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Relative phase effect of nonsequential double ionization in Ar by two-color elliptically polarized laser field*

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We investigated the nonsequential double ionization (NSDI) in Ar by two-color elliptically polarized laser field with a three-dimensional (3D) classical ensemble method. We study the relative phase effect of NSDI and distinguish two particular recollision channels in NSDI, which are recollision–impact ionization (RII) and recollision-induced excitation with subsequent ionization (RESI), according to the delay-time between the recollision and the final double ionization. The numerical results indicate that the ion momentum distribution is changed and the triangle structure is more obvious with the decrease of the relative phase. We also demonstrate that the RESI process always dominates in the whole double ionization process and the ratio of RESI and RII channels can be influenced by the relative phase.

Keywords: nonsequential double ionization, two-color elliptically polarized laser field, recollision-impact ionization, recollision-induced excitation with subsequent ionization

PACS: 32.80.Rm, 42.50.Hz, 42.65.Ky **DOI:** 10.1088/1674-1056/ab5d03

1. Introduction

As the development of the laser technology, people pay more attention to the dynamics of the atom and the molecule when they exposed to the strong laser field. There are many new physical phenomena can be found when the intense laser fields interact with atoms or molecules, such as high harmonic generation (HHG),[1] NSDI,[2] and above-threshold ionization (ATI).[3,4]

In the past decades, NSDI attracted much attention in strong field physics.[5,6] Because the correlation plays a significant role in the NSDI process, many researches about NSDI focused on electron correlation,[7] such as angular correlation[8–10] and recollision,[11–13] *etc*.

In recent years, various kinds of combination of the laser fields have been used to study the above phenomena. For example, the polarized two-color laser field allows us to control electron dynamics in NSDI.[2] The counter-rotating two-color circularly polarized (TCCP) laser field is also a hot topic due to achieving bright circularly polarized HHG[14–18] and low energy structures (LES)[19] as well as controlling electron–ion recattering.[20]

The NSDI process can be explained by a three-step model.[21,22] First, when an atom or molecule is exposed to a strong laser field, an electron can be liberated by tunneling ionization. Then the released electron is accelerated by the laser field and it may be driven back to the parent ion. The returning electron recollides with the parent ion in-elastically, resulting in NSDI.[23–25] The two electrons of the NSDI process are highly correlated due to the recollision process.

A great number of experimental and theoretical studies have been performed to explore the correlated dynamics of the electron pairs in NSDI. The two kinds of NSDI channels can be distinguished by the delay-time, which is defined as the time interval between the recollision and the final double ionization. The process for longer delay-time is defined as recollision-induced excitation with subsequent ionization (RESI). Accordingly, the direct ionization channel with prompt NSDI is defined as recollision–impact ionization (RII).[26,27]

The counter-rotating and co-rotating two-color laser fields with so many variable parameters, such as the relative intensity, the relative phase, and the polarization, enable us to shape the laser pulse and control electron ionization dynamics. Recently, the NSDI process in counter-rotating and co-rotating two-color laser fields are investigated experimentally and theoretically.[28–34] It is found that the intensity ratio and the intensity of the laser field could influence the double ionization probability and the correlation effect, whatever the counter-rotating or the co-rotating laser field.

2. Theoretical model

In this paper, we apply the 3D classical ensemble model to investigate the relative phase effect of Ar in NSDI by two-color elliptically polarized laser field. The classical ensemble

