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# Study on the interaction of intense femtosecond laser pulses with nanometre-sized hydrogen clusters*

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The interaction of intense femtosecond laser pulses with hydrogen clusters has been experimentally studied. The hydrogen clusters were produced from expansion of high-pressure hydrogen gas (backed up to $8\times10^6$Pa) into vacuum through a conical nozzle cryogenically cooled by liquid nitrogen. The average size of hydrogen clusters was estimated by Rayleigh scattering measurement and the maximum proton energy of up to 4.2keV has been obtained from the Coulomb explosion of hydrogen clusters under $2\times10^{16}$W/cm$^2$ laser irradiation. Dependence of the maximum proton energy on cluster size and laser intensity was investigated, indicating the correlation between the laser intensity and the cluster size. The maximum proton energy is found to be directly proportional to the laser intensity, which is consistent with the theoretical prediction.

**Keywords:** protons, hydrogen clusters, intense femtosecond laser pulse
**PACC:** 3640J, 5250J

## 1. Introduction

Since the availability of high-power ultrashort laser sources, their interaction with clusters has been an active research area for the interaction is extremely energetic and displays many features that are not observed when intense ultrashort laser pulses interact with atoms, molecules and solid targets. Rare-gas clusters, van der Waals bonded aggregates consisting of $10^3-10^6$ atoms, have proven to be a unique laser target that is very efficient in absorbing the incident laser energy. In Ref.[1] the experiment revealed that the laser energy absorption efficiency can even reach as high as 95% by using a xenon cluster jet, resulting from the near solid density within a single cluster and a gaseous average density of the jet without heat conductive dissipation into the cold solid substrate. This very high absorption efficiency implies that a large amount of laser energy is deposited in the cluster. Consequently, the explosion of the large rare-gas clusters leads to production of MeV ions,[2] keV electrons and soft x-ray photons.

Previous experiments have shown that when irradiated by an intense femtosecond laser pulse, deuterium clusters will eject fast deuterium ions that have sufficient kinetic energy to be harnessed to drive nuclear fusion which occurs in the D–D reaction of high-energy ions from adjacent clusters in a high-density gas plume.[3] The fusion neutron yield has been shown to be strongly dependent on the peak laser intensity,[4] cluster size,[5] and the average atomic density of the gas plume. Ions produced from the explosion of deuterium clusters have a broad energy distribution. Collisions between the ions with energies of a few keV or higher make D–D fusion events occur with high probability. The production of energetic ions from the interaction of the intense ultrashort laser pulse with clusters is dominated either by the hydrodynamic explosion[6] or Coulomb explosion,[7] depending on the laser intensity and the cluster species. For clusters composed of low $Z$ elements, such as hydrogen or deuterium clusters, when irradiated by an intense fem-

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tosecond laser pulse, all the field-ionized electrons can be directly removed very quickly by the laser pulse, making the individual cluster become a sphere of ions. Then the ionized hydrogen or deuterium clusters will explode under the strong Coulomb repulsion force and fast ions are ejected. It is indicated that under certain laser conditions which ensure cluster Coulomb explosion to take place, the ion energy increases with the square of the cluster radius.[8,9] A study of the dependence of the ion energy on the gas backing pressure and the laser intensity is required.

Hydrogen is an isotope of deuterium, a study of the interaction of an intense femtosecond laser pulse with hydrogen clusters[9,10] may show a close connection with nuclear fusion from explosions of femtosecond laser-heated deuterium clusters. In this paper, we present the diagnosis of the average hydrogen cluster size, the proton energy distribution from the laser–cluster interaction, and the dependence of the maximum proton energy on the cluster size (at a certain gas backing pressure) and the laser intensity. The explosion mechanism of hydrogen clusters is also discussed.

