Average kinetic energy of ions ejected from pure Coulomb explosions of methane clusters

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We use an electrostatic model to study the average kinetic energy of ions ejected from the pure Coulomb explosions of methane clusters $(CA_4)_n$ (light atom A=H and D). It is found that the ratio of the average kinetic energy of the ions to their initial average electrostatic potential energy is irrelevant to the cluster size. This finding implies that as long as the ratio is given, the average kinetic energies of the ions can be simply estimated from their initial average electrostatic potential energies, rather than from the time-consuming simulations. The ratios for the different charge states of carbon ions are presented.

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For more than a decade, the interaction of femtosecond intense laser pulses with clusters has been an active area of research and a lot of important findings were reported, such as X-ray radiation, energetic ions and electrons production, as well as the neutron production from laser driven nuclear fusion of deuterium clusters[1−3]. Most of the studies for the laser-cluster interaction are with the homonuclear clusters formed from a single species, such as rare gas clusters, hydrogen or deuterium clusters. Recently, Last and Jortner reveal through molecular dynamic simulations that the Coulomb explosions of heteronuclear clusters driven by a femtosecond ultra-intense laser pulse has advantages over that of homonuclear clusters, involving the enhancement of the kinetic energy of the light ions[4,5]. The kinetic energy of the light ions is enhanced by the energetic effect because the presence of multicharged high-Z heavy ions increases the initial electrostatic potential energy before cluster expansion, and by the kinematic effect which is related to the kinematic parameter of the light ions $\eta_A = q_A m_B / q_B m_A$, or the kinematic parameter of the heavy ions $\eta_B = q_B m_A / q_A m_B$, where q_A and m_A denote the charge and mass of the light ion A, and q_B and m_B are the charge and mass of the heavy ion B. For $\eta_A > 1$, because of the higher acceleration than that acquired by the heavy ions located at the same radial position in the cluster, the light ions overruns the heavy ions and attain stronger Coulombic repulsion during the cluster expansion. Consequently, the light ions acquires the kinetic energy larger than its initial electrostatic potential energy, while the kinetic energy of the heavy ions is lower than its initial electrostatic potential energy. Therefore, the heteronuclear clusters containing deuterium like $(CD_4)_n$ become a promising medium for the table-top laser driven nuclear fusion. Some related experiments with this species of cluster medium were reported previously[6−8]. Madison *et al.* demonstrated experimentally the Coulomb explosions of deuterated methane clusters $(CD_4)_n$, exhibiting higher ion energies than those from explosions of comparably sized neat deuterium clusters[7]. Hohenberger *et al.* presented an experimental confirmation of the kinematic effect by comparing the energy spectra from laser induced explosions of methane clusters $(CH_4)_n$ and deuterated methane clusters$(CD_4)_n$[8]. Being an important parameter, however, the average kinetic energy of the ions from the explosions of the heteronuclear clusters is difficult to be determined experimentally, because it is not fairly possible to distinguish the light ions from the carbon ions in the usual time-of-flight spectra in general[7]. Moreover, since the light ions can overrun the heavy ions during the Coulomb explosions of the heteronuclear clusters ($\eta_A > 1$), the average kinetic energy of the ions cannot be given analytically[4,5,9−12]. In this letter, we use an electrostatic model to study the kinetic energy of the ions ejected from the pure Coulomb explosions of heteronuclear methane clusters $(CA_4)_n$. Interestingly, it is found that the ratio of average kinetic energy of the ions to their initial average electrostatic potential energy is irrelevant to the initial cluster radius, instead, depending on the charge of the carbon ions. Consequently, for a given ratio, the average kinetic energy of the ions can be estimated from their initial average electrostatic potential energy.

