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Coulomb expansion of deuterated methane clusters irradiated by an ultrashort intense laser pulse*

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The simulations of three-dimensional particle dynamics show that when irradiated by an ultrashort intense laser pulse, the deuterated methane cluster expands and the majority of deuterons overrun the more slowly expanding carbon ions, resulting in the creation of two separated subclusters. The enhanced deuteron kinetic energy and a narrow peak around the energy maximum in the deuteron energy distribution make a considerable contribution to the efficiency of nuclear fusion compared with the case of homonuclear deuterium clusters. With the intense laser irradiation, the nuclear fusion yield increases with the increase of the cluster size, so that deuterated heteronuclear clusters with larger sizes are required to achieve a greater neutron yield.

Keywords: deuterated methane cluster, ultrashort intense laser pulse, Coulomb explosion, nuclear fusion
PACC: 3640J, 5250J, 2588

1. Introduction

The interaction of intense laser pulses with gas clusters has been one of the most interesting subjects in recent years due to the fact that the clusters can be used as the bridge systems between the solids and gases. Extraordinary phenomena originating from the cluster dynamics have been explored, such as very efficient absorption of laser energy,[1] the production of energetic ions with highly charged state,[2] the strong x-ray emission,[3,4] the high-order harmonic generation[5] and self-focusing.[6] The remarkable findings in wide applications are that the laser-driven deuterated clusters can provide energetic deuterons and display the potential as a table-top source of neutron bursts.[7] The neutron yield from the Coulomb explosion processes of laser-heated homonuclear deuterium clusters and heteronuclear clusters containing deuterium have been shown to be dependent on the cluster size and laser pulse characteristics,[8−11] such as the pulse width and the laser energy.

In addition to these extensive experimental investigations, various theoretical models have also been proposed to unveil the dynamic mechanisms of laser interaction with clusters, e.g. nanoplasma model,[12,13] ionization–ignition model,[14] molecular dynamic model[15,16] as well as particle in cell (PIC) code model.[17] It is believed that the laser–cluster interactions contain three primary sub-processes: ionization of atoms, absorption of laser energy, and expansion of clusters. Both the hydrodynamic pressure and Coulomb force play their roles in the interaction process, however, which of them is dominant depends strongly on the details of the laser-cluster coupling.

In this paper, the Coulomb expansion process of deuterated methane cluster driven by an ultrashort intense laser pulse is investigated by using the simulation of the classic particle dynamics. The dependences of the deuteron kinetic energy on the cluster size and the laser intensity as well as the contribution of deuteron energy distribution to the nuclear fusion yield are also obtained.

2. Theoretical model

At the early phase of an ultraintense laser pulse irradiating gas clusters, the atoms inside the clusters are

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ionized by the optical field and converted into charged ions and free electrons. Additionally, the electron impact ionization (EII) also contributes to the ionization level of atoms in the clusters.[15] The generated free electrons play a role of initiator in triggering the consequent ion and electron dynamics such as laser-driven heating, escape of electrons from the cluster, and the explosion process of cluster sphere. Some heated electrons with higher energies will first fly away from the cluster so that the cluster acquires positive charges to explode outwards while some electrons with lower energies have to remain in the clusters, adding some hydrodynamic factors to the cluster expansion.

The standard particle dynamics is developed herein by integrating the Newton equations of motion for all ions and free electrons over the duration of the laser pulse,

$$m_i \frac{\mathrm{d}\boldsymbol{v}_i(\boldsymbol{r}_i,t)}{\mathrm{d}t} = \sum_{j\neq i} \frac{q_i q_j e^2(\boldsymbol{r}_i-\boldsymbol{r}_j)}{4\pi\varepsilon_0\left(|\boldsymbol{r}_i-\boldsymbol{r}_j|^2+\alpha\right)^{3/2}} + q_i e\boldsymbol{F}(t), \quad (1)$$

where $\boldsymbol{r}_i$ and $\boldsymbol{v}_i$ denote respectively the expanding velocities and displacements of the particles (including ions and electrons) at the time instant t; q_i and m_i are respectively the charged state and the rest mass of the particle; e is the electron charge; ε_0 is the vacuum permittivity; $\boldsymbol{F}$ is the electric field strength of the laser pulse. According to the soft-core model, the smoothing parameter α is introduced to avoid the singularity of the Coulomb field.

