Knee structure in double ionization of noble atoms in circularly polarized laser fields

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Nonsequential double ionization is characterized by a knee structure in the plot of double-ionization probability versus laser intensity. In circularly polarized (CP) laser fields, this structure has only been observed for Mg atoms. By choosing laser fields according to a scaling law, we exhibit the knee structure in CP laser fields for Ar and He atoms. The collision of the ionized electron with the core enhances the ionization of the second electron and forms the knee structure. The electron recollision is universal in CP laser fields, but the ionization probability in the knee region decreases as the wavelength of the driven field increases. For experimental observations, it is beneficial to use target atoms with small ionization potentials and laser fields with short wavelengths.

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I. INTRODUCTION

When atoms and molecules are irradiated by intense laser fields, nonsequential double ionization (NSDI) may occur [1–4]. NSDI denotes a process whereby the two electrons are dependently ionized and hence the double-ionization yield is enhanced, owing to the electron correlation. There are several scenarios to explore the NSDI process, such as the shake-off mechanism [5], the collective-tunneling mechanism [6], and the rescattering mechanism [7]. NSDI predominates when the laser intensity falls below a certain value. When the laser intensity is further increased, the second electron will be directly ionized by the laser field and is independent of the first electron. Thus, the double ionization becomes sequential, and the ionization enhancement is suppressed. Hence, a knee structure appears in the plot of double-ionization probability versus laser intensity, and this is a signature of NSDI [8].

The knee structure was observed in linearly polarized (LP) laser fields for many atoms, but it is rarely observed in circularly polarized (CP) laser fields. A widely accepted picture is that, in the CP laser fields, the freed electron moves in a circle around the parent core and hence is less likely to encounter the parent core. Thus, recollision with the parent core is greatly suppressed. One exception is the knee structure observed in Mg atoms driven by the CP laser field [9], which has attracted much attention [10–13]. Mauger *et al.* [10] confirmed the return of the first electron and its subsequent recollision with the parent core. Fu *et al.* [11] disclosed the returning time of the first electron. Xu *et al.* [12] illustrated the role of the time interval in electron emission between the recollision and the final double ionization. These studies disclosed the electron recollision process in the CP laser fields.

Generally, in double ionization, the target atoms can be treated as structureless, and only the first and second ionization potentials are important [9]. This implies that when one electronic process occurs in one atom, it might occur in other atoms, provided that the driving laser fields satisfy certain conditions. Because the electron recollision in the CP laser field occurs for Mg atoms, one might expect the electron recollision as well as the resulting knee structure to be ubiquitous in the CP laser fields. However, the knee structure in the CP laser fields has not been observed for other atoms. Whether the structure exists or not in the CP laser fields is of common interest in the strong-field community. Once the knee structure is observed in the CP laser fields, it would verify the universality of the electron collision, which enriches our knowledge on the electron dynamics in intense laser fields. However, observing the knee structure is still an open question.

In 2003, a practical scaling law on photoionization was established [14] and was used to identify equivalent electronic process in different laser-driven atoms [15,16]. The scaling law holds for rescattering [17], high-order harmonics [18], and double ionization [19]. The scaling law states that for two systems of laser-driven atoms S (ω, I, I_p) and S' (ω', I', I'_p), the electronic processes are the same provided that we choose $\omega' = \omega(I'_p/I_p)$ and $I' = I(I'_p/I_p)^3$, where ω and I are the frequency and intensity of the laser field and I_p is the ionization potential of the atom. The values with prime denote those for another system. This suggests that, for two atoms, the principal electronic processes are the same if the laser fields are connected by the scaling law. This makes a way to exhibit the knee structure in the CP laser fields.

Here, we study the NSDI of Ar and He atoms in intense CP laser fields, by referring to the Mg atoms. We choose the driving laser fields according to the scaling law, and exhibit the knee structure for both Ar and He atoms driven by the CP laser fields. By tracking the time evolution of double-ionization events, we show the electron recollision and the resulting enhancement in double-ionization rate. We show that the electron recollision is universal in the CP laser fields, but the double-ionization probability in the knee region decreases as the driven wavelength increases. How to observe the knee structure is also discussed.

