Nonsequential double ionization of diatomic molecules by elliptically polarized laser pulses*

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Using the classical ensemble method, we investigate nonsequential double ionization (NSDI) of diatomic molecules by elliptically polarized laser pulses. The results show that the ellipticity of the laser field has a strong suppression effect on NSDI probabilities both in parallel and perpendicular alignments. The double ionization (DI) channel is commonly dominated by NSDI, and the NSDI channel changes with ellipticity. As ellipticity increases, more and more NSDIs occur through recollision excitation with subsequent field ionization (RESI). Moreover, like the case of linear polarization, the two electrons involved in NSDI for perpendicularly aligned molecules are more likely to emit into the opposite hemispheres as compared to the case of parallel alignment. Additionally, this alignment effect increases as ellipticity increases.

Keywords: nonsequential double ionization, electron correlation, molecular alignment effect

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1. Introduction

Nonsequential double ionization (NSDI) of atoms and molecules has been a hot topic in strong field physics since it can provide a profound understanding of dynamical electron correlation. Many experimental and theoretical studies have been performed to reveal the electron dynamics and characteristics in NSDI.[1–12] Nowadays, the most widely accepted mechanism to explain the NSDI is the recollision model,[13] in which the first ionized electron is driven back to its parent ion by the oscillating laser field, causing the ionization of the second electron by inelastic recollisions. This recollision process is also responsible for high-order harmonic generation[14–18] and high-order above-threshold ionization.[19]

Based on the polarization and intensity dependence of NSDI, in the past decades most of NSDI researches have focused on linearly polarized laser pulses and moderate laser intensities where NSDI dominates. Recently, investigations on DI by elliptical laser fields have received much attention since NSDI events have been observed for some molecules such as NO and O_2[20] and atomic magnesium[21] driven by circular laser pulses. These studies have revealed many characteristics of DI in elliptical laser fields. For example, recollision is also the underlying mechanism for NSDI, and it occurs through elliptical trajectories;[22] non-zero initial longitudinal velocity has a suppressing effect on single-return collision events in the DI process;[23] DI channel of diatomic molecules changes from sequential double ionization (SDI) to NSDI as internuclear distance increases.[24]

For molecules, molecular alignment is an influence factor in DI process. Experimental[25] and theoretical[26] investigations have shown that in the linearly polarized laser fields, the two electrons involved in NSDI are more likely to escape into the opposite hemispheres for perpendicular molecules than for parallel molecules, indicating the dependence of the electron correlation on molecular alignment. In this paper, we employ the classical ensemble model to investigate NSDI of diatomic molecules by elliptically polarized laser pulses. We analyze in detail the origin of the suppression of NSDI by the elliptical laser field and explore the ellipticity dependence of the molecular alignment effect on electron–electron correlation. The results show that the suppression of NSDI originates from the decrease in rescattering probability and the recollision energy. Like the linear laser field, in elliptical laser field the electron pairs prefer to emit into opposite hemisphere in perpendicular alignment more than in parallel alignment. Moreover, this alignment effect increases with increasing ellipticity.

2. Theoretical model

The classical model has been widely used to study NSDI[27–29] and even sequential double ionization[30] in intense laser fields. In this study, we use this model to investigate the NSDI of diatomic molecules by elliptically polarized laser fields. The evolution of the two-electron system is determined by Newton's classical motion equations (atomic units are used throughout this paper unless otherwise stated): $\mathrm{d}^2\boldsymbol{r}_i/\mathrm{d}t^2 = E(t) - \nabla V_{\mathrm{ne}}(\boldsymbol{r}_i) - \nabla V_{\mathrm{ee}}(\boldsymbol{r}_1, \boldsymbol{r}_2)$, where the subscript $i = 1, 2$ is the electron label. $V_{\mathrm{ne}}(\boldsymbol{r}_i)$ represents the ion–electron interaction, and $V_{\mathrm{ee}}(\boldsymbol{r}_1, \boldsymbol{r}_2)$ is the electron–