The laser pulse used in the simulation is linearly polarized with a Gaussian profile

$$\boldsymbol{F} = \boldsymbol{F}_0 \exp\left(-2\ln 2\frac{t^2}{\Delta t^2}\right)\sin(\omega t), \quad (2)$$

with $\boldsymbol{F}_0$, Δt and ω being the maximum amplitude, full width at half maximum, and frequency of the laser oscillation respectively. Generally, the sizes of clusters are much smaller than the laser wavelength so that the laser electric field is supposed to be independent of the spatial coordinates inside the cluster.

The geometric configuration of a deuterated methane cluster $(CD_4)_n$ is assumed in this paper to be a multiplayer icosahedron which is the stable frame taken by atomic gas clusters.[18] A k-layer closed-shell icosahedron of $(CD_4)_n$ should contain a well-defined total number of methane molecules

$$n(k) = 1 + \sum_{i=1}^{k}\left(10i^2+2\right). \quad (3)$$

In the molecule (CD_4), the C–C distance is 0.44 nm, the C–D distance is 0.109 nm[19] and the C–D angle is 109.5°.

In our simulations, the optical ionization rate induced by the combined (laser+inner) field is calculated using the cycle-averaged tunnel ionization rate given by Ammosov *et al.*[20] The EII mechanism is ignored in the simulation, for it does not appreciably affect the Coulomb explosion energetics.[15,21]

3. Results and discussion

The atoms inside the cluster can be ionized under the joint influence of the laser field and inner Coulomb field. The inner field becomes more intense with the increase of initial cluster size, so its effect on the ionization level of atoms, the so-called ignition effect,[14,15] is more distinct for larger clusters. Figure 1 shows the temporal evolution of average ionization level of carbon ions, with and without the inner field included separately. It can be seen that the exclusion of the inner field (represented by dotted line) which is equivalent to the case of a single atom makes the charged state of all carbon ions increase synchronously from a neutral state to a charged state of C^{4+} in a stepwise manner during a time period of 45 fs. However, when the inner field is taken into account (represented by the dashed line), the ionization process of the

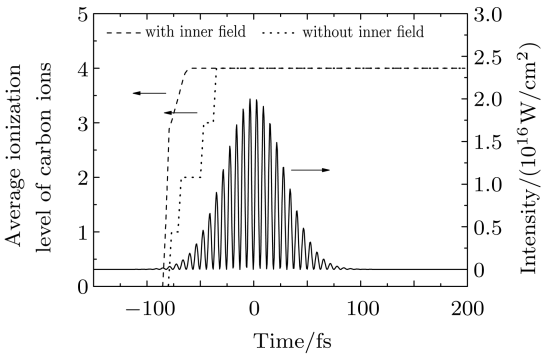


Fig.1. Temporal evolutions of average ionization level of carbon ions, with and without inner field included, represented by dash line and dotted line respectively. The cluster $(CD_4)_{1415}$ is irradiated by a Gaussian laser field represented by solid line. The peak intensity of the laser pulse is 2×10^{16} W/cm^2, its central wavelength is 800 nm and the pulse duration is 65 fs.

carbon ions starts up earlier and the total time for all neutral carbon atoms to reach a charged state of C^{4+} is shorter, i.e. 36 fs. Therefore, the laser intensity required to ionize the atoms in bigger clusters to a certain charged state is lower than that required in smaller clusters or a single atom. Owing to the inner Coulomb field increasing along the cluster radius, at a certain time instant the carbon ions on the outer layers of the cluster will have a higher probability of being ionized to a more highly charged level than those in the inner layers.[15] As a result, when the inner field is included the average ionization level is not likely to be an integer but a fraction and the trend in the increasing charged level as time goes on is like a smoothing profile rather than a stepwise profile.