In the present electrostatic model and under the pure Coulomb explosion approximation, all the electrons are assumed to be stripped out of the cluster (outer ionization) before the expansion of the ionic cluster takes place. The carbon ions and the light ions are assumed to be uniformly distributed in the ionic cluster and the proportion of the total number of the carbon ions to that of the light ions is 1:4. The cluster which has an initial radius R is divided into a number of spherical shells, and each shell is defined by its initial radius r_0 and a thickness $\mathrm{d}r_0$ at time $t = 0$. The number of the methane molecules in the $r_0 - (r_0 + \mathrm{d}r_0)$ shell is $4\pi\rho r_0^2 \mathrm{d}r_0$, where ρ is the methane molecular density in the cluster. In the calculation, the ion motion is assumed purely radial and the ions are assumed to be classical. From the Gaussian law, the radial electrostatic electric field $\mathbf{E}$ in the radial position r for an ionic sphere can be expressed as

$$\mathbf{E} = \frac{e}{4\pi\varepsilon_0 r^2}\left(\int_0^r \mathrm{d}N_\mathrm{A} + q_\mathrm{B}\int_0^r \mathrm{d}N_\mathrm{B}\right), \qquad (1)$$

where e is the electron charge, ε_0 is the dielectric constant, q_B is the charge of the carbon ions in e units (q_A is equal to 1), and $\int_0^r \mathrm{d}N_\mathrm{A}$, and $\int_0^r \mathrm{d}N_\mathrm{B}$ are the total number of the light ions A and that of the carbon ions in the sphere of radius r, respectively. Then the motion of the light ions and the carbon ions can be expressed by the Newton equations $m_\mathrm{A}\frac{\mathrm{d}^2 r_\mathrm{A}}{\mathrm{d}t^2} = e\mathbf{E}$ and $m_\mathrm{B}\frac{\mathrm{d}^2 r_\mathrm{B}}{\mathrm{d}t^2} = eq_\mathrm{B}\mathbf{E}$, respectively. In order to simulate the pure Coulomb explosion of the methane cluster, we have to track the carbon ions and the light ions in each spherical shell at each time interval. In our simulation, the time step is set to be 10^{-17} s and the expansion time is 4×10^{-13} s, being the same as those in Ref. [8]. When the cluster is fully exploded, i.e., both the kinetic energy of the light ions and the kinetic energy of the carbon ions keep nearly constant, respectively, with the expansion time further increasing, we can acquire the ion kinetic energy distribution $f(E) = \mathrm{d}N/\mathrm{d}E$. The average kinetic energy of the ions is given as $E_\mathrm{av} = \int Ef(E)\mathrm{d}E/\int f(E)\mathrm{d}E$. For the pure Coulomb explosion of the methane cluster, the initial average electrostatic potential energy of the ions can be given as [5]

$$E'_\mathrm{av} = 4\pi B\,(4+q_\mathrm{B})\,q\rho R^2/5, \qquad (2)$$

where ρ is taken to be 16 nm^{-3}, the constant B is 1.44 eV·nm, and q is equal to 1 for the light ions and to q_B for the carbon ions. Equation (2) shows that the initial average electrostatic potential energy of the ions increases with the charge of the carbon ions and is proportional to the square of the initial cluster radius R.

Using the electrostatic model, we calculate the energy spectra of the light ions ejected from the pure Coulomb explosions of the ionized methane clusters $(\mathrm{C^{4+}H_4^+})_n$, $(\mathrm{C^{4+}D_4^+})_n$, and $(\mathrm{C^{4+}T_4^+})_n$ with the cluster radius R of 3.18 nm which is equal to that in Ref. [5]. The ion energy spectra shown in Fig. 1(a) are very similar to those presented in Ref. [5]. Meanwhile, the average kinetic energy and the maximum kinetic energy of the deuterons ejected from the pure Coulomb explosion of the cluster $(\mathrm{C^{4+}D_4^+})_n$ with the cluster radius R of 4.29 nm are also calculated to be 10.86 and 14.47 keV, respectively. The results are close to the reported energies of 10.1 and 15.1 keV obtained using dynamics simulations for the same cluster radius[5]. These results indicate that the present model may provide a proper description of the pure Coulomb explosions of methane clusters.

