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Effect of electron excitation in nonsequential double ionization by intense laser fields*

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We study the double ionization process of atoms in intense laser fields. The momentum distributions of the correlated electrons are calculated. Contrary to the general expectation, we show an increasing proportion of the electrons ionized via excitation with the increasing laser intensity. These electrons generally have small energy thus they concentratedly distribute on the central region of the momentum diagram. Consequently, the central part of the momentum diagram becomes more notable in higher intensity laser fields. Further study suggests that this phenomenon is general in double ionization.

Keywords: electron excitation, electron collision, double ionization

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1. Introduction

When atoms and molecules are irradiated by intense laser fields, nonsequential double ionization (NSDI) may occur.[1] NSDI denotes such a process that an electron is freed and accelerated by the intense laser field, which may turn back to near its parent core when the electric field reverts. If the electron collides with the parent core, it may release another electron. As a result, the ionization probability of the second electron is greatly enhanced due to the collision of the returned electron. This process provides a simple prototype of electron–electron correlation, which lies in the core of many processes and has aroused extensive studies.[2–5]

NSDI can be further classified into two categories.[6] One is the recollision-induced double ionization (RII) process, in which the second electron is freed directly by the collision of the first electron. The other is the recollision-induced excitation with subsequent ionization (RESI) process, in which the second electron is firstly excited into high-lying states, then is freed by the subsequent laser field. Hence the excited states play a key role in the RESI process. To disclose the effect of the excited states, many investigations have been carried out in the double ionization process, especially in the RESI process.[7,8] Substantial evidence on the role of the excited states is still needed.

In double ionization, the kinetic energy of an electron depends on the laser intensity, and reaches a maximal value of about $3.2U_p$, where U_p is the ponderomotive energy. During collision, the returned electron transfers its kinetic energy to the remaining electrons. The energy obtained by the second electron also depends on the laser intensity. Generally, to excite an electron requires less energy than to ionize it directly. It is then expected that the RESI process becomes predominant when the laser intensity is low. When the laser intensity is high, the returned electron is energetic enough to ionize the second electron directly, so the RII process dominates the double ionization and the RESI process is suppressed.[9] This makes it possible to control the effects of the excited state by the laser fields.

Various theoretical approaches have been developed to deal with the NSDI phenomena.[10–12] The classical ensemble method based on the time-dependent Newton equation is one of them.[13–16] This method is widely used in the NSDI process and discloses the principal features of NSDI. Mauger *et al.* showed the characteristic knee structure in the ion yield by circular polarization.[17] Zhou *et al.*[18] reproduced the multiband structure observed by Pfeiffer *et al.*,[5] and distinguished the contribution of electrons ionized at different time to the cross structure.[4,19] Furthermore, by the back-trajectory technique, this method can be used to track subcycle electronic dynamics during the double ionization.[20,21]

In this paper, using the classical simulation method, we study the NSDI process of Ar and Ne atoms in intense laser fields. By identifying the individual contributions of the RII and RESI events to the correlated-electron momentum distribution (CMD), we find an electron-excitation dependent phenomenon in the NSDI process. We will show that, contrary to the general expectation, the RESI events take an increasing fraction in the NSDI events with the increasing laser intensity.

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This increase is identified as the increasing proportion of electron excitation in double ionization.

2. Simulation method

The light-free Hamiltonian of a two-active-electron atom can be written as (in atomic units)

$$H_{\rm e} = \sum_{i=1,2}\left[\frac{\boldsymbol{P}_i^2}{2} - 2V_{\rm sc}(r_i,a)\right] + V_{\rm sc}(\boldsymbol{r}_1-\boldsymbol{r}_2,b), \qquad (1)$$

where $\boldsymbol{r}_i$ and $\boldsymbol{p}_i$ denote, respectively, the position and momentum of the i-th electron; and $V_{\rm sc}$ is the soft-core Coulomb potential defined as $V_{\rm sc}(\boldsymbol{r},c) = (r^2+c^2)^{-0.5}$. The initial position space collection of the two electrons is populated by a Gaussian random series ensuring the total kinetic energy positive in Eq. (1). These electrons move freely for enough time until they keep the stable position and momentum distribution. Then we obtain the initial state of the ensemble.