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model has been used before.[26,32,35] It has been widely recognized as reliable and useful approaches in exploring electron dynamics in NSDI. In this model, the evolution of the three-particle system is described by the Newton's equations of motion (the atomic unit (a.u.) is used throughout until stated otherwise)

$$\frac{\mathrm{d}r_i}{\mathrm{d}t} = -\nabla[V_{\mathrm{ne}}(r_i) + V_{\mathrm{ee}}(r_1, r_2)] - E(t), \tag{1}$$

where the subscript $i = 1, 2$ is the label of the two electrons and r_i is the coordinate of the i-th electron. The potentials $V_{\mathrm{ne}}(r_i) = -2/\sqrt{r_i^2 + a^2}$ and $V_{\mathrm{ee}}(r_1, r_2) = 1/\sqrt{(r_1 - r_2)^2 + b^2}$ represent the ion–electron and the electron–electron Coulomb interactions. The softening parameter $a = 1.5$ is introduced here to avoid autoionization. The electron–electron softening parameter b is included primarily for numerical stability and here it is set to be 0.05.

We investigate the relative phase effect along z direction in each pulse of the two-color elliptically polarized laser field which can change the ellipticity of the laser field. The electric field of the two-color elliptically laser pulse is given by

$$\begin{aligned} E_{\mathrm{r}}(t) &= \frac{E_0}{1+\gamma_{\mathrm{E}}} f(t)[\cos(\omega_r t + \varphi_0)\boldsymbol{y} \\ &\quad + \sin(\omega_r t + \varphi_0 + \Delta\varphi_1)\boldsymbol{z}], \end{aligned} \tag{2}$$

$$\begin{aligned} E_{\mathrm{b}}(t) &= \frac{\gamma_{\mathrm{E}} E_0}{1+\gamma_{\mathrm{E}}} f(t)[\cos(\omega_{\mathrm{b}} t + 2\varphi_0)\boldsymbol{y} \\ &\quad - \sin(\omega_{\mathrm{b}} t + 2\varphi_0 + \Delta\varphi_2)\boldsymbol{z}], \end{aligned} \tag{3}$$

where $\boldsymbol{y}$ and $\boldsymbol{z}$ are the unit vectors along the y and z directions, respectively. E_0 is the laser intensity and γ_{E} is the electric field amplitude ratio between the second harmonic laser pulse and the fundamental laser pulse. The electric field amplitude ratio γ_{E} is chosen to be 1.4, which ensures that the probability of double ionization reaches the maximum.[36] $f(t) = \sin^2(\pi t/nT)$ is the envelope of the laser pulse and T is optical cycles (o.c.) of the fundamental laser field. The full duration is four optical cycles ($n = 4$). φ_0 is the carrier-envelope phase (CEP) which is set to be a random number, while $\Delta\varphi_1$ and $\Delta\varphi_2$ are relative phases between z direction and y direction in each pulse, we set $\Delta\varphi = \Delta\varphi_1 = \Delta\varphi_2$. $\omega_{\mathrm{r}} = 0.0584$ a.u. and $\omega_{\mathrm{b}} = 0.117$ a.u., which correspond to the wavelength of the fundamental (780 nm) and the second harmonic (390 nm) laser fields.

In our calculation, the initial condition for Eq. (1) is that evolution ensemble starts to form a classically allowed position of the energy. For Ar atom, the energy is -1.59 a.u., which is approximately equal to the sum of the first and second ionization energies. We can obtain the stable momentum and position of the two electrons by assigning the available kinetic energies and the directions of the momentum vectors of two electrons randomly in the field-free case. The double ionization event is defined as the energies of these two electrons are larger than zero at the end of the laser fields.

3. Results and discussion

Figure 1 shows the Lissajous figures of the electric field for three different relative phases. We can control the ellipticity and the shape of the laser field by changing the relative phase. From the Lissajous curves we can see that the electric field reaches maximum for three times in one optical cycle (o.c.) of the fundamental laser field. Figure 1(a) shows that the Lissajous figure traces out a triangle structure while one of the tips is short at the relative phase 0.4π. The short tip represents that the electric field is short-lived. Figure 1(b) shows that the triangle shape of the Lissajous curve is distinct although one of the tips is narrow at the relative phase 0.2π. However, the electric field value in the narrow tip is higher than that in the short tip as shown in Fig. 1(a), which results that the probability of the double ionization is enhanced. Figure 1(c) shows that the triangle shape of the Lissajous curve is distinct at the relative phase 0.0π, which means that the threefold symmetry of the electric field is pretty well.

