

Controlling nonsequential double ionization mechanisms of argon atoms by parallel two-color laser pulses

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ABSTRACT

Introduction: Recently, physicists have been interested in the nonsequential double ionization (NSDI) process since it provides information about the interaction between electrons in the atomic and molecular shells through the recollision process. Technically, the development of near-infrared laser pulses has made them one of the most useful tools for studying the electron correlation effect in atoms and molecules. Furthermore, the parallel two-color (PTC) laser pulse is a useful instrument for controlling the process of recollision in a strong laser field. In this paper, with the aim of investigating the role of ionization mechanisms governing the NSDI of argon atoms, we use a PTC laser pulse with wavelengths of 2400 nm and 1200 nm to control the motion of two electrons. We investigate the contribution of ionization mechanisms as well as the correlation dynamics of two electrons upon the change in relative phase between the two component lasers from 0 to 2π . **Method:** The two-dimensional classical ensemble model is used to simulate the NSDI process, in which the motion of two electrons is governed by Newton's law. **Results:** The correlated two-electron momentum distribution, recolliding time, and energy distribution after the returning electron's recolliding moment are extraordinarily sensitive to the relative phase of the two lasers. To modulate the ionization mechanisms, the relative phase between two lasers is tuned. **Conclusion:** The results demonstrate that parallel two-color laser pulses are an effective method of controlling the correlated electron dynamics in nonsequential double ionization.

Key words: Nonsequential double ionization, correlated two-electron momentum distribution, parallel two-color laser pulses, ionization mechanism

INTRODUCTION

The interaction process between the intense laser field (10^{13} – 10^{16} W/cm²) and atoms/molecules results in a number of fascinating physical phenomena, including the generation of high-order harmonics (HHG)^{1–3}, above threshold ionization (ATI)^{4,5} and double ionization (DI). There are two types of DI processes: sequential double ionization (SDI)^{6–8} and nonsequential double ionization (NSDI)^{9–11}. Physicists are particularly interested in the NSDI process because it provides information about the interaction between electrons in the atomic and molecular shells via the recollision process. Based on the energy of the two electrons immediately after the moment of recollision, the NSDI process occurs through four different mechanisms: recollision impact ionization (RII), ionization mechanism recollision-induced excitation with subsequent ionization (RESI), exchange-state ionization (ESI), and recollision-doubly induced excitation with subsequent ionization (RDESI)^{12–14}.

The laser wavelength is a crucial parameter for the recollision process because it influences the returning energy of the ionization electron at the moment

of recollision, which is proportional to the ponderomotive energy U_p ¹⁵. Near-infrared (NIR) laser pulses have been developed in recent decades and are regarded as one of the most effective tools for investigating the electron correlation effect in atoms and molecules^{9,10,16,17}. Y. Zhou *et al.* investigated the multiple recollision process of the Ar atom in the NSDI process using a laser pulse of low intensity and a wavelength of 780 nm in 2016¹⁸. They concluded that by increasing the laser's intensity and wavelength, the multiple recollision process can be reduced. Moreover, when investigating the NSDI process of the Xe atom, the authors demonstrated that the multiple recollision mechanism predominates when using a laser pulse with a mid-infrared region wavelength of 3200 nm (MIR)¹². When using a laser with a long wavelength, the initial returning energy of the electron is predetermined, resulting in a short interaction time between the two electrons, and a small portion of the recolliding electron's energy is transferred to the bounding electron for its transition to the excited state. Therefore, subsequent recollision triggers the NSDI procedure.

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In addition, a two-color laser pulse is a useful tool for controlling the recollision process in a strong laser field¹⁹, with the benefit of having multiple recollision-controlling parameters, such as the relative phase, intensity, and polarization of the two component lasers. Widely used to investigate electron dynamics in HHG²⁰, ATI²¹, and NSDI processes²² are two-color laser pulses. Several research conclusions, including the following, have been published: Using circularly polarized two-color laser pulses, the rate of double ionization events depends on the ratio of relative intensity between the two component lasers²³; the correlation or anticorrelation between the two electrons can be controlled by varying the relative phase between the two components of orthogonal two-color laser pulses^{24,25}; on the other hand, parallel two-color laser pulses (PTC) were used to investigate the NSDI process and demonstrated that the electron. However, the ionization mechanisms that govern the NSDI process when MIR-wavelength PTC laser pulses are employed have not yet been thoroughly investigated.

