Correlated electron dynamics in strong-field nonsequential double ionization of Mg
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I. INTRODUCTION

Strong-field nonsequential double ionization (NSDI) is one of the most fundamental processes among various phenomena in intense laser-matter interaction. It has attracted increasing attention since the observation of the knee structure in the curve of doubly charged ion yield versus laser intensity. Nowadays it is widely accepted that the mechanism for NSDI could be understood with the recollision model, i.e., the outermost electron tunnels through the distorted potential barrier formed by the atomic potential and the intense laser field; then it is accelerated by the oscillating laser field and returns back to the parent ion when the laser field changes its sign, leading to the ionization of the second electron. Intuitively, this recollision process only occurs in linearly polarized laser fields. For elliptically and circularly polarized laser fields, the first ionized electron is driven away from the core by a transverse drift velocity, and thus recollision is forbidden. This view was confirmed by experiments for strong-field double ionization of the noble atoms, such as He and Ar, where the knee structure disappears in elliptically and circularly polarized laser fields. However, it is surprising that the knee structure is still present in the ion yield curve for double ionization of Mg by circularly polarized laser fields. Recently, theoretical studies show that recollision is still the responsible mechanism for NSDI in circularly polarized laser fields. In this case, the initial transverse velocity of the first electron and the Coulomb attraction between the ion and ionized electron play a key role for recollision.

The correlated electron momentum spectrum (CEMS) of NSDI provided deeper insight into the recollision process. By analyzing the CEMS, it has been shown that NSDI could occur through two different pathways: recollision-induced direct ionization (RDI) and recollision-induced excitation with subsequent field ionization (RESI). In the past decades, a great number of experimental and theoretical studies have been performed aiming at revealing the microscopic electron dynamics in NSDI. For instance, it has been experimentally shown that the CEMSs exhibit a repulsion behavior in the first and third quadrants. Theoretical studies demonstrated that this subtle feature indicated the final-state electron-electron repulsion and the electron-ion attraction after recollision and the asymmetric energy sharing during recollision. Recently, the decay process of recollision-induced doubly excited states in NSDI has also been revealed from the CEMS in NSDI of Ar. Previous studies indicated that the details of the CEMSs and the correlated electron dynamics of NSDI strongly depended on the targets. It has been demonstrated that the CEMSs for NSDI of He and Ar showed a strong correlated behavior, while for Xe, the CEMSs are almost uniformly distributed in the four quadrants and no correlation was observed. For Mg, there is seldom study focused on the CEMSs neither experimentally nor theoretically (with one exception).

The microscopic electron dynamics in NSDI also strongly depends on the wavelength of the laser pulse. Most of the previous studies on NSDI are focused on the near-infrared region. The development of laser technology pushes the wavelength into the mid-infrared (MIR) region. For long wavelengths, the microscopic dynamic of correlated electrons in NSDI is very different from the case of the near-infrared region. It was observed that the double-hump structure in the ion momentum distributions for NSDI of the noble gases become more pronounced at the MIR region. For Xe, the electron distribution at the MIR region is mainly located in the first and third quadrants and appears as a pronounced cross-like behavior, which is very different from the situation of the near-infrared region. For Mg, no study has been performed in the MIR region. How the knee structure...
and the correlated electron dynamics in elliptically and circularly polarized laser pulses depend on the wavelength remains unknown.

In this paper, employing the three-dimensional classical ensemble model, we systematically investigated strong-field NSDI of Mg atoms by intense laser fields with different wavelengths. The double ionization probability of Mg as a function of laser intensity with different wavelengths driven by elliptically and circularly polarized laser fields is calculated. It is shown that the double ionization yield decreases sharply and the knee structure finally disappears as the laser wavelength increases. We also analyze the wavelength dependence of CEMSs at different laser intensities. Interestingly, the correlated behavior in the CEMSs becomes more pronounced as the wavelength increases. In addition, we find that the recollision distance of two electrons in circularly polarized laser fields becomes smaller with the wavelength increasing.

