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Research paper

# Enhancement of nonsequential double ionization in counter-rotating two-color circularly polarized laser fields

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ABSTRACT

Nonsequential double ionization is theoretically investigated in the counter-rotating two-color [frequencies $(\omega_1, \omega_2)$] circularly polarized laser fields with classical ensemble approach. It is found that remarkable ionization enhancement is obtained by adjusting the relative electric field amplitude ratios ($\gamma_E$). The result indicates that there is a broader steerable range of $\gamma_E$ to obtain higher ionization enhancement for $\omega_2 = 3\omega_1$ than that for $\omega_2 = 2\omega_1$. Through the analysis of trajectories, the recollision picture for $\omega_2 = 3\omega_1$ is complicated leading to slow change of double ionization, which is benefit to gentle control mechanism. However, the recollision ionization channel is easily distinguishable and sensitive to control double ionization in $\omega_2 = 2\omega_1$ laser fields and the travel time can be controlled in the order of magnitude of 100 attoseconds.

## 1. Introduction

In the past two decades, nonsequential double ionization (NSDI) has exerted a great attention, because the NSDI processes reflect the character of the electron correlation. NSDI of atoms that are exposed to intense laser fields has been studied numerous theoretically and experimentally. Experimentally, the early works began with measurements of the total doubly charged ion yield as a function of the laser intensity. The well-known general feature observed in the NSDI data is the appearance of a characteristic "knee structure": as the laser intensity increases, the double ionization is sequential. When the laser intensity is increased to a certain value, the double ionization probability presents a platform area and in high laser intensity area, the probability of double ionization accords with the ionization curve predicted by ADK theory, and it forms a "knee structure" as a whole [1,2]. This dramatic enhancement of doubly charged ion yields has been explained by the semiclassical three-step model [3]: the Coulomb potential is distorted by the intensive laser field and hence the single electron is liberated by tunnel ionization. After that the electron is accelerated by the laser field and may re-encounter the parent ion and rescatter, resulting in the ionization of the second electron. This NSDI processes perform the enhancement of the double-ionization (DI) yield.

The ionization enhancement is significantly sensitive to the laser intensity, laser polarization and laser wavelength. It is also commonly accepted that NSDI is significantly suppressed for helium and xenon in circularly polarized (CP) fields in contrast to linearly polarized (LP) fields because the recollision with the parent ion is greatly suppressed in CP laser fields [4,5]. Several studies have revealed that Mg atoms driven by CP laser fields seem to be an exception [6–10]. A velocity window for the recollision demonstrates the important role of the chaotic return electrons in triggering NSDI and the phase diagram for the NSDI unveil NSDI area related to first ionization energy and laser wavelength [8]. The DI probabilities as a function of laser intensity for Ar at 400 nm, 436 nm and 541 nm uncover that as the wavelength of the driving laser pulses decreases, the ionization probability in the knee region increases [11].

Recently, counter-rotating two-color circularly polarized (TCCP) laser fields have been used to explore new capabilities in high harmonic generation (HHG) [12–14]. It can also drive circularly polarized extreme ultraviolet (EUV) [15,16] and soft-X-ray beams [17] in such fields. It supplies a new insight into the ultrafast electron dynamics. The fact is that using counter-rotating TCCP laser fields, electrons can be driven in a two-dimensional (2D) plane and the tunneling and electron-ion rescattering processes occur at different angles [18]. Similarly, the novel phenomenons of strong-field ionization (SFI) can be also observed in TCCP [19,20]. High-energy rescattered electrons were observed in Ref. [19]. The theoretical and experimental results of NSDI in TCCP laser fields were

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performed in Refs. [21–23]. They found that the NSDI yield was significantly suppressed in co-rotating fields but enhanced in counter-rotating fields. The ionization enhancement depended on the two-color laser field intensity ratio. Additionally, the ionization enhancement yield was up to 700% compared with co-rotating TCCP fields. This effect was commonly for noble gas [24].

