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Energy correlation in above-threshold nonsequential double ionization at 800 nm

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We have investigated the energy correlation of the two electrons from nonsequential double ionization of helium atoms in 800-nm laser fields at intensities below the recollision threshold by quantum calculations. The circular arcs structure of the correlated electron momentum spectra reveals a resonant double-ionization process in which the two electrons transit from doubly excited states into continuum states by simultaneously absorbing and sharing excess energy in integer units of the photon energy. The effectiveness of Coulomb repulsion plays a decisive role in controlling the electronic dynamics in continuum states and is responsible for the dominant back-to-back electron emission and two intensity-independent cutoffs in the two-electron energy spectra.

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Electron-electron correlation in nonsequential double ionization (NSDI) of atoms and molecules by a short laser pulse at near-infrared (NIR) wavelengths has become the standard example for studies of dynamical electron correlations, which govern the dynamics of many phenomena in nature [1]. Thus, it has been investigated extensively in both experiment [1–6] and theory [8–11] in the past two decades. The physical mechanism responsible for NSDI is well established via the "recollision model" [12]. In this model, the first ionized electron is driven back to its parent ion by the oscillating laser field, causing ionization of the second electron in a direct $(e,2e)$-like encounter or indirectly via recollision-induced excitation of the ion with subsequent field ionization (RESI) [4]. At high laser intensities, two recent experiments [13,14] revealed some novel dynamical details in the $(e,2e)$ process. The fingerlike structure in the correlated electron momentum spectrum from NDSI of He is contributed by binary and recoil recollisions [13]. At a higher intensity, the correlated momentum spectrum exhibits a pronounced V-shaped structure, which is explained as a consequence of Coulomb repulsion and typical $(e,2e)$ kinematics [14]. At intensities below the recollision threshold, a very recent experiment of double ionization of Ar [15] found dominant back-to-back emission of the two electrons along the laser polarized direction, in striking contrast to previous findings at higher intensities. In addition, this experiment confirmed a predicted intensity-independent high-energy cutoff in the two-electron energy spectra [16]. However, the detailed microscopic dynamics in the RESI process remains unknown.

Quantum-mechanical calculations of multiphoton double ionization of He at extreme ultraviolet (XUV) wavelengths [17], as well as visible and ultraviolet (UV) wavelengths [18], demonstrated nonsequential double-electron above-threshold ionization (NS-DATI), from which the two-electron energy spectrum shows peaks separated by the photon energy. Recent experiments on double ionization of He and Ne by strong free-electron laser pulses [19,20] revealed a dominant NSDI process in which the two electrons can absorb two vacuum UV photons resonantly. Unlike NSDI at XUV and UV wavelengths, in which the two electrons transit directly from the ground state into continuum states, at NIR wavelengths they are ionized from excited states into continuum states after recollision for the RESI mechanism and this process is sequential according to Ref. [4]. Does the resonant double ionization also exist in NSDI of atoms at NIR wavelengths?

In this communication, we investigate the correlated electron momentum and energy spectra from NSDI of helium atoms with ultrashort 800-nm laser pulses at intensities below the recollision threshold by numerically solving the two-electron time-dependent Schrödinger equation (TDSE). Our calculations excellently reproduce the experimental results in [15] and, most importantly, reveal that a resonant double-ionization process dominates in the RESI mechanism. We draw a NSDI scenario which provides a clear and complete physical explanation for the back-to-back emission of the electrons and two intensity-dependent cutoffs in the two-electron kinetic energy spectra predicted in our calculations, as well as observed in the experiment [15].