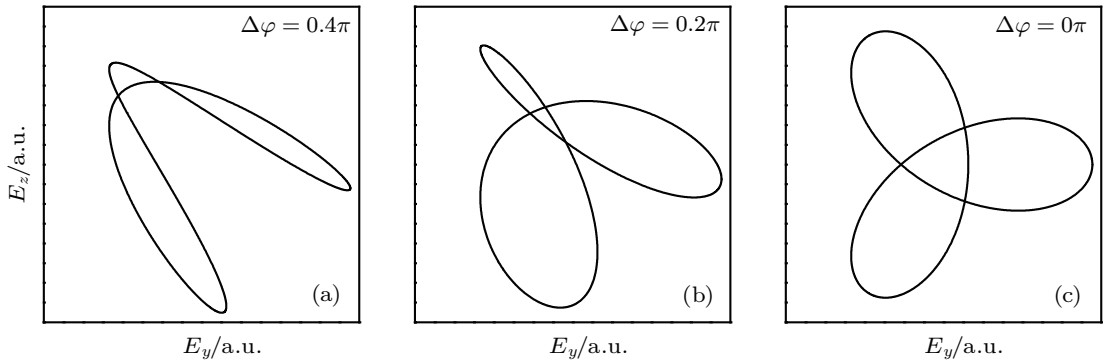


Fig. 1. The Lissajous figures of the electric field for three different relative phases.

Figure 2 shows the double ionization (DI) probability of Ar as a function of the laser intensity in two-color elliptically polarized laser field with different relative phases. We can see that a "knee" structure occurs and the shape of the knee structure is changed with varying the relative phase, which means that the relative phase can affect the double ioniza-

tion probability of Ar in counter-rotating two-color laser field. When the relative phase is 0.0π, the knee structure is narrow and increased sharply with the laser intensity increasing. When the relative phase is 0.2π, the knee structure is changed slowly compared with that at the relative phase 0.0π. When the relative phase is 0.4π, the changing of the knee structure is the most slow and a smoother platform in the knee structure emerges, which is in agreement with the Lissajous curves as shown in Fig. 1. There is a little bit difference with that demonstrated in Refs. [26,37], where the curves of the knee structure in the few-cycle elliptically polarized laser pulse and linearly polarized laser pulse for different CEPs twist together, which illustrates the CEP effect of NSDI.

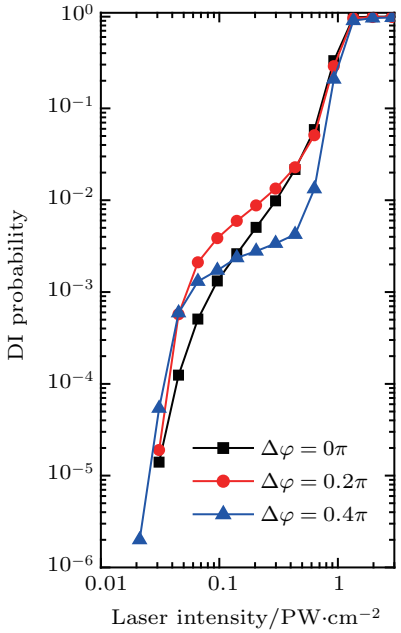


Fig. 2. Double ionization probability of Ar as a function of the laser intensity in counter-rotating two-color laser field with three different relative phases (logarithmic coordinate is used).