In order to investigate the kinematic effect during the methane cluster expansion, we calculate the average kinetic energy of the ions ejected from the pure Coulomb explosions of the methane clusters $(\mathrm{C^{4+}H_4^+})_n$ and the deuterated methane clusters $(\mathrm{C^{4+}D_4^+})_n$ with different cluster radii R. Meanwhile, the initial average electrostatic potential energies are also calculated from Eq. (2) and compared with the average kinetic energies in Fig. 1(b). It can be seen that the average kinetic

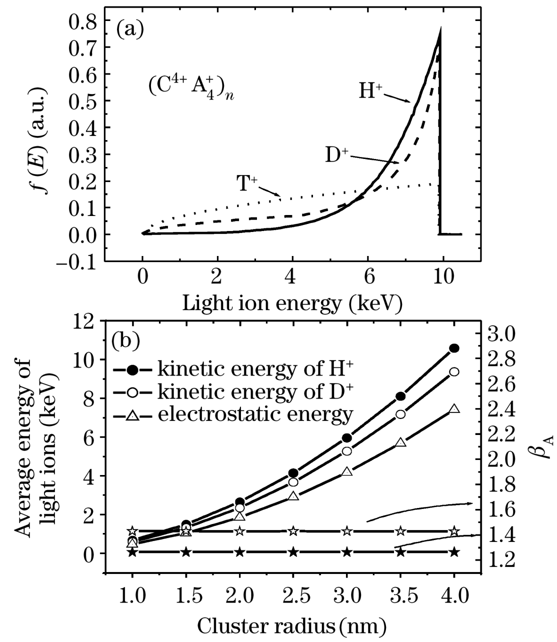

Fig. 1. (a) Energy spectra of the light ions ejected from the pure Coulomb explosions of the ionized methane clusters. The initial methane cluster radius is 3.18 nm. (b) the dependence of the average kinetic energy (circles) and the initial average potential energy (triangles) of the light ions, as well as the ratio β_A (stars) on the cluster radius. The carbon ions are assumed to be $\mathrm{C^{4+}}$.

energy of the protons is always higher than that of the deuterons for different cluster sizes. This is because that the protons are connected with the larger kinematic parameter η_A and then acquire higher acceleration than the deuterons during the cluster expansion. With the increase of the cluster size, the average kinetic energies of the protons and the deuterons increase consistently, but the difference of these average kinetic energies with the initial average potential energy becomes remarkable gradually. However, as the ratio of the average kinetic energy of the light ions to their initial average electrostatic potential energy is defined to be $\beta_\mathrm{A} = E_\mathrm{av}/E'_\mathrm{av}$, it is found that β_A keeps constant and is independent of the cluster radius. β_A is equal to 1.43 and 1.26 as shown in Fig. 1(b) for the protons and the deuterons at different cluster radii. Obviously, β_A can be used to describe the kinetic energy enhancement of the light ions quantitatively, which results from the kinematic effect.

To gain a further insight into the kinetic energy enhancement of the light ions due to the kinematic effect during the cluster expansion, the dependence of β_A on the charge of the carbon ions is studied. The ratio β_A calculated for different charges of the carbon ions for the pure Coulomb explosion of the methane cluster $(\mathrm{C^{q+}H_4^+})_n$ is shown in Fig. 2(a). The calculation shows that, within the calculation time scale ($0-4\times10^{-13}$ s), if the cluster is assumed to be divided into one hundred spherical shells, the protons located initially outside the 5th, 12th, 19th, 28th, and 37th shells can overrun the outmost carbon ions (located initially at the 100th shell) after the full expansion of the cluster for the charges of the carbon ions being 2, 3, 4, 5, and 6, correspondingly. It means that