In this paper, we demonstrate the NSDI processes in the two-color counter-rotating circularly polarized laser fields by using the classical ensemble approach [25–29]. A large quantity of ensembles of randomized model atoms is used to simulate some quantum effects, which results in excellent qualitative agreement with experimental observation. This theoretical calculations can provide clear insight to the underlying processes and guide future experiments. Through the analysis of electronic trajectories, we found that NSDI dependents on the relative electric field amplitude ratio. Several controllable laser parameters are used to control the ionization enhancement.

Atomic units are used in this paper, if no particular explanation is given.

## 2. Theoretical methods

In our simulation, the classical ensemble approach is employed which is proved to be useful in strong field ionization and especially in e-e correlation [25–29]. In order to maintain the stable initial ensemble, the initial positions and momenta of electrons are randomized which ensures the argon bound state energy and $10^7$ randomized ensemble trajectories are used. We use the 3D model of Ar by the following equation in the absence of external fields

$$H_0 = \sum_{\mathrm{i}}^{2}\left(\frac{|\boldsymbol{p}_{\mathrm{i}}|^2}{2} - \frac{2}{\sqrt{|\boldsymbol{r}_{\mathrm{i}}|^2 + a^2}}\right) + \frac{1}{\sqrt{|\boldsymbol{r}_1 - \boldsymbol{r}_2|^2 + b^2}}, \tag{1}$$

where $\boldsymbol{p}_{\mathrm{i}}$ is the electron momentum, and $\boldsymbol{r}_{\mathrm{i}}$ is the electron coordinate. The subscript i = 1, 2 labels the two electrons. In Eq. (1), two terms in bracket represent kinetic energy and soft-core Coulomb potential between the ion core and the ith electron, final term is the potential between the two electrons. The total energy of the Hamiltonian system whose negative value is equal to the sum of first and second electronic ionization potential. The Coulomb potential with softening terms is modified to avoid autoionization. The ionic and $e-e$ soften parameters are set to be $a = 1.5$ and $b = 0.1$, which models a real Ar atom. The Hamiltonian system (1) is solved numerically by symplectic method, which can preserve the symplectic structure of the system and be suitable for long-time calculations.

We consider DI by a counter-rotating TCCP laser field, the Hamiltonian is written by

$$H = H_0 + (\boldsymbol{r}_1 + \boldsymbol{r}_2)\cdot \boldsymbol{E}(\boldsymbol{t}), \tag{2}$$

and the electric field is

$$\boldsymbol{E}_1 = \frac{E_0}{1+\gamma_E} f(t)[\cos(\omega_1 t + \phi_0)\widehat{\boldsymbol{y}} + \sin(w_1 t + \phi_0)\widehat{\boldsymbol{z}}], \tag{3}$$

$$\boldsymbol{E}_2 = \frac{\gamma_E E_0}{1+\gamma_E} f(t)[\cos(\omega_2 t + 2\phi_0)\widehat{\boldsymbol{y}} - \sin(w_2 t + 2\phi_0)\widehat{\boldsymbol{z}}], \tag{4}$$

where $E_0$ is the combined electric field amplitude and the combined electric field is written as $\boldsymbol{E} = \boldsymbol{E}_1 + \boldsymbol{E}_2$. $\omega_1 = 0.057$, which corresponds a wavelength of 800 nm. $\omega_2 = 2\omega_1$ or $\omega_2 = 3\omega_1$. $\gamma_E$ is the electric field amplitude ratio between the second or third harmonic laser pulse and the fundamental laser pulse (the corresponding light intensity ratio $\gamma_I = \gamma_E^2$), and the sine-squared shape pulse envelope $f(t)$ and carrier-envelope phase (CEP) $\phi_0 = 0$ are used.

## 3. Numerical results and analysis

Fig. 1(a) demonstrates the DI probability of Ar atom versus laser intensity and the electric field amplitude ratio $\gamma_E$ for $\omega_2 = 2\omega_1$. It indicates that the DI probability for the larger field strength obeys ADK theory [30], which is the SDI mechanism, such as the dark red region shown in Fig. 1(a). When $\gamma_E$ comes to zero, the DI probability decreases sharply and the NSDI is significantly suppressed [8]. When $\gamma_E$ goes to ten, the yellow area indicates that there is also a knee structure [11]. In the sensitive area of $\gamma_E$ from 1.3 to 6.0, DI probability is controllable which has a common ionization enhancement.