II. KNEE STRUCTURE IN DOUBLE-IONIZATION YIELD

The simulation method used here is based on the numerical integration of the time-dependent canonical equation, which is very effective in dealing with the NSDI phenomena [20–25]. We model the atoms as explained in Ref. [19]. Briefly, an initial state of a two-active-electron atom satisfies the field-free Hamiltonian in which the Coulomb interaction uses the soft-core model. The initial state moves freely until the resulting ensemble maintains a stable position and momentum distributions. Thus, we obtain the initial ensemble. When the

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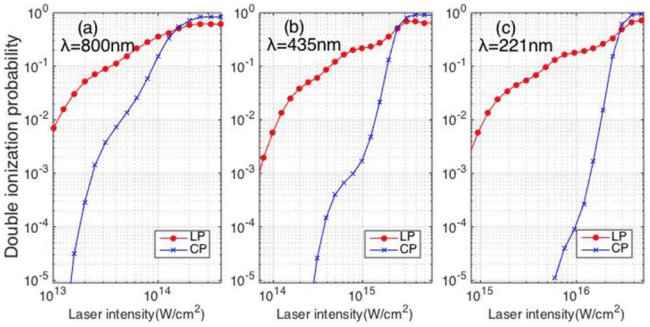

FIG. 1. Variation of double-ionization probabilities with the laser intensity for (a) Mg, (b) Ar, and (c) He atoms. The wavelengths of the driving laser field are stated in each plot.

laser pulse is switched on, a dipole interaction term is added on the field-free Hamiltonian. The motion of the two electrons is governed by the canonical equation, which is solved via the standard fourth- to fifth-order Runge-Kutta algorithm. A double-ionization event is counted when the energy of each electron is greater than zero at the end of the laser pulse. The double-ionization probability is calculated as the number of double-ionization events over that of the initial ensemble. In our calculations, the electric field $\mathbf{E}(t)$ of the CP laser pulse is given by $\mathbf{E}(t) = E_0 f(t)[\cos(\omega t + \phi)\hat{\mathbf{x}} + \sin(\omega t + \phi)\hat{\mathbf{y}}]$, where E_0 and ω are the laser amplitude and frequency, respectively, and ϕ is the carrier-envelope phase. The pulse envelope $f(t)$ has a five-cycle duration and is trapezoidal with one cycle in both the ramp on and the ramp off [26].

We first studied the double ionization of Mg atoms driven by 800-nm laser pulses. This was done to check our code and was used as a reference for further simulations. The double-ionization probability increases fast with laser intensity but keeps almost constant (i.e., ionization saturation) for laser intensities higher than 2.0×10^{14} W/cm^2 [Fig. 1(a)]. When the laser intensity is among $(3.0–8.0) \times 10^{13}$ W/cm^2, the double-ionization probability increases clearly more slowly than that in neighboring regions. This forms the characteristic knee structure, which is more notable in the CP case. The knee structure in the CP laser field appears in the same laser intensity range as that in the LP laser field, which agrees well with the experimental observation [9]. In the CP laser fields, the first ionized electron runs into some periodic orbits via saddle points. The electron in the periodic orbits returns to the parent core and then collides with the inner electrons [10]. The electron recollision enhances the ionization yield of the second electron to form a knee structure.

According to the scaling law, if the driving laser fields are of equal ponderomotive parameters, then the electron dynamics will be the same, as is the electron recollision in the CP laser fields. To keep the ponderomotive parameter (defined as $u_p = 0.25I/\omega^3$) constant, one should enlarge the laser intensity k^3 times the reference value when the laser frequency is enlarged k times. In double ionization, the scaling ratio k equals the ratio of the second ionization potentials [19]. Here, we study the double-ionization process by choosing the laser fields according to the scaling law.

We next calculate the double-ionization probability of Ar atoms. The second ionization potentials of Ar and Mg atoms are 27.63 and 15.04 eV, respectively, and the scaling ratio equals 1.84. Hence, the wavelength of the laser field driving the Ar atoms is chosen as 435.5 nm. We expect the knee structure to appear at an intensity range 1.84^3 times that for the Mg atoms. The calculated double-ionization probabilities are shown in Fig. 1(b) as a function of laser intensity. Clearly, the knee structure does appear and is obvious for the laser intensity near 5.0×10^{14} W/cm^2. The double-ionization probabilities in the LP laser fields exhibit a knee structure in the same laser intensity range. In the knee region, the double-ionization probabilities in the CP laser fields are two orders of magnitude lower than that in the LP laser fields.

