Theoretical study on non-sequential double ionization of carbon disulfide with different bond lengths in linearly polarizedlaser fields

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By using a two-dimensional Monte-Carlo classical ensemble method, we investigate the double ionization (DI) process of the CS_2 molecule with different bond lengths in an 800-nm intense laser field. The double ionization probability presents a "knee" structure with equilibrium internuclear distance $R = 2.9245$ a.u. (a.u. is short for atomic unit). As the bond length of CS increases, the DI probability is enhanced and the "knee" structure becomes less obvious. In addition, the momentum distribution of double ionized electrons is also investigated, which shows the momentum mostly distributed in the first and third quadrants with equilibrium internuclear distance $R = 2.9245$ a.u. As the bond length of CS increases, the electron momentum becomes evenly distributed in the four quadrants. Furthermore, the energy distributions and the corresponding trajectories of the double-ionized electrons versus time are also demonstrated, which show that the bond length of CS in the CS_2 molecule plays a key role in the DI process.

Keywords: intense laser field, non-sequential double ionization, classical ensemble method, the bond length

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1. Introduction

The non-sequential double ionization (NSDI) has attracted considerable interest since it can provide a better understanding of laser–matter interaction and electron correlation.[1–5] Recently, the recollision model[6] is widely accepted to describe the ionization events: one electron is free through tunneling ionization; then the free electron is accelerated in the laser fields; finally, when the direction of the field is reversed, the free electron revisits the core to release the second electron by collision.[7,8] According to the recollision mechanism, the atomic non-sequential double ionization process[9] in strong laser fields can occur either by recollision impact ionization (RII) or by recollision excitation with subsequent ionization (RESI).[10]

Compared to atoms, the mechanism of the ionization of polyatomic molecules is much more complex in laser fields because of their diverse molecular structure and additional nuclear degree of freedom. The evidences showed that NSDI also exists for molecules.[11–13] The rescattering picture was also used to explain the molecular NSDI process. Theoretical studies have been investigated with the simple diatomic molecules,[14–19] linearly triatomic molecules,[20–23] and even more complicated polyatomic molecules.[24]

Recently, the model of the CS_2 molecule has been successfully used[25] and the theoretical result is in excellent agreement with experimental data. Based on the above description, we investigate the DI process of two-dimensional CS_2 with different bond lengths by using the classical ensemble method. It is demonstrated that the probability of the double ionization increases with the increasing bond length. Besides, the corresponding momentum distribution of doubly ionized electrons shows that the momentums are mostly distributed in the first and third quadrants with equilibrium internuclear distance $R = 2.9245$ a.u. As the bond length increases, the electron momentum is evenly distributed in the four quadrants. Moreover, the time evolution of energies and the corresponding trajectories of the double-ionized electrons are also illustrated.

2. Theoretical model

The classical ensemble method proposed by Haan and Eberly *et al.* has been widely used to study the DI of atoms in intense laser fields.[26,27] In this study, we use this method to investigate the DI dynamics of the CS_2 molecule in linearly polarized laser fields. In our calculation, the molecular axis is along the x axis. By using the two-electron approximation, the classical Hamiltonian of the CS_2 molecule (with Born–Oppenheimer approximation) in an intense laser field can be given by

$$H(\boldsymbol{r}_1, \boldsymbol{r}_2; \boldsymbol{P}_1, \boldsymbol{P}_2; t) = \mathscr{T}(p) + \mathscr{V}(q,t), \tag{1}$$

where the kinetic energy $\mathscr{T}$ and the potential energy $\mathscr{V}$ are given by

$$\mathscr{T}(p) = \frac{P_1^2}{2} + \frac{P_2^2}{2}, \tag{2}$$

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$$\mathscr{V}(q,t) = -\sum_{i=1}^{2}\frac{Z_c}{\sqrt{x_i^2+y_i^2}} - \sum_{i=1}^{2}\frac{Z_s}{\sqrt{(x_i-R)^2+y_i^2}} - \sum_{i=1}^{2}\frac{Z_s}{\sqrt{(x_i+R)^2+y_i^2}} + \frac{1}{|\boldsymbol{r}_1-\boldsymbol{r}_2|} + (\boldsymbol{r}_1+\boldsymbol{r}_2)\boldsymbol{E}(t), \quad (3)$$