## 2. Experiment

In our experiment, hydrogen clusters were produced through the adiabatic expansion of high-pressure hydrogen gas through a conical nozzle ($d$=300$\mu$m, $2\alpha$=10°) into vacuum. Due to the very low bond-formation-related constant for hydrogen, clustering cannot occur at room temperature. To solve this problem, the cluster source was cooled by a cooling jacket reservoir which was full of liquid nitrogen. In this way, the temperature of the cluster source consisting of a pulsed valve and the nozzle was maintained at about 80K during the experiment. The mean cluster size was controlled by varying the gas backing pressure (up to $8\times10^6$Pa). Meanwhile, the pulsed valve was operated at a repetition rate of 0.2–1Hz, depending on the gas backing pressures in order to ensure a sufficient vacuum in the chamber during the experiment. The mean cluster size was measured by a Rayleigh scattering technique.[11,12] A 532nm laser beam from a frequency-doubled Q-switched Nd:YAG laser ($\sim$0.2$\mu$J , 10ns pulse duration) was roughly focused by a lens onto the cluster jet at right angles about 2mm from the nozzle exit. The scattered light from the cluster jet was then detected by a photomultiplier in the direction perpendicular to both the laser beam and the cluster jet. From the Rayleigh scattering measurement, a method commonly used,[11−13] the average cluster size can be estimated.

The laser utilized in our experiment was a Ti:sapphire laser system (TSA-25) based on chirped pulse amplification, producing 790nm pulses at a 10Hz repetition rate. The laser pulse energy delivered to the gas plume was estimated to be $\sim$8mJ and the pulse duration was 60fs. The laser was focused using an off-axis parabolic mirror with a focal length of 20cm, and the focal spot 2mm downstream from the nozzle was about 40$\mu$m in diameter, yielding a peak laser intensity of about $1\times10^{16}$W/cm$^2$. The protons produced from the exploding hydrogen clusters travelled along a 225cm long field-free time-of-flight (TOF) tube before reaching a detector. The TOF tube was arranged such that it was perpendicular to both the laser beam and the cluster jet. After passing through a skimmer, the protons were detected by a dual microchannel plate (DMCP) detector. The front plate of the DMCP was held at −1.5kV and the back plate was grounded. A grounded metal mesh was placed 5mm 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 proton energy was determined by a TOF measurement through $E=\frac{1}{2}m\left(\frac{l}{t}\right)^2$ ($m$ is the proton mass, $l$ is the length of the TOF tube, and $t$ is the flight time). The energy distribution $f(E)$ of the protons can be converted from the TOF spectrum of $f(t)$ via $f(E)=f(t)\times(\mathrm{d}E/\mathrm{d}t)^{-1}$) and the mean proton energy can be calculated through $\overline{E}=\dfrac{\int Ef(E)\mathrm{d}E}{\int f(E)\mathrm{d}E}$.

## 3. Results and discussion

Figure 1 shows the dependence of the peak intensity of the scattered light signal $S_\mathrm{R}$ from the clusters in the gas jet on the backing pressure $P_0$ at the cluster source temperature $\sim$80K. The scattered light signal $S_\mathrm{R}$ varies with $P_0$ pertaining to a power scaling $P_0^\beta$, with $\beta$ being 3.4 as shown in Fig.1. From $1\times10^6$ to $8\times10^6$Pa, the average cluster size increases from about 100 atoms to $n_\mathrm{c}\approx1.5\times10^4$ atoms. The corresponding radii of the cluster spheres being $r_\mathrm{c}$=0.83 and 4.4nm, respectively, estimated using the liquid hydrogen density given in Ref.[14].

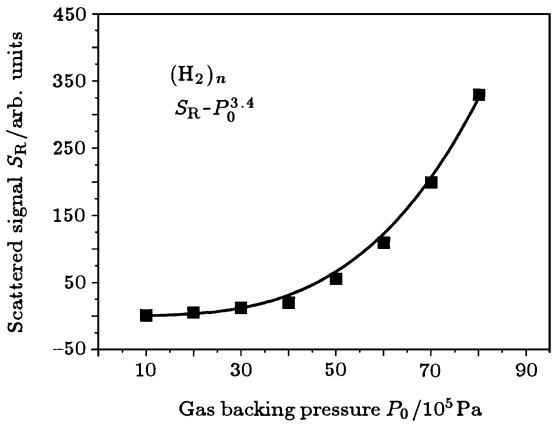



**Fig.1.** Scattered light signal $S_R$ as a function of the gas backing pressure $P_0$ at ~80K for hydrogen clusters, showing power scaling as $S_R - P_0^{3.4}$.