The Hamiltonian of the atom in a laser pulse is given by $H = H_{\rm e} + (\boldsymbol{r}_1+\boldsymbol{r}_2)\cdot\boldsymbol{E}(t)$, where $\boldsymbol{E}(t)$ is the electric field of the laser pulse. For linear polarization, it can be written as $\boldsymbol{E}(t) = E_0 f(t)\cos(\omega t)\hat{\boldsymbol{x}}$, where E_0 and ω are the amplitude and frequency of the electric field, respectively, $f(t)$ denotes the pulse envelope, and $\hat{\boldsymbol{x}}$ is a unit vector along the x-direction. In our simulation, the pulse envelope is chosen as a 16-cycle trapezoidal shape with 3 cycles in both the ramp on and off. The motion of the two electrons in the laser field is governed by the canonical equation

$$\frac{{\rm d}\boldsymbol{r}_i}{{\rm d}t} = \frac{\partial H}{\partial \boldsymbol{p}_i}, \quad \frac{{\rm d}\boldsymbol{p}_i}{{\rm d}t} = -\frac{\partial H}{\partial \boldsymbol{r}_i}. \qquad (2)$$

The above equation is solved by using the standard fourth to fifth-order Runge–Kutta algorithm. At the end of the pulse, a double ionization event is counted when the energy of each electron is greater than zero. A CMD is obtained statistically on all double ionization events.

According to the time interval between collision of two electrons and ionization of the second electron, the NSDI process is further classified into the RESI and RII processes. The time interval in the RII process is much shorter than that in the RESI process. In our simulation, the events of the time interval less than 0.1 optical cycle are classified into RII events, while those of the time interval greater than 0.3 optical cycle are classified into RESI events. It has been shown that the electron pairs ionized via the RESI process distribute symmetrically about the two axes in the momentum diagram. Because of momentum conservation, the corresponding recoil ions exhibit a single peak at zero in the momentum distribution. On the contrary, the momentum distribution of the recoil ions produced in the RII process exhibits two peaks located at both sides of zero.[22]

3. Variation of CMDs

The CMD denotes the number of two electrons as a function of their momenta along the laser polarization. The CMD is a principal tool to disclose the electron correlation effect and was frequently studied in NSDI. Many features, such as the finger-like structure[3,9] and the cross-shaped structure,[4] were observed in the CMDs. The study on the CMDs provides deep insight into the microscopic electron process in NSDI.[23–27]

We study the double ionization of Ar and Ne atoms driven by laser pulses of equal intensities and wavelengths. Both the first and the second ionization potentials of Ne atoms are larger than those of Ar atoms, thus it is expected that more electrons are ionized from Ne atoms via the RESI process. Figures 1(a) and 1(b) depict two CMDs for Ar and Ne atoms, respectively. The laser field is of 800 nm wavelength and 0.3 PW/cm^2 peak intensity. Figure 1(a) depicts the CMD for Ar atoms. The CMD exhibits large cyan bright regions near the plot center and small red structures highlighted from the bright regions. The bright regions distribute mainly along the diagonal of the plot, indicating that the electrons emit mainly in a side-by-side manner. The electron emissions are correlated. Figure 1(b) depicts the CMD for Ne atoms. The electrons from Ne atoms distribute dispersively around the center. The CMD exhibits two bright arc regions located at the first and the third quadrants, respectively, which is similar to that observed in Ref. [28]. Less bright structures are found in this CMD, and the bright structure in the central region becomes unnoticeable. These features differ from those of Ar atoms. What do these differences mean?

By the back-trajectory technique, the contributions of electrons from the RII and RESI processes to NSDI can be identified. This presents a way to identify the differences in the CMDs of Ar and Ne atoms. The momentum distributions for electrons ionized from Ar atoms via the RESI and RII processes are drawn in Figs. 1(c) and 1(d), respectively. The CMD of RESI electrons exhibits bright regions along the two axes. This denotes that at least one electron in each RESI electron pair has a small momentum. In the plot center, the bright regions overlap and thus form the red structures. Figure 1(d) depicts the CMD of RII electrons, in which the bright areas appear only in the first and the third quadrants, and the red structures keep away from the center. This demonstrates that most RII electrons are energetic. In brief, the bright regions far away from the center are mainly formed by the RII electrons, and those near the center are mainly by the RESI electrons.

