Cooling-induced increase of methane cluster size investigated under a Coulomb explosion scheme

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In this letter, we discuss the increase in the average cluster size by lowering the stagnation temperature of the methane (CH_4) gas. The Coulomb explosion experiments are conducted to estimate the cluster size and the size distribution. The average CH_4 cluster sizes N_{av} of 6 230 and 6 580 are acquired with the source conditions of 30 bars at 240 K and 60 bars at 296 K, respectively. Empirical estimation suggests a five-fold increase in the average size of the CH_4 clusters at 240 K compared with that at room temperature under a backing pressure of 30 bars. A strong nonlinear Hagena parameter relation ($\mathit{\Gamma}^* \propto T_0^{-3.3}$) for the CH_4 clusters is revealed. The results may be favorable for the production of large-sized clusters by using gases at low temperature and high back pressures.

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Clusters, with van der Waals bonded agglomerations of up to 10^7 atoms, have received considerable attention, and have been investigated because of their density characteristics in internal solid and general gas. Such clusters were first discovered by Becker *et al.*[1] in 1956 and were investigated systematically by Hagena[2]. Moreover, such clusters have been used for various applications, such as X-ray generation[3], energetic ion production[4], and nuclear fusion[5], after the invention of intense femtosecond laser systems. In 1999, Ditmire *et al.* demonstrated the deuterium-deuterium (D-D) nuclear fusion in deuterium cluster jets under the irradiation of intense femtosecond laser pulses[5]. Thereafter, intensive studies on laser-cluster interaction have been conducted to generate higher energetic ions and consequently increase fusion neutron yields[6−14]. Theoretical results have revealed the essence of laser-cluster interaction and have proposed the use of heteronuclear clusters, such as deuterated methane (CD_4) and heavy water (D_2O), as targets for generating more energetic light ions compared with homonuclear clusters such as D_2 with the same sizes because of the energetic and kinematic effects[15−18]. The proposal paves the way for future research on the table-top laser-driven fusion[13,19], which can provide high-flux sources of clean fusion neutrons and short pulse durations. A petawatt-laser-produced[20] fusion neutron source may be of considerable importance to studies on radiation-induced damage on materials, as well as benefit fast neutron radiography, in which the small source size could lead to a high spatial resolution[21].

We have recently demonstrated an efficient fusion neutron generation by using CD_4 clusters under the irradiation of 120-mJ and 60-fs laser pulses[22]. The key factors for generating fusion neutrons includes the energetic deuterons and the density of deuterons inside and around the plasma channel[19,23]. Larger average cluster sizes are required to further increase the average kinetic energy (KE) of exploded deuterons.

Gaseous clusters are typically produced through the supersonic expansion of a high backing pressure gas into a vacuum through a conical nozzle[2]. Hagena parameter ($\mathit{\Gamma}^* \propto P_0 T_0^{-2.29}$) was introduced to describe cluster formation through $N = 33(\mathit{\Gamma}^*/1\,000)^{2.35}$ for monatomic and diatomic clusters[24]. Usually, hydrogen clusters were produced[25−27] under very low temperature and can be analytically described by Hagena parameter. Despite the limited knowledge on polyatomic cluster formation, particularly at low temperatures, large CH_4 clusters can be produced with high backing pressure and low temperature considering the similarity of the hydrodynamic process of cluster formation process with that of monatomic and diatomic clusters. In previous studies, we increased the CH_4 cluster sizes by increasing the backing pressures at room temperature[22,23,28]. However, the increase in backing pressure is expected to cause serious vacuum loading in the pumping systems, and decrease cluster formation efficiency[28].