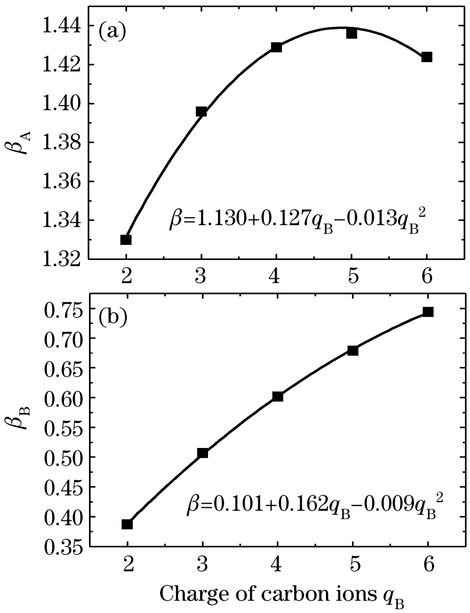

Fig. 2. Dependence of the ratios $\beta = E_{\rm av}/E'_{\rm av}$ on the charge of carbon ions (a) for the protons and (b) for the carbon ions ejected from the pure Coulomb explosions of the ionized methane clusters $(C^{q+}H^{4+})_n$.

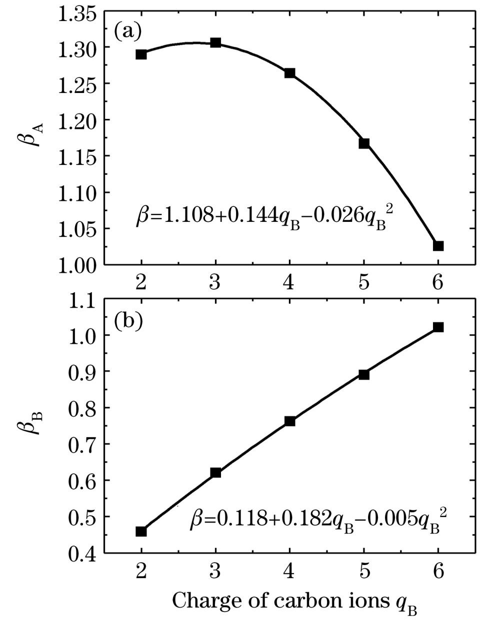

Fig. 3. Dependence of the ratios $\beta = E_{\rm av}/E'_{\rm av}$ on the charge of carbon ions (a) for the deuterons and (b) for the carbon ions ejected from the pure Coulomb explosions of the ionized methane clusters $(C^{q+}D^{4+})_n$.

with the increase of the charge of the carbon ions (the kinematic parameter η_A of the protons decreases gradually), the number of protons that overrun the outmost carbon ions decreases gradually. However, due to the increase of the charge of the carbon ions, the protons overrunning the carbon ions are more strongly repelled and thus acquire higher energies. Consequently, not only the kinematic parameter but also the charge of the carbon ions has an impact on the energy enhancement resulting from the kinematic effect. As shown in Fig. 2(a), when $q_{\rm B}$ is increased to 5 ($\eta_{\rm A} > 2.4$), though the kinematic parameter $\eta_{\rm A}$ of the protons gradually decreases with the increase of $q_{\rm B}$, the kinetic energy enhancement resulting from the kinematic effect is still large and the ratio $\beta_{\rm A}$ increases continually until $q_{\rm B} = 5$. However, when $q_{\rm B}$ is increased to 6 (the kinematic parameter $\eta_{\rm A}$ of the protons is decreased to 2.0), $\beta_{\rm A}$ begins to decrease. Meanwhile, the calculation on the ratio $\beta_{\rm B}$ of the average kinetic energy of the carbon ions to their initial average electrostatic potential energy indicates that $\beta_{\rm B}$ is also independent of the cluster radius. $\beta_{\rm B}$ increases monotonously with the charge $q_{\rm B}$ increasing as shown in Fig. 2(b). As a matter of fact, the calculated $\beta_{\rm B}$ from our model is in accordance with that calculated from the expression $1 - 4(\beta_{\rm A} - 1)/q_{\rm B}$ deduced from the energy conservation law and the Eq. (2). By fitting the calculated ratios $\beta_{\rm A}$ and $\beta_{\rm B}$ for the pure Coulomb explosions of the methane clusters $(C^{q+}H_4^+)_n$, we can approximately express the dependence of the ratios $\beta_{\rm A}$ and $\beta_{\rm B}$ on the charge of the carbon ions as

$$\beta_{\rm A} = 1.130 + 0.127q_{\rm B} - 0.013q_{\rm B}^2, \quad \text{(for protons)}, \quad (3a)$$

and

$$\beta_{\rm B} = 0.101 + 0.162q_{\rm B} - 0.009q_{\rm B}^2, \quad \text{(for carbon ions)}. \quad (3b)$$