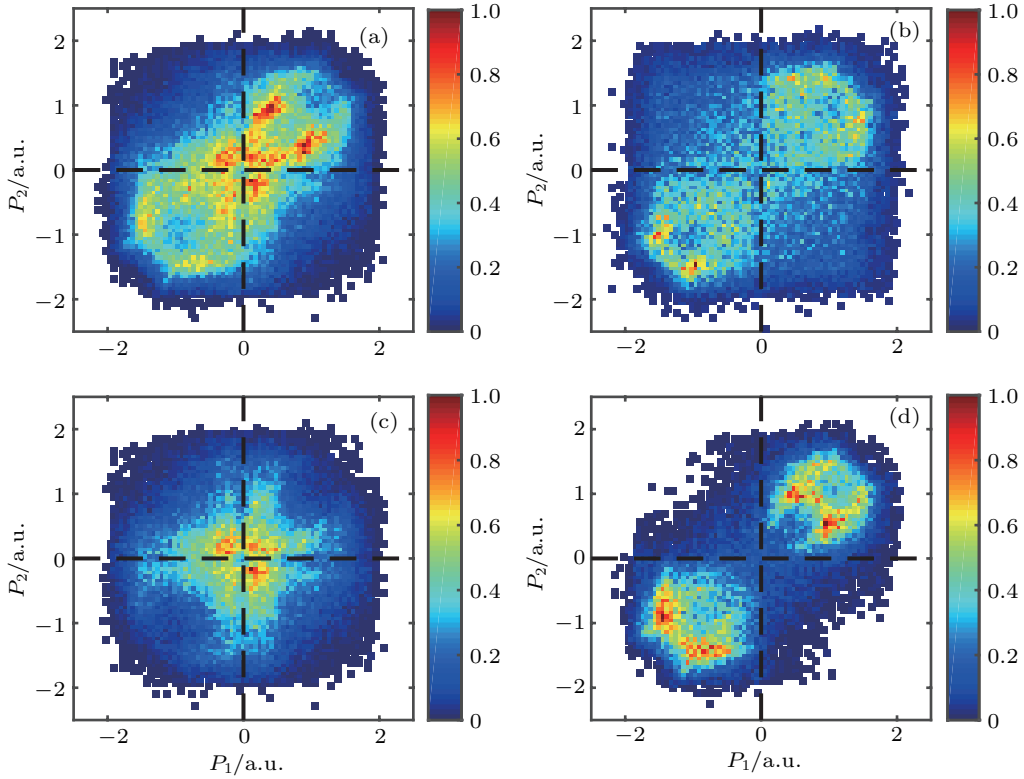


Fig. 1. The CMDs from (a) Ar and (b) Ne atoms irradiated by the laser field of intensity 0.3 PW/cm^2 and wavelength 800 nm. (c) and (d) The CMDs from the Ar atoms via the RESI and RII processes, respectively.

According to this knowledge, we judge that less electrons are produced from Ne atoms via the RESI process, thus no bright structure appears in the central region of the plot. In order to verify this judgement, we draw the CMD of RESI electrons from Ne atoms. Unfortunately, the plot is dim because the number of RESI electrons is too small. This contrasts with the general expectation that more RESI electrons are produced from Ne atoms. However, in an experiment performed with 800 nm, 0.24 PW/cm^2 laser pulses, similar features have been found.[29] The momentum distribution of Ar^{2+} cations exhibits two trivial peaks and a shallow valley at zero, which suggests that both the RESI and RII electrons contribute much to the NSDI electrons. While, at the same intensity, the momentum distribution of Ne^{2+} cations exhibits two sharp peaks, which indicates that the RII process dominates the NSDI process of Ne atoms. This feature agrees well with our calculation.

How to increase the proportion of the electrons ionized via the RESI process in the Ne atoms? This can be done in two ways. One is to change the laser fields according to a scaling law,[24] i.e., changing the laser intensity k^3 times and the laser frequency k times to those of the Ar atoms, where $k = 1.482$ is the ratio of the second ionization potential of the Ne atoms to that of the Ar atoms. For example, increasing the laser intensity to 0.98 PW/cm^2 and increasing the laser frequency to 0.083 a.u. (wavelength 540 nm), one may obtain a CMD which is the same as the one shown in Fig. 1(a). Thus the RESI electrons from the Ne atoms have the same contribution as those in the Ar atoms. Another way is to keep the laser frequency unchanged but vary the laser intensity. The first way is known so we focus on the second way in this paper.

We then calculate the CMDs from the Ne atoms at different laser intensities. The double ionization rate in low laser intensities is small, and the corresponding CMDs become invisible and thus are hard to be identified. For high laser intensities, the CMDs are easy to identify. Figures 2(a) and 2(b) depict two CMDs from Ne atoms calculated at high intensities. We obverse from this figure that, as the laser intensity increases, the central region becomes brighter and brighter. In Fig. 2(b), the bright structures in the central region dominate the CMD. According to the above discussion, the central region is formed by the electrons ionized via the RESI process. In order to confirm this, we depict the CMDs of the RESI and RII electrons in Figs. 2(c) and 2(d), respectively. We obverse again that the RII electrons mainly form the structures far away from the center, and that the RESI electrons mainly form the structures near the center. It was observed that the momentum distribution of Ne^{2+} cations shows two sharp peaks, and that the peak-to-valley ratio increases as the laser intensity is higher than 0.6 PW/cm^2.[30] This implies more electrons ionized via the RESI process being freed in more intense laser fields, which agrees qualitatively with our numerical results. Further studies disclose that these features remain for other frequencies and pulse durations.