Finally, we calculate the double-ionization probability for He atoms. He atoms are those atoms with large ionization potentials. The scaling ratio equals 3.62, and the wavelength of the driving laser field is 221 nm [27]. The calculated double-ionization probabilities for both the CP and LP laser fields are shown in Fig. 1(c). The knee structure does appear and is evident around 1×10^{16} W/cm^2 for both the CP and LP laser fields. At this region, the double-ionization probabilities in the CP laser field are three orders of magnitude lower than that in the LP laser fields.

III. ORIGIN OF KNEE STRUCTURE

The knee structure in the double-ionization probability of noble atoms driven by the CP fields does exist and appears at the same intensity range as that in the LP laser fields. This gives rise to several questions: What causes the enhancement of the double-ionization probability in the CP laser fields? We can refer to the electronic dynamics for the Mg atoms in which the knee structure is caused by the electron recollision at certain time windows [10,11]. Does the electron recollision exist for noble atoms in the CP fields? When and how does the electron recollision occur? To answer these, we studied the time evolution of electron trajectories.

The time evolution of the repulsion energy distribution between two electrons is depicted in Fig. 2. As a reference, Fig. 2(a) shows the case for Ar atoms in the LP laser fields.

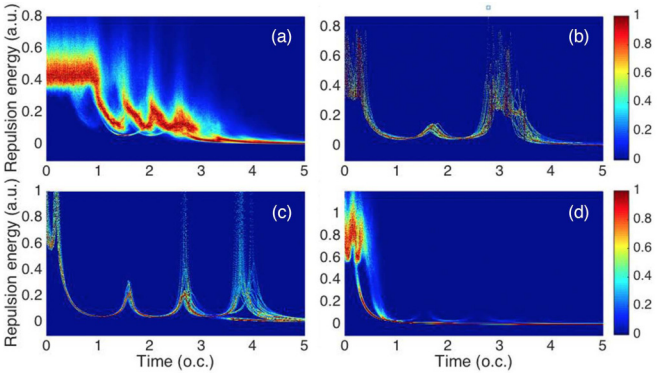

FIG. 2. Time evolution of the repulsion energy distribution between two electrons: (a) Ar atoms driven by the LP laser pulse at intensity 7.80×10^{13} W/cm^2, (b) Ar atoms driven by the CP laser pulse at intensity 2.47×10^{14} W/cm^2, (c) He atoms driven by the CP laser pulse at intensity 5.96×10^{15} W/cm^2, and (d) He atoms driven by the CP laser pulse at intensity 1.50×10^{16} W/cm^2. The term o.c. in the abscissa denotes the optical cycle of the laser fields.

In the first optical cycle, the two electrons are both near the parent core, because the laser field is weak. Thus, the repulsion energy is large. The electric field is strong from the second cycle and drives the electron away from the parent core. The two electrons become distant, and the repulsion energy decreases. However, when the electric field inverts, it pulls the two electrons back to the parent core, so the two electrons become close again. This generally occurs after half of an optical cycle, and the repulsion energy exhibits several peaks separated by a half-cycle interval. The electrons collide when they are close and then the Coulomb force pushes them apart. At the chosen intensity, the electron recollision finishes within two optical cycles. Finally, the two electrons become distant and are not pulled back again. Thus, the repulsion energy is always small. Additional optical cycles do not result in electron recollisions, implying that the ionization probability in the used short pulses is the same as that in the long pulses. This ensures that our results hold for longer pulses.

The peaks in the repulsion energy distribution denote that the two electrons are close and the electron recollision may occur at this time. Based on this, Fig. 2(b) shows that the electron recollision does occur for Ar atoms driven by the CP laser field: two peaks appear in the pulse-flat region, and the latter one is more prominent. The second peak is as high as 0.8 a.u. and the two electrons may be close to 1.25 a.u. after one electron is ionized. This suggests that the electron recollision does occur and the latter peak corresponds to the time of electron recollision. The prior peak is not so notable, indicating that the electrons are not very close to each other. The electron recollision occurs mainly in the second peak.

The time evolution of the repulsion energy for He atoms is similar [Fig. 2(c)] at a laser intensity of 5.96×10^{15} W/cm^2. Three peaks are evident in the pulse-flat region, suggesting that the electron recollision does occur. Compared with Fig. 2(b), the bright curves are slim, indicating fewer recollision events and a smaller probability of double ionization. This is caused by the large ionization potentials of He atoms. Of the three peaks, the third one is the most prominent. This suggests that the recollision events occur mainly in the third cycle of the pulse flat. The time interval of the adjacent peaks is one optical cycle, implying that the electron return occurs only once in an optical period. This differs from the LP case. In high-intensity laser fields [Fig. 2(d)], no peak in the pulse-flat region is seen, denoting that there are fewer recollision events. The double ionization is in a sequential region; hence, the electron recollision is not yet important.