If a laser field has a high intensity as well as a short rising time such that the total electrons inside a cluster can be stripped out of the cluster immediately, the cluster expansion is governed by the Coulomb repulsive forces between the positive charged ions and thus the expansion process can be simplified as a pure Coulomb explosion (PCE). For a small deuterated methane cluster, when the intensity of an ultrashort laser pulse grows to higher than $10^{16}\mathrm{W/cm^2}$, the deuterium can be completely ionized and the carbon can be ionized to a charged state of C^{4+}. All unbound electrons are removed from the system before the cluster expansion and thus the PCE can take place easily. Figure 2 shows a two-dimensional image of the spatial configuration of $(C^{4+}D_4^+)_{147}$ at the time instant $t = 40$ fs after the PCE has taken place. It is shown that the cluster expansion is isotropic. The carbon ions distribute uniformly in the inner space while a large number of deuterons overtake the more slowly expanding carbon ions and assemble outside to form a sphere shell. It is because the charge-to-mass ratio of deuterons is higher than that of carbon ions, and then the deuterons are accelerated by the multicharged carbon ions to acquire a higher kinetic energy. The overrunning effect further causes the formation of the two subclusters with different radially expanding velocities as the explosion evolves.

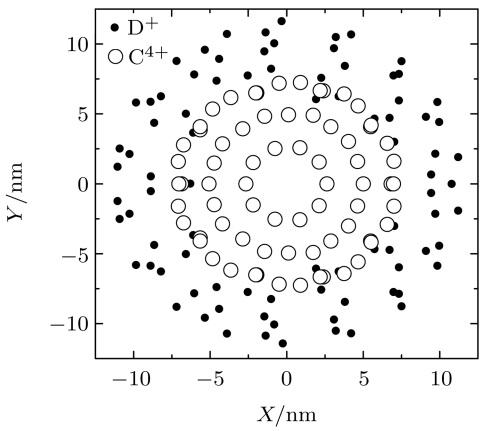


Fig.2. Two-dimensional image of the spatial configuration of Coulomb exploding $(C^{4+}D_4^+)_{147}$ at the time instant $t = 40$ fs after the pure Coulomb explosion has taken place.

Figure 3 presents the radial distributions of the deuterons and the carbon ions of a laser-heated $(CD_4)_{3871}$ at some time instants. Although the deuterons and the carbon ions initially overlap together inside the cluster (shown in an inset), some deuterons will overtake the more slowly expanding carbon ions after the cluster has started to expand due to the velocity difference between the two ions, and two expanding subclusters form with their borders separated from each other. The spatial overlap of two subclusters decreases as time goes on. The overrunning effect indirectly leads a large portion of deuterons to distribute in the high-energy region which is represented with solid line in Fig.4. As can be seen from this figure, the energy distribution of carbon ions (represented by dash line) is roughly proportional to the square root of the ion energy, which is similar to the ion energy distribution profile of homonuclear clusters[22] (represented by dotted line). The effective collisions between fast deuterons from such a narrow high-energy region can make a prominent contribution to driving d-d fusions among neighbouring clusters.

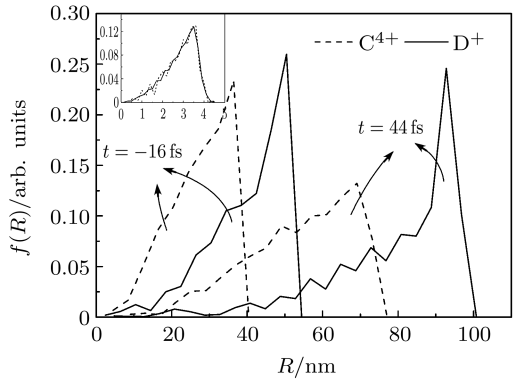


Fig.3. Radial distributions of the deuterons and carbon ions of a laser-heated cluster $(CD_4)_{3871}$ at times $t = -16$ and 44 fs. Inset is for the initial radial distribution of ions. The laser peak intensity is $3 \times 10^{17}\,\mathrm{W/cm^2}$ at zero time. The central wavelength and the duration of the laser pulse are the same as those in Fig.1.