When irradiated by the $1 \times 10^{16}$W/cm$^2$ intense femtosecond laser pulse, the hydrogen clusters were rapidly heated and explosion was followed consequently, ejecting fast electrons and protons. Figure 2(a) gives the raw TOF spectrum of protons at the gas backing pressure of $6\times10^6$Pa ($r_c$=3.5nm), and the corresponding proton energy distribution converted from the TOF spectrum is shown in Fig.2(b). The left sharp spike in Fig.2(a) at $t$=0 coincides with the photoelectron ejection from the DMCP detector which is illuminated by UV radiation emitted from the laser-created plasma. The broad signal is a result of the arrival of the protons. As the field-free TOF tube is long enough (225cm), the proton signal can be separated completely from the photoelectron signal. From the TOF spectrum, the information of the proton energy, the proton energy distribution, and the average and maximum proton energies can be obtained. The average and maximum proton energies at the gas backing pressure of $6\times10^6$Pa are calculated to be 0.6 and 1.8keV, respectively.

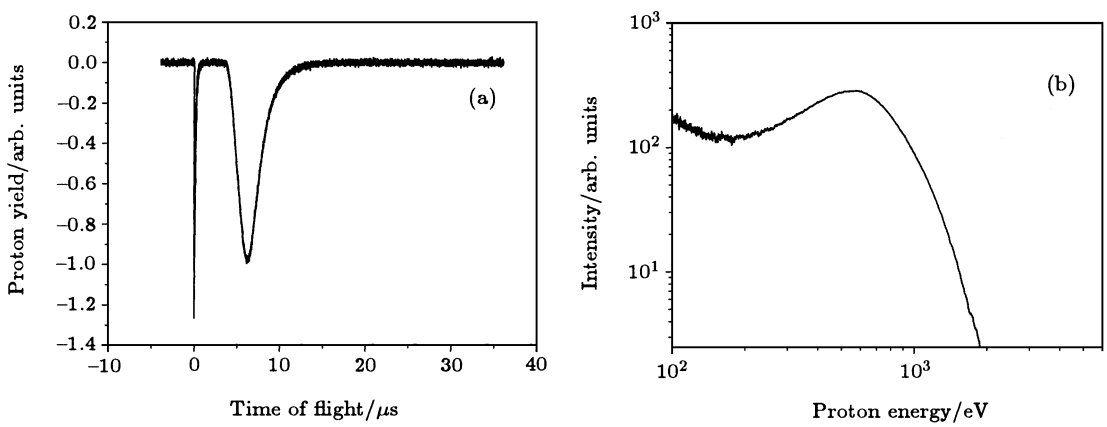



**Fig.2.** (a) TOF spectrum and (b) the corresponding energy distribution of protons produced by a 60 fs laser pulse with a $1 \times 10^{16}$W/cm$^2$ intensity which interacts with a hydrogen gas jet backed with a pressure of $6\times10^6$Pa.

Figure 3(a) shows the dependence of the maximum proton energy on the gas backing pressure. As can be seen, the maximum proton energy increases from 0.7 to 1.8 keV, as the gas backing pressure increases from $2\times10^6$ to $6\times10^6$Pa. It trends to become saturated when the hydrogen gas backing pressure is higher than $5\times10^6$Pa. This phenomenon of energy saturation, which takes place when the gas backing pressure is higher than a certain value, is similar to that reported by Sakabe *et al*,[9] due to the limited laser intensity compared with the large cluster size. A theoretical study reported by Parks *et al* indicates that for hydrogen or deuterium clusters pure Coulomb explosions occur only when the cluster radius $r_c$ is less than a certain maximum value for a fixed laser intensity $I_0$, i.e., $r_c < r_{max}(\text{nm}) \equiv 1.653 \times 10^{-8} I_0^{1/2}(\text{W/cm}^2)$.[8] In our experiment the laser intensity $I_0 = 1 \times 10^{16}$W/cm$^2$, then the corresponding maximum cluster radius for the pure Coulomb explosion to occur is $r_{max}$=1.653nm. According to our Rayleigh scattering measurement, clusters with an average size of $r_c$=1.653nm are produced at the gas backing pressure of $2.4\times10^6$Pa. The Coulomb explosion of the clusters of this size will lead to the production of ions with a maximum energy of 0.8keV, about a factor of 2 less than that reported in the experiment. An unambiguous explanation for this difference in the maximum energy is difficult. However, Parks *et al* indicate

