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# Attosecond control of correlated electron dynamics in strong-field nonsequential double ionization by parallel two-color pulses

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## ABSTRACT

We theoretically investigate the correlated electron dynamics in strong-field nonsequential double ionization (NSDI) of Ar by the parallel two-color pulses consisting of 800- and 400-nm laser fields with the semiclassical ensemble model. Our calculations show that the momentum distributions of Ar<sup>2+</sup> in the direction parallel to the polarization of laser field exhibit the single- or double-hump structure, depending on the relative phase of the two-color fields. Back analysis of the NSDI trajectories reveals that recollision of returning electron is well confined in a time window of several attoseconds, and the position of which can be controlled with attosecond precision by changing the relative phase of the two-color fields. Thus, the recollision energy is accurately controlled with the two-color fields. As a consequence, the time delay between the double ionization and recollision changes with the relative phase, resulting in the relative-phase-dependent ion momentum distributions. More interestingly, the different peaks of ion momentum spectra result from the NSDI events with different time delays between double ionization and recollision. Thus, the corresponding correlated electron dynamics for different peaks in the ion momentum distributions can be directly determined in the two-color fields.

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## 1. Introduction

Tunneling of atoms or molecules in strong laser field usually is accompanied by recollision, where the tunneling electron is accelerated in the laser field and then returns back to the parent ion when the electric field of the driving laser pulse changes its direction [1,2]. This recollision leads to various interesting phenomena, such as high harmonic generation (HHG) [3–5], high-energy above-threshold ionization (HATI) [6–10], and nonsequential double ionization (NSDI) [11–14]. Among these phenomena, NSDI has attracted special attention due to the highly correlated behavior of the ionized electron pairs during the inelastic recollision [15]. In past decades, a great number of experimental [16–20] as well as theoretical studies [21–32] have been performed to explore the correlated dynamics of the electron pairs in NSDI.

Previous studies have shown that the correlated electron dynamics in NSDI is rather complicated and sensitively depends on the driving laser pulses. Generally, NSDI could occur through two different pathways, recollision impact ionization (RII) and recollision-excitation with subsequent field ionization (RESI) [33].

At low laser intensities where the maximum recollision energy of the first electron is not enough to ionize the second electron directly, NSDI occurs through the RESI pathway. At high laser intensities, the recollision energy of the first electron is higher than the ionization potential of the second electron and thus double ionization can occur through the RII pathway. Usually, in the NSDI experiments the recollision energy of the returning electron spans over a wide range and thus both RII and RESI pathways make contribution, complicating the analysis of the dynamics in NSDI. Additionally, in the multi-cycle laser pulses, the multiple-recollision process also contributes to NSDI [34,35]. Moreover, the multiple-returning-recollision process, where recollision occurs at the later other than the first returning of the tunneled electron, has significant contribution to the total NSDI yields, especially in the mid-infrared laser fields [36–38]. The combined contribution of these processes hampers a full exploration of the detailed microscopic dynamics in NSDI. Revealing and ultimately controlling the correlated electron dynamics are the central tasks in current studies on strong-field NSDI.

With the development of the laser technology, ultrashort few-cycle laser pulses with stabilized carrier-envelope phase are widely available in laboratory [39]. In few-cycle pulses, the multiple-recollision and multiple-returning-recollision processes are

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strongly suppressed and the recollision are well confined in single optical cycle [40]. Thus the few-cycle pulses enable one to study the microscopic electron dynamics of NSDI in a much more clean way [41]. Additionally, in the multi-cycle pulses, there are considerable NSDI events occurring through the RESI pathway with a time delay longer than one optical cycle between double ionization and recollision [42,43]. While in the few-cycle pulses, this time delay is well within one optical cycle. Thus, the subcycle electron dynamics can be revealed with the few-cycle pulses. Taking these advantages of the few-cycle pulses, for example, the decay dynamics of the doubly excited state formed by recollision in NSDI is unambiguously traced [44]. The subcycle ionization dynamics of RESI in the few-cycle is clearly visualized and the asymmetric energy sharing during recollision is also revealed [41]. Moreover, the few-cycle pulse provides a feasible way to control NSDI. It has been reported that the ion momentum distributions as well as the correlated electron momentum spectra can be manipulated by changing the carrier-envelope phase of the few-cycle pulses and more details of the correlated electron dynamics have been uncovered [40,45]. Recently, control of NSDI with the few-cycle pulses has draw extensive attentions.

The two-color laser pulse is another powerful tool in controlling the recollision processes in strong laser fields [46,47]. Compared with the few-cycle pulses, it has more parameters for control, for example, the relative phase, relative intensity, polarizations et al. The scheme of two-color pulses has been widely employed to control the electron dynamics in HHG and ATI [48,49]. Recently, the two-color field has also been used to control the electron correlations in strong-field NSDI [50]. For example, with the orthogonal two-color pulses, the electron pairs can be controlled to exhibit correlated or anticorrelated behaviors [51]. With the circularly polarized two-color laser pulses, the double ionization yields have been manipulated by adjusting the relative intensity ratio of the two fields [52,53]. In this paper, using a semiclassical ensemble model, we theoretically study the correlated electron dynamics in the parallel two-color fields consisting of an 800-nm and a 400-nm laser pulses. We focus on the RESI pathway of NSDI. Our results show that the subcycle decay process of the excited states induced by recollision in RESI pathway can be controlled by changing the relative phase of the two-color fields. Back analysis of the NSDI trajectories shows that this is based on the attosecond control of the recollision time and thus the recollision energy of the tunneled electron. The controlling of the subcycle decay process results in the relative phase dependence of the experimentally observable ion momentum distributions. Interestingly, in the ion momentum distributions, different peaks originate from the NSDI events with different time delays between double ionization and recollision. Thus, the correlated electron dynamics of NSDI can be directly judged from the observable ion momentum distributions.