It is clear that the counter-rotating two-color laser field has no simple clearly polarized direction, so we explore the electron correlation by recoil momentum distributions of the ion. Because the net momentum of the electroneutral ion–electron system is zero, so we can obtain the relationship $\boldsymbol{p}_{Ar^{2+}} = -(\boldsymbol{p}_{e1} + \boldsymbol{p}_{e2})$.[36] Figure 3 shows the ion momentum distribution with three different relative phases at the end of the laser pulse in counter-rotating two-color laser fields at the laser peak intensity $I_0 = 0.2$ PW/cm^2, and the red lines represent the negative vector potential $-A(t)$ of the electric field. For the relative phase $\Delta\varphi = 0.4\pi$, figure 3(a) shows that the shape of the ion momentum distribution is consistent with the negative vector potential which looks like number "8" with a small titled angle. Figure 3(b) indicates that one of the side of the 8 shape of the negative vector potential is wide and the other side is narrow for the relative phase $\Delta\varphi = 0.2\pi$. It also shows that the distribution of the ion momentum is coherent with the shape of the negative vector potential. For the relative phase $\Delta\varphi = 0.0\pi$, figure 3(c) shows that the shape of the ion momentum distribution depicts a triangle structure which is in agreement with the shape of the negative vector potential. The similar phenomenon is demonstrated in Ref. [32] when the amplitude ratio of the electric field between the second harmonic laser pulse and the fundamental laser pulse is changed in the TCCP laser field. It is illustrated that the shape of the ion momentum distribution coincides with the shape of negative vector potential.

The Coulomb repulsion energy will increased instantaneously when the recollision process happened between the two electrons. We define the recollision time (t_r) as the instant when the repulsion energy reaches its maximum. The final double ionization time (t_{DI}) is defined as the instant when the two electrons finally become free. The delay-time ($t_{DI} - t_r$) is defined as the time interval between the recollision and the final double ionization.[26,37]

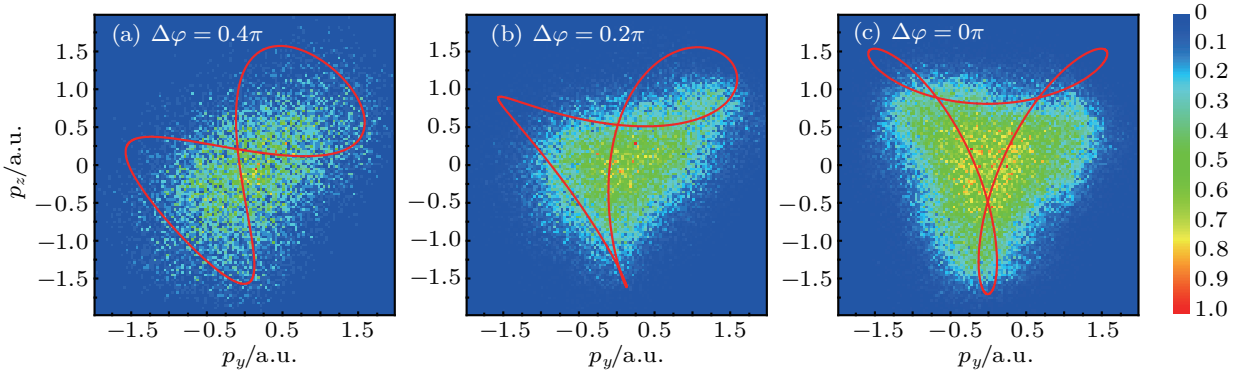


Fig. 3. Ion momentum distributions at the end of the laser pulse with three different relative phases in counter-rotating two-color laser fields at the laser peak intensity $I_0 = 0.2$ PW/cm^2. The red lines represent the negative vector potential $-A(t)$ of the electric field.