To show how the electrons collide in the CP fields, we depict the time evolution of the distance between the electrons and the parent core in Fig. 3. In the LP laser fields [Fig. 3(a)], the bright region in the third and fourth cycles denotes double ionization, and the ionized electrons generally leave the core more than 10 a.u. In the CP laser fields shown in the other three panels, the trajectories can be classified into two categories: one is the red line located at the bottom of each panel and denoting that most electrons move in a small circle around the core. The other is the light-blue curve that exhibits several minima and maxima denoting that the electron may be very near and far away from the core. When the electron moves near the parent core, it may collide with and transfer energy to the second electron. This enhances the ionization probability of the second

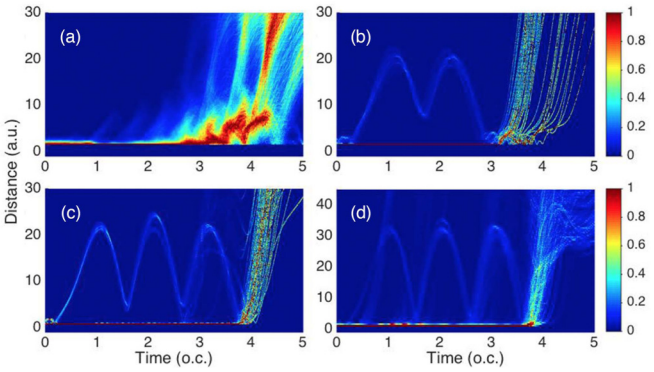

FIG. 3. Same as in Fig. 2, but for the time evolution of the distance between the electron and the parent core. The term o.c. denotes the optical cycle of the laser fields.

electron. Figure 3(b) suggests that the electron recollision happens mainly during the fourth cycles for Ar atoms, while Fig. 3(c) indicates that the electron recollision occurs in the end of the fourth cycles for He atoms. In the sequential ionization region [Fig. 3(d)], the light-blue curve is still notable and oscillates in a larger amplitude. This curve exhibits several minima about zero, denoting that electron collisions still occur at high-intensity CP fields, but the recollision events make only minor contributions to the double ionization. Most electrons are ionized directly by the laser field.

Electrons return to the core through some periodic orbits [10]. Stark-like saddle points exist in the combined field of the Coulomb attraction of the core and the electric force of the CP laser field. If the electron is ionized through the saddle point, then it may move into a periodic orbit [28]. The trajectories of the electrons in these periodical orbits compose the light-blue curves shown in Fig. 3. These curves generally oscillate in a period of one optical cycle, which suggests that the electrons in these orbits return to the core in each optical cycle. In Ref. [11], the electrons' return in Mg atoms has been shown to occur at certain time windows separated by one optical cycle. The periodic orbits depend on the laser intensity, which is seen upon comparison of Figs. 3(c) and 3(d).

To get a deep insight into the electron motion in the periodic orbits, Fig. 4 depicts two typical trajectories of the ionized electrons and the time evolution of the total energy, i.e., the kinetic energy plus the Coulomb potentials. The left-hand column is for Ar atoms driven by a 1.96×10^{15} W/cm^2 laser pulse. The electron moves in a cylindrical spiral after its ionization, and the corresponding energy is nearly constant before colliding. The electron collides with the parent core in the middle of the third cycle, and its energy decreases suddenly and then increases promptly. The time evolution of the total energy distribution is a bright line with three light-blue peaks. The bright line has a trivial variation in the pulse-flat region, indicating the energies of all electrons in the orbits vary slightly in one optical cycle. The three peaks become distinct in turn, implying that the electron collisions occur from the second cycle of the laser pulse but mainly in the fourth cycle. The right-hand column is for He atoms driven by a 9.45×10^{15} W/cm^2 laser pulse. Similar features are recovered. The electron trajectories suggest that the electron

