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## Research paper Recollision induced excitation-ionization with counter-rotating two-color circularly polarized laser field

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## ABSTRACT

Nonsequential double ionization of Ar by a counter-rotating two-color circularly polarized laser field is theoretically investigated. At the combined intensity in the "knee" structure range, the double ionization occurs mainly through recollision induced excitation followed by subsequent ionization of Ar<sup>++</sup>. By tracing the history of the recollision trajectories, we explain how the relative intensity ratio of the two colors controls the correlated electron dynamics and optimizes the ionization yields. The major channels contributing to enhancing the double ionization are through the elliptical trajectories with smaller travel time but not through the triangle shape or the other long cycle trajectories. Furthermore, the correlated electron dynamics could be limited to the attosecond time scale by adjusting the relative intensity ratio. Finally, the double ionization from doubly excited complex at low laser intensity is qualitatively discussed.

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With the progress of the ultrafast strong laser technology, the possibility of visualizing and steering electrons on the attosecond time scale and subnanometer dimension is realized. One can get more and more information about atomic and molecular structure or electron-electron correlation by analyzing the light-matter interaction as well as the strong field phenomena, such as high harmonic generation (HHG) [1], nonsequential double ionization (NSDI) [2] and above threshold ionization (ATI) [3]. Besides simple linearly polarized (LP) fields, a laser pulse composed of two harmonic colors has been widely used to explore those fundamental phenomena in recent years. For example, the orthogonally polarized two-color (OTC) laser fields allow us to realize subcycle control of electron dynamics in NSDI [4–6]. The counter-rotating two-color circularly polarized (TCCP) laser fields have also attracted much attention due to achieving bright circularly polarized HHG [7-11] and low-energy structures (LES) [12] as well as controlling electron-ion rescattering [13]. The underlying mechanism in those works can be explained by three-step model proposed by Corkum [14], in which the fact that ionizing electrons could reencounter their parent ion also shows the possibility in controlling NSDI by this kind of laser pulse.

Very recently, the theoretical work by Chaloupka and Hickstein [15] as well as the experimental observations by Mancuso et al. [16] and by Eckart et al. [17] show that the NSDI yield depends

\* Corresponding author. *E-mail address:* liuxs@jlu.edu.cn (X.-S. Liu). strongly on the relative intensity of the two colors. Meanwhile a beam of nearly monoenergy recolliding electrons can be generated. In Refs. [15,16], the NSDI yield is controlled by changing the relative intensity ratio or is suppressed by co-rotating TCCP laser fields. The observation using an electron-ion coincidence spectrometer in Ref. [17] shows that the recollision induced excitation process plays a key role at some certain intensities (at the platform of the "knee" structure). The reason is that there are no recolliding electrons with energies above the  $Ar^+$  ionization threshold. However, the recollision energies can be larger than the excitation energy of  $Ar^+$ . Meanwhile, recollision impact ionization (RII) channel may be suppressed. In addition, electron-emission dynamics on the attosecond time scale is also recorded in the counter-rotating TCCP laser fields.

The counter-rotating TCCP fields with so many tunable parameters (e.g. relative intensity, relative phase, and polarization) enable us to shape the laser pulse and control electron ionization dynamics artificially. In this paper, we investigate the relative intensitydependent NSDI process using 10 optical cycles counter-rotating TCCP laser fields. In recollision induced excitation with subsequent ionization (RESI) [18] of Ar atom, electron-emission dynamics could be limited to the attosecond time scale by changing the relative intensity ratio of the two colors. The major recollision events in larger range of relative intensity ratio are through the elliptical shorter recollision trajectories (written as SRT) [see, e.g., Fig. 6(a) or Fig. 6(e)] by nearly monoenergy returning electrons and spend shorter time (about 450~1300 attoseconds). The longer recollision







trajectories are written as LRT accordingly, which are more complicated due to too many cycles traveled. Our results indicate that changing relative intensity can simplify the RESI picture through the SRT and optimize the NSDI yield.

The 3D classical ensemble method is employed for simulation, which is proved to be useful in investigation of the electron correlation in double ionization under strong field [19–26] and is successful in simulating the NSDI in the two-color circularly fields (see, e.g., Refs. [15,16]). Our simulated results agree well with the experimental measurements (see Fig. 1).

In classical ensemble method, the softened Coulomb potential is employed. The ionic and *e-e* soften parameters is set to be a = 1.5and b = 0.05, respectively, which could avoid auto-ionization. In our simulations, an ensemble of ten million is generated by randomly assigning initial positions and momenta to each electron, while ensuring that the total energy of the system is equal to the ground state energy for argon. To obtain the stable initial ensemble, the system is allowed to evolve sufficient long time in the absence of the external laser field by classical equations of motion.