Many previous works have shown that one-dimensional models are able to qualitatively reveal the mechanisms of NSDI and reproduce the features of NSDI [9,18,21,22]. Furthermore, the experiment [15] has shown that at intensities below the recollision threshold no effect of Coulomb repulsion between the two electrons is found in the direction perpendicular to the laser polarization. Hence, we employ a "one-plus-one"-dimensional model of an helium atom with soft Coulomb interactions, where the motion of both electrons is restricted to the laser polarization direction [9]. We use the split-operator spectral method [23] to numerically solve the two-electron TDSE (in atomic units): $-i\frac{\partial}{\partial t}\Psi(z_1,z_2,t) = H(z_1,z_2,t)\Psi(z_1,z_2,t)$, where z_1,z_2 are the electron coordinates. $H(z_1,z_2,t)$ is the total Hamiltonian and reads $H(z_1,z_2,t) = -\frac{1}{2}\frac{\partial^2}{\partial z_1^2} - \frac{1}{2}\frac{\partial^2}{\partial z_2^2} - \frac{2}{\sqrt{z_1^2+1}} - \frac{2}{\sqrt{z_2^2+1}} + \frac{1}{\sqrt{(z_1-z_2)^2+1}} + (z_1+z_2)E(t)$. $E(t)$ is the electric field of a laser pulse. Following Ref. [9], the two-dimensional space is partitioned into two outer regions, (A) $\{|z_1| < a\}$, or $\{|z_2| < a\}$ and (B) $\{|z_1|,|z_2| \geqslant a\}$, with $a = 150$ a.u. The final results are insensitive to the choice of a ranging from 100 to 200 a.u. In region A, the wave function is propagated exactly in the presence of combined Coulomb and laser field potentials. In region B, which corresponds to double ionization, all the Coulomb interactions are neglected and the time evolution of the wave function can be performed simply by multiplications in momentum space. The two regions are smoothly divided by a splitting technique [24]. At the end

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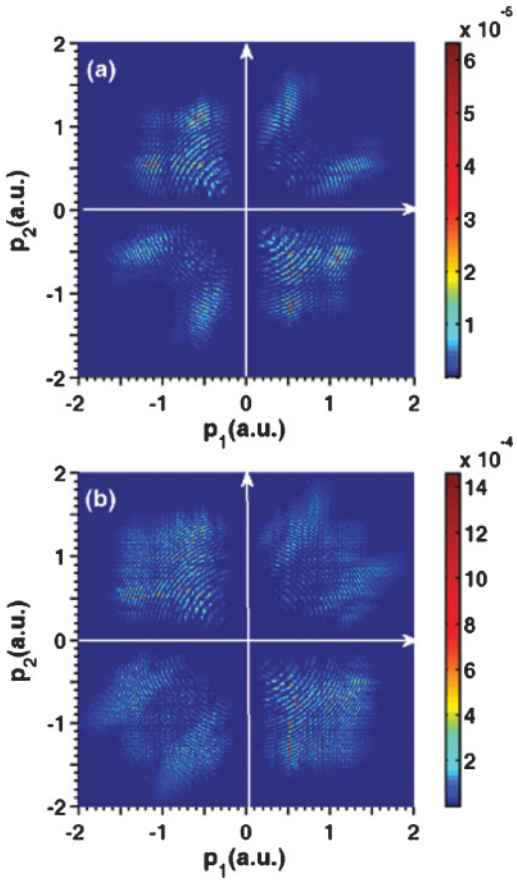

FIG. 1. (Color online) Correlated electron momentum spectra for double ionization of He at the intensities (a) 0.1 PW/cm^2 and (b) 0.15 PW/cm^2. The units are arbitrary.

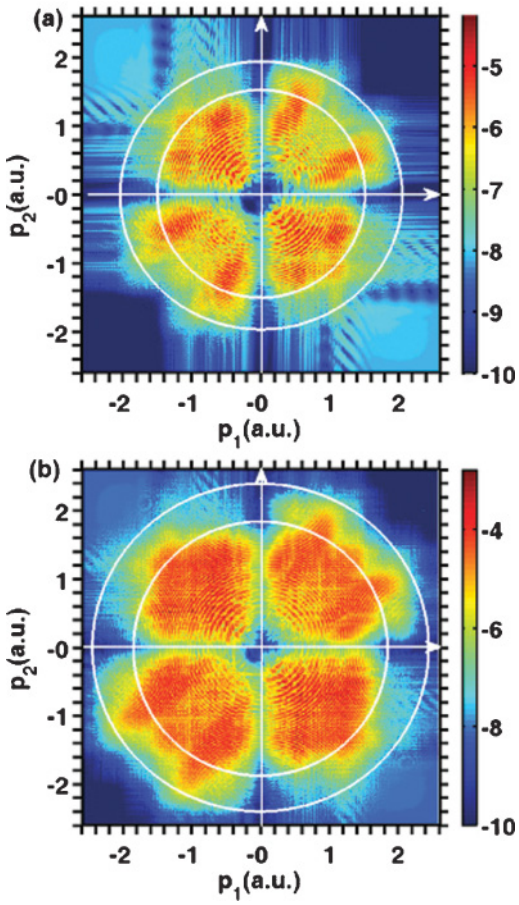

FIG. 2. (Color online) Log plot of the correlated electron momentum spectra for double ionization of He at the intensities (a) 0.1 PW/cm^2 and (b) 0.15 PW/cm^2. The units are arbitrary.

of the propagation, the wave function in region B yields the two-electron momentum and energy spectra.