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electron interaction. In our calculations, the motions of two electrons are restricted in the x–y polarization plane for simplicity since the out-of-plane effects are negligible.[31] We use the fixed-nuclei model[32] and align the molecular axis along the x axis. So, $V_{\rm ne}(\boldsymbol{r}_i) = -1/\sqrt{(x_i - R/2)^2 + y_i^2 + a^2}$, $V_{\rm ee}(\boldsymbol{r}_1, \boldsymbol{r}_2) = 1/\sqrt{(x_1 - x_2)^2 + (y_1 - y_2)^2 + b^2}$. We employ N_2 as our model diatomic molecules and assume the internuclear distance R to be 2.0 a.u., the unit a.u. is the abbreviation for atomic unit, (which is the internuclear distance of N_2). To obtain the initial value, the ensemble is populated starting from a classically allowed position for the ground state energy of our model diatomic molecule N_2 (-1.67 a.u.). The softening parameter a is used to avoid autoionization, which sets the lower limit of a. There is also an upper limit for a, which is determined by the condition that there is a classically allowed region for the two electrons with the total energy of the ground state of the target. For the target investigated in this paper, the lower and the upper limits of a are about 0.85 and 1.8 respectively. Here we set $a = 1.25$. Note that a change of the parameter a has little influence on the statistical results presented in our paper. The parameter b is included to avoid the mathematical singularity in our calculations. Indeed, the value of b has no physical meaning. It could be set to equal any small value. We set $b = 0.1$ here. The available kinetic energy is distributed between the electrons randomly. Then the electrons are allowed to evolve for a sufficiently long time in the absence of the laser field to obtain stable position and momentum distributions. When the initial ensemble is obtained, the electric field is turned on and the process is determined by Newton's motion equation presented above. The electric field $\boldsymbol{E}(t)$ is an 800-nm elliptically polarized electric field. When the long axis is parallel to the molecular axis, the laser field is written as

$$\boldsymbol{E}(t) = E_0 f(t) \left[\frac{1}{\sqrt{1+\varepsilon^2}} \sin(\omega t)\hat{x} + \frac{\varepsilon}{\sqrt{1+\varepsilon^2}} \cos(\omega t)\hat{y} \right],$$

where $\hat{x}$ and $\hat{y}$ represent the unit vectors along long axis and short axis of the laser polarization plane; ε is the ellipticity; $f(t)$ is the laser envelope, which turns on and turns off linearly during two cycles and keeps full strength for six cycles; E_0 and ω are the amplitude and frequency of the electric field respectively. The intensity of the elliptically polarized laser field is 2×10^{14} W/cm^2.

3. Results and discussion

After the laser field is turned off, we examine the DI yields. We define that the diatomic molecule is doubly ionized if the energies of both electrons are positive and singly ionized (SI) if either of the two electrons first possesses positive energy, where the energy of each electron is composed of the kinetic energy, potential energy of the electron–ion interaction, and half electron–electron repulsive interaction. We define recollision if the distance between the two electrons is less than $d = 3$ a.u. after the ionization of the first electron. The statistical results change very little when the value of d ranges from 2 a.u. to 5 a.u. If recollision occurs, DI is classified into NSDI. Otherwise DI is classified into SDI.

According to the above definitions of DI and NSDI, we calculate the total DI probability, NSDI probability, and the ratio of the NSDI probability to the DI probability, each as a function of ε, which are shown in Fig. 1. From Figs. 1(a) and 1(b) it follows that both DI probability and NSDI probability decrease fast with ellipticity increasing, showing the strong suppression of DI and NSDI yields by the elliptical laser field. Figure 1(c) shows that the ratio of the NSDI probability to the DI probability is negligibly dependent on the ellipticity and that all the ratios are larger than 0.9, meaning that double ionization of the diatomic molecules both in the linear laser field and the elliptical laser field are dominated by NSDI.