where $q=(\boldsymbol{r}_1,\boldsymbol{r}_2)$ represents the position of the two electrons, and $p=(\boldsymbol{P}_1,\boldsymbol{P}_2)$ stands for the corresponding conjugate momenta. $E(t)$ is the external electric field. $Z_s=2/3$ and $Z_c=2/3$ are effective nuclear charge. R is the bond length of CS. The subscript $i=1,2$ represents two electrons, respectively; x_i, y_i are the x- and y axes of the electrons, respectively. Here the soft core potential is used to remove the singularity as follows:

$$\mathscr{V}(q,t) = -\sum_{i=1}^{2}\frac{Z_c}{\sqrt{x_i^2+y_i^2+a_c^2}} - \sum_{i=1}^{2}\frac{Z_s}{\sqrt{(x_i-R)^2+y_i^2+a_s{}^2}} - \sum_{i=1}^{2}\frac{Z_s}{\sqrt{(x_i+R)^2+y_i^2+a_s{}^2}} + \frac{1}{\sqrt{(x_1-x_2)^2+(y_1-y_2)^2+q_e^2}} + (\boldsymbol{r}_1+\boldsymbol{r}_2)\boldsymbol{E}(t), \quad (4)$$

where the soft core parameters are $a_s=1.2$, $a_c=1.4$, and $q_e=0.05$.

The canonical system of equations for the CS_2 molecule is

$$\frac{\mathrm{d}p}{\mathrm{d}t} = -\frac{\partial\mathscr{V}(q,t)}{\partial q}, \quad (5)$$

$$\frac{\mathrm{d}q}{\mathrm{d}t} = \frac{\partial\mathscr{T}(p)}{\partial p}. \quad (6)$$

The symplectic method is suitable for the long-time and many-step calculations and preserves the symplectic structure of the system. We can obtain the time evolution of the electron position and the corresponding momenta by solving the above canonical equations numerically with the symplectic method. Since the Hamiltonian system (1) is a separable Hamiltonian system in the sense that q and p are contained separately in $\mathscr{V}(q,t)$ and $\mathscr{T}(p)$. We may use an explicit symplectic scheme to solve the corresponding Hamiltonian equation in order to obtain the classical trajectories of electrons in the CS_2 molecule in linearly polarized fields. For instance, we can integrate the Hamiltonian canonical equation by adopting the four-stage fourth-order explicit symplectic scheme.[28–30]

In the field-free case, the system has the total energy (E_{tot}) whose negative value is approximately equal to the sum of the first and second ionization energies of the CS_2 molecule. For each R, we calculated the total energy, the first and second ionization energies of the CS_2 molecule by using the coupled-cluster singles and doubles model (CCSD), as listed in Table 1. To obtain the initial classical ensemble, the two-electron system is allowed to evolve a sufficiently long time in the absence of the laser field.[26]

Table 1. The first and second ionization energies of CS_2 (I_{p1}, I_{p2}) and the total energy of two electrons of CS_2 (E_{tot}) with different bond lengths (R).

R	2.9245 a.u.	3.3962 a.u.	3.7735 a.u.
I_{p1}	0.62198	0.60606	0.5925
I_{p2}	0.37178	0.36854	0.3633
E_{tot}	−0.99376	−0.9746	−0.9558

3. Results and discussion

In our calculation, the linearly polarized electric field is chosen as $\boldsymbol{E}(t)=E_0 f(t)\sin(\omega_0 t)$, where $\omega_0=0.057$ a.u. is the laser frequency; E_0 is the maximum field strength of the linearly polarized electric field; $f(t)=\sin^2(\pi t/T_0)$ is the laser envelope; the pulse duration is 4 o.c. (optical cycle). Figure 1 shows the DI probability of the CS_2 molecule as functions of the laser intensity with different bond lengths in linearly polarized laser fields. As shown in Fig. 1, we can see that the double ionization probability of the CS_2 molecule in the range of 15 TW/cm^2 to 150 TW/cm^2 with equilibrium internuclear distance is in good agreement with experimental data. As the bond length increases, the DI probability also increases. Besides, there exists a "knee" structure with the equilibrium internuclear distance $R=2.9245$ a.u. around the laser intensity 0.05 PW/cm^2. This means that some ionized electrons can come back to the core and collide with the inner one, which leads to the NSDI process. However, we find that the "knee" structure becomes less obvious with the increasing bond length.

