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Exploration of strong-field double ionization of C_3H_6 with the structures of propene and cyclopropane in intense laser fields*

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By using classical ensemble method, we investigate the double ionization of C_3H_6 molecule with different structures (propene and cyclopropane) in intense laser fields. The numerical results show that the non-sequential double ionization occurs in propene molecule rather than cyclopropane molecule in 1200 nm laser field. To further explain this interesting phenomenon, the momentum distribution of double ionized electrons is presented and the result presents the "finger-like" structure at about 30 TW/cm^2 of propene molecule, and this structure is more obvious than that in cyclopropane molecule. The above phenomena are also demonstrated by analysing the energy distributions of double-ionized electrons versus time. Moreover, we also investigated the angular distribution at the end of pulse, which is different between propene and cyclopropane.

Keywords: C_3H_6 molecule, non-sequential double ionization, momentum distribution

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1. Introduction

The physical sight of double ionization processes of atoms and molecules has attracted both practical and fundamental interest.[1–3] Dynamics of simple systems has been studied thoroughly both theoretically and experimentally.[4,5] Corkum[6] proposed a rescattering model that one electron ionizes first and revisits the core to let the second electron free by collision, which is widely accepted to explain the non-sequential double ionization (NSDI). With the development of laser technology, the tracing of electron motion in atoms and molecules in strong-field also has been a hot topic recently and its physical insight still needs further exploration.[7,8] Recently, it turns out that semi-classical or classical simulations are useful to treat the strong-field ionization with very strong two-electron correlation, such as NSDI.[9–14,21]

Comparing with atoms, molecules exhibit much more complicated processes in strong-field ionization, owing to their diverse molecular structure and additional nuclear degree of freedom. The underlying mechanism of molecular double ionization (DI) is still not clearly understood, which stimulates further investigations on the topic. There exists evidence that has shown that NSDI is also contributed to DI of molecules when subjected to strong laser fields. The "knee" structure, which is viewed as a signature of NSDI of atoms, has been observed in simple diatomic[15–18] and linear triatomic molecules,[19–21] and even more complicated polyatomic molecules.[22] Bandrauk *et al.*[23,24] also investigated the HHG and ionization process of symmetric and unsymmetric molecules. Moreover, the bichromatic counterrotating circularly polarized laser fields attracts a lot of attention and there have many excellent works on the dynamics of atoms and molecules in the bichromatic counterrotating circularly polarized laser fields.[25–30]

Thus, in this paper, we will investigate the double ionization mechanism of C_3H_6 with different structures and compare them together. We will illustrate how non-sequential double ionization of propene happens at 1200 nm intense laser fields. Particularly, we will investigate the corresponding momentum and energy distributions of doubly ionized electrons to study the DI process of propene and cyclopropane molecule.

2. Theoretical model

In this paper we use the classical ensemble method, which has previously been used[11,12,21] successfully to explore the ionization dynamics of atoms and three-atom molecule CS_2[31] in intense laser fields. In C_3H_6 with different structures, the bond angle between C–C–C are 109.5 degree and 60 degree in propene and cyclopropane, respectively. The structure of cyclopropane is shown in Fig. 1(a). The triangular structure of cyclopropane requires the bond angles between carbon–carbon bonds to be 60 degree. This is far less than the thermodynamically most stable angle of 109.5 degree (for bonds between atoms with sp3 hybridised orbitals) and leads to significant ring strain. The molecule also has torsional strain due to the eclipsed conformation of its hydrogen atoms. As such, the bonds between the carbon atoms are consider-

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ably weaker than in a typical alkane, resulting in much higher reactivity.

Propene, also known as propylene or methyl ethylene, is an unsaturated organic compound having the chemical formula C_3H_6, which structure is shown in Fig. 1(b). It has one double bond, and is the second simplest member of the alkene class of hydrocarbons. Propene is produced from fossil fuels petroleum, natural gas, and, to a much lesser extent, coal. Propene is a byproduct of oil refining and natural gas processing. During oil refining, ethylene, propene, and other compounds are produced as a result of cracking larger hydrocarbon molecules to produce hydrocarbons more in demand. A major source of propene is naphtha cracking intended to produce ethylene. Propene can be separated by fractional distillation from hydrocarbon mixtures obtained from cracking and other refining processes. Refinery-grade propene is about 50 to 70 percent.