II. METHOD

Due to the huge computational demand of numerically solving the time-dependent Schrödinger equation for multi-electron systems in strong laser fields, in the past decades numerous theoretical studies on NSDI have resorted to the classical models. It has been demonstrated that the classical models are very successful not only in explaining the experimental results, but also in predicting new phenomena in strong-field double ionization. For example, with the classical ensemble models, the repulsion behavior in the CEMSs for NSDI of He was well reproduced and explained. Based on the numerical results for a classical model, it was predicted that the electron pairs could be controlled to exhibit correlated or anticorrelated behavior with the two-color field, which was experimentally confirmed. Here, we employ the well-established three-dimensional classical ensemble model to study NSDI of Mg. In this model, the evolution of the two-electron system is determined by Newton’s equation of motion (atomic units are used throughout until stated otherwise),

\[
\frac{d^2 \mathbf{r}_i}{dt^2} = -\nabla[V_{ne}(\mathbf{r}_i) + V_{ee}(\mathbf{r}_1, \mathbf{r}_2)] - \mathbf{E}(t),
\]

where the subscript \( i \) is the label of the two electrons and \( \mathbf{r}_i \) is the coordinate of the \( i \)-th electron. \( \mathbf{E}(t) = \frac{E_0 f(t)}{\sqrt{1 + \varepsilon^2}} [\hat{x} \cos(\omega t) + \hat{y} \varepsilon \sin(\omega t)] \) is the electric field of the laser pulses, where \( E_0, \omega, \) and \( \varepsilon \) are the electric amplitude, the frequency, the ellipticity of the laser pulse, respectively. \( f(t) \) is the envelope of the laser pulse which has a trapezoidal shape with two-cycle turn on, six cycles at full strength, and two-cycle turn off. \( V_{ne}(\mathbf{r}_i) = -2/\sqrt{\mathbf{r}_i^2 + a^2} \) represents the ion-electron interaction potential, and the soft parameter \( a = 3.0 \) is introduced here to avoid autoionization. The range for the choice of the value is addressed in detail in Refs. 60 and 66. \( V_{ee}(\mathbf{r}_1, \mathbf{r}_2) = 1/\sqrt{(\mathbf{r}_1 - \mathbf{r}_2)^2 + b^2} \) is the potential for the electron-electron interaction, and parameter \( b \) is set to be 0.05. This parameter does not affect our results as long as it is small enough.

To obtain the initial condition, the ensemble is populated starting from a classically allowed position for the energy of \(-0.8 \) a.u., corresponding to the sum of the first and second ionization potentials of Mg. The available kinetic energy is distributed between the electrons randomly in momentum space. Then the system is allowed to evolve in the absence of the laser field for a sufficiently long time to obtain stable positions and momentum distributions. Once the initial ensemble is obtained, the laser field is turned on and all trajectories are evolved in the combined Coulomb and laser fields. We check the energies of the two electrons at the end of the laser field, and the double ionization (DI) event is determined if the energies of both electrons are positive, where the energy of each electron contains the kinetic energy, potential energy of the electron-ion interaction, and half electron-electron repulsion. In this study, the double ionization is identified as NSDI if recollision occurs during the ionization process. We mention that in the classical model, no tunneling occurs and the first ionization of the recollision electron ionizes over the suppressed potential barrier.

III. RESULTS AND DISCUSSIONS

Figure 1 depicts the double ionization probability of Mg as a function of laser intensity in linearly, elliptically (\( \varepsilon = 0.5 \)), and circularly (\( \varepsilon = 1 \)) polarized laser fields. For circularly polarized laser fields, the curve shows the distinct knee structure at 800 nm, consistent with experimental data. When