The DI probability versus laser intensity and the electric field amplitude ratio $\gamma_E$ for $\omega_2 = 3\omega_1$ is shown in Fig. 1(d). There is a same tendency in $\omega_2 = 3\omega_1$ laser fields compared with that in $\omega_2 = 2\omega_1$ laser fields at the beginning of $\gamma_E$. However, there is a remarkable difference for large $\gamma_E$ where the wavelength comes to 267 nm. According to the picture illustrated in Ref. [11], there is a significant knee structure for Ar near 267 nm wavelength. It is indicated that there is a broader steerable range of $\gamma_E$ to obtain higher ionization enhancement for $\omega_2 = 3\omega_1$ than that for $\omega_2 = 2\omega_1$.

We pick up the prominent DI probability with two typical $\gamma_E$ ($\gamma_E = 1.6$ and $\gamma_E = 8.0$) for $\omega_2 = 2\omega_1$ laser fields in Fig. 1(b) and that with two typical $\gamma_E$ ($\gamma_E = 2.5$ and $\gamma_E = 8.0$) for $\omega_2 = 3\omega_1$ laser fields in Fig. 1(e) to illustrate the classical knee structure. $\gamma_E = 1.6$ and $\gamma_E = 2.5$ represent typical smaller $\gamma_E$ and $\gamma_E = 8.0$ is the typical representative selected from larger $\gamma_E$. The experimental observations of single- and double-ionization yields present the kneelike structures with different values of $\gamma_E$ in $\omega_2 = 2\omega_1$ laser fields. We theoretically simulate the kneelike shapes which are similar with the experimental results [22,23] in Fig. 1(b). We also find that when the value of $\gamma_E$ is larger, the knee structure will be significantly weakened. However, the significant differences are that for larger $\gamma_E$, the platform of the knee structure is higher in $\omega_2 = 3\omega_1$ laser fields than that in $\omega_2 = 2\omega_1$ laser fields.

In order to provide intuitive insights to the NSDI, we present the DI probability as a function of $\gamma_E$ for $\omega_2 = 2\omega_1$ in Fig. 1(c) and $\omega_2 = 3\omega_1$ in Fig. 1(f) at $I_0$ = 0.08 PW/cm$^2$, 0.1 PW/cm$^2$, 0.12 PW/cm$^2$, 0.14 PW/cm$^2$ and 0.16 PW/cm$^2$. The DI probabilities increase first and then decrease as the value of $\gamma_E$ increases. At the ends of $\gamma_E$, the DI probabilities tend to small which is in agreement with that in circularly polarized laser fields. The maximal enhancement of DI probabilities is around $\gamma_E = 1.6$ for $\omega_2 = 2\omega_1$ (which has been illustrated in Refs. [21–23]) and around $\gamma_E = 2.5 \sim 3.0$ for $\omega_2 = 3\omega_1$. Especially, our results in Fig. 1(c) are qualitatively in good accord with the experimental and theoretical results [21–23,29,31].

As shown in Fig. 1(c) or (f), the tendency of DI probability and the maximum ionization enhancement position at different laser intensities are similar, which demonstrates that the laser intensity has little effect on the $\gamma_E$ control mechanism. In essence, the shape of laser field (i.e. the value of $\gamma_E$) is more important to the ionization enhancement or suppression. In addition, the DI probability decreases much faster when $\gamma_E$ decreases than that when $\gamma_E$ increases. It indicates that the laser shape for smaller $\gamma_E$ is very sensitive to DI control. This provides experimentalists with sensitive and gentle controls. But, the gentle control is more obvious in $\omega_2 = 3\omega_1$ laser fields than that in $\omega_2 = 2\omega_1$.

In order to further illustrate the NSDI dependence on the value of $\gamma_E$, we exhibit the electron return energy which includes kinetic energy only to the parent ion in Fig. 2. We calculate the electron returning energy spectra using semi-classical approach [22,32]. In general, the electrons are driven back to the parent ion with energies below the second ionization potential of Ar (1.01 a.u.),