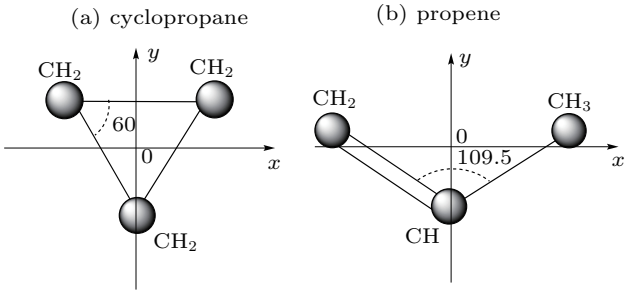


Fig. 1. The structures of cyclocropane and cropene.

In our calculation, the coordinate of mass center is used and the classical Hamiltonian of C_3H_6 molecules in an intense laser field can be given by (atomic units are used throughout unless otherwise stated)

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

where $\mathcal{T}$ is the kinetic energy and $\mathcal{V}$ is the potential energy of C_3H_6, which is different between cyclopropane and propene.

The canonical system of equations for CS_2 molecule is

$$\frac{\mathrm{d}p}{\mathrm{d}t} = -\frac{\partial \mathcal{V}(q,t)}{\partial q}, \tag{2}$$

$$\frac{\mathrm{d}q}{\mathrm{d}t} = \frac{\partial \mathcal{T}(p)}{\partial p}. \tag{3}$$

The symplectic method is the difference method that preserves the symplectic structure and especially suitable for the long-time many-step calculations. We choose a set of initial stable states $\{r_i(0),P_i(0)\}_{i=1}^{2}$ by the Monte Carlo method and solved the above canonical equations numerically in order to obtain the time evolutions of the electron positions and the corresponding momenta $\{r_i(t),P_i(t)\}_{i=1}^{2}$. Since the Hamiltonian system (1) is a separable Hamiltonian system in the sense that q and p are contained separately in $\mathcal{V}(q,t)$ and $\mathcal{T}(p)$, and the Hamiltonian function contains the time variable, we may use the four-stage fourth-order explicit symplectic scheme to solve it, so that we can obtain the classical trajectories of C_3H_6 molecule in an intense laser field.[16]

In our calculation, we assume that the initial condition has the same energy approximately equal to the sum of first and second ionization energy of C_3H_6 molecule (0.7864 for cyclopropane and 0.7945 for propene). Comparing with the molecular structure of the hydrogen molecule (0.5668 for H_2, 0.56837 for D_2, and 0.56756 for HD),[32] the solution of C_3H_6 molecule is a little bit bigger than the ionization energy of hydrogen molecules. Besides, many researchers have obtained values for the dissociation energies of C_3H_6 molecule (0.3748 for cyclopropane and 0.3546 for propene) and of hydrogen molecule (0.5393 for H_2, 0.5423 for D_2, and 0.5406 for HD) by comparison.[32,33] Obviously, the dissociation energies of C_3H_6 molecule is a little bit smaller than hydrogen molecules. We defined the energy of each electron as $E_1(t)$ and $E_2(t)$, respectively. If both $E_1(t)$ and $E_2(t)$ are greater than zero at the end of the laser pulse, the double ionization occurs.

3. Results and discussion

We investigate the ionization process of the C_3H_6 molecule in intense laser field. The linearly polarized electric field is chosen as $\boldsymbol{E}_1(t) = E_0 f(t)\sin(\omega_0 t)$. The counter-rotating field $(\omega+2\omega)$ is expressed as $\boldsymbol{E}_2(t) = E_0 f(t)\hat{x}[\cos(\omega_0 t)+\cos(2\omega_0 t)]+E_0 f(t)\hat{y}[\sin(\omega_0 t)-\sin(2\omega_0 t)]$, and the counter-rotating field $(\omega+3\omega)$ is $\boldsymbol{E}_3(t) = E_0 f(t)\hat{x}[\cos(\omega_0 t)+\cos(3\omega_0 t)]+E_0 f(t)\hat{y}[\sin(\omega_0 t)-\sin(3\omega_0 t)]$, where ω_0 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, and the pulse duration is 4 optical cycle, shown in Fig. 2. In this work, we utilize a micro-canonical ensemble which consists of 1×10^6 two-electron "trajectories".