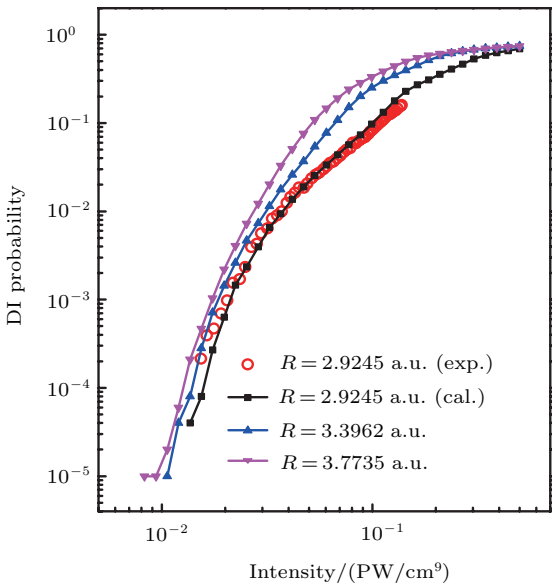


Fig. 1. (color online) The double ionization probability of the CS_2 molecule as a function of laser intensity in linearly polarized laser field with bond length $R=2.9245$ a.u. (black curve with squares), 3.3962 a.u. (blue curve with up triangles), 3.7736 a.u. (pink curve with down triangles), respectively. The curve with red rings is the experimental data. The wavelength of the linearly polarized laser field is 800 nm.

To explain the influence of bond length of CS on the DI process, in Fig. 2 we plot potential energy curves of the CS_2 molecule

$$V_{\rm ne}(x_1) = -\frac{Z_{\rm c}}{\sqrt{x_1^2 + a_{\rm c}^2}} - \frac{Z_{\rm s}}{\sqrt{(x_1 - R)^2 + {a_{\rm s}}^2}} - \frac{Z_{\rm s}}{\sqrt{(x_1 + R)^2 + {a_{\rm s}}^2}} + x_1 E_0$$

with $E_0 = 0.038$ a.u. (E_0 is the maximum field strength of the laser with intensity $I = 0.05$ PW/cm^2) for $R = 2.9245$ a.u., 3.3962 a.u., and 3.7735 a.u., respectively. We can see from Fig. 2 and Table 1 that as the bond length of CS increases, the external suppressed potential barrier becomes lower and the second ionization energy also decreases, the second electron is easier to be pulled out by the external field with the bond length increasing. Consequently, the probability of the double ionization also increases.

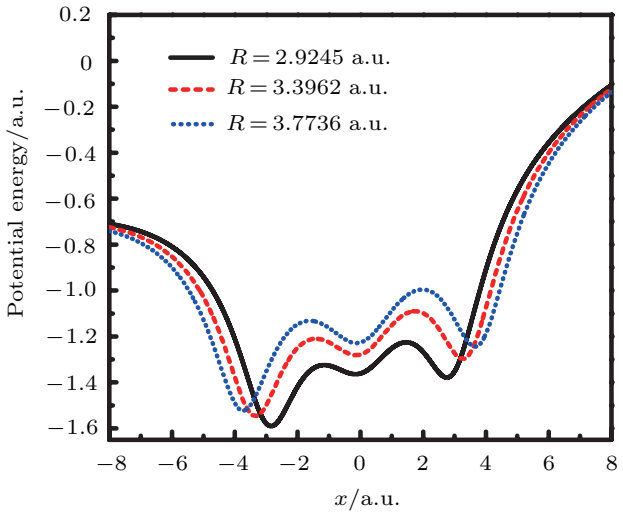


Fig. 2. (color online) The soft-core Coulomb potential of the CS_2 molecule along the laser polarization with bond lengths $R = 2.9245$ a.u. (black solid curve), 3.3962 a.u. (red dashed curve), 3.7736 a.u. (blue dotted curve), respectively. The laser intensity is 0.05 PW/cm^2.

Figure 3 shows the correlated momentum distribution of two electrons in linearly polarized laser fields with different bond lengths ($R = 2.9245$ a.u., 3.396 a.u., and 3.7735 a.u.). In the sequential double ionization (SDI) process, two electrons ionize independently, and the probability of electrons emitted to the same direction is supposed to be equal to that emitted to the opposite direction.[29] In the NSDI process, because of the recollision in the DI process, there is a big chance that both electrons are emitted at almost the same time and fly into the same direction.[31] Thus, we can see from Fig. 3 that the correlated momentum distribution is mostly distributed in the first and third quadrants, and presents a "finger-like" structure, which indicates that the NSDI process is predominant. As the bond length increases, the momentum of the two electrons becomes evenly distributed in the four quadrants [see Figs. 3(b) and 3(c)], thus the NSDI process is not the dominant process.

