Energetic Protons Emitted from Coulomb Explosion Dynamics of Large-Sized Hydrogen Clusters Driven by an Ultrashort Intense Laser Pulse *

LI Hong-Yu(李洪玉)**, LIU Jian-Sheng(刘建胜), WANG Cheng(王成), NI Guo-Quan(倪国权), LI Ru-Xin(李儒新), XU Zhi-Zhan(徐至展)***

State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800

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Protons with very high kinetic energy of about 10 keV and the saturation effect of proton energy for laser intensity have been observed in the interaction of an ultrashort intense laser pulse with large-sized hydrogen clusters. Including the cluster-size distribution as well as the laser-intensity distribution on the focus spot, the theoretical calculations based on a simplified Coulomb explosion model have been compared with our experimental measurements, which are in good agreement with each other.

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The interactions of high-intensity lasers with clusters have been extensively explored over the past few years. As bridge systems between isolated molecules and condensed phase, clusters can provide optimum conditions for such processes as very efficient laser energy absorption,[1] incoherent and coherent x-ray emission,[2,3] production of energetic ions with highly charged states.[4,5] Especially it is pleasing that the laser-heated explosion dynamics of homonuclear or heteronuclear clusters containing deuterium can generate fast deuterium ions with high kinetic energy to efficiently overcome the Coulomb repulsive barrier and drive DD nuclear fusion.[6−9] Quite a lot of correlative researches have been carried out not only theoretically but also experimentally to investigate the dynamics of laser-cluster interaction as well as to produce table-top neutron sources.

In this Letter, we report the observation of highly energetic protons with maximum energy about 10 keV and average energy about 2 keV produced in the Coulomb explosion process of large-sized hydrogen clusters irradiated by an intense femtosecond laser pulse. In order to explain the generation of highly energetic protons in our experiments, a simplified Coulomb explosion model is employed to analyse the explosion dynamics. Taking the size distribution of clusters and intensity distribution of the laser focus spot into account, we compare the simulated proton energy spectrum and the saturation effect of proton energy with our experimental results, and obtain a good agreement between them. The investigation of the Coulomb explosion of hydrogen clusters which are analogues of deuterium clusters is very helpful for further research of deuterium cluster-laser interaction to obtain deuterium ions with keV-range energy sufficient to drive nuclear fusion.

The explosion of hydrogen clusters irradiated by ultra intense short laser pulses has been studied experimentally. In our experiment, the hydrogen clusters are produced by an adiabatic expansion of high-pressure low-temperature (about 80 K) hydrogen gas through a conical nozzle into vacuum. A pulsed sonic gas jet with a nozzle diameter of 0.3 mm and an expansion angle of 10° is employed in the experiment. The operation repetition of the jet is 1 Hz, and the vacuum is 2×10^{-3} Pa, which is good for cluster formation. The mean cluster size is controlled by varying the gas backing pressure. By means of the Rayleigh scattering technique,[10] the mean cluster size is estimated to be about 5.3 nm at the gas backing pressure of 80 bar. The laser pulses employed for the interaction with hydrogen clusters are delivered from a Ti: sapphire laser system operating at 800 nm, 200 mJ, and 60 fs in pulse duration with chirped-pulse amplification. The laser beam propagates in a vacuum tube before it enters into the target chamber and thus a good profile of the laser beam can be maintained. Focused by an off-axis $f/5$ parabolic mirror into a gas jet of hydrogen clusters, the radius of the focus spot is measured to be about 7.5 μm. The energy spectrum of protons produced from the exploded hydrogen clusters is measured by a 225-cm-long time-of-flight (TOF) spectrometer. The detection direction is perpendicular to the propagation of the laser beam. The measured energy spectra of protons at different laser intensities are shown in Fig. 1, in which the proton yield increases and then decreases slowly as the proton energy increases. Protons with high energy of about 10 keV have been observed. The average kinetic energy of protons as a function of laser intensity can

* Supported by the National Natural Science Foundation of China under Grant No 10535070.
** Email: lihongyu@mail.siom.ac.cn
*** Email: zzxu@mail.shcnc.ac.cn
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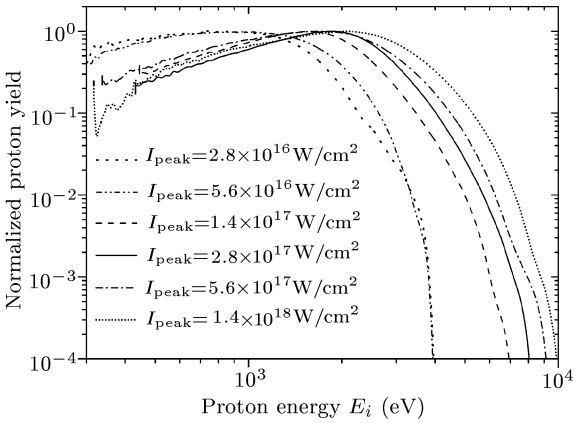


Fig. 1. Experimental energy distribution of protons emitted from hydrogen clusters at different laser peak intensities I_{peak}. The gas backing pressure for forming the clusters is 80 bar and the pulse duration is 60 fs (FWHM).