Figure 3 shows the double ionization probability of propene and cyclopropane molecule as a function of laser intensity. We can see that in 800 nm and 1200 nm laser fields the ionization probability in counter-rotating field is about 2 order higher than that in linearly polarized field. Figure 3(a) presents that in the 800-nm case the ionization probability of cyclopropane and propene increases as the increasing laser intensity and does not have any "knee" structure for all the laser fields. However, in Fig. 3(b) we can see for the 1200-nm case a remarkably "knee" structure of propene and cyclopropane molecule in intense laser field at the range of 20–60 TW. Besides, the ionization probability of propene is larger than cyclopropane for all the intensity we considered. Thus, we choose the wavelength of 1200 nm and the intensity of 30 TW to further explore the underlying DI mechanism in cyclopropane and propene molecule.

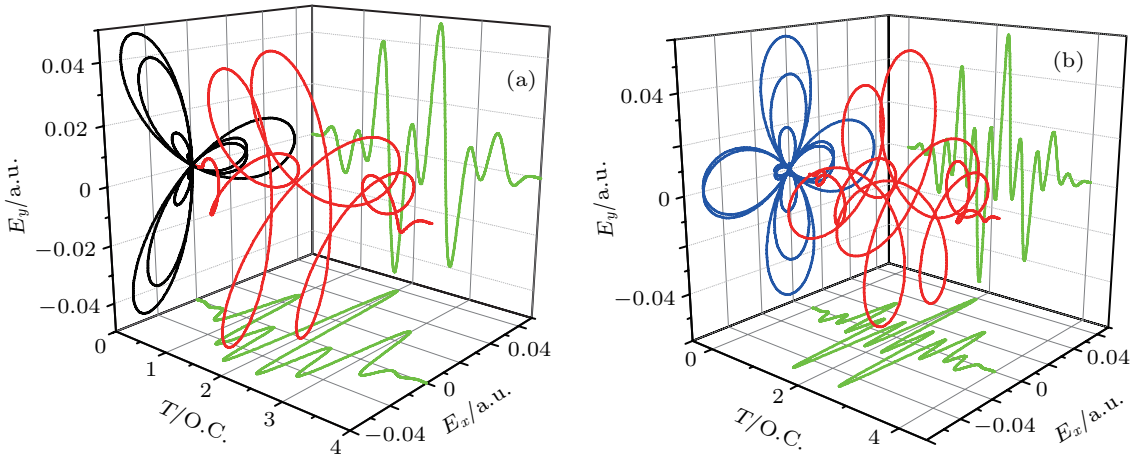


Fig. 2. (color online) The frame of counter-rotating laser fields with $\omega+2\omega$ and $\omega+3\omega$, respectively. The laser wavelength is 1200 nm.