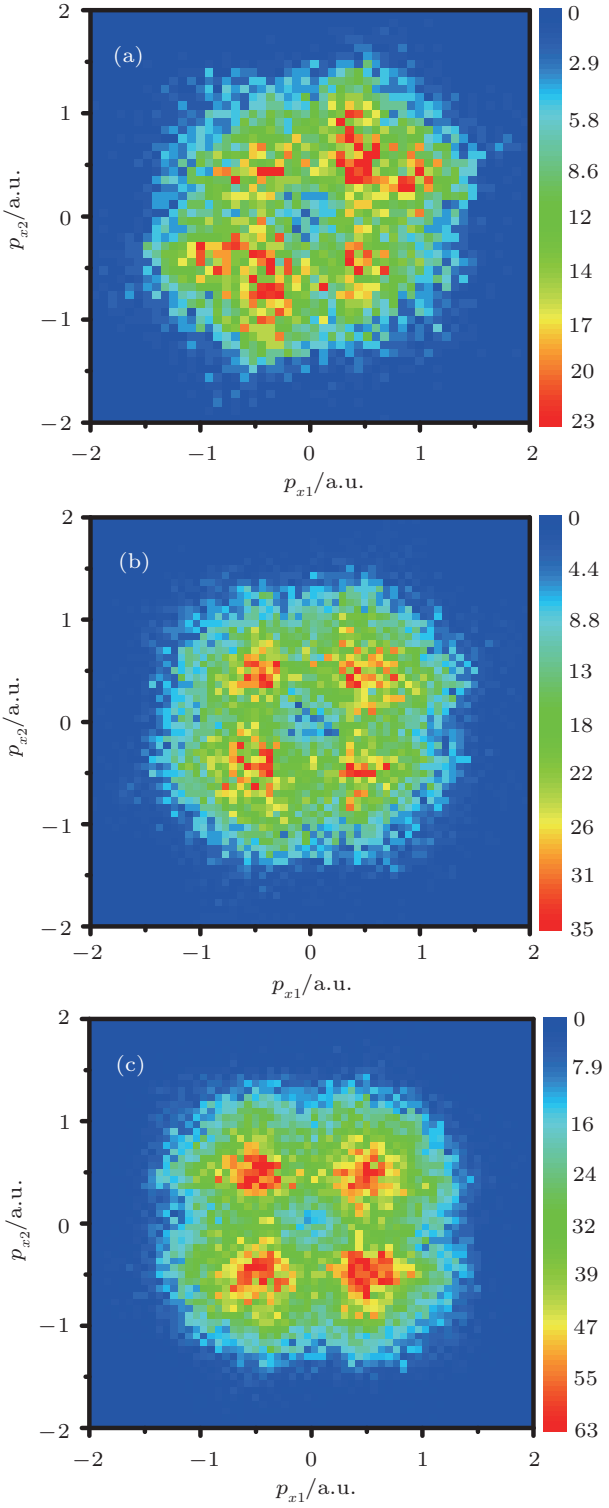


Fig. 3. (color online) The correlated momentum distribution of the CS_2 molecule along the x direction (molecular axis) for different bond lengths: (a) $R = 2.9245$ a.u., (b) $R = 3.396$ a.u., and (c) $R = 3.7735$ a.u., respectively. The wavelength of the linearly polarized laser field is 800 nm.

Figure 4 shows the energy and repulsion energy of the doubled ionized electrons as functions of time for the cases of the bond lengths $R = 2.9245$ a.u. and $R = 3.7735$ a.u. [shown in Figs. 4(e) and 4(f)], respectively. Panels (a), (c), and (e) show the time evolution of the energy distribution of the double ionized electrons. We can see clearly three stages: (i) there is an initial period of a steady distribution at the nega-

tive energy region, corresponding to pre-ionization where the two electrons exchange energy frequently; (ii) the distribution splits into two branches, one of which emerges from the negative energy region while the other drops to a lower energy region, which depicts the first ionization process, with one electron escaped and an ion core formed; and (iii) after a period of time, the remaining distribution also emerges from the negative energy region, which corresponds to the second ionization. In addition, we can also see a widely oscillatory distribution, which corresponds to the jitter motion of a free electron in laser fields. It is shown that, with the bond length of the CS increasing, the recollision becomes weaker [see Figs. 4(c) and 4(e)] and the number of the energy peak is decreased. By comparing Figs. 4(a), 4(c), and 4(e) together, we can see that the NSDI with the equilibrium internuclear distance $R = 2.9245$ a.u. [shown in Fig. 4(a)] is more obvious than that with $R = 3.396$ a.u. and $R = 3.7735$ a.u. [shown in Figs. 4(c) and 4(e)].