## 2. Method

Accurately describing NSDI in strong laser fields needs numerically solving the time-dependent Schrödinger equation. However, for the two- and multi-electron systems, the computational demand of this method is huge [21,54,55]. In the past decades numerous studies have resorted to the classical and semiclassical models, which have been very successful in explaining various phenomena in NSDI, for example, the V-like [27,56,57] and cross-like structures [41,58] in the correlated electron momentum spectra, and the anticorrelation of the electron pairs in low laser intensities [59]. The (semi) classical models are also successful in predicting phenomena in strong-field NSDI [51,60,61]. Moreover, with the (semi) classical model, the electron dynamics in NSDI can be intuitively revealed by back tracing the NSDI trajectories

[29,62,63]. Therefore, here we employ the semiclassical ensemble model [27,64] to investigate the correlated electron dynamics of NSDI by the parallel two-color pulses. In the semiclassical model, the first electron is ionized through tunneling with the rate given by tunneling theory [65]. The subsequent evolutions of the tunneled and the bound electrons are described by the Newton's classical motion equation (atomic units are used throughout until stated otherwise):

$$\frac{d^2 \mathbf{r}_i}{dt^2} = -\nabla[V_{ne}(\mathbf{r}_i) + V_{ee}(\mathbf{r}_1, \mathbf{r}_2)] - \mathbf{E}(t), \quad (1)$$

where the indices  $i = 1, 2$  refer to the first and the second electrons, respectively.  $V_{ne}(\mathbf{r}_i) = -2/\sqrt{\mathbf{r}_i^2 + a^2}$  and  $V_{ee}(\mathbf{r}_1, \mathbf{r}_2) = 1/\sqrt{(\mathbf{r}_1 - \mathbf{r}_2)^2 + b^2}$  represent the ion-electron and electron-electron coulomb interaction potentials, respectively. A soft parameter  $a$  is employed to avoid autoionization of the two-electron system [56]. Here we set  $a = 1.5$ . The parameter  $b$  in the electron-electron interaction potential is not important as long as it is small enough. Here we set  $b = 0.01$ . The electric field of the parallel two-color pulse is written as  $\mathbf{E}(t) = f(t)[E_0 \cos(\omega t)\hat{\mathbf{x}} + \sqrt{\varepsilon}E_0 \cos(2\omega t + \Delta\phi)\hat{\mathbf{y}}]$ , where the pulse envelope  $f(t)$  has a constant amplitude for the first eight cycles and is linearly turned off with a two-cycle ramp.  $E_0$  is the amplitude of the 800-nm field and  $\varepsilon$  is the intensity ratio of the 800-nm and 400-nm fields.  $\Delta\phi$  is the relative phase between the 400-nm and 800-nm fields.  $\omega$  is the frequency of the 800-nm laser field.

The initial conditions of the two electrons for Eq. (1) are obtained as follows. The first tunneling electron has zero parallel (with respect to the direction of electric field) velocity and a Gaussian-like transverse velocity distribution. The weight of each trajectory is  $w(t_0, v_{\perp 0}) = w(t_0)w(v_{\perp 0})$  [64], in which

$$w(t_0) = \frac{2(2I_{p1})^{1/2}}{|\mathbf{E}(t_0)|} \frac{1}{\sqrt{2I_{p1}}} \exp\left[\frac{-2(2I_{p1})^{3/2}}{3|\mathbf{E}(t_0)|}\right], \quad (2)$$

$$w(v_{\perp 0}) = \frac{1}{|\mathbf{E}(t_0)|} \exp\left[-\frac{v_{\perp 0}^2(2I_{p1})^{1/2}}{|\mathbf{E}(t_0)|}\right], \quad (3)$$

where  $w(t_0)$  is the instantaneous tunneling probability and  $w(v_{\perp 0})$  is the distribution of the initial transverse momentum  $v_{\perp 0}$  of the first electron. For the second electron, the initial position and momentum are depicted by microcanonical distribution of ground state energy of  $\text{Ar}^+$ . In our calculations, the first and second ionization potentials are chosen as  $I_{p1} = 0.58$  a.u. and  $I_{p2} = 1.01$  a.u., respectively, to match those of Ar. Several millions weighted classical two-electron trajectories are traced from the tunneling moment  $t_0$  to the end of the pulse. The double ionization events are determined if the energies of both electrons are positive after the laser field is turned off.

## 3. Results and discussions

We first show the ratio of  $\text{Ar}^{2+} : \text{Ar}^+$  as a function of the relative phase in Fig. 1(a). Here we consider three cases:  $I_{800} = 1.0 \times 10^{14}$  W/cm<sup>2</sup> with the intensity ratio  $\varepsilon = 0.1$  (i),  $\varepsilon = 0.2$  (ii) and  $I_{800} = 0.6 \times 10^{14}$  W/cm<sup>2</sup> with  $\varepsilon = 0.1$  (iii). It shows that the ratio of  $\text{Ar}^{2+} : \text{Ar}^+$  exhibits an oscillating behavior with a period of  $\pi$  and this behavior is similar for these three cases. It indicates that NSDI has indeed been controlled by the linear two-color laser fields. More information of the two-electron dynamics can be obtained from the momentum distribution. In Fig. 1(b)–(d), we show the momentum distributions of the ion along the laser polarization direction as a function of the relative phase. In Fig. 1(b), the

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