The aim of this article is to investigate the role of ionization mechanisms in the NSDI process by controlling the motion of two electrons using PTC laser pulses with MIR wavelengths. The effect of ionization mechanisms and the correlated dynamics of the two electrons on the relative phase of the two component lasers is investigated. Methods and outcomes are discussed in Methods and Results, respectively.

METHODS

In this paper, we use a two-dimensional classical ensemble model to simulate the NSDI process^{13,26,27}. According to the classical model, the motion of two electrons is governed by Newton's law

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

where $V_{ne}(\vec{r}_i) = \frac{-2}{\sqrt{\vec{r}_i^2 + a^2}}$ is the ion-electron potential, $V_{ee}(\vec{r}_1, \vec{r}_2) = \frac{1}{\sqrt{(\vec{r}_1 - \vec{r}_2)^2 + b^2}}$ is the electron-electron repulsive potential, i is the label of two electrons and \vec{r}_i corresponds to the position of each electron. In this study, the linearly polarized laser pulse used to investigate the NSDI process has the form $\vec{E} = E_1 \hat{i} + E_2 \hat{i}$. In addition, soft parameters $a = 1.5$ and $b = 0.1$ are used to prevent possible automatic ionization and Coulomb potential singularities^{27,28}.

To investigate the NSDI process using the classical ensemble model, the initial state of the system must be determined by solving equation (1) prior to turning on the laser. According to the classical model, the

initial state of the atomic system is established when all the particles are in the ground state with an energy level of -1.5911 a.u., which is the summation of the first and second ionization potentials of the argon atom. The kinetic energy of the two electrons is randomly distributed in momentum space. Before the laser is activated, two electrons are allowed to move in the atomic nuclear field for a sufficient amount of time (approximately 200 a.u.) to acquire a stable momentum and position distribution of the system (the initial condition)²⁷.

In the following phase, we solve equation (1) for each atom in the presence of the laser electric field. At the end of the interaction between the atom and the laser field, the kinetic energy, ion-electron potential, and half of the electron-electron potential of each electron are analyzed. An atom is considered to have an NSDI process if both electrons' energies are currently positive. To reduce statistical errors and obtain stable results, we examine a set of thirty million atoms.

RESULTS

In this paper, we use parallel two-color laser pulses with wavelengths $\lambda_1 = 2400$ nm and $\lambda_2 = 1200$ nm. The electric field of each laser is described by the equations