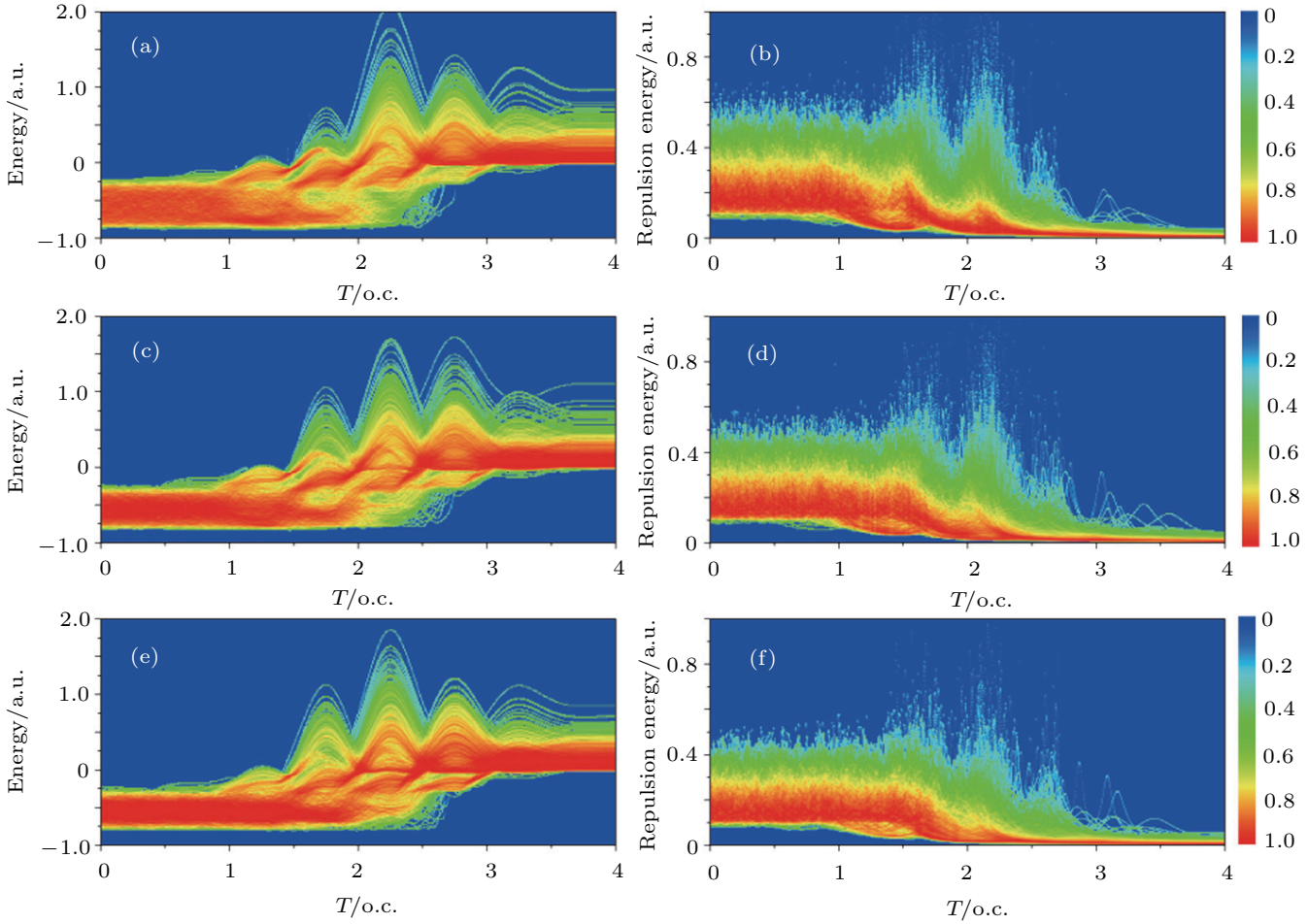


Fig. 4. (color online) The electron energy distribution and the repulsion energy of double ionized electrons in CS_2 molecule as a function of time for different bond lengths. (a) and (b) $R = 2.9524$ a.u.; (c) and (d) $R = 3.396$ a.u.; and (e) and (f) $R = 3.7735$ a.u., respectively. The wavelength of the linearly polarized laser field is 800 nm.

