 a.u., which is slight higher than the binding energy of the first excited states of $Ar^+[I_p(3s3p^6) \sim 0.5$ a.u.]. Once one of electrons is released, the other electrons, denoted as the collisionally excited electrons, prefer to move in elliptical orbits and will continue mediated by the combined doubly charged ionic potential and the laser field. The statistical binding energy of the collisionally excited electron changes slightly after the first electron is released from the doubly excited state. The period of collisionally excited electrons in an elliptical orbital can be estimated with $T = 2\pi(1/2E)^{3/2} \sim 10.7$ a.u. (for $E = I_p \sim 0.35$ a.u.). The temporal period is an important signature for the ionization sequence, which is much less than the laser period (110 a.u.). The collisionally excited electrons will be either released by the laser field with the similar phase with the first ionized electron, producing the side-by-side emission, or later escape when the laser field is opposite, producing the back-to-back emission.

We have learned the temporal and energy information of the excited electrons. The further question we should settle is the physical origin of high energy photoelectrons for both types of electron correlation. It is a very intuitive picture that two electrons will be sequentially released through the doubly excited states. Here, we can assume the two electrons occupy the doubly excited level with an ionization potential $I_p = 0.35$ a.u. We first ignore the effect of Coulomb potential after the tunneling and solve the Newton's equations of motion: $(d^2r_i/dt^2) = -\varepsilon(t)$. The initial conditions of two electrons at the tunnel exit are prescribed by the ADK theory [26]. We can obtain the final momentum of two electrons as $p_{||}^i = (\varepsilon/\omega)\,\sin\omega t_i$ (t_i the ionization time of the two electrons), respectively. The calculated momentum distributions of the fast electron and slow electron of the side-by-side and back-to-back emission are shown in Figs. 4(a) and 4(b) (dashed curves). The ionization probability of the back-to-back emission is mainly populated with a peak near zero momentum. The cutoff of struck and bounded electrons is about $2Up$ as predicted with the simple man model when ignoring the Coulomb potential. The simple Coulomb-free sequential tunneling model does not reproduce the results of both the experiment and *ab initio* semiclassical simulation

In reality, a more accurate consideration should include the effect of the doubly charged ionic potential in the above simple model. Including the Coulomb effect, the final momenta can be approximated with $p_{||}^i = -\int_{t_i}^{t_f}(z_i/r_i^3)dt + (\varepsilon/\omega)\,\sin\omega t_i$. Here $\vec{r}_i(t) = [x_i(t), y_i(t), z_i(t)]$, and $r_i = \sqrt{x_i^2 + y_i^2 + z_i^2}$, which are obtained by solving Newton's equations of motion: $(d^2r_i/dt^2) = -\varepsilon(t) - (2/r_i^2)$. In doubly excited states, those electrons feel the potential

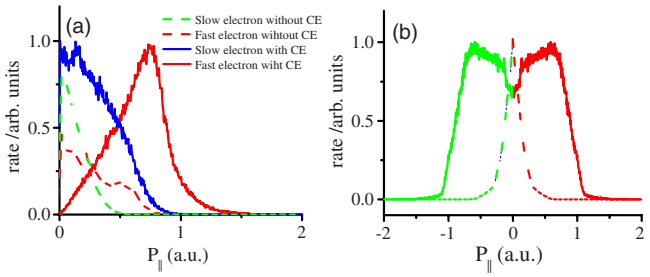

FIG. 4 (color online). The calculated momentum distributions of the fast and slow electrons for the side-by-side (a) and back-to-back (b) emission with the models of the Coulomb-free sequential tunneling (dash lines) and the Coulomb-corrected sequential ionization (solid lines) from doubly excited states.

of a doubly charged ion, i.e., $\sim 2/r_i$. Under these approximations, we then obtain the momentum distributions of the fast electron and slow electron of the side-by-side and back-to-back emission, as shown in Figs. 4(a) and 4(b) (solid curves), respectively. The calculated longitudinal momentum spectra with the Coulomb-corrected sequential tunneling model agree with the experiment and the *ab initio* semiclassical two-electron calculation. Different with the result in few-cycle laser fields [28], the collisionally excited electrons can achieve much higher momentum because of the Coulomb scattering with doubly charged ions in a long laser pulse.

The essential question is what is the role of electron correlation? Our analysis reveals that strong electron correlation takes place at an effective hard recollision to facilitate the formation of doubly excited states. The sequential ionization from doubly excited states with subsequent Coulomb scattering effect in the combined laser and doubly charged ionic fields are responsible for those high energy photoelectrons as much as 6.5 Up.

In conclusion, we have presented a triple coincidence study on strong-field double ionization of Ar atoms at a low laser intensity, where the momentum distribution of two outgoing electrons for the side-by-side and back-to-back emission are fully determined. We have traced back the ionization dynamics with the two-electron semiclassical atomic-ensemble model. By comparing the simple Coulomb-corrected and Coulomb-free sequential tunneling ionization models with an *ab initio* semiclassical model, we have found that the doubly excited states are largely populated after the laser-assisted recollision and large amounts of double ionization dominantly takes place through sequential ionization of doubly excited states. Electron correlation manifests its importance at the instant of formation of doubly excited states. The collision and ionization of the correlated electron pairs reveal regular temporal dynamics. The high energy electrons from double ionization are associated with the Coulomb scattering with the doubly charged ionic potential in the laser field.

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