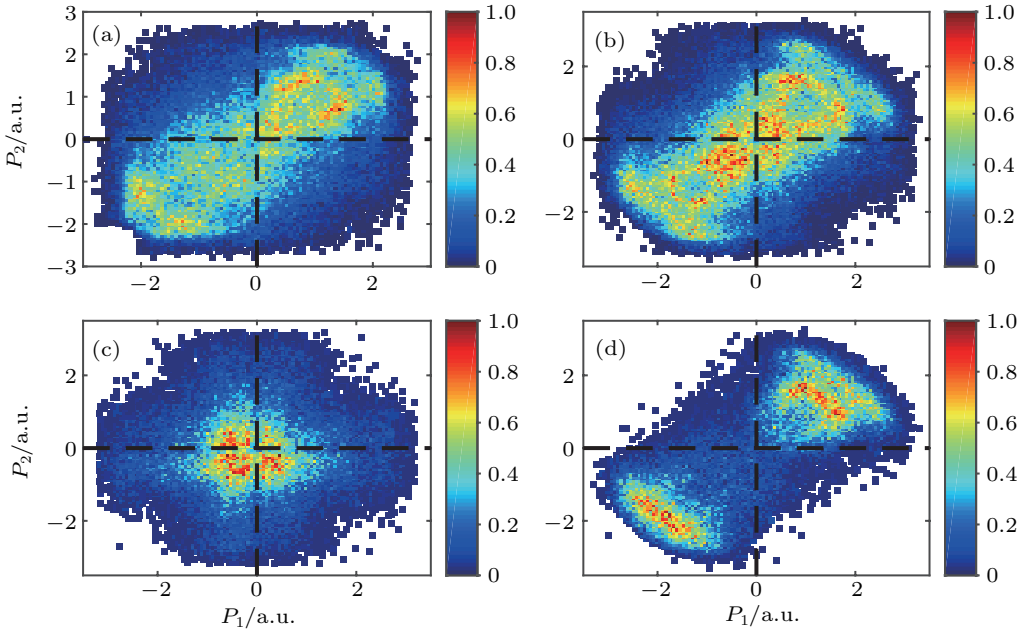


Fig. 2. The CMDs from the Ne atoms in the laser field of intensities (a) 0.6 PW/cm^2 and (b) 0.9 PW/cm^2. (c) and (d) The CMDs of the RII and RESI electrons at 0.9 PW/cm^2, respectively. Laser wavelength is 800 nm.

4. Physical explanations

Three questions are left unanswered: one is why the RESI electrons distribute mainly along the two axes, antoher is why most RII electrons are of relative high energy, and the final one is why the RESI electrons take an increasing proportion with the increasing laser intensity.

We look for answers from the momentum distribution of the electrons. The end-of-pulse momentum of an electron freed by the laser pulse can be written as

$$\boldsymbol{P} = \boldsymbol{P}_0 - \int_{t_0} \boldsymbol{E}_0 \cos(\omega t)\,\mathrm{d}t \simeq \boldsymbol{P}_0 + \frac{\boldsymbol{E}_0}{\omega}\sin(\omega t_0), \qquad (3)$$

where the first term $\boldsymbol{P}_0$ is the electrons' initial momentum, and the second term is the drift momentum obtained by the free electron from the laser field, and t_0 denotes the time when the electron is free. For the returned electron, $\boldsymbol{P}_0$ can be regarded as its momentum after collision. The second electron can be treated as ionized with zero velocity, so the end-of-pulse momentum depends critically on the ionization time t_0.

In the RII process, the second electron is ionized promptly after the collision. Thus the ionization time of the second electron can be treated as the collision time approximately. The collision generally occurs at the zero crossing of the laser field, i.e., the moments satisfy $\boldsymbol{E}(t_0) = 0$. According to Eq. (3), the drift momentum is large at the zero crossing of the laser field, so is the end-of-pulse momentum of the second electron. For the returned electron, both its initial and final momenta vary with the laser intensity. If the laser intensity is low, the energy of the returned electron is small, and the electron transfers a large proportion of its energy to the remaining electrons for the second ionization. Thus the momentum of the returned electron is greatly decreased. It will change its motion direction shortly after the collision and thus can be accelerated for a long time. As a result, the kinetic energy of the first electron is large. If the laser intensity is high, the returned electron has a large kinetic energy, and the energy transferred to the second electron takes a small proportion. So the kinetic energy of the first electron is still large. In the between, the kinetic energy of the first electron varies in a large range.