In this letter, we increase the average cluster size by lowering the stagnation temperature of the CH_4 gas before its expansion into a vacuum through a conical nozzle. Moreover, analytical calculations are performed to evaluate the average cluster size and the size distribution. Finally, an average size of $N_{av} = 6\,230$ (with a distribution width σ of 1.43) is obtained by lowering the stagnation temperature to 240 K under the backing pressure of 30 bars. Otherwise, an average CH_4 cluster size of $N_{av} = 6\,580$ (with a distribution width σ of 1.18) is obtained at room temperature with the backing pressure of 60 bars. The similar average-sized clusters are acquired under half of the backing pressure at a temperature of 56 K lower compared with room temperature conditions. In the experiment, a rough scaling of $\mathit{\Gamma}^* \propto T_0^{-3.3}$ is determined for the CH_4 clusters. Thus, larger CH_4 clusters can be made under relatively lower backing pressures at low temperatures.

The experiments were conducted in the CPA-based Ti-Sapphire femtosecond laser facility at the State Key Laboratory of High Field Laser Physics of Shanghai Institute of Optics and Fine Mechanics. A 160-mJ, 60-fs laser pulse was delivered to the target at a 10-Hz repetition rate from the laser system. The experimental setup had been reported in Ref. [23] and was described briefly in the present study. In the present experiments, the CH_4 clusters were produced using a solenoid valve and a conical nozzle with a throat diameter of 310 μm, a length of 26 mm, and a half opening angle of 4.6°. The valve was tightly surrounded by a coolant jacket that delivers liquid nitrogen through the inside pipe. The valve temperature was monitored using a platinum thermo-resistor buried in the valve body. The laser pulse was focused approximately 0.8 mm before the central axis of the cluster jet and 1 mm beneath the nozzle exit by using an $f/4$ off-axis parabolic mirror (f= 200 mm). The laser focus spot size was measured to be approximately 9 μm in diameter, resulting in a peak intensity of 4×10^{18} W/cm^2 in vacuum. The average proton KEs were measured using a time-of-flight (TOF) mass spectrometer with a 3.25-m long free flight tube ended with dual micro-channel-plates (DMCP). The TOF spectra were recorded using a digital oscilloscope (LeCroy Wave runner 42Xs) and then stored.

CH_4 clusters were produced at room temperature (296 K) with a backing pressure of 60 bars and were irradiated by a 160-mJ, 60-fs laser pulse. The KE spectra of the protons were converted from the recorded TOF spectra as

$$E_{\mathrm{k}} = \frac{1}{2}m\left(\frac{L}{t}\right)^2, \tag{1}$$

where m is the mass of the protons, L is the length of the flight tube, and t is the recorded TOF. For comparison, another experiment was conducted with the valve cooled down to 240 K to generate CH_4 clusters under a backing pressure of 30 bars. At room temperature, the average proton KEs were adequately small, and thus, they were hardly detected with the TOF mass spectrometer.

The TOF spectrum of the exploded ions detected using the DMCP for the experiment at room temperature was shown in Fig. 1(a), and the calculated KE spectrum was shown in Fig. 1(b). The average proton KE of 8.9 keV was calculated by integrating the measured spectrum $f(E)$ over the whole energy range E as

$$E_{\mathrm{av}} = \int Ef(E)\mathrm{d}E\Big/\int f(E)\mathrm{d}E. \tag{2}$$

The TOF and the KE spectra of the protons were shown in Figs. 2(a) and 2(b), for the experiment under a backing pressure of 30 bars at 240 K, respectively. As can be seen in Fig. 2(b), the average proton KE was 8.1 keV, which is slightly lower compared with that produced at room temperature under a backing pressure of 60 bars.

The cluster size N nonlinearly scales as the backing pressure P_0 and presents an empirical scaling of

$$N \propto P_0^{\beta}. \tag{3}$$

A previous study yielded $\beta = 3.0$ for $0< P_0 \leqslant 44$ bar and $\beta = 1.7$ for 44 bar $\leqslant P_0 \leqslant 84$ bar with an equivalent throat diameter $d_{\mathrm{eq}}^{[24,28]}$ of 3.89 mm at room temperature[28]. In the present experiments, d_{eq} was calculated to be 3.85 mm. Empirically, the results can be used to estimate the average cluster size at room temperature. However, the actual average cluster size should be determined exactly for an accurate comparison. Thus, a cluster size characterization scheme was proposed using a spherical model[23], in which clusters explode layer by layer[29] with the measured KE spectra.