FIG. 1. The double ionization probability of Mg as a function of the laser intensity in (a) linearly, (b) elliptically (\( \varepsilon = 0.5 \)), and (c) circularly (\( \varepsilon = 1 \)) polarized laser fields. The vertical dashed lines indicate the intensity above which sequential double ionization becomes more prevalent.
the laser wavelength increases to 1600 nm, the knee structure completely vanishes, as shown in Fig. 1(c). The result is in agreement with the previous prediction by the semiclassical model, which forecasts that the knee structure disappears as the wavelength increases to 1500 nm. For elliptically polarized laser fields shown in Fig. 1(b), the curves also show the knee structure and the double ionization yield is higher than circularly polarized laser fields. The knee structure still exists at the wavelength of 1600 nm. In addition, Figs. 1(b) and 1(c) show that at the intermediate laser intensities $[2.0 \times 10^{13} \sim 7.0 \times 10^{13} \text{ W/cm}^2$ in Fig. 1(b) and $7.0 \times 10^{13} \sim 2.0 \times 10^{14} \text{ W/cm}^2$ in Fig. 1(c)], the double ionization probability reduces quickly as the laser wavelength increases. While for the linear laser, the yield only changes slightly, as shown in Fig. 1(a). When the laser intensity decreases to the lower intensity (below $2.0 \times 10^{13} \text{ W/cm}^2$), the curves get closer. We could expect that the curves may cross as the laser intensity decreases further. This behavior is understood as follows. The NSDI yield is mainly affected by two factors: the recollision energy and the recollision probability of the first ionized electron. At high laser intensities, the recollision energy is higher than the ionization potential of the second electron, and thus double ionization is restrained by the recollision probability. In elliptically and circularly polarized laser fields, longer wavelengths require a larger initial transverse momentum for the first tunneled electron to return back. Consequently, the double ionization yield decreases sharply with the laser wavelength increasing. For linearly polarized laser fields, the initial transverse momentum should be nearly zero for all of the wavelengths in linearly polarized laser fields, and thus the double ionization yield is not sensitive on the wavelength. While at low laser intensities, the recollision energy plays the vital role, which is higher for longer wavelengths. The combination of the two effects leads to the double ionization yield curves for different wavelengths approaching as the laser intensity decreases.

In order to explore more details of the correlated electron dynamics and its wavelength dependence for strong-field NSDI of Mg, we analyze the CEMSs from our numerical results. In the following, we separately study the correlated electron dynamics of NSDI in linearly, elliptically, and circularly polarized laser fields.

First, we focus on linearly polarized laser fields. In Fig. 2, we present the CEMSs for 800 nm and 2000 nm. At low laser intensities [Figs. 2(a) and 2(c)], for both 800 nm and 2000 nm laser fields, the population is mainly located in the second and fourth quadrants, showing a clear anticorrelated behavior. This behavior is similar to the results for NSDI of Ar at low laser intensities. It has been explained that at low laser intensities, NSDI occurs through recollision-induced excited states. Back tracing of the classical trajectories shows that the anticorrelation behavior for NSDI of Mg at the low laser intensity originates from this dynamics. In Fig. 3, we show two representative trajectories to provide an intuitive picture on the correlated and anticorrelated behaviors. Figures 3(a) and 3(b) show the time evolution of the energies and momenta of the two electrons.
of the two electrons for the two NSDI events. Figures 3(c) and 3(d) show the time evolution of the momentum parallel to the laser polarization direction of the corresponding trajectories. For the first one (left side), the time difference between ionization of the two electrons after recollision is about 0.35 T. In this event, the two electrons achieve final momenta with the same direction. For the other NSDI event (right side), the ionization time of the two electrons after recollision is about 1.25 T, and finally the two electrons achieve momenta with different directions.

At the higher laser intensity, the population is almost equally distributed in the four quadrants for the 800 nm case, as shown in Fig. 2(b). This is different from the case of He and Ar but similar to NSDI of Xe at 800 nm.38 At the 2000 nm laser field and the intensity of $1.0 \times 10^{13}$ W/cm$^2$, the distribution is mainly distributed in the first and third quadrants and presents an obvious valley with respect to the main diagonal, as shown in Fig. 2(d). This valley indicates that the two electrons seldom acquire the same final momentum. According to the recollision model, the maximal energy of the returning electron is $3.17 U_p$ ($U_p$ is the ponderomotive energy).9 At this laser intensity, the maximum recollision energy of $3.17 U_p = 0.43$ a.u. is just below the second ionization potential ($I_{p2} = 0.55$ a.u.) of Mg. In this case, the second electron is excited by recollision while the returning electron itself still stays in the continuous state. The excited electron is ionized by the laser field just before the first electric field peak after recollision. The ionization time difference of the two electrons leads to the two electrons acquire different final momenta,65 and thus the CEMS shows a valley behavior. We mention that this structure in the CEMS has been observed in NSDI of Ar by 800 nm laser pulses.36 Our calculations indicate that the electron correlation in NSDI of Mg becomes more pronounced as the laser wavelength increases.

We mention that in the classical model, the excited states of the atom are continuous. In quantum mechanism, the bound states are discrete. However, in the strong laser fields, these highly excited states are quasi-continuous, and thus the classical model could correctly catch most of the features in strong-field NSDI, even at these laser intensity ranges where the dynamics of the excited states plays important roles.