Figures 4(b), 4(d), and 4(f) show the repulsion energy distributions between the doubly ionized electrons as functions of time. Firstly, the repulsion energy oscillates from 0.15 a.u. to 0.6 a.u. Then the repulsion energy decreases, which means that the single ionization occurs. After that, the repulsion energy becomes comparable with the repulsion energy around 1 o.c., which means that the ionized electron returns to the ion core. In comparison with the case of the equilibrium internuclear distance $R = 2.9245$ a.u. [shown in Fig. 4(b)], we can see that as the bond length of the CS increases, the number of the ionized electrons which return to cores is reduced, and the correlation between the two electrons becomes weaker. When the bond length is 3.7735 a.u. [see Fig. 4(f)], the repulsion between the two electrons after the single ionization becomes the weakest.

In order to distinguish the energy spectrum and the repulsion energy distribution with different bond lengths, in the following Fig. 5 we pick up all the recollision trajectory of the double ionization. We can see that the recollision is more obvious in the case of $R = 2.9245$ a.u. than in the cases of $R = 3.396$ a.u. and $R = 3.7735$ a.u., which means that the strong correlation between the two electrons exists at $R = 2.9245$ a.u. However, the number of the recollision trajectories decreases with the increasing R. Thus, as R increases, the dominant double ionization channel changes from the non-sequential double ionization to the sequential double ionization.