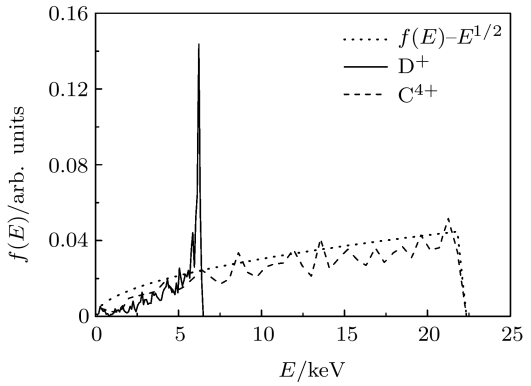


Fig.4. Energy distributions of deuterons and carbon ions of cluster $(CD_4)_{3871}$ under the irradiation of a laser pulse. The laser parameters are the same as those in Fig.3.

The laser-heated clusters explode outward and form a hot plasma filament with a diameter determined by the laser focus spot and a length dependent on the absorption depth of the laser pulse into the cluster gas. The d-d fusion yield produced in the collisions among the energetic deuterons from neighbouring clusters inside the cluster plasma filament is roughly estimated at[8,9]

$$Y_{\mathrm{IC}} = \frac{1}{2}\bar{\rho}^2 L_{\mathrm{IC}} V_{\mathrm{r}} \langle\sigma\rangle_{\mathrm{IC}} . \tag{4}$$

In this expression, the factor of 1/2 comes out of the consideration of the double counting of ions in number, and the parameter $\bar{\rho}$ is the average number density of nuclei inside the heated cluster filament. According to the experimental condition,[7] the cluster filament is assumed to be cylindrical with a radius R_{r} and a height H_{r}, so the volume is $V_{\mathrm{r}} = \pi R_{\mathrm{r}}^2 H_{\mathrm{r}}$; L_{IC} is a characteristic distance free streaming ions will traverse inside the cluster filament and is estimated at the plasma filament diameter. The averaged intercluster fusion cross section $\langle\sigma\rangle_{\mathrm{IC}}$ is evaluated from

$$\langle\sigma\rangle_{\mathrm{IC}} = \frac{1}{2}\int_0^\infty f(E_1)\,\mathrm{d}E_1 \int_0^\infty f(E_2)\,\mathrm{d}E_2 \times \int_0^\pi \sigma(E_{\mathrm{coll}})\sin\beta \mathrm{d}\beta, \tag{5}$$

where $E_{\mathrm{coll}} = E_1 + E_2 - 2(E_1 E_2)^{1/2}\cos\beta$ is the binary collision energy for deuterons with kinetic energy $E_1 = m_{\mathrm{D}} v_1^2/2, E_2 = m_{\mathrm{D}} v_2^2/2$ and collision angle $\beta \cdot f(E)$ is the normalized distribution of ion energy.

It is assumed that the energy distribution of deuterons from a deuterium cluster $(D_2)_n$ has the same energy range from zero to a maximum energy $E_{\max}$ (about 6.5 keV) as that of a deuterated methane cluster $(CD_4)_n$ (Fig.4). Applying the typical parameters of $\bar{\rho} = 2\times10^{19}\,\mathrm{cm}^{-3}$, $R_{\mathrm{r}} = 0.01\,\mathrm{cm}$, $H_{\mathrm{r}} = 0.1\,\mathrm{cm}$, and $L_{\mathrm{IC}} = 0.02\,\mathrm{cm}$,[7] the fusion reaction yield in the two cases can be roughly estimated. In the case of $(D_2)_n$, the average intercluster fusion cross section $\langle\sigma\rangle_{\mathrm{IC}}$ is $2.1\times10^{-29}\,\mathrm{cm}^2$ while the cross section is $4.8\times10^{-29}\,\mathrm{cm}^2$ in the case of $(CD_4)_n$. The total fusion yield in the latter case can reach 6.0×10^3, which is twice as high as that in the former case (2.6×10^3). It can be concluded that for the same range of deuteron energy, a sharp peak around the energy maximum in the deuteron kinetic energy distribution from a heteronuclear cluster provides a supplementary contribution to the efficiency of nuclear fusion compared with the homonuclear clusters.[23]

Figure 5 illustrates the deuteron kinetic energy and the intercluster fusion yield versus the initial size of deuterated methane clusters. It is shown that for a given intense laser field, the maximum and the average energies of deuterons from the cluster expansion increases linearly with the squared initial radius of cluster, which is the same as the relationship obtained from homonuclear clusters.[22] Correspondingly, the d-d fusion yield increases as the cluster becomes bigger. It is revealed that heteronuclear clusters with larger sizes are required to achieve a greater neutron yield.