Figure 4 shows the double ionization time t_{DI} *versus* the recollision time t_r for three different relative phases. The black solid line is the diagonal of the picture which represents the recollision time is equal to the double ionization time. The detail of the graph is shown in the inset of Figs. 4(a)–4(c). Figure 4(a) shows that the whole cluster is far away from the diagonal at the relative phase $\Delta\varphi = 0.4\pi$. Figure 4(b) shows that the whole cluster is split into several stripes and one of

the stripes is near the diagonal as indicated in the red arrow at the relative phase $\Delta\varphi = 0.2\pi$, which means that the delay time can be separated to several parts. The recollision time near the double ionization time and far from the double ionization time can be recognized by the delay-time. In Fig. 4(c) we can also see that the whole cluster is split into several stripes and one of the stripes is closer to the diagonal as indicated in the red arrow at the relative phase $\Delta\varphi = 0.0\pi$. From Fig. 4 we can conclude that the relative phase will affect the double ionization event.

Figure 5 shows the counts of the delay-time with three different relative phases. It indicates that the counts reach its first minimum (as shown by the red arrows) around 0.1 o.c.–0.2 o.c. at three different relative phases, which separates the shorter and longer delay-time clearly. Therefore we define the RII channel as the ionization process with the delay-time shorter than 0.18 o.c., while the RESI channel is defined as the ionization process with the delay-time longer than 0.18 o.c. as demonstrated in Refs. [26,37].

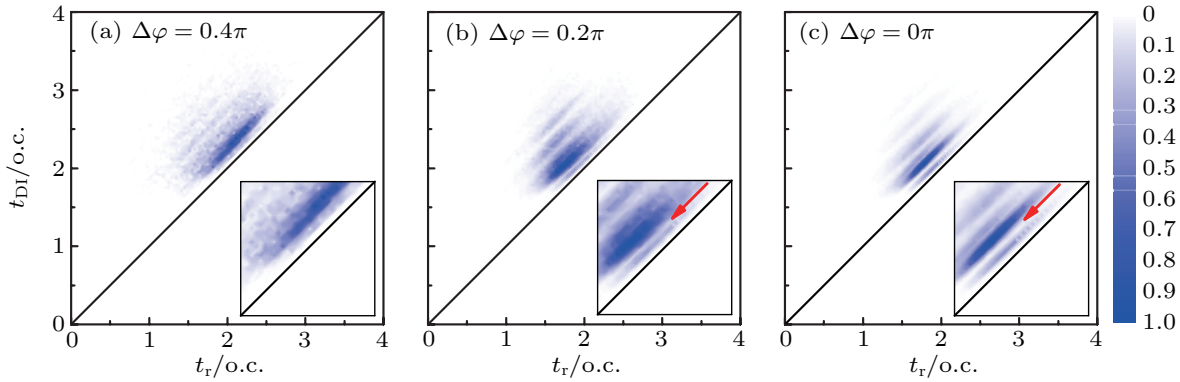


Fig. 4. The double ionization time ($t_{\rm DI}$) *versus* the recollision time ($t_{\rm r}$) for three different relative phases. The insets show the details of the graph.

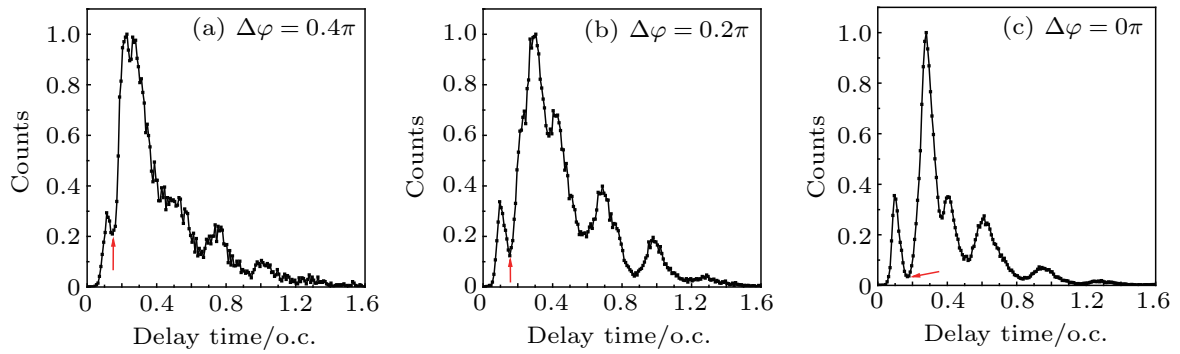


Fig. 5. Counts of the delay time for three different relative phases. The RII and RESI channels can be distinguished by the first minimum of the statistic results. The first minimum position is indicated by the red arrow.