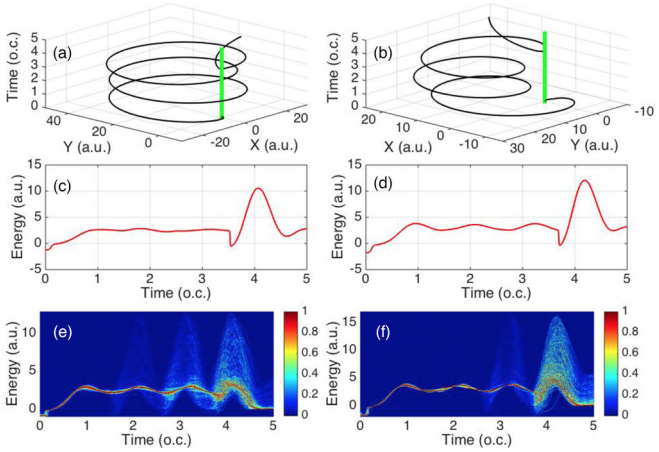

FIG. 4. Typical trajectory of an ionized electron and the corresponding energy for (a, c) Ar and (b, d) He atoms, respectively. (e, f) Time evolution of the electron's energy distributions. The green lines in the top two panels denote the position of the core. The term o.c. in the abscissa denotes the optical cycle of the laser fields.

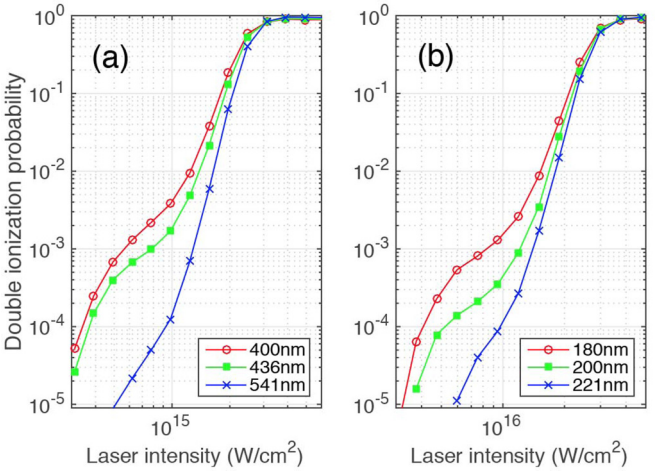

FIG. 5. The double-ionization probabilities as a function of laser intensity for (a) Ar atoms and (b) He atoms. The wavelengths are marked in each plot.

moves mainly in one side of the core, and tends to leave from the core. The electron collides with the core when the other electron lies in the opposite side of the core. This suggests that the electron-core collision occurs via other electrons.

IV. DISCUSSIONS

The electron recollision is universal in the CP laser fields, and it enhances the double-ionization probability. One might wonder why the knee structure in the CP fields is never observed for noble atoms. Figure 5 depicts the calculated double-ionization probabilities at several wavelengths. In each panel, the knee structures are evident in general and appear at the same intensity range. As the laser wavelength increases, the ionization probability in the knee region decreases; hence, the knee structure becomes not as striking as that in the short-wavelength case. This suggests that the double-ionization probability in the knee region is too low to be observed experimentally in the 800-nm CP laser field. To observe the knee structure in the CP laser fields, the wavelength of the driving laser field should be sufficiently short. In comparing Figs. 5(a) and 5(b), we find that the deeper the ionization potential, the smaller the double-ionization probability in the knee region. This indicates that target atoms with small ionization potentials, such as Xe and Ar atoms, are the proper candidates to observe the knee structure in the CP laser fields.

In Ref. [11], based on a semiclassical quasistatic treatment, Fu *et al.* made a criterion for observing the knee structure that the laser frequency should be larger than $0.18(I_p)^{5/4}$, where I_p is the first ionization potential. Correspondingly, some alkali-metal atoms are predicted to show the knee structure at 800 nm wavelength while all the rare gas atoms are expected to show the knee structure for wavelengths shorter than 800 nm. This agrees with our statement. In Ref. [28], the knee structure for Xe atoms was shown at 780 nm wavelength, but the double-ionization yield at the knee region is low. For convenience of observation, one may choose the driving wavelength of about 600 nm.

V. CONCLUSIONS

We studied the double ionization in CP laser fields using a classical simulation method. This exhibits the characteristic knee structures for Ar and He atoms for the first time. We find that the electron recollision is universal in the CP laser fields, and the knee structure does exist for noble atoms. The ionization probability in the knee region decreases as the wavelength of the driving laser pulses increases. It is lower for target atoms of large ionization potentials. To observe the knee structure in the CP laser fields, it is beneficial to use target atoms with small ionization potentials and to use driving laser fields with short wavelengths.

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