We turn to elliptically ($\varepsilon = 0.5$) polarized laser fields. Figure 4 shows the CEMSs of Mg in the direction along the major axis ($\hat{x}$ axis) of the polarization plane. Here we show the results for 800 nm and 1200 nm fields. The double ionization yield for longer wavelengths is very low, and the number of the collected NSDI events is not large enough.

\[ \text{FIG. 4. The correlated electron momentum distributions of Mg in the direction of the major elliptical axis ($\hat{x}$ axis) of elliptically polarized laser fields ($\varepsilon = 0.5$). The laser wavelengths are [a] and [b] $\lambda = 800$ nm and [c] and [d] $\lambda = 1200$ nm. The laser intensities are [a] and [c] $2.0 \times 10^{13}$ W/cm$^2$ and [b] and [d] $4.0 \times 10^{13}$ W/cm$^2$.} \]
for us to analyze the features in the CEMSs. For the 800 nm laser pulses, the distribution exhibits an anticorrelated behavior at the lower laser intensity of $2.0 \times 10^{13}$ W/cm$^2$ [Fig. 4(a)]. As the laser intensity increases, more electrons are distributed in the first and third quadrants, as shown in Fig. 4(b). For the 1200 nm fields, the distributions exhibit more prominent correlated behavior at both intensities, as shown in Figs. 4(c) and 4(d).

In order to explore the underlying dynamics, we trace the classical trajectories and perform statistical analysis.\textsuperscript{32,34,35,60} We find out the recollision time and the double ionization time for each NSDI event. Here, the recollision time is defined as the instant of the closest approach of the two electrons after departure of one electron from the core, and the double ionization time is defined as the instant when both electrons achieve positive energies after collision.\textsuperscript{33,34} Figure 5 shows the time delay between double ionization and recollision. The distributions show very different behaviors. For the 800 nm field, the main peak is located between 0.5 T and 1.0 T (T is the laser cycle) at $2.0 \times 10^{13}$ W/cm$^2$. At the higher laser intensity, the peak in this interval becomes weaker. At the 1200 nm fields, the distributions exhibit more prominent correlated behavior for both wavelengths, as shown in Fig. 5(c). At the higher intensity of $1.0 \times 10^{14}$ W/cm$^2$, the distribution is mainly located in the second and fourth quadrants and exhibits a clear arc-like shape for the 800 nm laser pulse.\textsuperscript{39} While for the 1000 nm laser pulse, the distribution exhibits a very strong correlated behavior. We mention that the CEMSs in the other direction (y-axis) of the laser polarization plane are the same as those in Fig. 7 due to the symmetry of the circular laser pulses.

In Fig. 8, we display the time delay between double ionization and recollision for circularly polarized laser fields. At the lower laser intensity of $5.0 \times 10^{13}$ W/cm$^2$, the peak distributions of the time delay are located around 0.5 T for both wavelengths, as shown in Fig. 8(a). A small difference is visible at the time delay larger than 0.6 T, where the tail of the distribution decreases slower for the 800 nm laser field. We divide the time delay distribution into two parts: the time delay less than 0.6 T and larger than 0.6 T and separately show the CEMSs. For the time delay less than 0.6 T, the events are almost uniformly distributed in four quadrants for both wavelengths, as shown in Figs. 9(a) and 9(c). While for the time delay larger than 0.6 T, due to the subtle difference in the delay time distribution, the faint difference in

FIG. 7. The correlated electron momentum distributions of Mg in circularly polarized laser fields. The laser wavelengths are [(a) and (b)] $\lambda = 800$ nm and [(c) and (d)] $\lambda = 1000$ nm. The laser intensities are [(a) and (c)] $5.0 \times 10^{13}$ W/cm$^2$ and [(b) and (d)] $1.0 \times 10^{14}$ W/cm$^2$.