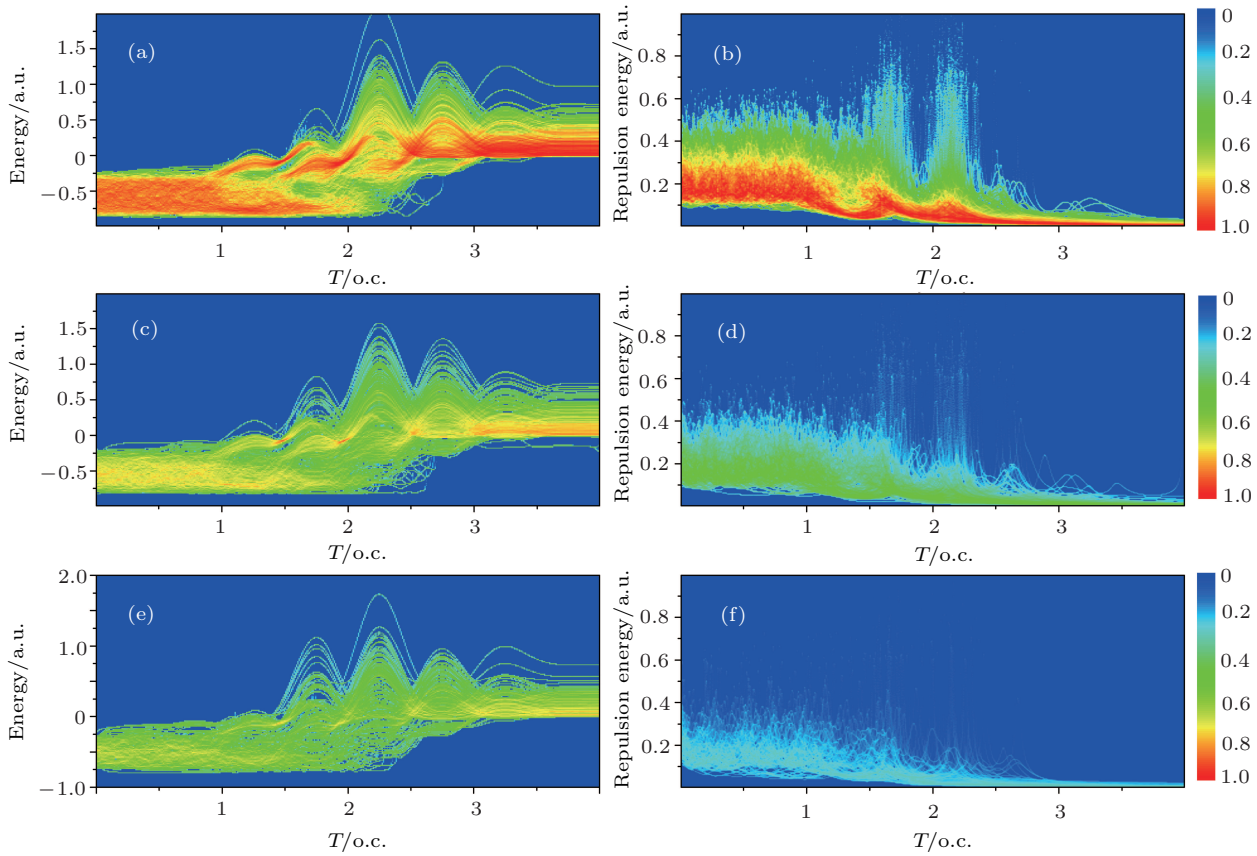
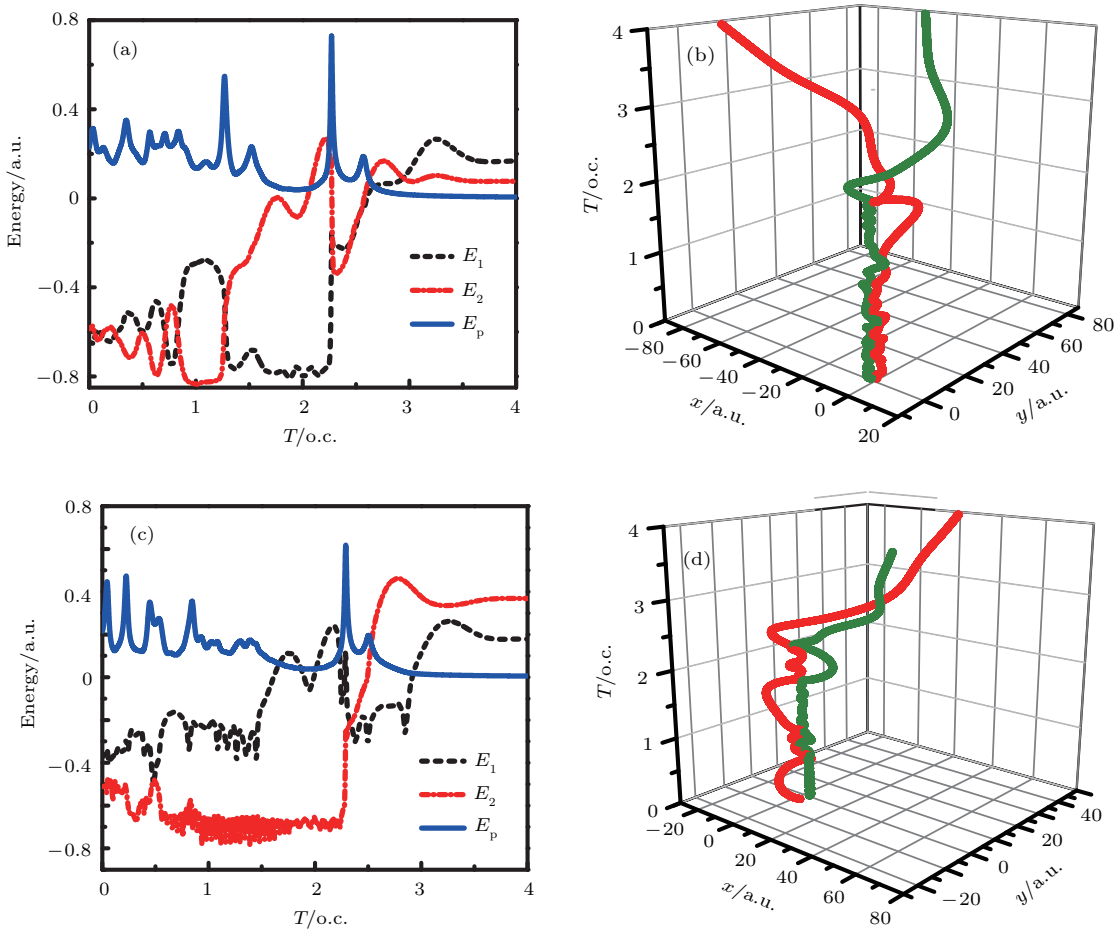


Fig. 5. (color online) The electron energy distribution and the repulsion energy of all the recollision trajectories of double ionized electrons in the CS_2 molecule as functions of time for different bond lengths. (a) and (b) $R = 2.9524$ a.u.; (c) and (d) $R = 3.396$ a.u.; and (e) and (f) $R = 3.7735$ a.u., respectively. The wavelength of the linearly polarized laser field is 800 nm.