The ion momentum distributions of double-ionized electron shown in Fig. 3 can be further separated into two parts (RII channel and RESI channel), which is shown in Fig. 6. The upper row shows the ion momentum distributions that the delay time is longer than 0.18 o.c., while the lower row shows the ion momentum distribution for which the delay time is shorter than 0.18 o.c., they correspond to the RESI and RII channels respectively. For the RESI channel [as shown in Figs. 6(a)–6(c)], most of the ion momentum is mainly distributed around the origin, it clearly shows an anticorrelation between the two electrons. For the RII channel [as shown in Figs. 6(d)–6(f)], most of the ion momentum is mainly distributed far away from the origin which indicates that the two electrons are in the correlation. The relative phase could influence the delay time, as discussed above. The shorter delay time makes the two electrons escape from the core almost with the same time, and they are both driven by the external field, thus the ionized electrons are in the correlation.

To explain the difference mechanisms in the two channels of NSDI clearly, figures 7(a) and 7(b) show the typical energy trajectories of the RESI and RII at the relative phase 0.4π, the red dashed line and blue dotted line mark the energies of the two electrons, respectively. Figure 7 also shows the time evolution of the repulsion energy which marked by the black solid line. The recollision occurs where the repulsion energy reaches the maximum. From Fig. 7(a) we can find that one electron with the positive-energy emerges and another electron oscillates at the excited bound state after recollision. And then the double ionization occurs, which is the RESI process. From Fig. 7(b) we can find that the two electrons with positive-energy emerge after recollision, which shows that the returning electron collides strongly with other electron and transfers enough energy to make it ionized directly in the RII process.