FIG. 8. Time delay between double ionization and recollision for 800 nm and 1000 nm laser pulses with intensities of (a) $5.0 \times 10^{13}$ W/cm$^2$ and (b) $1.0 \times 10^{14}$ W/cm$^2$. and the corresponding CEMSs are shown in Fig. 6. It was shown that the distributions are mainly located in the first and third quadrants for the events with time delay less than 0.5 T [Figs. 6(a)–6(d)] and mainly located in the second and fourth quadrants for the events with time delay larger than 0.5 T [Figs. 6(e)–6(h)]. It indicates that in elliptically polarized laser fields, it is the time delay that determines the correlated behavior of the two electrons in the CEMSs, for both wavelengths. At the longer wavelength, due to the high returning energy, the second electron is ionized more quickly and thus electron pairs exhibit more pronounced correlation in the CEMSs.
FIG. 9. The correlated electron momentum distributions of Mg in circularly polarized laser fields at the laser intensity of $5.0 \times 10^{13}$ W/cm$^2$. The laser wavelengths are [(a) and (b)] 800 nm and [(c) and (d)] 1000 nm. [(a) and (c)] The distributions correspond to the events with time delay less than 0.6 T. [(b) and (d)] The distributions correspond to the events with time delay larger than 0.6 T.

The CEMSs for 800 nm and 1000 nm laser fields is distinguishable, as shown in Figs. 9(b) and 9(d). This difference of the time delay can be ascribed to the wavelength-dependent returning energy. Longer wavelength corresponds to higher returning energy which makes the two electrons ionize more quickly.\(^{35}\)

For the higher intensity of $1.0 \times 10^{14}$ W/cm$^2$, the time delay is much smaller than the lower laser intensity and the distributions become narrower, as shown in Fig. 8(b). To achieve more details, we also divide the total electron correlated momentum spectrum into two parts according to the time delay, as shown in Fig. 10. For the time delay less than 0.35 T, the spectrum exhibits a corrected behavior, as shown in Figs. 10(a) and 10(c). For the time delay larger than 0.35 T, more electron pairs distribute in the second and fourth quadrants, as shown in Figs. 10(b) and 10(d). For 800 nm, more electrons are distributed in the range of the time delay larger than 0.35 T, while more electrons are distributed in the range of the time delay less than 0.35 T for 1000 nm. As shown in Fig. 10(b), an arc-like shape appears in the second and fourth quadrants, and the structure is the same as that in Fig. 7(b). And the momentum distribution in Fig. 10(c) is similar to Fig. 7(d). This well exhibits that the time delay is responsible for the formation of the CEMS.

Finally, we show a very interesting issue explored by tracing the classical trajectories of the NSDI events. We record the recollision distance of two electrons which is defined as the closest approach of the two electrons during recollision, as displayed in Fig. 11. Figure 11(a) shows this distance for the 800 nm and 2000 nm linearly polarized laser fields. The peaks of the two distributions locate at the same distance of about 2.5 a.u. However, for circularly polarized laser fields, the recollision distance is smaller for the longer wavelength, as clearly shown in Fig. 11(b). This behavior could be ascribed to the wavelength-dependent Coulomb focusing effect\(^{69,70}\) and the initial velocity window for NSDI.\(^{15}\) This wavelength-dependent recollision distance could partially account for the difference in the CEMSs for different wavelengths of linearly and circularly polarized laser pulses. For the longer wavelength, the recollision distance is smaller and thus the returning electron transfers energy to the bounded electron more efficiently. This leads to double ionization that occurs more quickly for the longer wavelength.

FIG. 10. The correlated electron momentum distributions of Mg in circularly polarized laser fields at the laser intensity of $1.0 \times 10^{14}$ W/cm$^2$. The laser wavelengths are [(a) and (b)] 800 nm and [(c) and (d)] 1000 nm. [(a) and (c)] The distributions correspond to the events with time delay less than 0.35 T. [(b) and (d)] The distributions correspond to the events with time delay larger than 0.35 T.

FIG. 11. The closest distance at the time of recollision in (a) linearly polarized laser fields and (b) circularly polarized laser fields.
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IV. CONCLUSION

We investigate the wavelength dependence of NSDI of Mg by linearly, elliptically, and circularly polarized laser fields. The curves of doubly charged ion yield versus laser intensity show the clear knee structure in elliptically and circularly polarized laser fields. At the relatively high laser intensity, the double ionization yield decreases quickly as the wavelength increases and approaches the low intensity regime. This is due to the competition between the diffusion of the returning electron wavepacket and the recollision energy. Our calculations show that the correlation in the CEMSs becomes more pronounced as the wavelength increases. Interestingly, we find that the recollision distance in the circularly polarized laser field becomes smaller as the wavelength increases, indicating more efficient energy transfer during recollision. The higher recollision energy and the more efficient energy transfer during recollision at longer wavelengths give rise to more pronounced correlation in CEMSs at longer wavelengths of circular laser pulses.