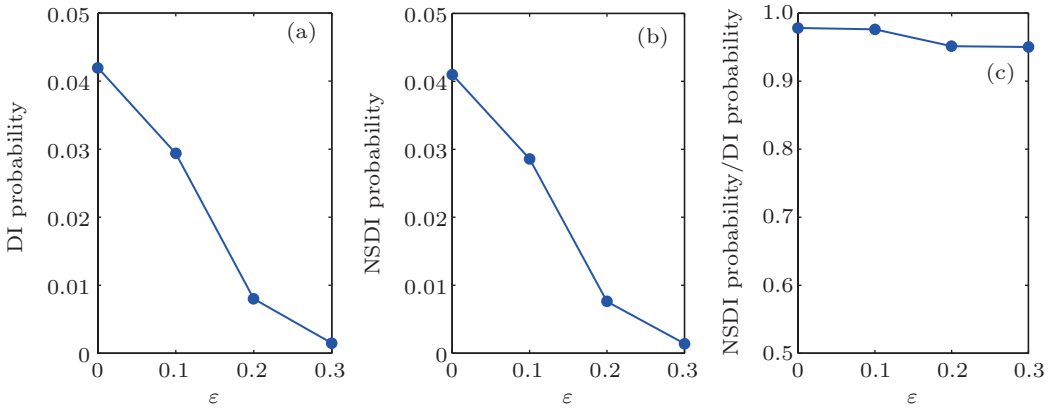


Fig. 1. (color online) (a) ε-dependent double ionization probability, (b) ε-dependent nonsequential double ionization probability, (c) ε-dependent ratio of nonsequential double ionization yield to double ionization yield.

The origin of the above suppression of DI and NSDI yields by elliptical laser field is analyzed as follows. As is well known, NSDI results from the recollision between the returning electron and the parent ion. Therefore, the NSDI probability is determined by SI probability, the rescattering probability and the recollision energy of the first ionized electron. Since almost all DI events belong to NDDI, DI probability is also determined by the above factors.

The ε-dependent SI probability is plotted in Fig. 2(a), which is insensitive to the ellipticity. As ellipticity increases, SI probability increases slightly. By analyzing a large number of SI trajectories, we find that in the elliptical laser field, the time window of SI increases slightly with the increasing of ellipticity, leading to the slight increase of SI probability with the increase of ellipticity. Compared with the other two factors, SI probability makes a negligible effect on NSDI probability. Based on the same statistical criterion as that for NSDI, the rescattering probability of the first ionized electron as a function of ε is plotted in Fig. 2(b), which decreases fast with ε increasing. The recollision energies obtained by back analysis are displayed in Fig. 3, in which curves are normalized to unity at maximum. From Fig. 3 it follows that there are two maximum recollision energies (about 1 a.u. and 1.5 a.u.) for each of all the ellipticity values. As ellipticity increases, the distribution of recollision energy around 1 a.u. increases while that around 1.5 a.u. decreases. By back analysis of DI trajectories, we find that the recollision energies around 1.5 a.u. and 1 a.u. come from recollisions at the first returning and the second returning respectively.

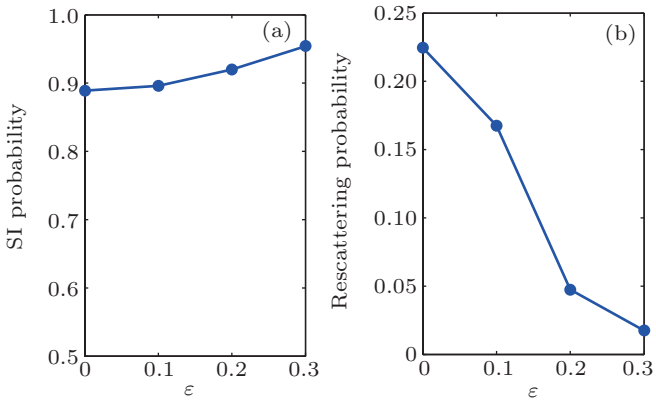


Fig. 2. (color online) (a) ε-dependent single ionization probability, (b) ε-dependent rescattering probability of the first ionized electron.

From the variation of above factors with the ellipticity, it is easy to know that the suppression of the DI and NSDI yields originates from the decrease of the rescattering probability and the recollision energy of the first ionized electron.