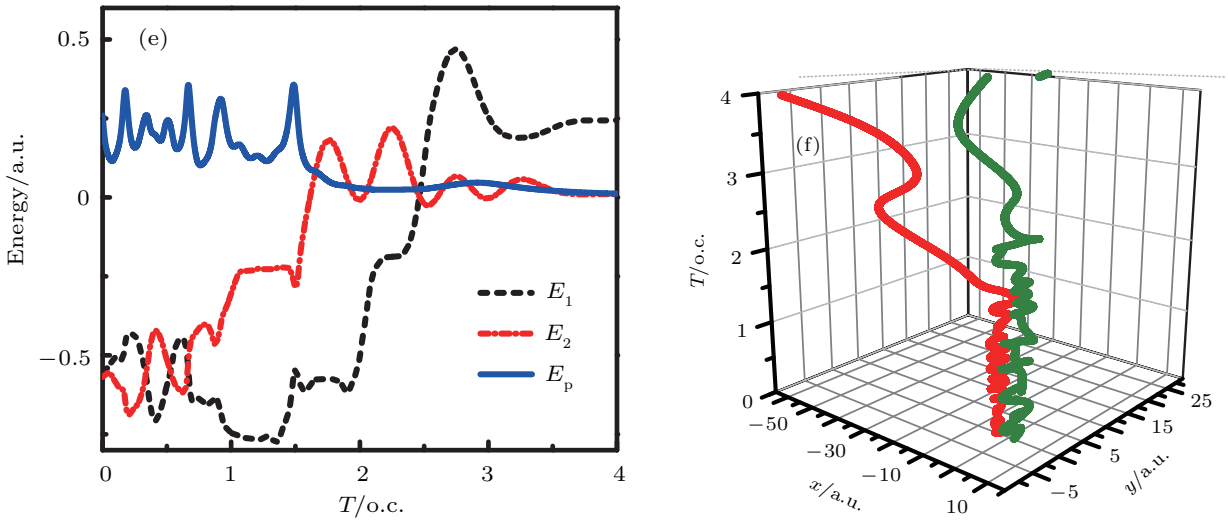


Fig. 6. (color online) The presentive examples of energy and position distribution of the double ionized electrons in the CS_2 molecule as a function of time for different bond lengths. Panels (a) and (b) for $R = 2.9245$ a.u.; (c) and (d) for $R = 3.396$ a.u.; (e) and (f) for $R = 3.7735$ a.u. in linearly polarized laser fields. (Left column) E_1, E_2 represent the energies of the first electron (black dashed curve), the second electron (the red dashed and dotted curves), and the E_p represents the repulsion energies between two electrons (blue solid curve). (Right column) The positions of the first electron (red curve) and the second electron (green curve) are shown. The laser wavelength is 800 nm.

Figure 6 shows the presentative examples of energies and the corresponding trajectories of the double ionized electrons with increasing bond lengths as functions of time. The E_1 and E_2 represent the energies of the two electrons. The E_p represents the Coulomb repulsive energy between the two electrons. If the recollision occurs at time t, there must be a peak in E_p at the same time due to the e–e correlation. From Fig. 6(a) ($R = 2.9245$ a.u.), we can find a peak in E_p at about 2.3 o.c., which means the recollision between the two electrons occurs, which is also in accordance with the electrons' trajectories in Fig. 6(b). As the bond length of CS increases, the correlation between the two electrons becomes weaker but the recollision excitation with subsequent ionization still exists for $R = 3.396$ a.u. [as shown in Figs. 6(c) and 6(d)]. As the bond length increases up to 3.7735 a.u. [as shown in Figs. 6(e) and 6(f)], the E_p is very weak around the ionization time, which indicates that the two electrons ionize one after another subsequently.

4. Conclusions

In summary, we investigated the NSDI process of CS_2 with different bond lengths in linearly polarized laser fields by using the classical ensemble method. By analyzing the double ionized probability as functions of the laser intensity and the potential curves of the CS_2 molecule with different bond lengths, we can find a "knee" structure with equilibrium internuclear distance $R = 2.9245$ a.u., which is in good agreement with the experimental result. As the bond length of CS increases, the probability of the double ionization increases and the "knee" structure becomes less obvious. The momentum distribution of the two electrons is mostly distributed in the first and third quadrants at the equilibrium internuclear distance $R = 2.9245$ a.u.; however, for $R = 3.7735$ a.u., the momentum distribution is distributed evenly in the four quadrants. In addition, by analyzing the energy distributions and the corresponding trajectories of the double ionized electrons versus time, we find that as R increases, the dominant double ionization channel changes from the non-sequential double ionization to the sequential double ionization in linearly polarized fields.

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