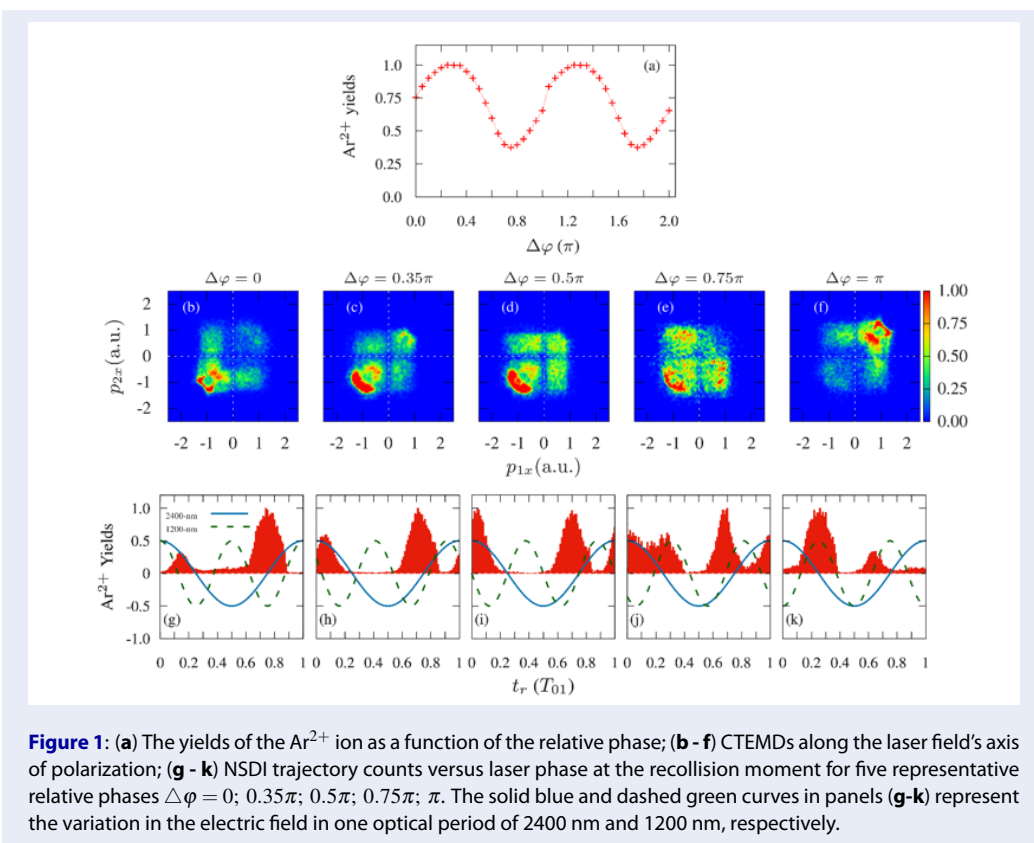
$$E_1 = E_1 \cos(\omega_1 t) \cdot f(t), \quad (2)$$

$$E_2 = E_2 \cos(\omega_2 t + \Delta\phi) \cdot f(t), \quad (3)$$

where $\Delta\phi$ is the relative phase between the two component lasers and $f(t)$ is the envelope of a trapezoidal laser pulse, including a two-cycle turn on, six cycles at full strength, and a two-cycle turn off. The peak intensity of the two component lasers is $I_1 = 4I_2 = 1.6 \cdot 10^{13}$ W/cm².

Figure 1 (a) illustrates the yields of the Ar²⁺ ion as a function of the relative phase $\Delta\phi$. The result demonstrates a significant dependence of the Ar²⁺ yields on $\Delta\phi$ with periodicity π and is comparable to that of the theoretical work¹³ investigating the NSDI process of the Ar atom with a 400-800 nm PTC laser pulse. The maximum and minimal yields of Ar²⁺ occur around the relative phases $(0.35 + n)\pi$ and $(0.75 + n)\pi$, with $n \in R$, respectively.

The correlation between the momentum distributions of two electrons provides insight into their dynamics. Consequently, we analyze the correlated two-electron momentum distribution (CTEMD) along the polarization direction of the laser (Ox axis) corresponding to $\Delta\phi = 0; 0.35\pi; 0.5\pi; 0.75\pi; \pi$ in Figure 1(b) — 1(f). In the case of $\Delta\phi = 0$, the majority of the CTEM along the Ox axis is concentrated on the third quadrant, indicating that the two



electrons depart from the parent ion in parallel along with a negative momenta (Figure 1(b)). When $\Delta\varphi$ changes to 0.5π , the CTMD gradually expands to all four quadrants, whereas the preponderance of the CTMD events continues to be concentrated on the third quadrant. Meanwhile, for $\Delta\varphi = \pi$, the CTMD shifts to the first quadrant, meaning that both electrons ionize in parallel with the positive momenta. The aforementioned findings show that the CTMD strongly depends on the relative phase of the PTC laser field, which is consistent with previous research¹³.