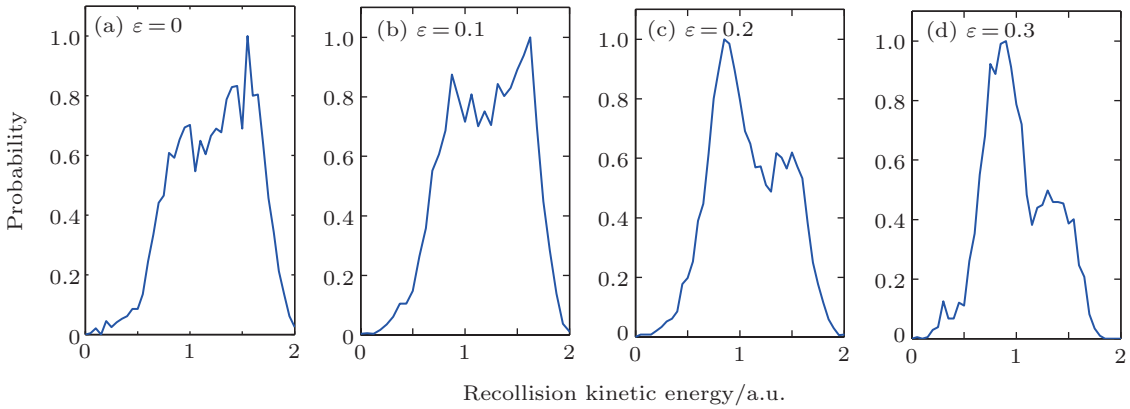


Fig. 3. (color online) Probabilities versus recollision kinetic energy of the first ionized electron for the values of ellipticity $\varepsilon = 0$ (a), 0.1 (b), 0.2 (c), and 0.3 (d). Curves are normalized to unity at maximum.

Figure 4 shows the correlated electron momentum distributions when the long axis of the laser polarization plane is parallel to the molecular axis (parallel alignment). The distributions of the upper row correspond to the correlated electron momentum in the direction parallel to the molecular axis, and those of the lower row correspond to the electron momentum in the direction perpendicular to the molecular axis. p_{xi} and p_{yi} represent the momentum components of the two electrons parallel and perpendicular to the molecular axis respectively, where the subscript $i = 1, 2$ is the electron label. The ellipticity values are 0, 0.1, 0.2, and 0.3 from left to right. In the direction parallel to the molecular axis (the x axis), the correlated electron momentum distribution for each of all the ellipticity values dominates the first and third quadrants, showing the same characteristic of strong correlation [see Figs. 4(a)–4(d)]. However, as the ellipticity increases, the populations of the correlated momenta move gradually to the second and fourth quadrants, showing that the correlation pattern weakens. In the direction perpendicular to the molecular axis (the y axis), the correlated momentum distributions are similar and dominate the second and fourth quadrants, showing an obvious anticorrelation pattern.

