Dynamic Explosions of Laser-Irradiated Hydrogen and Methane Clusters

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In recent years, the interaction of clusters with intensity ultrashort laser has attracted great attention. The most fascinating characteristics in this unique interaction are the observations of high charge states and very energetic ions [1-3]. The latter directly leads to the idea and implantation of table-top neutron sources [4]. Most studies have centered on single species homonuclear clusters such as Van der Waals bonded rare gas or hydrogen clusters. At enough intensity, particularly in small, low-Z clusters, an intense, ultrashort laser pulse can ionize and expel most of the electrons from cluster. This results in a pure Coulomb explosion, in which the ions expand nearly isotropically by mutual repulsion in the charged sphere, gaining kinetic energies simply related to their initial potential energy at their equilibrium position in the cluster[5]. Last and Jortner first revealed through simulation that Coulomb explosions of heteronuclear clusters subject into intense ultrashort laser irradiation has advantages over the homonuclear clusters, involving the considerably increased light ions (such as D⁺, H⁺) kinetic energy[6,7]. This energy enhancement is due to the presence of the multicharged high-Z ions which increases the Coulomb potential and besides the acceleration of the light ions can be enhanced if they overtake heavy ions during the explosion and are thus more strongly repelled. Very recently, Hohenberger et al first present experimental confirmation of Last-Jortner’s predictions [8].

In this letter, we show more evidences for the dynamic explosion of hydrogen and methane clusters. In our experiments, the hydrogen and methane clusters were produced by the expansion of high pressure H₂ and CH₄ gases into vacuum through a conical nozzle. For the production of the hydrogen clusters, the cluster source assembly consisting of a pulsed valve and the nozzle was cryogenically cooled to T₀~80K by liquid hydrogen in a reservoir which tightly connected to the assembly. Meanwhile, the methane clusters were generated at room
temperature ($T_0 \sim 295$K). For a more reliable estimation of the cluster sizes, Rayleigh scattering measurements[9] were made. The scattered light $S_R$ shows a power scaling with the gas backing pressure $P_0$, as indicated by the $S_R \sim P_0^\beta$ curves in Fig.1 (a) and (b). For the hydrogen and methane clusters, $\beta$ is 3.4 and 4.0, respectively.

**Fig.1** Rayleigh scattered light signal $S_R$ as a function of the gas backing pressure $P_0$ (a) for the hydrogen clusters, and (b) for the methane clusters. A nozzle with a 300$\mu$m orifice used.

The laser used is a chirped pulse amplification Ti:sapphire laser which delivers 60 fs pulses at the wavelength of 790 nm with a 10 Hz repetition rate. The laser intensity at the focal spot 2 mm downstream from the nozzle is about $1 \times 10^{16}$W/cm$^2$ and $1 \times 10^{17}$W/cm$^2$. The ions expelled from the clusters propagated along a 225 cm field-free flight tube after a skimmer and were detected by a dual microchannel plate detector (DMCP). The ion energies were determined by time-of-flight (TOF) measurements through $E = (1/2)ml^2/t^2$ ($m$ is the ion mass, $l$ is the length of the flight tube and $t$ is the flight time) and the energy distribution $f(E)$ of the ions can be obtained by conversion of the TOF spectrum $f(t)$ via $f(E) = f(t)(dE/dt)^{-1}$.

**Fig.2.** Dependence of the maximum proton energy $E_{\text{max}}$ of the exploding on the square of the cluster size $r_c^2$.

The maximum energies of protons produced in the explosions of hydrogen and methane clusters irradiated by a $1 \times 10^{16}$W/cm$^2$ laser pulse were measured and showed in Fig.2. The
maximum proton energy is linearly dependent on the square of the cluster size for both hydrogen and methane cluster, which indicated that the intense laser-irradiated hydrogen and methane clusters were Coulomb exploded, in consistence with the theoretical predictions[5,7].

![Fig.3](image1.png)

**Fig.3.** Energy distribution of ions from the exploding methane clusters, it is set to mass of H$^+$ in the transformation from time-of-flight spectrum.

![Fig.4](image2.png)

**Fig.4.** Dependence of the percent of H$^+$ only to the total distribution on the square of cluster radius $r_c^2(nm^2)$ at $I \times 10^{16} W/cm^2$ and $1 \times 10^{17} W/cm^2$.

When the heteromolecular clusters irradiated by laser, the acceleration of the light ions can be enhanced if they overrun heavy ions during the expansion, which occurs if the kinematic parameter for ion species A, $\eta_A=q_Am_B/q_Bm_A$, is greater than 1 (where $q$ and $m$ are the charge and mass of the light ion A and heavy ion B). In this case, the light ions explode in an outer shell with an average energy higher than what would be expected from the purely energetic argument based on the initial potential energy of the ions. A laser of intensity $\sim1\times10^{17} W/cm^2$ is sufficient for the clusters of radius $\sim3nm$ to be inner ionized to a charge state of C$^{4+}$H$_4^+$ and for CVI approximation to be valid [10]. For C$^{4+}$H$_4^+$, $\eta_H=3$, the dynamic effect is stronger. At a lower laser intensity of $\sim1\times10^{16} W/cm^2$, in Fig.2 shows the maximum H$^+$ energy is about twice for methane cluster compare to hydrogen cluster. As it is known, for pure coulomb explosion of clusters, the maximum ion energy of a cluster with density $n$ and radius $R$ is given by $E_{max}=4\pi Z^2e^2nR^2/3$, $n$ is $6\times10^{22}$ ions/cm$^3$ for (CH$_4$)$_n$ and $4.22\times10^{22}$ ions/cm$^3$ for (H$_2$)$_n$. So, it can be estimated at $I\sim1\times10^{16} W/cm^2$, C ions has an average charge state of 2. For (C$^{2+}$H$_4^+$)$_n$, although $\eta_H=6$, due to less charged, H$^+$ ions less benefit from the dynamic
acceleration effects. Any ions of energy larger than $E=\frac{E_{C}m_{H}}{m_{C}}$, denoted in Fig.3 by the line can be attributed to protons only. Here $E_{C}$ is the maximum energy of carbon ions which is estimated to be about 3 times the $E_{H^{+},\text{max}}$ [8]. The percent of the $H^{+}$ only to the total distribution were calculated at two laser intensity of $1 \times 10^{16}$W/cm$^{2}$ and $1 \times 10^{17}$W/cm$^{2}$, and showing in Fig.4. The small methane clusters has almost the same percent and larger methane clusters has a higher percent at $1 \times 10^{17}$W/cm$^{2}$ which intensity predicted to have stronger dynamic acceleration effects, which is consistent with the kinetic energy enhancement.

In conclusions, the maximum energy $E_{\text{max}}$ of the protons produced is proportional to $r_{c}^{2}$, where $r_{c}$ is the cluster size, which revealed that the hydrogen and methane clusters are Coulomb exploded irradiated by intense laser. A detailed analysis of the proton energy distribution from explosions of methane clusters at various laser intensity presents confirmation of proton energy enhancement in methane clusters which theoretically predicted by Last and Jortner.

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References