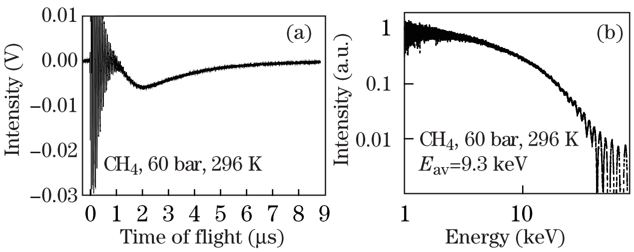

Fig. 1. (a) TOF and (b) KE spectra of the exploded ions under the irradiation of the 160-mJ, 60-fs laser pulse under a backing pressure of 60 bars at room temperature.

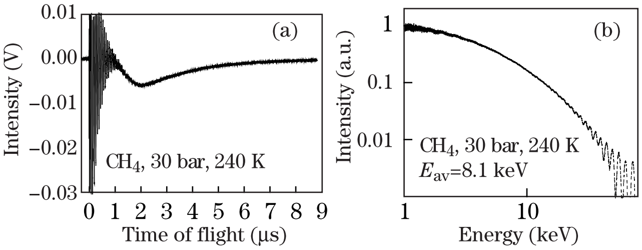

Fig. 2. (a) TOF and (b) KE spectra of the exploded ions under the irradiation of the 160-mJ, 60-fs laser pulse under a backing pressure of 30 bars at 240 K.

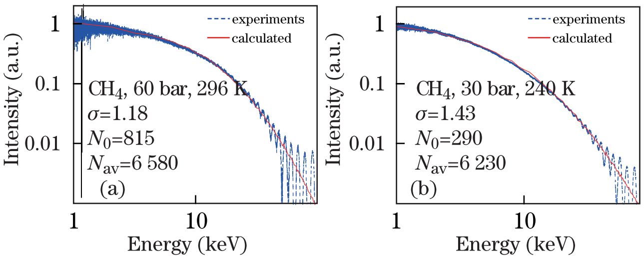

Fig. 3. (Color online) Calculated and measured KE spectra of ions produced under (a) 60 bars at room temperature and (b) 30 bars at 240 K. N_0 and N_{av} represent the peak and average sizes of the log-normal distributed clusters, respectively.

In the proposed scheme, the ions, which are originally located in the radius r in a given single-sized heteronuclear CH_4 cluster with radius R and internal molecular density of $\rho = 1.6 \times 10^{22}\mathrm{cm}^{-3[19]}$, are accelerated during a Coulomb explosion to acquire the final average KE as

$$E_{\mathrm{P}}(r,R) = \frac{1}{6\varepsilon_0}e^2\rho\left(8r^2 + 3q_{\mathrm{C}}R^2 - q_{\mathrm{C}}r^2\right) \text{ (for proton)} \tag{4a}$$

and

$$E_{\mathrm{C}}(r,R) = \frac{1}{3\varepsilon_0}q_{\mathrm{C}}e^2\rho(q_{\mathrm{C}}+4)r^2 \text{(for carbon).} \tag{4b}$$

where q_{C} is the average charge state of each carbon ion and ε_0 is the dielectric constant.

The size distribution of the clusters produced through the supersonic expansion of gas in vacuum can be described using the following log-normal-shaped formula:

$$f(N) = \frac{1}{N\sigma\sqrt{2\pi}} \exp[-\frac{(\ln N - \mu)^2}{2\sigma^2}], \qquad (5)$$

where μ and σ are the mean and standard deviation of the logarithm of size N, respectively. N is the number of molecules in a given cluster, which is related to the radius R through $N = 4\pi R^3\rho/3$. Assuming that the peak size of the distribution is N_0, we have the relation $\mu = \ln N_0 + \sigma^2$. If N_0 and σ are known, the cluster size distribution is defined. The integration of the KE spectrum of the protons over the whole size range of the clusters in the jets may result in a theoretical spectrum of the proton KE for a given size distribution of CH_4 clusters under the conditions of a pure Coulomb explosion.