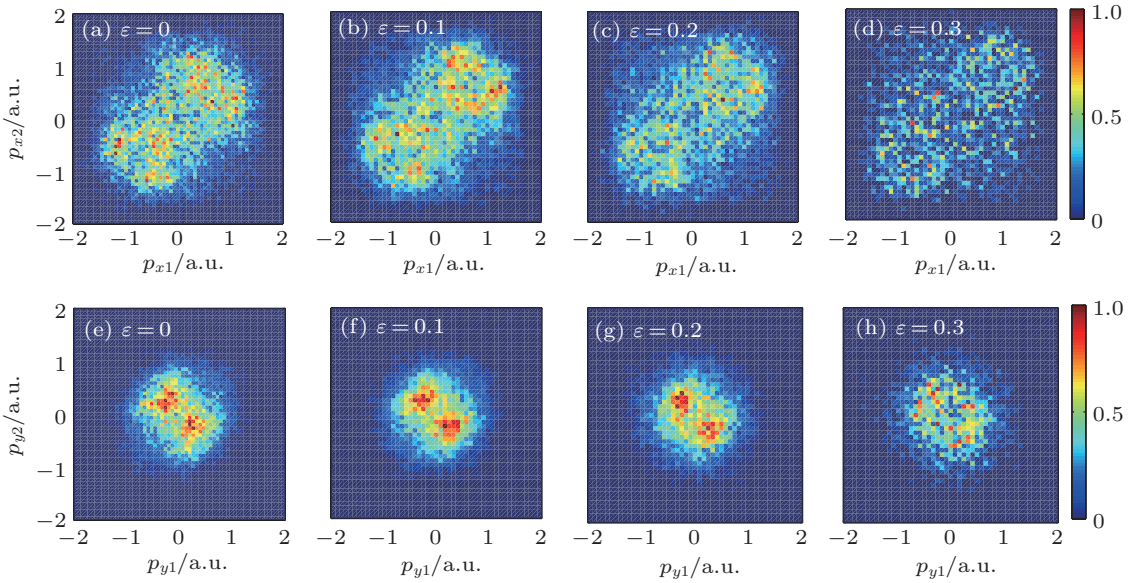


Fig. 4. (color online) Correlated electron momentum distributions of diatomic molecules along the x axis, i.e., the long axis of the laser polarization plane (upper row) and along the y axis, i.e., the short axis of the laser polarization plane (lower row) for the values of ellipticity $\varepsilon = 0$ ((a), (e)), 0.1 ((b), (f)), 0.2 ((c), (g)), and 0.3 ((d), (h)).

Back analysis of the DI trajectories is a valid method to reveal the relevant microscopic electron dynamics in the electron correlations.[33] By back analysis, we find the recollision time and the ionization time of DI event. For each DI trajectory, the recollision time is defined as the instant of the closest approach after first departure of one electron from the parent ion. The DI time is defined as the instant when the energy of an electron becomes positive for the first time.

Figure 5 shows the DI time versus recollision time for different ellipticity values. For linear polarization ($\varepsilon = 0$) [Fig. 5(a)], there are two dominant parts of the population. One is along the diagonal $t_{\rm DI} = t_r$, which implies that some DIs occur immediately after recollision, and the DI events belong to recollision ionization (RCI).[7] The other is that the final ionizations occur with a considerable time delay after recollision, belonging to recollision excitation with subsequent field ionization (RESI).[34] From Fig. 5 it follows that this correlation between DI time and recollision time changes with ellipticity. As the ellipticity increases, the population along the diagonal $t_{\rm DI} = t_r$ decreases. When $\varepsilon = 0.3$, almost no population populates on the diagonal and nearly all DIs belong to RESI [see Fig. 5(d)]. So, the contribution of RESI increases with the ellipticity increasing. As a result, more electron pairs emit into the opposite hemispheres as ellipticity increases.

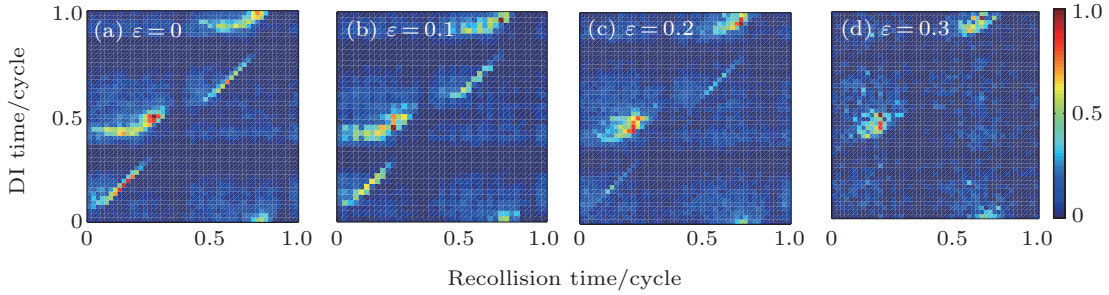


Fig. 5. (color online) Double ionization times versus recollision time (in laser cycle) for the values of ellipticity $\varepsilon = 0$ (a), 0.1 (b), 0.2 (c), and 0.3 (d).

To gain a comprehensive understanding of the evolutions of the electron–electron correlated momentum spectra when the long axis of the polarization plane is parallel to the molecular axis, we analyze the time lag between DI and recollision for different ellipticity values. Figure 6 shows the total fraction of double ionization versus delay time between DI and recollision. In Fig. 6, the delay time is plotted in units of optical cycle. For $\varepsilon = 0$, about 53% of the DI occurs after 0.25 cycle from recollision, which means that RCI and RESI make comparable contributions to the total DI event. This proportion increases to about 56% for $\varepsilon = 0.1$, 62% for $\varepsilon = 0.2$ and 66% for $\varepsilon = 0.3$, which demonstrates that more and more DIs occur through the RESI mechanism as ellipticity increases.