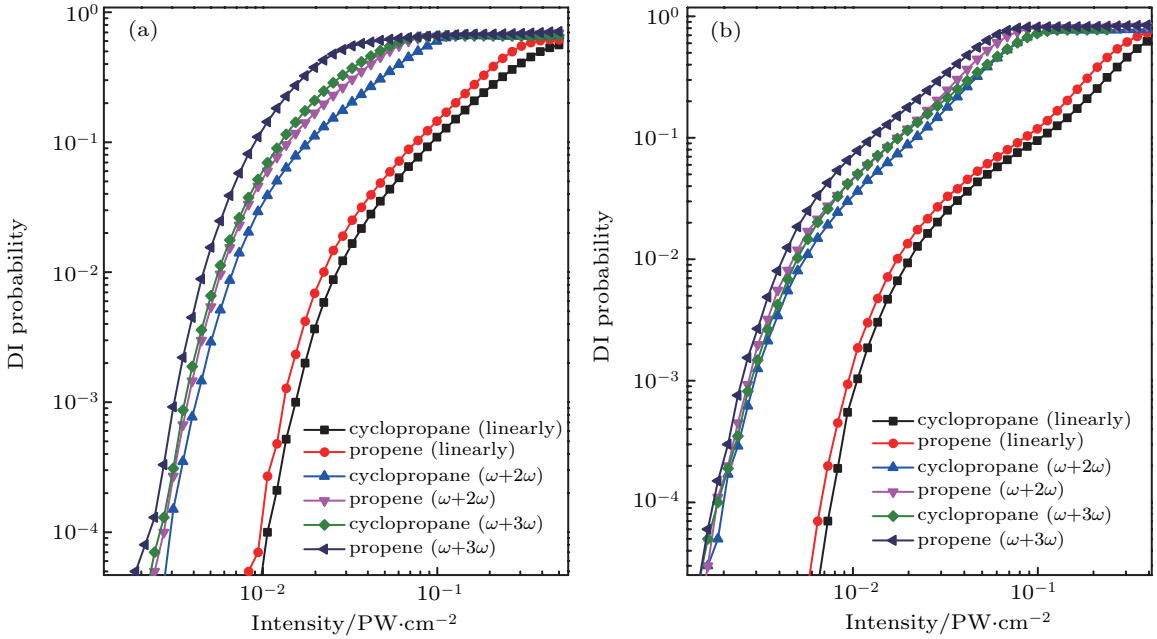


Fig. 3. (color online) The double ionization probability of C_3H_6 with the structure of cyclopropane and propene as a function of laser intensity in (a) 800 nm and (b) 1200 nm intense laser fields.

In order to further understand or identify whether the non-sequential double ionization mechanism of C_3H_6 molecule exists, we calculated the corresponding momentum distribution of correlated electrons of C_3H_6 molecule with the structures of cyclopropane and propene at the laser intensity 30 TW/cm^2, shown in Fig. 4. We first presents the momentum distribution of cyclopropane molecule in intense laser fields in Figs. 4(a1)–4(c1), which shows that in linearly polarized laser field the momentum distribution of correlated electrons of cyclopropane is mostly distributed in first and third quadrants, which indicates that the front scattering mechanism is predominant, whereas in counter-rotating $(\omega+2\omega)$ field the momentum is mostly distributed in second and fourth quadrants, which indicates that back-scattering is predominant. This phenomenon is even more obvious in the case of counter-rotating $(\omega+2\omega)$ field. Meanwhile, compared with the cyclopropane case, for propene case of Figs. 4(a2)–4(c2), in linearly case the correlated momentum is mostly distributed in first and third quadrants and has a more obvious "finger-like" structure, which is the signature of front scattering, and in counter-rotating $(\omega+2\omega)$ field the momentum is mostly distributed in second and fourth quadrants, which is also more obvious than that of cyclopropane case. In the case of counter-rotating $(\omega+3\omega)$ field, the momentum has a large distribution in four quadrant, which means that both front scattering and back scattering play an important role in double ionization process.

The classical limits lead to a well-known cutoff laws: for direct above-threshold ionization (ATI), where the electron once ionized does not significantly interact anymore with the ion, its maximum energy is $2\sqrt{U_p}$,[19] where U_p is the pon-

deromotive energy. So we can get the conclusion that the momenta which are greater than $2\sqrt{U_p}$ arise from the interaction between the electron and ions. Furthermore, in our case the maximum energy is comparable with the E_0/ω_0, which means that the recollisions are possible to occur. We can seen from Figs. 4(a1) and 4(a2) that for both cases a lot of doubly charged ions distributed out the range of 0.77 a.u. ($2\sqrt{U_p} = 0.77$ a.u.), which means that there exits strong correlation between two electrons. However, there are much more doubly charged ions distributed out the range of 0.77 a.u. in propene than that in cyclopropane, which is why the "knee" structure is much more obvious in the case of propene than that of cyclopropane.