In the RESI process, the second electron is ionized by the laser field from an excited state. The field-induced ionization generally occurs near the field maxima, say the moments satisfy $\cos(\omega t_0) \to 1$. The drift momentum as well as the end-of-pulse momentum of the second electron is small, because $\sin(\omega t_0) \to 0$. That is to say, the second electron has a small momentum. For the first electron, its momentum changes in a similar way to that in the RII process. The difference is that the energy transferred to the second electron is smaller than that in the RII process, because to excite an electron needs less energy than to ionized it. The kinetic energy of the first electron varies with the laser intensity, and distributes in a large range. Consequently, the RESI electrons distribute mainly along the two axes in the CMDs.

In order to present a substantial evidence, we count the numbers of single ionization, electron collision, and double ionization events as functions of time. Statistical data for two laser intensities are shown in Fig. 3. Generally, the first ionization occurs at the field maxima, while the collision of two electrons occurs at the zero crossing of the electric field. The ionization time brings forward as the laser intensity increases. At a lower laser intensity (Fig. 3(a)), the blue curve exhibits several notable peaks in the pulse front, indicating the ionization time of the first electron. At a higher laser intensity

(Fig. 3(b)), the first peak becomes notable, and the peaks behind the third peak become trivial, so the second peak becomes the dominated one. However, the ionization time of the second electron depends greatly on the laser intensity. In Fig. 3(a), the green curve exhibits several broad peaks. These green peaks follow closely with the red peaks, which is more evident for the peaks in the third and the fourth optical cycles. This feature discloses that the RII process dominates the double ionization in low laser intensities. When the laser intensity is high, as shown in Fig. 3(b), the green curve exhibits many sharp peaks, and these peaks lag behind the red peaks, indicating that most double ionization events occur with notable time delay after the electron collision. This suggests that the RESI process becomes dominated as the laser intensity increases.

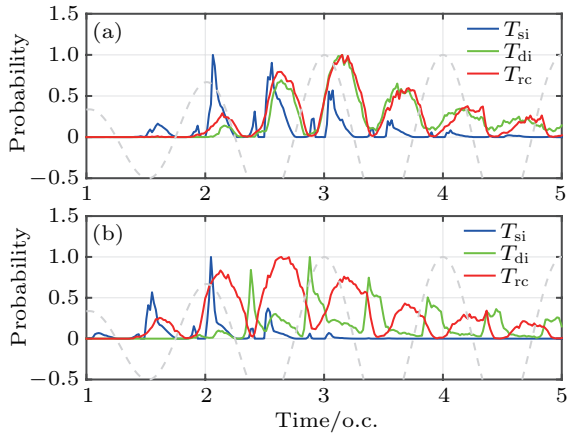


Fig. 3. Time-resolved counts of three events from the Ne atoms in (a) 0.3 PW/cm^2 and (b) 0.6 PW/cm^2, respectively. T_{si} denotes the time of single ionization, which is the ionization time of the first electron. T_{di} denotes the time of double ionization, which is the ionization time of the second electron. T_{rc} denotes the time of recollision. For clarity, each curve is normalized by its maximum. The laser wavelength is 800 nm. The dot line denotes the electric field of the laser pulse.

The last but the most important question is why the electrons ionized via the RESI process become dominated at high laser intensities. In the RESI process, the second electron is firstly excited into a high-lying state by collision and then is ionized by the laser field, thus the first-excited state should be populated primarily. Hence we look for an answer in the energy distribution of electrons.

The energy distribution of two electrons and the results are shown in Fig. 4. The energy of an electron includes the kinetic energy and the Coulomb energy and is taken at the moment of 0.1 optical cycle after the electron collision. Generally, each curve exhibits two peaks and a valley between them. The right peak locates at about zero, while the left peak locates at about -0.67 a.u. and shifts linearly with the laser intensity. The field-free energy of the first-excited state ($2s2p^6$) of the Ne^{2+} ion is about -0.516 a.u. Thus, the left peak indicates the number of electrons populated on this state, which is shifted by the laser field due to the AC stark effect.[31]

At a low laser intensity (Fig. 4(a)), the two peaks in the green curve are of equal heights, while the left peak in the blue curve is dim and thus the curve exhibits only one notable peak. This denotes that, when the laser intensity is low, the returned electron transfers most of its energy to the remaining electrons during collision. Consequently, one remaining electron is ionized directly, most likely with zero energy, and the returned electron is firstly excited and then ionized by the subsequent laser field. This is the RESI process. If the returned electron is sufficiently energetic, its energy stays positive after the remaining electron has been freed by the collision. The returned electrons are still free and they form the second peak in the green curve. This is the RII process. The amount of RESI events can be counted as the covered area in the left side of the valley, and that of RII events can be counted as the covered area in the right side. Judged from the covered areas in both sides of the valley, we find that although the two peaks are of equal heights in Fig. 4(a), the number of RII electrons is much larger than that of RESI electrons, owing to the blue curve also contributing much to the RII events. This feature agrees with our earlier statements.