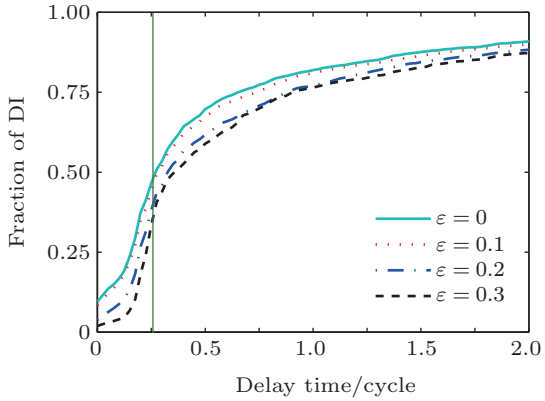


Fig. 6. (color online) Accumulated fraction of the double ionization events versus delay time between the closest recollision and the double ionization for parallel molecules with $\varepsilon = 0, 0.1, 0.2, 0.3$. The solid line (0.25 cycle) is drawn for guiding the eye.

We also calculate the double ionization of the model diatomic molecule when the long axis of the polarization plane is perpendicular to the molecular axis (perpendicular alignment). We change the molecular alignment by exchanging the directions of the long axis and short axis of the elliptically polarized laser pulses. In the perpendicular alignment, the effect of ellipticity on DI probability, i.e., DI mechanism is similar to in parallel alignment. As expected, for the same ellipticity, the DI probability in perpendicular alignment is smaller than in parallel alignment.

Finally, we investigate the alignment effect on electron correlation in an elliptically polarized laser field. We compare the electron correlations for $\varepsilon = 0$, 0.1, 0.2, and 0.3 in our calculations, here we use the case of $\varepsilon = 0.2$ as an example. Figure 7 displays the correlated momentum distributions for $\varepsilon = 0.2$ for both alignments. In each of Figs. 7(a) and 7(b), the long axis of the elliptical laser pulse is parallel to the molecular axis, while perpendicular to the molecular axis in each of Figs. 7(c) and 7(d). A comparison between Figs. 7(a) and 7(c) shows that more electron pairs emit into the second and fourth quadrants in perpendicular alignment than in parallel alignment. This phenomenon is consistent with the alignment effect in the linear laser field. From the comparison between Figs. 7(b) and 7(d) it follows that the anticorrelation pattern in Fig. 7(b) is more prominent than in Fig. 7(d).

The alignment effect on the electron correlation with $\varepsilon = 0.2$ can also be analyzed by the delay time between DI and recollision. Figure 8 shows the total fraction of double ionization versus delay time for the parallel and perpendicular alignments with $\varepsilon = 0.2$. Figure 8 shows that the fraction of DI occurring within a delay time of 0.25 cycle after recollision in the perpendicular alignment is obviously less than in the parallel alignment. So more DIs belong to RESI in the perpendicular alignment than in the parallel alignment, which is responsible for the above result that anticorrelated emissions in perpendicular alignment are more than in the parallel alignment.