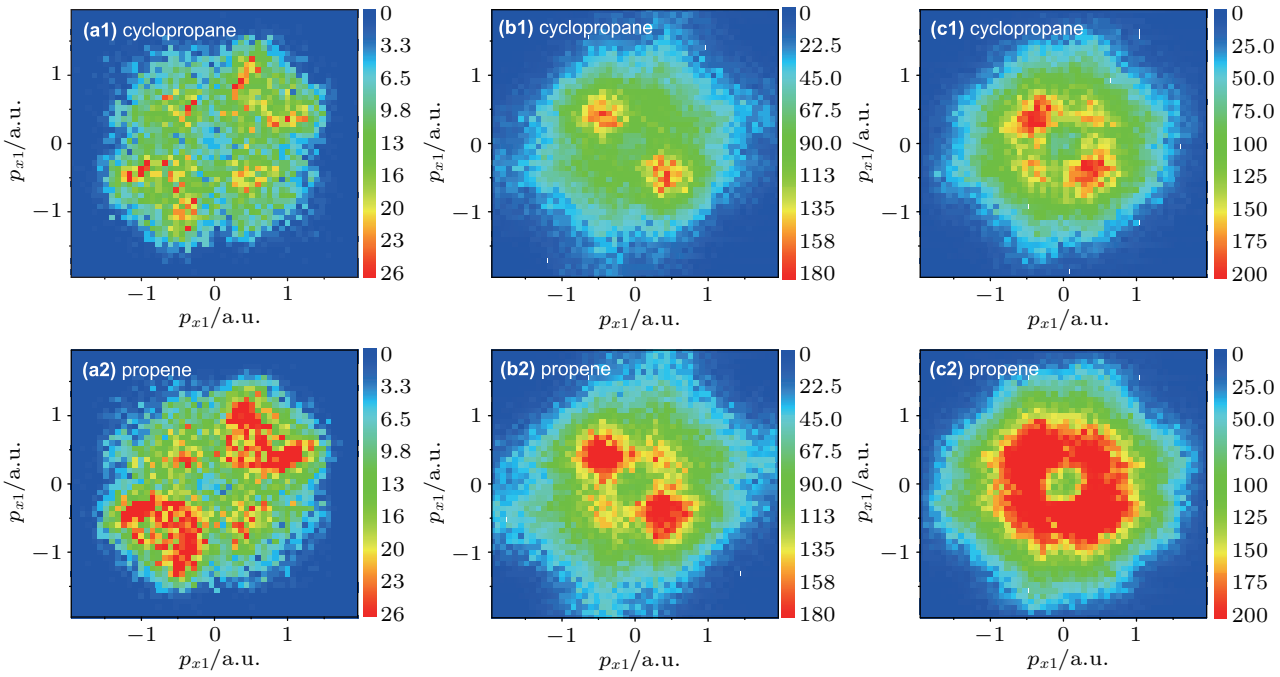


Fig. 4. (color online) Upper: the momentum distribution of C_3H_6 molecule with the structure of cyclopropane at the end of laser pulse at laser intensity 30 TW/cm^2 with (a1) linearly, (b1) counter-rotating ($\omega+2\omega$), and (c1) counter-rotating ($\omega+3\omega$) laser field. Lower: the momentum distribution of C_3H_6 molecule with the structure of propene with (a2) linearly, (b2) counter-rotating ($\omega+2\omega$), and (c2) counter-rotating ($\omega+3\omega$) laser field at laser intensity 30 TW/cm^2.