As the laser intensity increases, the left peak in the green curve grows taller, and the second peak becomes flat and shifts to the right side (Figs. 4(b) and 4(c)). This change is more notable in Fig. 4(c). The growth in the first peak denotes that more electrons are excited and the subsequent RESI events also increase in amount. The shift of the right peak denotes that the remaining energy of the returned electron after collision increases with the laser intensity. The expansion of the left peak suggests that the remaining energy of the returned electron increases and then covers a large range. It also denotes a large number of returned electrons remaining in the free state. The blue curve also varies with the laser intensity. A notable change is that the left peak becomes distinctive. This peak denotes the excitation of the second electron. Its growth indicates that more remaining electrons are excited. This does not occur at low laser intensities.

Comparing the two curves in the above figure, we obtain an RESI picture as follows. When the firstly ionized electron returns, it collides with its parent core and frees the second electron. After collision, the first electron loses most of its energy and then is bounded again, most likely, in the first-excited state. It is ionized by the subsequent laser field. This is the RESI process. If the returned electron transfers less energy to the remaining electrons during collision, it remains free, and one remaining electron is collided into the first-excited state. The second electron is ionized by the subsequent laser field. This opens another RESI channel, and occurs less in low-intensity laser fields. These RESI electrons form the bright structures in the central region of CMDs. Hence, the RESI electrons dominating the CMDs is an electron excitation effect in double ionization.

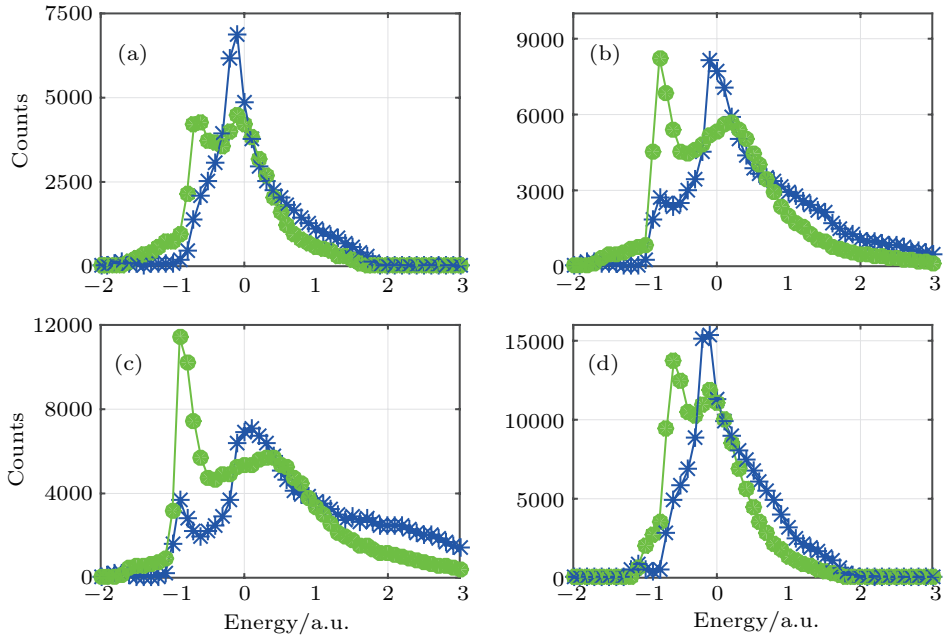


Fig. 4. Energy distributions of two electrons from the Ne atoms at 0.1 optical cycle after recollision at laser intensities of (a) 0.30 PW/cm^2, (b) 0.60 PW/cm^2, and (c) 0.90 PW/cm^2; (d) from Ar atoms at laser intensity 0.30 PW/cm^2. The laser wavelength is 800 nm. Green and blue lines denote the returned and the recollided electrons, respectively.

In order to check our statements, we draw the energy distribution of electron pairs produced from the Ar atoms in Fig. 4(d). Similar curves are found to those in Fig. 4(a), because the laser fields are the same and the returning electrons have similar kinetic energy. The blue curve exhibits a single peak, suggesting that the remaining electron is firstly ionized. The difference lies in the position of the left peak. For the Ar^{2+} ions, the left peak locates at about -0.58 a.u., which is the energy of the first-excited state ($3s3p^6$, -0.52 a.u.) shifted by the laser field due to the AC stark effect. Thus the left peak in the green curve denotes the population on the first-excited state of the Ar^{2+} ions. This peak is a little higher than the right one, which indicates that the RESI process in the Ar atoms takes a larger proportion. This causes the bright structures to be more notable in the central region in Fig. 1(a).