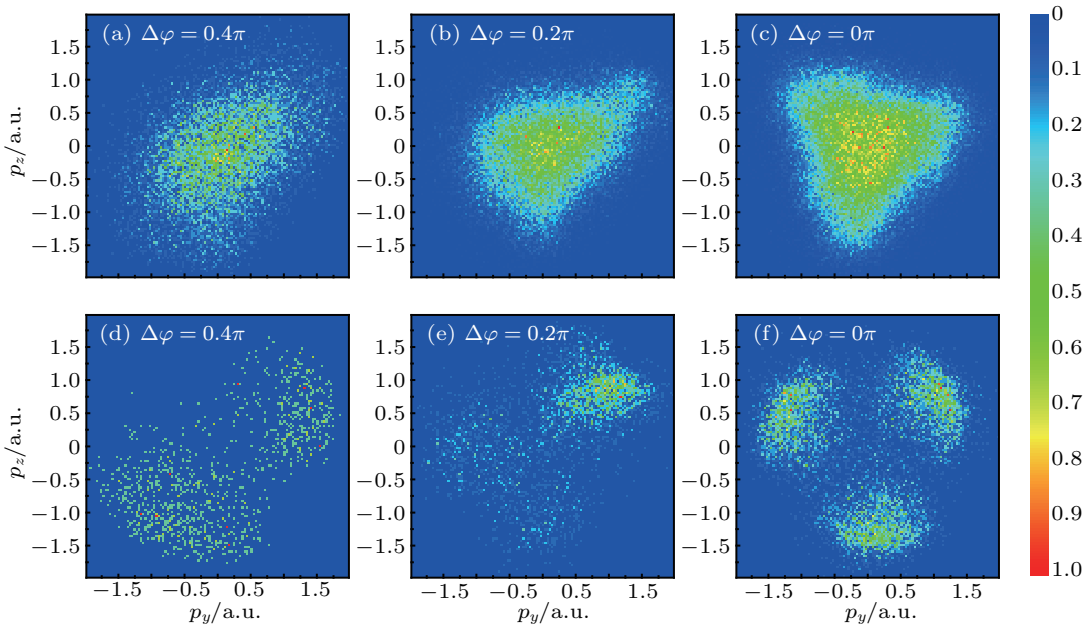


Fig. 6. Ion momentum distributions for different relative phases. The upper row shows the RESI channel and the lower row shows the RII channel.

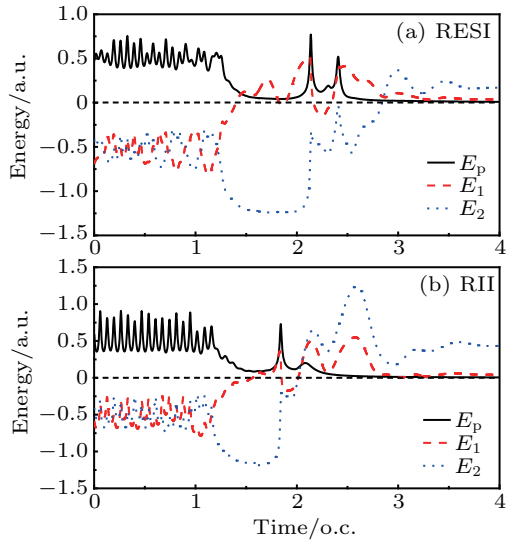


Fig. 7. Two typical energy trajectories of RESI and RII channels. The black solid line marks the repulsion energy between the two electrons. The red-dashed line and blue-dotted line mark the energies of the two electrons, respectively.

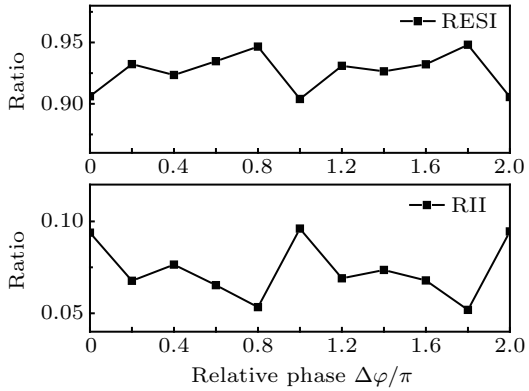


Fig. 8. Ratio between definite ionization mechanism and total ionization *versus* relative phases. The dominant of the RESI is obvious.

Figure 8 shows the ratio between the RESI (RII) and the total ionization *versus* relative phases. It is shown that the RESI process always dominates in the whole double ionization and the RESI channel (the RII) channel can be influenced by changing the relative phase. For example, at the relative phase 0.4π, the ratio of the RESI channel reaches its minimum and the ratio of the RII channel reaches its maximum; at the relative phase 0.8π, the ratio of the RESI channel reaches its maximum and the RII channel reaches its minimum. There is a little difference with that demonstrated in Ref. [26], where the ratio between definite ionization mechanism and the total ionization in few-cycle elliptically polarized laser can be controlled by changing CEPs and the two ratios are comparable.

4. Conclusions

In conclusion, with the 3D classical ensemble method, we investigated the mechanism in NSDI of Ar by few-cycle counter-rotating two-color laser field. The Lissajous figures show that the amplitude of the electric field is changed with the relative phases, which will influence the double ionization probability. The numerical results show that ion momentum distribution for three different relative phases is in agreement with the shape of the negative vector potential, which is used to illustrate the correlated electron emission in the NSDI process. To further investigate the relative phase effect, we separated the ion momentum to RII channel and RESI channel due to the delay time. The ratio of RII channel and RESI channel can be influenced by the relative phases, while the RESI process always dominates in the whole double ionization.

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