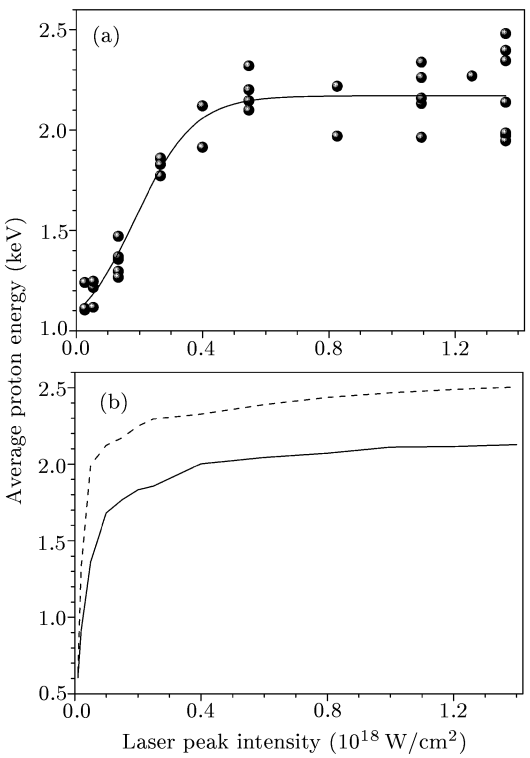


Fig. 2. (a) Dependence of measured average proton energy on laser peak intensity. The solid line represents a fit of the measured data. The gas backing pressure for forming the clusters is 80 bar. (b) Dependence of simulated average proton energy on laser peak intensity. The solid line represents the case including both the cluster-size distribution and the intensity distribution on the laser focus spot. The dashed line is under the consideration with only cluster-size distribution included.

be calculated and is given in Fig. 2(a). The average proton energy increases with the laser intensity as long as the intensity is lower than 0.6×10^{18} W/cm^2. Over the intensity, the average energy increases little and then reaches a plateau at about 2.2 keV.

The cluster expansion dynamics can be calculated by employing a simplified Coulomb explosion model without losing the main physical results. The interaction process is simplified as three ordinal sub-processes like inner ionization, outer ionization and Coulomb explosion process. The atoms inside the clusters will be ionized by optical field ionization (OFI) at the leading edge of the laser pulse. The removal of electrons from their host atoms is called inner ionization. It is assumed that no free electrons exist in the cluster until the laser field rises to the threshold intensity for inner ionization which can be estimated according to the one-dimensional barrier suppression model.[11] These unbound electrons generated by OFI will be heated and gain more kinetic energy from the laser field via the laser driven collisional process. Some parts of the heated electrons which have higher energies and reside on the outer layer of the cluster will first escape out of the cluster so that the cluster acquires positive charge. The process that free electrons escape out of the cluster is called outer ionization. If their energies are not high enough to overcome the positive potential barrier of the cluster ball, free electrons have to remain inside the cluster. Generally, the outer ionization begins simultaneously with the inner ionization, while in this model the outer ionization is assumed to take place after the inner ionization. As a consequence of the outer ionization, ions inside the cluster repulse each other by the Coulomb repulsive forces and the explosion of the cluster initiates. The movement of protons on the surface of a hydrogen cluster with an initial radius of R_0 can be expressed by the Newton equation of motion if the cluster is supposed as a uniform sphere,

$$m_p \frac{d^2R(t)}{dt^2} = \frac{N_e(t)e^2}{4\pi\varepsilon_0 R^2(t)} = \frac{N_c q(t) e^2}{4\pi\varepsilon_0 R^2(t)}, \qquad (1)$$

where m_p is the rest mass of a proton, e is the electron charge, $R(t)$ is the outer radius of the expanding cluster, and $N_e(t)$ is the amount of electrons removed from the cluster at the time instant t by the laser field. Here we define $N_e(t) = q(t)N_c$, while $N_c = 4\pi R_0^3\rho/3$ is the total number of hydrogen atoms inside the cluster, and ρ is atomic density of the hydrogen cluster. Therefore $q(t)$ is the average net charge state (outer ionization) of hydrogen atoms inside the cluster. $N_e(t)$ can be derived by assuming that the electrons can be expelled from the cluster if the ponderomotive energy U_p that the electrons acquire in the laser field with intensity I, and the central wavelength λ is higher than the Coulomb potential energy U_c of the charged cluster,[6,12,13] i.e.