Also, the dependence of the ratios $\beta_{\rm A}$ and $\beta_{\rm B}$ on the charge of the carbon ions for the ionic deuterated methane clusters $(C^{q+}D_4^+)_n$ is studied and the results are presented in Fig. 3. Because of the smaller kinematic parameter of the deuterons, the number of the deuterons which can overrun the outmost carbon ions is less than that of the protons under the same conditions. For example, within the calculation time scale ($0-4\times10^{-13}$ s), the calculation shows that among the deuterons located initially outside the 21st, 38th, 58th, 78th shells, none of the deuterons can overrun the outmost carbon ions for the charges of the carbon ions being 2, 3, 4, 5, and 6, correspondingly. The curve in Fig. 3(a) demonstrates that when $q_{\rm B}$ is higher than 3 (the kinematic parameter of the deuterons is less than 2.0), the ratio $\beta_{\rm A}$ for the deuterons begins to decline and approaches 1 for $q_{\rm B}$ is 6 ($\eta_{\rm A} = 1$). In contrast with the difference in $\beta_{\rm A}$ for the explosions of $(C^{q+}H_4^+)_n$ and $(C^{q+}D_4^+)_n$, the dependence of the ratio $\beta_{\rm B}$ for carbon ions on the charge of the carbon ions is very similar for the two cases. The ratios $\beta_{\rm A}$ and $\beta_{\rm B}$ as functions of $q_{\rm B}$ for the explosions of the clusters $(C^{q+}D_4^+)_n$ are given to be

$$\beta_{\rm A} = 1.108 + 0.144q_{\rm B} - 0.026q_{\rm B}^2, \quad \text{(for deuterons)}, \quad (4a)$$

and

$$\beta_{\rm B} = 0.118 + 0.182q_{\rm B} - 0.005q_{\rm B}^2, \quad \text{(for carbon ions)}. \quad (4b)$$

From Eqs. (2)—(4), the average kinetic energies of the protons, deuterons, and carbon ions can be estimated for a specific cluster size of the methane cluster and the given charge of the carbon ions. For example, for the pure Coulomb explosion of cluster $(C^{4+}D_4^+)_n$ with the initial cluster radius of 3.0 nm, it can be obtained from Eqs. (2) and (4a) that the ratio $\beta_{\rm A}$ is 1.27 and the initial average electrostatic potential energy of the deuterons

$E'_{\rm av}$ is 4.17 keV, resulting in the average kinetic energy of the deuterons $E_{\rm av} = 5.29$ keV.

In summary, using an electrostatic model, we investigate the enhancement of the kinetic energy of the light ions resulting from the kinematic effect during the pure Coulomb explosions of the methane clusters $(\mathrm{C}^{q+}\mathrm{H}_4^+)_n$ and the deuterated methane clusters $(\mathrm{C}^{q+}\mathrm{D}_4^+)_n$. It is found that the ratio of the average kinetic energy of the ions to their initial average electrostatic potential energy before cluster expansion is dependent on the kinematic parameter and the charge of the carbon ions, but independent on the cluster radius. The ratios $\beta_{\rm A}$ and $\beta_{\rm B}$ as functions of the charge of the carbon ions are presented. For the pure Coulomb explosions of the methane clusters $(\mathrm{C}^{q+}\mathrm{H}_4^+)_n$ and the deuterated methane clusters $(\mathrm{C}^{q+}\mathrm{D}_4^+)_n$, the initial average electrostatic potential energy of the ions before cluster expansion can be given analytically. Consequently, based on the presented ratios, the average kinetic energy of the protons, deuterons, and carbon ions can be estimated from their initial average electrostatic potential energies.

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