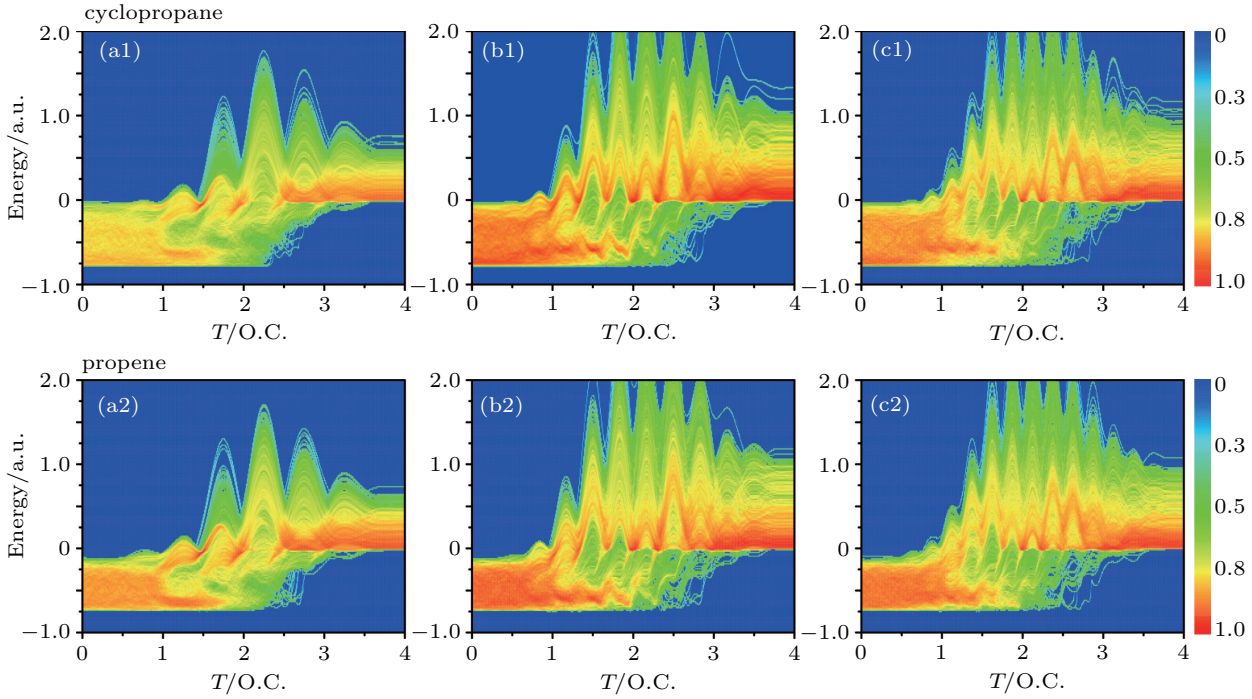


Fig. 5. (color online) Upper: the electron energy distribution of C_3H_6 with the structure of cyclopropane in (a1) linearly, (b1)counter-rotating ($\omega+2\omega$), and (c1) counter-rotating ($\omega+3\omega$) laser field. Lower: the electron energy distribution of C_3H_6 with the structure of propene with (a2) linearly, (b2) counter-rotating ($\omega+2\omega$), and (c2) counter-rotating ($\omega+3\omega$) laser field. The laser wavelength is 1200 nm and the intensity is 30 TW/cm^2.

In the classical ensemble approach, it is convenient to investigate NSDI and DI by tracing the classical energy trajectories. In order to reveal the DI dynamics of C_3H_6 molecule, we take into account all the two-electron energy trajectories that lead to DI events and make statistics at every moment of the time evolution, and furthermore we choose the recollision trajectories from them. Then the energy distribution of DI electrons as a function of time in intense laser fields can be obtained, as shown in Fig. 5. For each case, a set of two-electron energy trajectories is plotted to guide the distribution. From Fig. 5 we can see that there are more recollision double ionized trajectories in propene case than that in cyclopropane case. Figures 5(a1)–5(c1) illustrate that for cyclopropane, in linearly polarization case there are about 3 peaks that come back to the core, in counter-rotating case $(\omega+2\omega)$ the energy distribution has 7 peaks, which indicates that the multi-recollision occurs, and in counter-rotating case $(\omega+3\omega)$ there are even more peaks, about 9 or more peaks, which is because in counter-rotating field $(\omega+2\omega)$ or $(\omega+3\omega)$, there are 3 or 4 peaks in one cycle, which make the electrons have more chance to come back to the core, thus the recollision peaks increase. Figures 5(a2)–5(c2) illustrate that for propene the energy distribution presents similar tendency, but the initial energy range is lower than that of cyclopropane.

In addition, we also calculate the angular distributions at the end of pulse of propene and cyclopropane in Fig. 6 with linearly polarized laser field, counter-rotating field $(\omega+2\omega)$ and $(\omega+3\omega)$. For linearly polarized case, the angular distribution of propene and cyclopropane behaviors like a dumbbell shape, which is symmetric along x-axis. The difference is that in the case of propene molecule the angular distribution is more closer to the x-axis and more sharp at two ends. As the laser field changes from linearly polarized pulse to counter-rotating field, the angular distribution of propene does not change much, but for cyclopropane case it changes a lot, from dumbbell shape to circular shape, which means the ionization process from tunneling to multi-photon ionization. This is also due to the fact that the structure of cyclopropane has a triangle shape, which is similar with the counter-rotating field. All the above told us that the molecular structure plays an important role in double ionization process of multi-atom molecules.