From Fig. 4, we conclude that the RESI events increase in both the amount and the ratio to the RII events in more intense laser fields. As the laser intensity increases, the first-ionized electron is more energetic. However, the collision cross section decreases if the electron is of high velocity. Consequently, the energy obtained by the second electron via collision decreases and becomes more likely to be excited. This change also lies in the momentum distribution of recoil ions. Because of momentum conservation, the recoil-ion momentum distribution is peaked at zero in the RESI process, while that in the RII process exhibits two sharp peaks located symmetrically at both sides of zero momentum. Hence, the RII process dominating the NSDI process means two notable peaks around zero momentum, while the RESI process becomes predominated means that the valley gets shallow as the laser intensity increases. This feature can be found experimentally.[30] It is also expected that the number of RESI events exceeds that of RII events if the laser intensity is further increased, so the RESI process predominates NSDI. In such a case, the momentum distribution of recoil ions shows a predominant peak at zero momentum, which was observed in Ref. [32] at 2.0 PW/cm^2.

5. Discussion

Based on the above discussion, we conclude that the RESI process takes over an increasing proportion with the increasing laser intensity. The increase is caused by more electrons being excited in higher intensity laser fields. This exceeds the general expectation. It is generally regarded that the returned electron is energetic enough to ionize the second electron directly when the laser intensity is high, so the RESI process is greatly suppressed.[9] However, other treatments and experimental observations provide multiple support to our statement.

According to our study, the electrons ionized via the RII process are energetic and distribute far away from the central region in the momentum diagram. This feature is generally accepted.[3,9] The electrons ionized via the RESI process are of lower kinetic energy, thus they occupy the central part of the momentum diagram. The increasing RESI fraction means more electrons distributing in the central region. This feature can be found in many theoretical treatments. In Ref. [7], the RESI process of He atoms is studied by a semianalytic treatment. The calculated CMDs distribute along the two axes, and shrink towards the center region as the laser intensity increases. We notice that an excited state is involved in this treatment, so this finding supports our conclusion. In a quantum treatment on the energy correlation in NSDI, two excited

states are involved.[8] The calculated CMDs also exhibit notable convergence towards zero momentum as the laser intensity increases. This feature agrees with our statement qualitatively.

Several experimental observations also support our statements. A recent experiment performed on the Ar atoms shows that more electrons concentratedly distribute in the central part of CMDs in more intense laser fields.[33] This confirms that the electrons ionized via the RESI process take an increasing proportion with the increasing laser intensity. However, substantial support for our results comes from an earlier experimental observation.[32] It was observed that the momentum distribution of Ne^{2+} ions exhibits two sharp peaks at a lower laser intensity but shows only one peak located at zero momentum at 2.0 PW/cm^2. This indicates that the RESI process predominates the whole NSDI process at this high intensity. It was also observed that, for the recoil Ar^{2+} ions, similar changes occur in relative low laser intensities. These observations provide strong support to our statements.

6. Conclusions

Using a classical simulation method, we study the double ionization process of Ar and Ne atoms in intense laser fields. We distinguish the individual contribution of electrons ionized via the RII and RESI processes to the CMDs. We find that the electrons ionized via the RESI process generally are of lower kinetic energy and make an increasing contribution to the CMDs with the increasing laser intensity. Correspondingly, the central part of CMDs becomes more and more notable. In producing these RESI electrons, the first-excited state plays a key role. This contrasts to the general expectation that the RESI electrons contribute less in high-intensity laser fields. This unexpected phenomenon indicates that more electrons are firstly excited, owing to the electron collision, and then are ionized in higher intensity laser fields. This phenomenon is generally in the double ionization process when the laser intensity is higher than the threshold.

References

[1] Becker W, Liu X, Ho P and Eberly J H 2012 *Rev. Mod. Phys.* **84** 1011

[2] Weber Th, Giessen H, Weckenbrock M, Urbasch G, Staudte A, Spielberger L, Jagutzki O, Mergel V, Vollmer M and Döner R 2000 *Nature* (London) **405** 658

[3] Staudte A, Ruiz C, Schofler M, Schossler S, Zeidler D, Weber Th, Meckel M, Villeneuve D M, Corkum P B, Becker A and Döner R 2007 *Phys. Rev. Lett.* **99** 263002

[4] Bergues B, Kūel M, Johnson N G, Fischer B, Camus N, Betsch K J, Herrwerth O, Senftleben A, Sayler A M, Rathje T, Ben-Itzhak I, Jones R R, Paulus G G, Krausz F, Moshammer R, Ullrich J and Kling M F 2012 *Nat. Commun.* **3** 813