The pure Coulomb explosion may not occur with comparatively large clusters at a given laser intensity. However, specific considerations should be included in the calculation. For the clusters with radii larger than the radius $R_0^{(I)}$, which is defined as the border radius for a given laser intensity I and is determined as

$$R_0^{(I)}(\text{nm}) = 55.195 \left(\frac{I/10^{18}\text{Wcm}^{-2}}{n/10^{22}\text{cm}^{-3}}\right)^{1/2} \cdot (\lambda/\mu\text{m}), \quad (6)$$

the charge state of carbon is lower than a given charge state, e.g., +4. In Eq. (6), $n = \rho\,(q_\text{C} + 4)$ is the electron density inside the cluster and λ is the laser wavelength. Instead of the original density ρ, a uniform distribution of the molecular density $\rho' = \rho R_0^{(I)}/R$ is assumed for the approximation of the charge distribution inside the cluster. In the present experiment, $q_\text{C} = 4$ was assumed for the laser intensity at 4×10^{18} W/cm^2. The border radius can then be estimated using the formula $R_0^{(I)}$(nm)$=$ $1.234 \times 10^{-8} I^{1/2}$(W/cm^2), thus yielding a border radius of 24.7 nm.

Finally, the average cluster size and size distribution can be numerically determined with referring to the experimental detected KE spectra. The red solid line in Fig. 3(a) represents the calculated energy spectrum of protons for the clusters with a log-normal distribution $f(N)$ of $\sigma = 1.18$ and $N_0 = 815$. An average size of $N_\text{av} = 6\,580$ was calculated by integrating over the whole distribution $N_\text{av} = \int Nf(N)\text{d}N/\int f(N)\text{d}N$. An average size of $N_\text{av} = 1\,235$ was calculated using Eq. (3) under 30 bars at room temperature. A similar calculation was performed for the spectrum of the protons produced at 240 K under 30 bars, and an average cluster size of $N_\text{av} = 6\,230$ was obtained. The generated average cluster size at 240 K was five times larger than that produced at room temperature under the same backing pressure. $N \propto a\Gamma^{*b} \propto c(P_0 T_0^\alpha)^b$ was assumed, considering a similar relation between cluster size and the source backing pressure as well as the source temperature for the polyatomic clusters. The relation $\alpha = \ln(P_1/P_2)/\ln(T_2/T_1)$ was derived when a similar average cluster size was obtained under different source conditions of stagnation temperature and backing pressure. Thus, $\alpha = -3.3$ and $\Gamma^* \propto T_0^{-3.3}$ was obtained despite the small differences in the measured average cluster sizes in the present experiments. The results show a stronger nonlinear relation between Γ^* and T_0 for the CH_4 clusters than between the monatomic or diatomic clusters. A further decrease in the stagnation temperature may result in a larger average cluster size given that the efficiency of the cluster condensation rate does not decrease with temperature.

In conclusion, this study experimentally investigates cluster formation at both of the room temperature and the low temperature. Similar average sizes are acquired for the CH_4 clusters under a backing pressure of 60 bars at room temperature and under a backing pressure of 30 bars at 240 K. By comparing the results, a remarkable increase in CH_4 cluster average size (five times in N_{av}) is achieved by lowering the stagnation temperature to 240 K. Moreover, a strong nonlinear relation of the Hagena parameter $\Gamma^* \propto T_0^{-3.3}$ for CH_4 clusters is revealed. The results demonstrate an efficient method of generating larger clusters under a relatively low backing pressure, which may favor the efficient generation of fusion neutrons during the interaction of intense femtosecond laser pulses with the CD_4 clusters.

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