Size dependence of the maximum energy of protons from Coulomb explosion of methane clusters under intense femtosecond laser irradiation

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Received January 24, 2006

An investigation of the cluster size dependence of the maximum energy of protons ejected from explosion of methane clusters in an intense femtosecond laser field has been conducted on the basis of the cluster size estimation by Rayleigh scattering measurements. The interaction of a 2×10^{16}-W/cm^2 intense laser pulse (790 nm, 60 fs) with the methane clusters revealed that the clusters were Coulomb exploded and the maximum energy ($E_{\max}$) of the protons produced was linearly proportional to the square of the cluster radius (r_c^2). In a cluster size range, with the methane cluster radii up to about 3 nm, the established relation of $E_{\max}$ and r_c^2 was found to be $E_{\max}$ (keV) = $3.3 + 0.75 r_c^2$ (nm^2), in good agreement with the simulation results. This demonstrated that Coulomb explosion of ionic clusters $(\mathrm{C}^{+4}\mathrm{H}_4^+)_n$ took place following the cluster vertical ionization in the laser-cluster interaction.

OCIS codes: 020.2070, 320.2250, 350.5400.

For more than a decade, the ultrashort intense laser interaction with clusters has become an active research frontier of high-field laser physics. The production of 1-MeV ions in the interaction of an intense femtosecond laser pulse with xenon clusters is surprising indeed[1]. From this finding, the authors pointed out that a plasma formed from explosion of deuterium clusters may make table-top nuclear fusion possible[1]. In 1999, Ditmire *et al.* reported the first results of table-top nuclear fusion from explosion of femtosecond laser-heated deuterium clusters in Lawrence Livermore National Laboratory[2], following the successful preparation of a cryogenically cooled high-pressure deuterium cluster jet[3]. The realization of nuclear fusion in which a table-top femtosecond laser system is used has stimulated much experimental and theoretical efforts, aiming mainly at the increase of the neutron yield in order to make possible material damage studies with neutron fluxes of $\geq 10^9/\mathrm{cm}^2\mathrm{s}$[4]. While the experimental studies showed the critical importance of the increase of the cluster size[5] , the laser intensity[6] and the enhancement of the laser-cluster interaction which will result in the increase of the neutron yield, Last and Jortner revealed in theory that Coulomb explosions of heteronuclear clusters under intense laser irradiation has advantages over the homonuclear clusters, involving the considerably increased D^+ kinetic energy and a narrow high-energy distribution of deuterons[7]. Soon later, Grillon *et al.* reported the first experimental results of table-top nuclear fusion of laser-heated heteronuclear clusters (CD_4)[8]. Meanwhile, Last and Jortner published a theoretical study of nuclear fusion driven by Coulomb explosions of methane clusters $(\mathrm{CA}_4)_n$, A = H, D, and T[9]. These studies have indicated the increase in deuteron energy with the use of the heteronuclear clusters. As a consequence, the neutron yield of nuclear fusion increases due to the increase of the cross section of the D-D reaction. Recently, Madison *et al.* reported an experimental study in which a comparison has been made of the fusion neutron yields and the ion energies for the deuterium cluster- and the deuterated methane cluster-plasmas, showing that the deuteron energy is larger for CD_4 clusters than for D_2 clusters, and the neutron production from the laser driven CD_4 clusters is also superior to that from the D_2 clusters when the incident laser energy is less than about 300 mJ[10]. Very recently, Hohenberger *et al.* studied the dynamic acceleration effects in explosion of laser-irradiated heteronuclear CH_4 and CD_4 clusters in low density cluster beams[11]. The two simulation results predicted that the maximum energy $E_{\max}$ of protons produced in the intense femtosecond laser interaction with methane clusters is proportional to the square of cluster radius (r_c^2), with the proportion coefficients lying between ~ 0.80[9] and ~ 0.60[11], whereas the experimental data are scarce currently. In this letter, we report an experimental investigation of cluster size dependence of the maximum kinetical energy of protons created from the ultrashort intense laser interaction with methane clusters. The results confirm that the maximum proton energy $E_{\max}$ is proportional to the square of the cluster radius r_c^2 with $E_{\max} \sim 0.75 r_c^2$, indicating the Coulomb explosion of the ionized methane clusters $(\mathrm{C}^{+4}\mathrm{H}_4^+)_n$ and the validity of the cluster vertical ionization (CVI) approximation[7,12] under the present experimental conditions.