[5] Pfeiffer A N, Cirelli C, Smolarski M, Döner R and Keller U 2011 *Nat. Phys.* **7** 428

[6] Feuerstein B, Moshammer R, Fischer D, Dorn A, Schroter C D, Deipenwisch J, Crespo Lopez-Urrutia J R, Hohr C, Neumayer P, Ullrich J, Rottke H, Trump C, Wittmann M, Korn G and Sandner W 2001 *Phys. Rev. Lett.* **87** 043003

[7] Shaaran T, Nygren M T and Figueira de Morisson Faria C 2010 *Phys. Rev. A* **81** 063413

[8] Liao Q and Lu P 2010 *Phys. Rev. A* **82** 021403

[9] Rudenko A, de JesusV L B, Ergler Th, Zrost K, Feuerstein B, Schroter C D, Moshammer R and Ullrich J 2007 *Phys. Rev. Lett.* **99** 263003

[10] Becker A and Faisal F H M 2002 *Phys. Rev. Lett.* **89** 193003

[11] Chen Z, Liang Y and Lin C D 2010 *Phys. Rev. Lett.* **104** 253201

[12] Liu W C, Eberly J H, Haan S L and Grobe R 1999 *Phys. Rev. Lett.* **83** 520

[13] Su Q and Eberly J H 1991 *Phys. Rev. A* **44** 5997

[14] Panfili R, Haan S L and Eberly J H 2002 *Phys. Rev. Lett.* **89** 113001

[15] Wang X and Eberly J H 2009 *Phys. Rev. Lett.* **103** 103007

[16] Haan S L and Smith Z S 2007 *Phys. Rev. A* **76** 053412

[17] Mauger F, Chandre C and Uzer T 2010 *Phys. Rev. Lett.* **105** 083002

[18] Zhou Y, Huang C, Liao Q and Lu P 2012 *Phys. Rev. Lett.* **109** 053004

[19] Huang C, Zhou Y, Zhang Q and Lu P 2013 *Opt. Express* **21** 11382

[20] Haan S L, Breen L, Karim A and Eberly J H 2006 *Phys. Rev. Lett.* **97** 103008

[21] Zhang L, Xie X, Roither S, Zhou Y, Lu P, Kartashov D, Schoffler M, Shafir D, Corkum P B, Baltuška A, Staudte A and Kitzler M 2014 *Phys. Rev. Lett.* **112** 193002

[22] Zhang Z, Zhang J, Bai L and Wang X 2015 *Opt. Express* **23** 7044

[23] Liu Y, Tschuch S, RudenkoA, Durr M, Siegel M, Morgner U, Moshammer R and Ullrich J 2008 *Phys. Rev. Lett.* **101** 053001

[24] Dong S, Zhang Z, Bai L and Zhang J 2015 *Phys. Rev. A* **92** 033409

[25] Hu S L, Chen J and Hao X L 2017 *Chin. Phys. Lett.* **30** 043201

[26] Xin G G, Zhao Q and Liu J 2012 *Acta Phys. Sin.* **61** 133201 (in Chinese)

[27] Dong S, Han Q and Zhang J 2017 *Chin. Phys. B* **26** 023202

[28] Weckenbrock M, Zeidler D, Staudte A, Weber Th, Schoffler M, Meckel M, Kammer S, Smolarski M, Jagutzki O, Bhardwaj V R, Rayner D M, Villeneuve D M, Corkum P B and Dorner R 2004 *Phys. Rev. Lett.* **92** 213002

[29] Eremina E, Liu X, Rottke H, Sandner W, Dreischuh A, Lindner F, Grasbon F, Paulus G G, Walther H, Moshammer R, Feuerstein B and Ullrich J 2003 *J. Phys. B* **36** 3269

[30] de Jesus V LB, Feuerstein B, Zrost K, Fischer D, Rudenko A, Afaneh F, Schroter C D, Moshammer R and Ullrich J 2004 *J. Phys. B* **37** L161

[31] Wang Y, Zhang J, Xu Z, Wu Y S, Wang J T and Guo D S 2009 *Phys. Rev. A* **80** 053417

[32] Rudenko A, Zrost K, Feuerstein B, de Jesus V L B, Schroter C D, Moshammer R and Ullrich J 2004 *Phys. Rev. Lett.* **93** 253001

[33] Kubel M, Burger C, Kling N G, Pischke T, Beaufore L, Ben-Itzhak I, Paulus G G, Ullrich J, Pfeifer T, Moshammer R, Kling M F and Bergues B 2016 *Phys. Rev. A* **93** 053422