Our calculations use trapezoidally shaped 800-nm laser pulses with a total duration of 10 optical cycles, switched on and off linearly over 2 optical cycles. The very large grid size of 5000 × 5000 a.u. with a spatial step of 0.3 a.u. is used, while the time step is 0.1 a.u. The very large grid provides sufficiently dense continuum states [18] to yield highly accurate two-electron momentum and energy spectra. The initiate wave function is the two-electron ground state of He obtained by imaginary-time propagation. After the end of the pulse, the wave function is allowed to propagate without a laser field for the additional time of 10 optical cycles. The final results do not change any further even though the wave function propagates without a laser field for a longer additional time.

Figure 1 shows the resulting two-electron momentum spectra from double ionization of He at various intensities below the recollision threshold. These correlated momentum spectra exhibit several significant features. The first and most striking feature is serried concentric arcs, which are most pronounced on a logarithmic scale (see Fig. 2). These concentric arcs satisfy $p_1^2 + p_2^2 =$ constant, and it is shown here that the energy separation between adjacent arcs is the laser photon energy $\hbar\omega$. This essentially reveals a resonant double-ionization process [17] in which the two electrons are strongly correlated. The second feature is that the momentum distributions deviate significantly from the diagonals, which is a result of the mutual Coulomb repulsion between the two electrons in continuum states. The other two features of the momentum spectra in Fig. 1 that have been observed in the experiment [15] are the dominance of back-to-back emission of the two electrons and the clear minimum in a significant area around the origin. The authors of [15] explained that multiple recollision-induced continuous excitations with subsequent field ionization is responsible for the dominant back-to-back emission. However, according to [4] the two electrons due to the RESI mechanism are emitted independently, either into the same direction or back to back. Hence, multiple recollisions may explain the back-to-back emission but cannot explain the dominance of the back-to-back emission. Here, we consider the effectiveness of strong Coulomb repulsion between the two electrons in continuum states to be responsible for the last three features. More details of our explanation are given in the proposed scenario below.

The quantum calculations of double ionization of atoms by strong laser pulses at XUV [17] and UV [18] wavelengths, have shown concentric circles in the correlated electron momentum spectra. However, no effect of Coulomb repulsion is found in the correlated momentum spectra. These concentric circles correspond to a resonant double ionization process, in which the strongly correlated two electrons simultaneously absorb

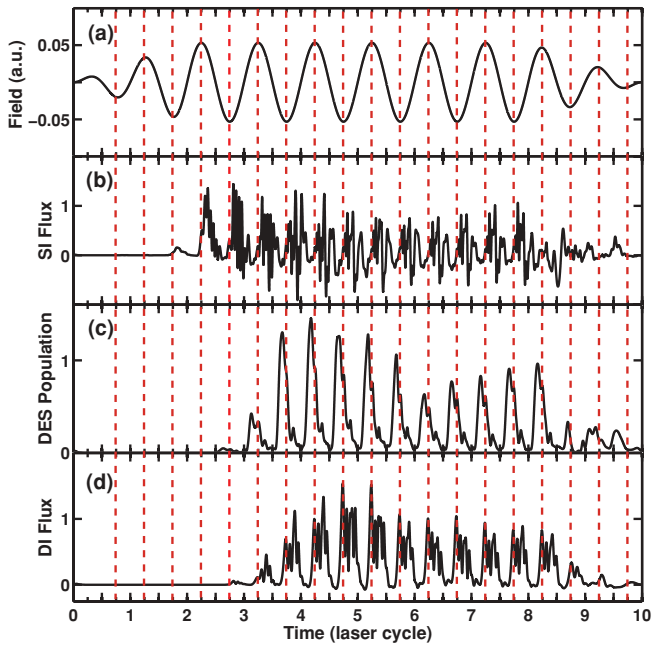

FIG. 3. (Color online) (a) Laser field for the intensity of 0.1 PW/cm^2. (b, d) Population fluxes of single and double ionization, respectively. (c) Population of doubly excited states.