The methane clusters were produced at room temperature by the supersonic expansion of a high pressure CH_4 gas into vacuum through a conical nozzle (26 mm in length, 500 μm orifice, and a half opening angle of 5°). For the estimation of the cluster size, Rayleigh scattering measurements were performed. The method used for the cluster size estimation was generally the same as that described previously[13]. Briefly, a 532-nm laser beam ($\sim$ 10 μJ, 20 ns) from a pulsed Nd:YAG laser was roughly focused by a lens onto the cluster jet at right angles. The focus was about 2 mm from the nozzle exit. The scattered light signal S_R from the clusters in the jet was then detected by a photomultiplier in the direction perpendicular to both the laser beam and the cluster jet. To well characterize the cluster size, some measures were taken to ensure the substantial suppression of the background light reflected from the vacuum chamber walls and the other items in the chamber. The scattered light signal S_R from the methane clusters in the gas plume is a function of the gas backing pressure P_0. The scattered light S_R shows a power scaling with the gas backing pressure P_0, as indicated by the $S_R \sim {P_0}^{4.0}$ curve in Fig. 1, in the measurements a 300-μm conical nozzle was used. An assumption is usually being made that the scattered light signal begins to emerge when the cluster size n (the number of atoms per cluster) is equal to about 100. This criterion is not precise enough indeed, however, it was generally accepted for size calibration in Rayleigh scattering measurements[3,13]. Referred to the Hagena relations[14], $\Gamma^* = k\frac{(d/\mathrm{tg}\alpha)^{0.85}}{T_0^{2.29}}P_0$, $n = 33\left(\frac{\Gamma^*}{1000}\right)^{2.35}$, and $r_c = \frac{r_0}{2}(n)^{1/3}$, where Γ^* is the Hagena parameter, k is a constant related to bond formation, d is the diameter of nozzle orifice (μm), α is the nozzle expansion half angle, T_0 and P_0 are the gas stagnation temperature (Kelvin) and pressure (mbar), respectively, and r_0 is the mutual interatomic spacing of the atoms within a cluster. We have cluster radius $r_c \sim d^{0.67}$. A factor of increment in cluster radius is then estimated to be about 1.4 for the nozzle orifice varied from 300 to 500 μm. The 500-μm nozzle was used in the present laser-cluster interaction experiment.

In our experiment, a Ti:sapphire laser (TSA-25) was used based on chirped pulse amplification, producing 790-nm pulses at a 10-Hz repetition rate. The laser pulse energy delivered to the gas plume was estimated to be ~ 15 mJ and the pulse duration was 60 fs. The laser was focused using an off-axis parabolic mirror with a focal length of 20 cm. The 2-mm focal spot downstream from the nozzle was about 40 μm in diameter, yielding a peak laser intensity of about 2×10^{16} W/cm^2. The ions produced from the exploded methane clusters, after passing through a skimmer, travelled along a 225-cm long field-free time-of-flight (TOF) tube which was perpendicular to both the laser beam and the cluster jet, and then were detected by a dual microchannel plate (DMCP) detector. The front plate of the DMCP was held at −1.5 kV and the back was grounded. A grounded metal mesh was placed 5 mm before the front plate of the DMCP, ensuring that the TOF tube was field free. The signal from the DMCP detector was recorded by a digital oscilloscope (TDS3052). The ion energy is determined by a TOF measurement through $E = \frac{1}{2}m\left(\frac{l}{t}\right)^2$ (m is the ion mass, l is the length of the TOF tube, and t is the flight time).

A TOF spectrum and the corresponding energy distribution of ions generated from explosion of methane clusters under 2×10^{16}-W/cm^2 laser irradiation at a backing pressure of 35 bar are shown in Fig. 2, in the experiment a 500-μm conical nozzle was used. In Fig. 2(a), the left sharp peak is due to photoelectrons produced by illumination of the DMCP detector by X-rays generated in the laser interaction with the cluster jet. The start point of this sharp peak serves to mark the beginning of the flight of the ions. The latter broad lobe results from the arrival of the ions to the DMCP detector. For the methane clusters, the ions contributing to the overall yield must involve C^{z+} besides protons. In the present experimental conditions, it is impossible to fully separate the carbon ions from the protons. However, the maximum proton energy can be read out in the energy distribution of the ions (Fig. 2(b)) unambiguously since the fastest protons should be the first to reach the DMCP detector and they are responsible for the start point of the lobe. Significantly, any energies larger than $E = E_c * m_A/m_c$, denoted in Fig. 2(b) by the dotted line, can be attributed to the detection of protons only[11]. Here m_A and m_c are the masses of hydrogen and carbon respectively, and E_c is the maximum energy of carbon ions which is estimated to be about 3 times the

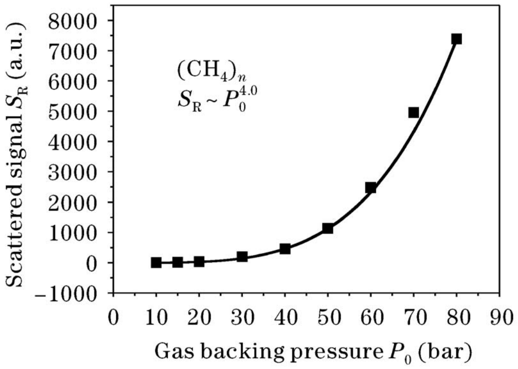

Fig. 1. Rayleigh scattered light signal S_R versus methane gas backing pressure P_0. In the measurements a 300-μm conical nozzle was used.