$$U_p \Big[= \frac{e^2\lambda^2 I(t)}{8\pi^2 c^3 \varepsilon_0 m_e} = 933\lambda^2(\mu\mathrm{m})I(t)(10^{16}\,\mathrm{W/cm^2})\mathrm{eV}\Big]$$

$$\geq U_c \Big[= \frac{e^2 N_e(t)}{4\pi\varepsilon_0 R(t)} = 1.44 \frac{N_e(t)}{R(t)(\mathrm{nm}} \mathrm{eV} \Big], \qquad (2)$$

where c is the speed of light, ε_0 is the vacuum permittivity, and m_e is the electron rest mass. The motion of protons on the surface of the cluster can be calculated by solving Eqs. (1) and (2) numerically for different laser intensities. Under the consideration of uniform expansion, the radial expanding velocity of a proton inside the cluster increases linearly to its radial position; hence the motions of other protons on the inner layer of cluster can also been obtained. It has to be pointed out that in experimental realities, hydrogen clusters are produced with structure which is considered generally as a multiplayer icosahedron similar to that of rare gas microclusters.[14] Although a few large clusters with anomalous structure may be generated in experiments, the model with the assumption of uniform sphere is applicable for most hydrogen clusters with different sizes from small size (about 1 nm) to very large size (> 10 nm) whose structures are similar to a ball.

If a laser field has such a high intensity as well as a short rising time that the total electrons can be stripped out of the hydrogen cluster immediately, the explosion process of the cluster can be further simplified as a pure Coulomb explosion. The critical laser intensity required for expelling all electrons out of the cluster can be calculated from Eq. (2). By analytically solving Eq. (1) with the relation of $q = 1$, the motion trajectory of the protons on the surface of a fully charged hydrogen cluster is obtained as $t = \sqrt{3m_p\varepsilon_0}[\sqrt{R(t)/R_0(R(t)/R_0 - 1)} + \ln(\sqrt{R(t)/R_0} + \sqrt{R(t)/R_0 - 1})]/e\sqrt{2\rho}$. When the cluster explosion ends at infinity, owing to the energy conservation the protons on the surface of the cluster will gain a maximum kinetic energy transferred from its initial Coulomb electrostatic potential

$$E_{\max} = \frac{1}{4\pi\varepsilon_0}\frac{4\pi}{3} e^2 \rho R_0^2 = 0.181 R_0^2(\mathrm{nm})\mathrm{keV}, \qquad (3)$$

where the atomic density of a hydrogen cluster ρ is chosen as $3 \times 10^{22}/\mathrm{cm}^3$.[6] Given a uniform explosion, the proton energy distribution function is calculated as $dN_c/dE_i = 6\pi\varepsilon_0\sqrt{3\varepsilon_0 E_i/\rho}/e^3 \propto \sqrt{E_i}$, where E_i is the proton kinetic energy. The average proton energy is

$$\bar{E} = \frac{1}{N_c}\int E_i dN_c = \frac{3}{5} E_{\max}. \qquad (4)$$

The above analytical description indicates that the proton energy distribution is proportional to the square root of the energy. The proton yield increases as the proton energy increases to the maximum given by Eq. (3). However, the proton yield decreases sharply to zero as the energy is higher than the maximum energy, which is plotted as the dot-dot-dashed line in Fig. 3. This analytical result is quite different from the measured proton energy spectra shown in Fig. 1, where the yield of high-energy protons does not decrease sharply. The main cause is that the above analytical model considers only a single cluster size and the laser intensity distribution in the detection area is not included either. In fact, it is known that the formation of clusters does produce a wide size distribution.

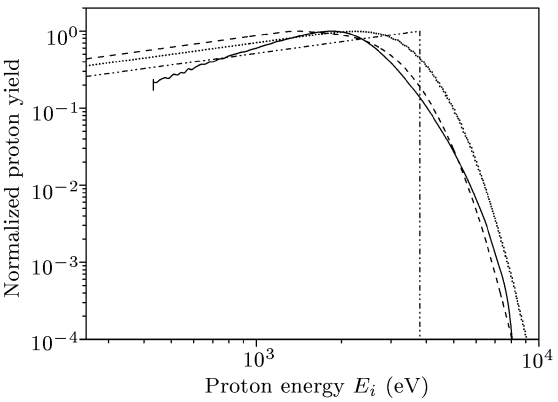


Fig. 3. Energy distribution of protons calculated with different considerations, compared with the experimental result (solid line) for the laser peak intensity of $2.8 \times 10^{17}\,\mathrm{W/cm^2}$. The dashed, dotted, dot-dot-dashed lines represent the results with cluster-size distribution and laser intensity distribution included, with only cluster-size distribution included, and without the two distributions included, respectively.