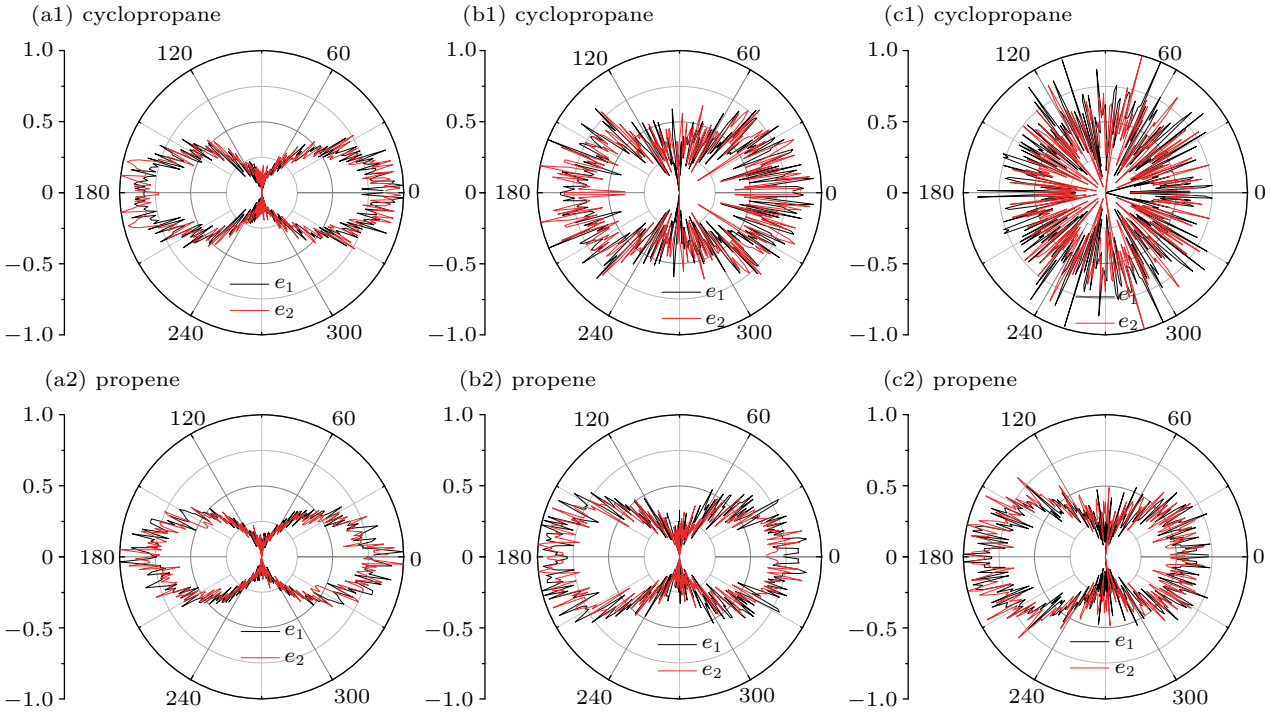


Fig. 6. (color online) Upper: the angular distribution of doubly ionized electrons of C_3H_6 as function of time with the structure of cyclopropane in (a1) linearly, (b1) counter-rotating $(\omega+2\omega)$, and (c1) counter-rotating $(\omega+3\omega)$ laser field. Lower: the angular distribution of doubly ionized electrons of C_3H_6 as function of time with the structure of cropene at the end of laser pulse in (a2) linearly, (b2) counter-rotating $(\omega+2\omega)$, and (c2) counter-rotating $(\omega+3\omega)$ laser field. The laser wavelength is 1200 nm and the intensity is 30 TW/cm^2.

4. Conclusion

We performed a theoretical study on strong-field DI process of C_3H_6 molecule with the structure of cyclopropane and propene in 800 nm and 1200 nm laser fields. A "knee" structure occurs in the DI yield in 1200 nm linearly polarized laser fields and counter-cotating laser fields, showing the evidence of NSDI in strong-field ionization of propene. By employing the classical ensemble method, we showed electron–electron correlations in the calculated momentum distributions and recollision of the first ionized electron with the ion core in the energy trajectories after DI in linearly polarized laser fields. In

order to reveal the DI dynamics of cyclopropane and propene molecule, we take into account all the recollision trajectories, which shows that both recollision impact ionization (RII) and recollision excitation with subsequent ionization (RESI) mechanisms may be contributed to the molecular strong-field NSDI, because we can see the direct recollision trajectories, multi-recollision trajectories, and laser-induced trajectories. We hope this work would stimulate further investigations to reveal molecular structure effect and quantum features in the ionization process.

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