In order to have the maximum combined electric field amplitude  $E_0$  corresponded to a fixed laser intensity  $I_0$  at different relative intensity, the fundamental ("red") and the second harmonic ("blue") laser pulses are expressed as

$$\boldsymbol{E}_{\mathbf{r}} = \frac{E_0}{1 + \gamma_{\rm E}} f(t) [\cos\left(\omega_{\rm r} t + \phi_0\right) \hat{\boldsymbol{y}} + \sin\left(\omega_{\rm r} t + \phi_0\right) \hat{\boldsymbol{z}}],\tag{1}$$

$$\boldsymbol{E}_{\mathbf{b}} = \frac{\gamma_{\mathrm{E}} E_{0}}{1 + \gamma_{\mathrm{E}}} f(t) [\cos\left(\omega_{\mathrm{b}} t + 2\phi_{0}\right) \hat{\boldsymbol{y}} - \sin\left(\omega_{\mathrm{b}} t + 2\phi_{0}\right) \hat{\boldsymbol{z}}], \tag{2}$$

Here,  $\gamma_{\rm E}$  is the electric field amplitude ratio between the second harmonic laser pulse and the fundamental laser pulse (the intensity ratio can be written as  $\gamma_{\rm I} = \gamma_{\rm E}^2$  accordingly). Naturally, the combined electric field is written as  $\mathbf{E} = \mathbf{E}_{\rm r} + \mathbf{E}_{\rm b}$ .  $\omega_{\rm r} = 0.0584$  a.u. and  $\omega_{\rm b} = 0.117$  a.u. correspond to the wavelength of the fundamental (780 nm) and second harmonic (390 nm) fields. f(t) is the sine-squared shape pulse envelope with random carrier-envelope phase (CEP)  $\phi_0$ .

We plot the double ionization (DI) probability of Ar atom versus intensity with three typical electric field amplitude ratio  $\gamma_E = 0.8, 1.4, 3.3$  in the counter-rotating TCCP fields and the circular polarized (CP) laser fields [Fig. 2(a)]. Generally, the NSDI will be significantly suppressed in the CP laser field and DI probability agrees with the ADK theory [27]. However, Fig. 2(a) indicates that the DI probability curves in counter-rotating TCCP fields show a prominent "knee" structure. NSDI becomes main mechanism in this range of intensity. Fig. 2(b) shows the relative yield of double to single ionization versus  $\gamma_E$  at 0.4 PW/cm<sup>2</sup>. It reaches maximum at about  $\gamma_E = 1.4$ , and tends to become zero if the ratio is too large or too small because the combined fields will tend to become CP laser fields.

In order to illustrate how the relative intensity controls the NSDI and optimizes the yields, we show the dependence of the return energy on ionization and return time by the three step model with  $\gamma_E = 2.0$  and 1.6 at  $I_0 = 0.4$  PW/cm<sup>2</sup> (Fig. 3). The similar analysis always used in studying the time-frequency distributions of HHG [28–30]. The wave curves show the combined electric field (normalized in the region marked by the yellow dashed lines) for two optic cycles (o.c.) of the fundamental laser. We have the 2N/6 o.c. and (2N + 1)/6 o.c. (N = 0, 1, 2...) been the maxima and minima of the electric field, respectively. Ionization time range just covered 2/6 o.c. (shown by the yellow range of the electric field curves) is taken into account due to the periodic equivalency.

Fig. 3 shows that the electron ionized just after the peak of the electric field can return with a larger energy. The return energy is below the  $Ar^+$  ionization threshold (1.01 a.u.) but more than the relevant excitation energy (0.63 a.u.), which corresponds to the SRT and shows an elliptical shape [see Fig. 6(a) or Fig. 6(e)]. Meanwhile, this range of return energy shows that NSDI process is more likely to occur through RESI but not RII.

The travel time of electrons (defined as  $\Delta \tau$ ) through SRT is approximately smaller than 3/6 o.c. for large range of  $\gamma_{\rm E}$ . All the other kinds of recollision trajectories with longer travel time are defined as the LRT. However, the LRT have little contributions to



**Fig. 1.** The relative yield of double to single ionization versus the field ratio and electron momentum distributions for double ionization for a field ratio of 1.42 at a combined intensity of 0.45 PW/cm<sup>2</sup>. The first row shows the simulated results; while the second row shows the experimental measurements, which are from Fig. 1 and Fig. 3 of Ref. [17]. All of the parameters used in calculation are the same as experimental conditions.

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