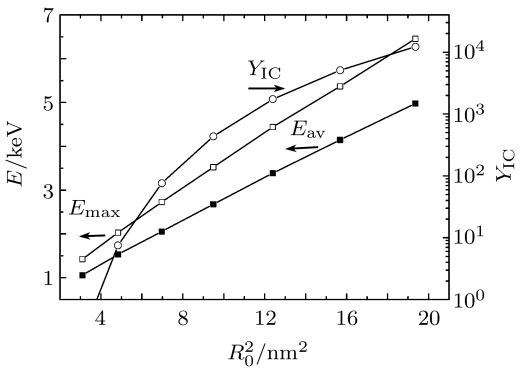


Fig.5. The maximum deuteron energy $E_{\max}$, average deuteron energy E_{av} and the intercluster fusion yield Y_{IC} versus the squared radius R_0 of cluster. The laser parameters are the same as those in Fig.3.

The dependence of the average deuteron energy of $(CD_4)_{1415}$ on the laser peak intensity is illustrated in Fig.6. The deuteron energy increases sharply as the laser intensity rises when the laser intensity is relatively low. However, this trend towards increasing slows down after the laser intensity grows to higher than $3\times10^{17}\,\mathrm{W/cm}^2$. In the chosen laser intensities ranging from $2\times10^{15}\,\mathrm{W/cm}^2$ to $2\times10^{18}\,\mathrm{W/cm}^2$, the carbon atoms can be ionized to a charged state of C^{4+}. With higher laser intensities the ionization process of carbon ions will be finished earlier and the electrons

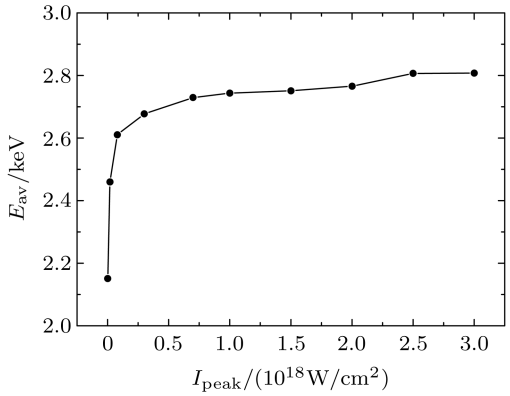


Fig.6. Dependence of the average deuteron energy E_{av} of $(CD_4)_{1415}$ on the laser peak intensity I_{peak}. The central wavelength and the duration of the laser pulse are the same as those in Fig.1.

will be expelled out of the cluster in a shorter time, so that the deuteron energy from the cluster expansion approaches to a saturation value and thus varies slightly with the increase of laser intensity. When the laser intensity increases over a peak intensity of $2.0 \times 10^{18}\,W/cm^2$, the carbon atoms can be ionized to a highly charged state of C^{5+}, and consequently the deuteron energy is remarkably enhanced due to the boosting effect of multicharged carbon ions on the kinetic energy of deuterons.

4. Conclusion

The simulation results of particle dynamics reveal that in the expansion of cluster $(CD_4)_n$ driven by an intense laser pulse, a large number of deuterons overtake the more slowly expanding carbon ions, resulting in the formation of two subclusters with different radially expanding velocities. It is also shown that the inner Coulomb field of the cluster is helpful for improving the ionization level of atoms in clusters, especially large clusters. The kinetic energy of deuterons from the cluster expansion varies linearly with the squared initial radius of cluster, and the corresponding nuclear fusion yield increases as the cluster size becomes larger. Heteronuclear deuterated clusters are better candidates as fast neutron sources than homonuclear deuterium clusters due to the boosting effect of multicharged ions on the kinetic energy of light ions as well as the accelerating effect on the light ion velocity in the overrunning process of light ions relative to heavy ions within the expanding heteronuclear clusters.

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