and share energy in integer units of the photon energy, transiting from the two-electron ground state into continuum states with the assistance of the Coulomb potential between the two electrons. Likewise, the first feature shown in Fig. 1 also reveals a similar resonant double ionization process. However, the two resonant processes are distinct because of their different mechanisms. For NIR wavelengths and laser intensities below the recollision threshold, it is well known that RESI is the mechanism of NSDI [4]. Conclusively, the two electrons transit exclusively from the doubly excited states [26], which are first populated via recollision, into continuum states in the resonant double-ionization process for NIR wavelengths. The doubly excited states are verified by observing the population in region 1: $\{7 < |z_1, z_2| < 10\}$ a.u. [7]. Regions 2 $\{|z_1| < 7, |z_2| > 12.5$ or $|z_1| > 12.5, |z_2| < 7\}$ a.u. and 3 $\{|z_1| > 12.5, |z_2| > 12.5\}$ a.u. correspond to singly and doubly ionization, respectively. Figure 3 shows the population of doubly excited states and the population fluxes of single and double ionization as functions of time. The maximum of single ionization occurs shortly after the extremum field strength. Recollision often occurs at zero field, that is, about 0.75 laser cycle after single ionization. Figure 3(c) shows doubly excited states bursting between 0.75 and 1 laser cycle after single ionization and decay sharply when the next extremum field strength reaches. Shortly thereafter, double ionization appears. Note that the population of region 1 is dominated by doubly excited states during the laser pulse, since double ionization cannot occur near zero field at intensities below the recollision threshold. During the pulse switch-off process, there is little double-ionization signal even though the population of doubly excited states is still significant, because the field is not strong enough to ionize doubly excited states [see Figs. 3(c) and 3(d) from 9 to 10 laser cycles].

According to these dominant features in Fig. 1, we draw a scenario for the corresponding double-ionization process. At intensities below recollision threshold, since the kinetic energy of the recolliding electron is not enough to directly free the second one, strongly correlated, doubly excited states are populated via recollision. Then the two electrons transit from doubly excited states into continuum states by simultaneously absorbing a number of NIR photons and sharing excess energy in units of the photon energy. Finally, and most importantly, the effectiveness of Coulomb repulsion plays a decisive role in the electronic dynamics in continuum states. With the same restricted Hamiltonian, it has been shown that the effect of Coulomb repulsion in continuum states along the laser polarization direction does not exist in NSDI process for the UV case [18]. This implies that the effect of Coulomb repulsion essentially results from the mechanism of NSDI for the NIR case, rather than the use of the restricted Hamiltonian. Near the field maxima the resonant double ionization is most probable and the electrons acquire small drift momenta from the laser field. Consequently, the strong Coulomb repulsion leads to the dominance of back-to-back emission and the small drift momenta accruing from the laser field lead to the momentum distribution in the second and fourth quadrants deviating from the minor diagonal. While freed near the zero field, the electrons can acquire the same large drift momenta from the laser field. Coulomb repulsion leads to the momentum distribution in the first and third quadrants off the main diagonal. Moreover, Coulomb repulsion is also responsible for the minimum at the origin in the momentum distribution. Actually, this minimum implies that the total kinetic energy of the two electrons (see Fig. 4) is often higher than 0 and essentially reveals that the two electrons already possess significant energy when they transit into continuum states. Exclusively, this energy is provided by the Coulomb repulsion energy. The minimum has a radius of about 0.45 a.u. for 0.1 PW/cm^2 and 0.6 a.u. for 0.15 PW/cm^2, implying a lowest Coulomb repulsion energy of 0.1 and 0.18 a.u., respectively, when the electrons are re-emitted at the field maximum. Figure 1(a) shows clear maxima for back-to-back emitted electrons with momenta of about 0.55 and 1.1 a.u.,

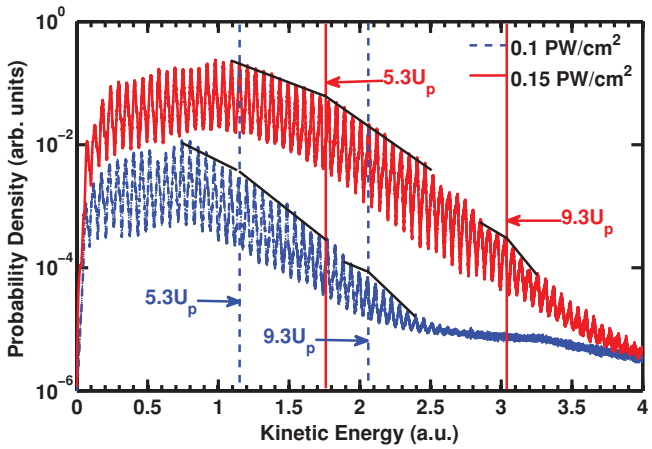

FIG. 4. (Color online) Two-electron kinetic energy spectra for double ionization of He shown in Fig. 1. The dashed (blue) line and the solid (red) line correspond to 0.1 and 0.15 PW/cm^2, respectively.