To investigate two-electron dynamics, the trajectories of DI events are analyzed. Previous research has demonstrated that trajectory analysis is an effective technique for investigating the two-electron correlation dynamics in the NSDI process^{25,26,29}. This analysis allows for the calculation of the first electron's ionization time (t_{SI}), the recollision time (t_r), and the double ionization time (t_{DI}). In our calculation, t_{SI} is the moment when the energy of the first electron turns positive, and t_r is the moment when the distance between the first ionized electron and the parent ion is less than 3 a.u. with the condition $t_r > t_{SI}$ ¹³, and t_{DI} is the moment when the energies of the two

electrons reach positive values^{12,26}. Note that we only consider the recollision time of the recolliding electron with the parent ion in one cycle of the 2400 nm laser field. Within this article, we refer to the first ionization electron as the recolliding electron and the second ionization electron as the bounding electron. Figure 1(g) — Figure 1(k) depict the DI yield versus t_r corresponding to the trajectories from Figure 1(b) — Figure 1(f), in which the solid blue and dashed green curves represent the variation in the electric field in one optical period of 2400 nm and 1200 nm, respectively. Previous works have demonstrated that the electron velocity at time t_i is governed by electric field $E = E_0 \cos \omega t$ consisting of two components: the first component is the initial velocity v_0 at the ionization time, while the second is the velocity caused by the electric field $\left[\left(-\frac{E_0}{\omega} \right) \sin \omega t_i \right]$ ^{24,30}. The results show that for the NSDI process induced by a PTC laser pulse, the recollision moments are concentrated in a narrower time range, and their positions change with the relative phase. This indicates that the recollision time between the recolliding electron and the parent ion can be manipulated by the PTC laser. In the case of $\Delta\varphi = 0$ (Figure 1(g)), the recollision time is clustered near the zero intersection in the second half-

cycle of the 2400 nm electric field, so v_0 is negligible, and the final momenta of the two electrons along the polarization of the 2400 nm electric field are determined by the second component and have the same value. Additionally, the repetition period of t_r is not an optical half-cycle, so the CTEM is asymmetric in the third quadrant. As the relative phase increases, the recollision time progressively shifts to the peak of the electric field, so that the competition between the two velocity components determines the final momenta of the two electrons. For $\Delta\varphi = 0.5\pi$, there are two recollision time peaks at the zero-intersection and after the peak of the 2400 nm electric field, so that the momentum spectrum is distributed across all four quadrants, as shown in Figure 1(i). When $\Delta\varphi$ changes to π , the recollision time is concentrated near the zero-intersection in the first half cycle of the 2400 nm electric field and almost does not repeat in the second half, so the CTEM is asymmetrically distributed in the first quadrant (Figure 1(k)).

In the NSDI process, the electron dynamics after the recollision moment are strongly dependent on the returning energy³¹. Figure 2 shows the returning energy of the recolliding electron just before the recollision moment and the ratio of ionization mechanisms governing the NSDI process. In Figures 2(b) – 2(f), the green vertical dashed line represents the ionization potential of Ar^+ (I_{P2}). These results indicate that in cases of $\Delta\varphi = 0$ and $\Delta\varphi = \pi$ (Figure 2(b), Figure 2(f)), the returning energy of the recolliding electron is less than I_{P2} . Therefore, when the recollision event occurs, this energy is insufficient to directly ionize the bound electron, and the RII mechanism hardly dominates the NSDI process (see Figure 2(a)). For $\Delta\varphi = 0.35\pi$ (Figure 2(c)), the returning energy expands and is larger than I_{P2} . Nevertheless, according to the simple man model¹⁵, when the electron returns with larger energy, the interaction time with the parent ion is shorter, so the bound electron receives only a small amount of energy, which is insufficient for direct ionization. Consequently, the RII mechanism contributes only partially to the NSDI process. When $\Delta\varphi$ changes to 0.5π (Figure 2(d)), the returning energy has two peaks, one smaller than I_{P2} and the other at approximately 35 eV. Therefore, similar to $\Delta\varphi = 0.35\pi$, the RII mechanism contributes a negligible amount to the NSDI process. In the case of $\Delta\varphi = 0.75\pi$ (Figure 2(e)), the returning energy of the recolliding electron is concentrated on the range of 10 eV to 20 eV, so the energy imparted to the bound electron is insufficient to directly ionize it. In addition, after transferring energy to the bounding electron, the

electron's remaining energy has a small value, resulting in a recombination effect between the recolliding electron and the parent ion, and then the electron is in an excited state. Thus, the proportion of NSDI cases that occur through the RDESI mechanism predominates.