In order to compare with the measured energy spectra of protons, we include the effect of cluster distribution as well as the laser intensity distribution in the simulation. The cluster size distribution is assumed as a log-normal distribution function $f(N_c) \propto \exp[-\ln^2(N_c/N_0)/2w^2]$, where N_0 is the modal cluster size and w is proportional to the full width half maximum of the distribution.[15] The width of the distribution is assumed approximately equal to N_0 and thus w is 0.4087, accordingly. The mean cluster size $\bar{N}$ is related to N_0 by $N_0 = \bar{N}/1.29$. At the same time, the laser intensity distribution as a Gaussian shape on the focus spot plane is also considered.

We take the measured energy distribution of protons at the laser intensity of about $2.8 \times 10^{17}\,\mathrm{W/cm^2}$ as an example for comparison with the theoretical calculation, as shown in Fig. 3. Including the two distributions mentioned above, we find that the calculated proton energy spectrum (represented by dashed line in Fig. 3) can best fit the measured one (represented by solid line) when the calculated mean cluster radius R_{mean} (defined by $\bar{N} = 4\pi R_{\mathrm{mean}}^3\rho/3$) is chosen to be 5.5 nm, close to the estimated radius of 5.3 nm which is measured by Rayleigh scattering. It can be seen from Fig. 3 that the calculated proton yield increases and then decreases slowly as the proton energy increases,

in good agreement with the measurement especially in the high-energy range. However, the calculated proton yield in the low energy range is higher than that in the measurement. The discrepancy is partially because the electron efficiency of our micro-channel plate installed in the TOF spectrometer might be lower for protons with low energy compared to that for highly energetic protons. Another possible cause is that after expansion of the cluster some part of protons with low energy may recombine with electrons and become neutral hydrogen atoms, failing to arrive at the detecting system. If we include only the cluster size distribution but not the intensity distribution, the proton energy spectrum (dotted line) moves towards the higher energy region and the maximum proton energy (defined as the energy value where the proton yield is four orders lower than the maximum proton yield) of 9 keV is slightly higher than the measured value of 8 keV.

It can be seen from Fig. 2(a) that the average proton energy produced in the experiments increases with the laser intensity and then reaches a plateau at about 2.2 keV. It means that the proton energy becomes of saturation when the laser energy continues to increase and therefore the minimum laser intensity exists for maximizing proton energy. At this laser intensity, most clusters have already realized full Coulomb explosion and the highest Coulomb potential of the cluster has been transferred to the kinetic energy of protons. Including the cluster size and the laser intensity distribution, we calculate the average proton energy as a function of laser intensity, shown by solid line in Fig. 2(b). The tendency of proton energy to saturation value has a good similarity to the experimental trend, but slightly sharper. A peak laser intensity around $0.6 \times 10^{18}\,\mathrm{W/cm^2}$ is strong enough for saturating the proton energy in our simulation, close to the intensity required in the experiment. In addition, the calculated saturation value of the average proton energy is about 2.1 keV, in good agreement with the measured one. However, if the intensity distribution is not included, the increasing trend of proton energy as the laser intensity increases grows even more sharply and the average energy is higher than the experimental value, illustrated by dashed line in Fig. 2(b). It indicates that our consideration about the laser intensity distribution in the detection area is reasonable. The simulated and the measured average proton energies are lower than the analytically calculated energy value of 3.3 keV from Eqs. (3) and (4) for a pure Coulomb explosion of a hydrogen cluster with radius of 5.5 nm. The difference mainly originates from the effects of cluster size distribution as well as the laser pulse duration, which are not included in the analytical description.

In summary, high-energy proton emission from the interaction of an ultrashort-intense laser-pulse with large-size hydrogen clusters is measured. It has also been found that the cluster-size distribution as well as the laser-intensity distribution on the focus spot should be considered in order to compare the theoretical calculations with our experimental results such as the measured proton energy spectrum and the saturation effect of proton energy. In view of the similarity between hydrogen and deuterium clusters, these discussions are very helpful for further research of laser–deuterium cluster interaction to obtain ideal neutron sources.

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