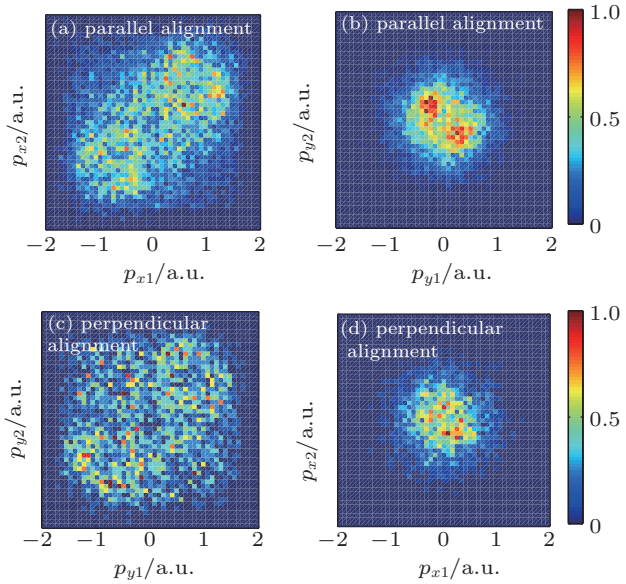


Fig. 7. (color online) Correlated electron momentum distributions of diatomic molecules for $\varepsilon = 0.2$ for both alignments: (a) and (b) the long axis of the elliptical laser pulse is parallel to the molecular axis, (c) and (d) the long axis of the elliptical laser pulse is perpendicular to the molecular axis.

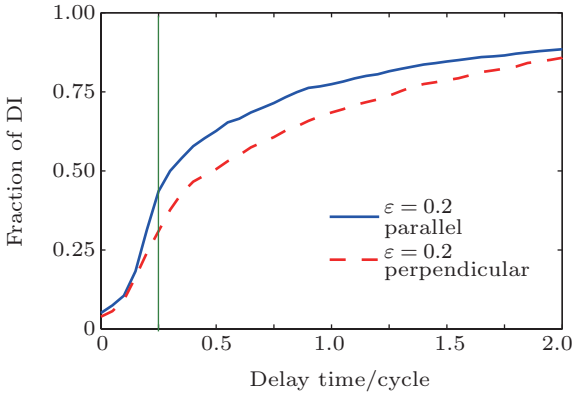


Fig. 8. (color online) Accumulated fractions of the double ionization event versus delay time between the closest recollision and the double ionization for parallel and perpendicular alignments with $\varepsilon = 0.2$. The solid line (0.25 cycle) is drawn for guiding the eye.

We further study the dependence of the alignment effect on the ellipticity of the laser field. In our calculations the ratios of anticorrelated emission for perpendicular molecules to that for parallel molecules are increased by 6.2%, 7.3%, 18.1%, and 35.6% for the values of ellipticity $\varepsilon = 0$, 0.1, 0.2, and 0.3 respectively. The data are obtained when the DI events from the two cases are normalized to the same number. n_1 and n_2 represent anticorrelated emission numbers from the perpendicular molecules and the parallel molecules respectively. The squares are the calculated data. The solid line is drawn for guiding the eye. Figure 9 shows the ellipticity dependence of the increase of anticorrelated emissions from the perpendicular molecules as compared with the parallel molecules. We can

easily find that the effect of molecular alignment on electron–electron momentum correlation increases with the ellipticity increasing.

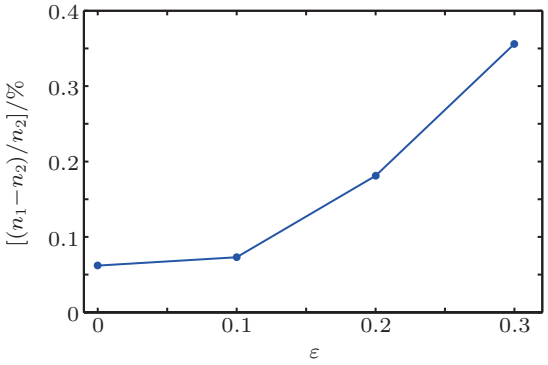


Fig. 9. (color online) Ellipticity dependence of the increase of anticorrelated emissions from the perpendicular molecules as compared with the parallel molecules. The data are obtained when the DI events from the two cases are normalized to the same number. n_1 and n_2 represent anticorrelated emission numbers from the perpendicular molecules and the parallel molecules respectively. The solid circles denote the calculated data, and the solid line is drawn for guiding the eye.

4. Conclusion

In this paper, using the classical ensemble model, we investigated DIs of parallel and perpendicularly aligned diatomic molecules by elliptically polarized laser pulses. We found that DI is commonly dominated by NSDI while NSDI yields decrease fast with the ellipticity increasing, which originates from the decrease in rescattering probability and the recollision energy. Moreover, NSDI channel changes with ellipticity: the contribution from RESI increases as ellipticity increases. Additionally, we explored the ellipticity dependence of the alignment effect on electron correlation, which increases as ellipticity increases.

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