The preceding results demonstrate that four ionization mechanisms contribute to the NSDI process in all phases. In addition, it is possible to control the ionization mechanism as well as the interaction between two electrons in the atomic shell by adjusting the relative phase of the PTC laser. Figure 2(a) demonstrates that the RDESI ionization mechanism dominates the NSDI process of the Ar atom, whereas the RII mechanism is responsible for a negligible number of DI events. Specifically, when altering the relative phase between the two component lasers for the RII mechanism, this ratio reaches its maximum at $\Delta\varphi = 0.35\pi$ (4.26%) and decreases as the relative phase continues to increase to π . The RESI and ESI ionization mechanisms follow the same trend, with the ratio attaining its maximum value at relative phase $\Delta\varphi = 0.25\pi$ and minimum value at relative phase $\Delta\varphi = 0.8\pi$. For the RDESI ionization mechanism, the ratio reaches the maximum and minimum values at $\Delta\varphi = 0.85\pi$ (85.00%) and $\Delta\varphi = 0.25\pi$ (45.53%), respectively.

Finally, we proceed to analyze the role of ionization mechanisms in CTEM at two relative phases $\Delta\varphi = 0.35\pi$ and $\Delta\varphi = 0.75\pi$, which correspond to the cases where the yields of the Ar^{2+} ion as a function of the relative phase reach maximum and minimum values, as shown in Figure 3. In both relative phases of the RII mechanism, two electrons ionize at the same time at zero electric field. This means that the velocities of the two electrons are similar, and the CTEM gathers on the main diagonal of the third quadrant (Figure 3(a) and Figure 3(e)). In the RESI and ESI mechanisms, at the recollision time, one electron is ionized, and the other electron is in the excited state, which is then ionized after a certain time interval, so the CTEM has a difference (Figure 3(b), Figure 3(f) and Figure 3(c), Figure 3(g)). After the recollision time, the two electrons in the RDESI mechanism are in a double excited state, and after a certain time interval, they ionize, so the CTEM is spread out evenly in all four quadrants (Figure 3(d), Figure 3(h)).

CONCLUSION

Using the classical ensemble model, we investigate in detail the two-electron dynamics in an atom under the influence of a PTC mid-infrared laser pulse. The results demonstrate that the correlated two-electron