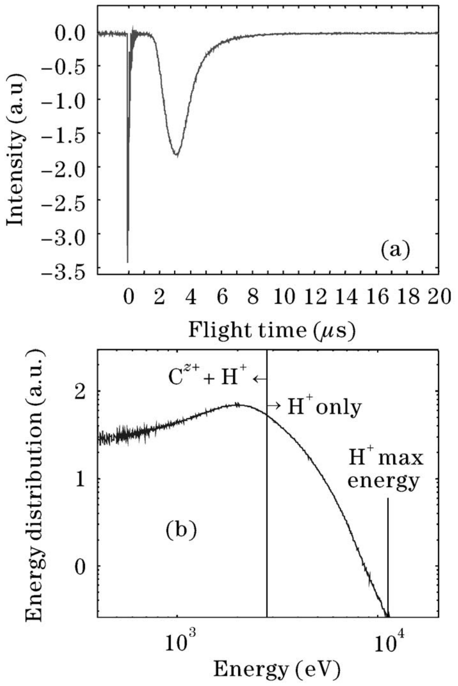

Fig. 2. Time-of-flight spectrum and corresponding energy distribution of ions from the exploding methane clusters. The gas backing pressure of 35 bar and a 500-μm conical nozzle were used.

maximum proton energy[11]. The errors of the maximum proton energies obtained from the energy spectra are estimated to be within ±10%.

In the experiment, the maximum energy of protons produced in the explosion of methane clusters irradiated by a 2×10^{16}-W/cm^2 laser pulse was measured as a function of the gas backing pressure and is shown in Fig. 3(a). Referring to the Rayleigh scattering measurements of the cluster size and by taking into account the different nozzles that were used in the cluster size measurements (orifice 300 μm) and in the laser-cluster interaction experiment (orifice 500 μm), we obtained a linear dependence of the maximum ion energy $E_{\max}$ on the square of cluster radius r_c^2. The maximum energy is linearly proportional to the square of the cluster radius, given as $E_{\max}$ (keV) $= 3.3 + 0.75 r_c^2$ (nm^2) (Fig. 3(b)). The slope of the line which defines the $E_{\max} \sim 0.75 r_c^2$ relation is sensitive to the measured maximum proton energies and the cluster radii, and also a signature of the Coulomb explosion of $(\mathrm{C}^{+k}\mathrm{H}^{+})_n$ where $k = 1 - 6$, depending on the laser intensity and the cluster size. The value of the slope here is 0.75, it is in good agreement with the simulation results reported by Last and Jortner[9], and, very recently by Hohenberger *et al.*[11]. The agreement may indicate clearly that the methane clusters with the radii of up to about 3 nm were Coulomb exploded under the irradiation of a 2×10^{16}-W/cm^2 laser pulse. At this laser intensity, the methane clusters were ionized to $(\mathrm{C}^{+4}\mathrm{H}^{+})_n$ before the strong Coulomb explosion took place. The good agreement of this experimental result with the simulation results may also indicate that the cluster size estimation made in the present Rayleigh scattering measurements seems fairly reliable. However, the maximum proton energy measured is a little higher than the simulation results[9,11]. This issue is remaining for open discussions and will be subjected to a further study in which the cluster size distribution in a gas jet should be considered in the theoretical treatment.

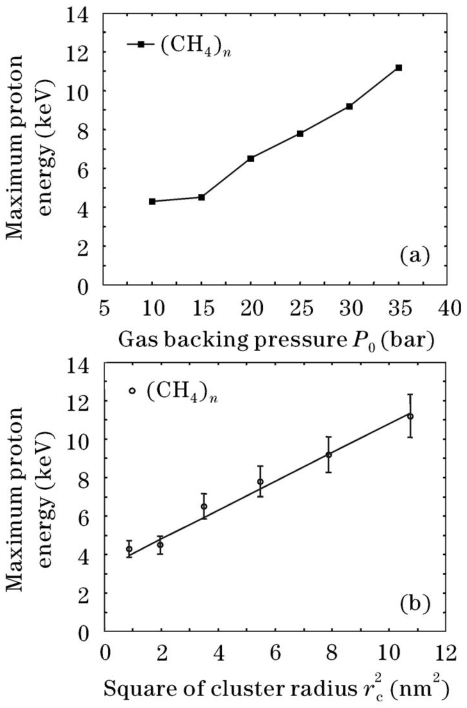

Fig. 3. Dependence of the maximum proton energy $E_{\max}$ of the exploding methane clusters on the gas backing pressure (a) and the square of the cluster size r_c^2 (b). $E_{\max}$ (keV) $= 3.3 + 0.75 r_c^2$ (nm^2) was obtained to characterize the line.

In conclusion, a study of the dependence of the maximum proton energy on the cluster size has been made in the intense femtosecond laser interaction with methane clusters based on the cluster size estimation by Rayleigh scattering measurements. The maximum proton energy $E_{\max}$ of the Coulomb explosion for methane clusters was found to be linearly proportional to the square of the cluster radius, and consistent well with the simulation results. The production of more energetic protons in the intense laser interactions with methane clusters is expected if a more intense laser pulse is employed to interact with larger-sized methane clusters.

This work was supported by the National Key Basic Research Special Foundation of China (No. G1999075200) and the National Natural Science Foundation of China (No. 10535070). G. Ni is the author to whom the correspondence should be addressed, his e-mail address is gqni@mail.shcnc.ac.cn.

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