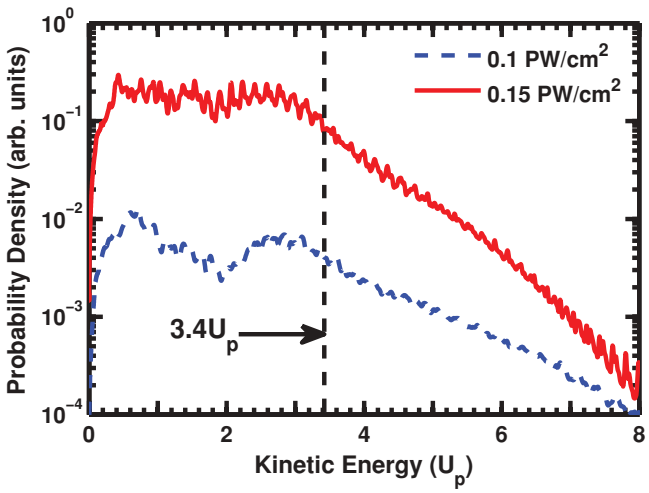

FIG. 5. (Color online) Kinetic energy spectra of one ionized electron from double ionization of He shown in Fig. 1. The dashed (blue) line and the solid (red) line correspond to 0.1 and 0.15 PW/cm^2, respectively.

respectively. These maxima correspond to a momentum of 0.825 a.u. delivered by Coulomb repulsion and a momentum of 0.275 a.u. acquired from the laser field. The momentum of 0.275 a.u. corresponds to a re-emission phase of 17° off the field maximum [1,6].

The double-ionization process based on this scenario can be called recollision-induced double excitation with subsequent above-threshold double ionization. Within this double-ionization scenario, the high-energy cutoffs in the two-electron kinetic energy spectrum can be well explained. Figure 4 shows the total kinetic energy spectra of both emitted electrons from Fig. 1. Very similar to the above-threshold ionization electron spectrum, the two-electron spectrum also exhibits peaks spaced by the photon energy, implying a strong energy correlation of the two electrons. Moreover, a plateau with an intensity-independent cutoff energy of about $5.3U_p$ is evident in the spectra. Beyond this plateau the spectra decay exponentially until another intensity-independent cutoff energy of about $9.3U_p$. Beyond $9.3U_p$, the multiphoton effect vanishes quickly and a background plateau follows. The two cutoffs are more prominent in Fig. 2, indicated by the two circles, respectively.

The $5.3U_p$ cutoff has been predicted at 390 nm [16] and observed at 800 nm (see Fig. 1(c) in [15]). The $9.3U_p$ cutoff is also observable in Fig. 1(c) in [15] but is not pointed out explicitly by the authors. The difference between the two cutoffs is $4U_p$, just twice the maximum energy a "free" electron can gain from an oscillating field. Surprisingly, the spectra ranging from $5.3U_p$ to $9.3U_p$ decay exponentially, very similar to single-ionization spectra ranging from 0 to $2U_p$ [12,25]. Thus, we propose that the maximum Coulomb repulsion energy between the electrons corresponds to the $5.3U_p$ cutoff. An additional energy of $4U_p$, the maximum energy the two electrons can gain from the oscillating field, plus the maximum Coulomb repulsion energy, is responsible for the $9.3U_p$ cutoff. Figure 5 shows an evident intensity-independent cutoff energy of about $3.4U_p$ in the spectra of one electron from double ionization. This reveals that for the first plateau region, one electron can share a maximum energy of $3.4U_p$, while the maximum energy of the other can share is $1.9U_p$ [16].

In summary, we have found a resonant double-ionization process at 800 nm and intensities below the recollision threshold, in which both electrons simultaneously absorb and share energy in integer units of the photon energy, transiting from doubly excited states into continuum states. The electronic dynamics in continuum states is controlled by the effectiveness of Coulomb repulsion. Our proposed double-ionization scenario provides a fundamental physical explanation for the dominant features in the correlated electron momentum spectra and the two intensity-independent cutoffs in the two-electron kinetic energy spectra. The peak structure and the two cutoffs in the energy spectra are signatures of a strong energy correlation of both electrons in the transition process and continuum states, respectively.

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