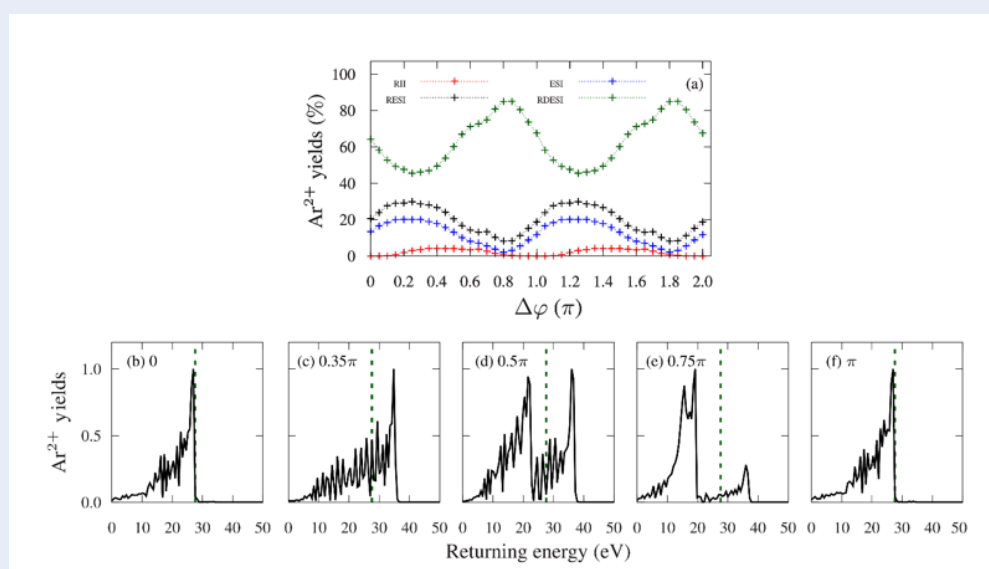


Figure 2: (a) The yields of the Ar²⁺ ion as a function of the relative phase $\Delta\varphi$ for four ionization mechanisms; (b - f) Returning energy distribution before the recollision moment for five representative relative phase phases $\Delta\varphi = 0; 0.35\pi; 0.5\pi; 0.75\pi; \pi$

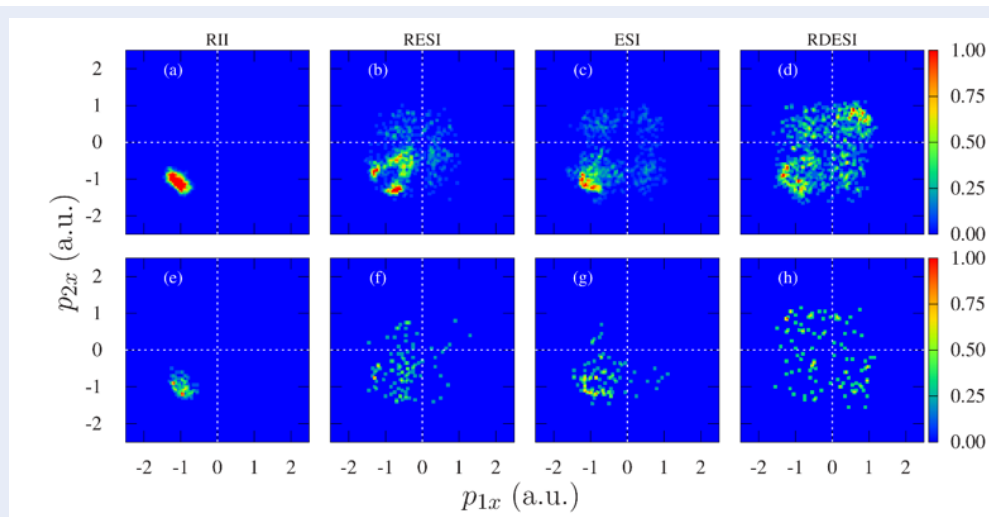


Figure 3: CTMD along the laser polarization axis at relative phases $\Delta\varphi = 0.35\pi$ (a - d) and $\Delta\varphi = 0.75\pi$ (e - h), corresponding to four ionization mechanisms.

momentum distribution, returning energy, and especially ionization mechanisms governing the NSDI process are dependent on the relative phases of the two lasers. In our calculations, the contribution of the direct ionization mechanism to the NSDI process is negligible, whereas the ionization mechanism via the double excited state predominates in all cases. These findings demonstrate that PTC laser pulses are a useful tool for regulating the NSDI process of atoms and

molecules.

ABBREVIATIONS

- NSDI: NonSequential Double Ionization
- PTC: Parallel Two-Color
- HHG: High-order Hamornic Generation
- ATI: Above Threshold Ionization
- DI: Double Ionization
- RII: Recollision Impact Ionization

RESI: Recollision-induced Excitation with Subsequent Ionization

RDESI: Recollision-Doubly induced Excitation with Subsequent Ionization

NIR: Near-Infrared Region

MIR: Mid-Infrared Region

CTEMD: Correlated Two-Electron Momentum Distribution

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AUTHOR'S CONTRIBUTIONS

Thu D. H. Truong and Vinh N. T. Pham did the simulation calculations, analysed the results, and wrote the manuscript. Hanh H. Nguyen and Hieu V. Tran helped with maths and drew figures to show results. All authors read and approved the final manuscript.

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COMPETING INTERESTS

The authors declare that they have no competing interests.

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