that the boundary in terms of cluster radius between the fully (Coulomb) exploded clusters ($r_c < r_{max}$) and the unexploded clusters ($r_c > r_{max}$) is actually fuzzy, and, the critical cluster radius ($r_{max}$) is not so well defined since the laser–cluster interaction for $r_c \approx r_{max}$ is very complicated. They emphasize that the partial liberation of some electrons from a cluster with radius $r_c > r_{max}$ must be considered. As a consequence, an increase of 0.1–0.3keV for the maximum ion energy $E_{max}$ is estimated.[8] Besides, the clusters formed in the gas expansion process are characteristic of a broad size distribution that has been found to be best described by a log–normal distribution.[15] This results in the intense laser interaction with clusters of different sizes, not with a single defined one. The calculation made by Sakabe *et al* demonstrates a little bit broader ion energy distribution when the cluster size distribution is taken into account,[9] leading to a larger maximum proton energy. Then, within the limit of the experimental accuracy the discrepancy between the experimental data and the theoretically predicted value in the maximum proton energy could be explained. Actually, the same discrepancy can be found in Ref.[3] if the $r_c$(max) criterion is invoked.

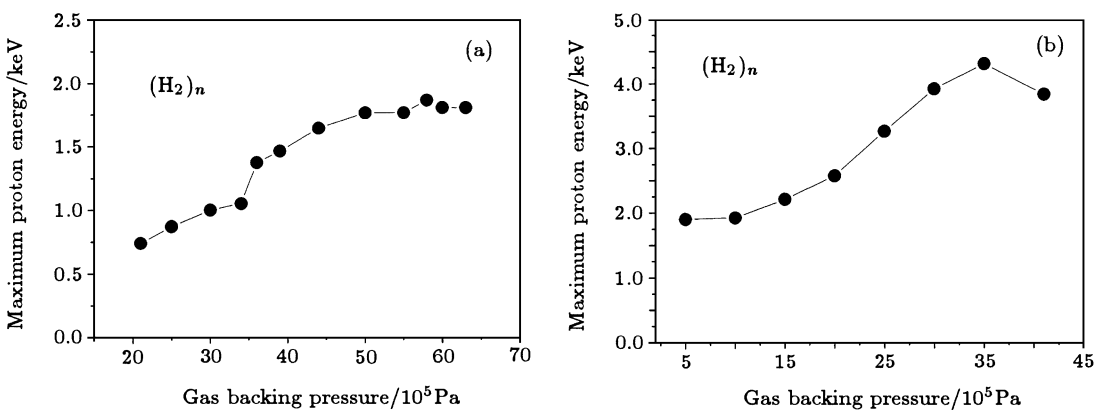



**Fig.3.** Dependence of the maximum proton energy on the hydrogen gas backing pressure. (a) with a peak laser intensity of $1 \times 10^{16}$W/cm$^2$ and a conical nozzle of 300$\mu$m in diameter, and (b) laser intensity of $2 \times 10^{16}$W/cm$^2$ and a conical nozzle of 500$\mu$m in diameter.

The above experimental result shows that the maximum energy of protons produced in the intense laser pulse interaction with hydrogen clusters is restricted by the limited laser intensity of $1 \times 10^{16}$W/cm$^2$. An experiment was conducted in which a more intense laser pulse with a $2 \times 10^{16}$W/cm$^2$ intensity was used to test the proton energy dependence on the laser intensity. The result shown in Fig.3(b) gives a maximum proton energy of 4.2keV which is about a factor 2 larger than that obtained when the $1 \times 10^{16}$W/cm$^2$ laser intensity was employed. This is consistent with the theoretical prediction that the maximum energy of ions produced from laser-irradiated clusters is directly proportional to the laser intensity in the case that the Coulomb explosion of the clusters is ensured. Along with the $2 \times 10^{16}$W/cm$^2$ laser, a conical nozzle with a larger orifice (500$\mu$m, $2\alpha$=10°) was used. It is estimated that a factor of 1.4 increase in cluster radius is realized with the nozzle orifice increased from 300 to 500$\mu$m while the other cluster source parameters are identical. Consequently, the maximum proton energy slightly declines for the gas backing pressure higher than $3.5 \times 10^6$Pa when the laser intensity has increased twice that in Fig.3(a).

## 4. Conclusion

In conclusion, the interaction of intense femtosecond laser pulses with hydrogen clusters has been studied. We measured the average hydrogen cluster size with a Rayleigh scattering method and detected the production of protons of a maximum energy of 4.2keV from the laser–cluster interaction. The dependence of the maximum proton energy on the gas backing pressure and the laser intensity is presented. The results show that for a given laser intensity the resulting maximum proton energy will saturate as the cluster size exceeds a certain value. The maximum proton energy is found to be proportional to the laser intensity when the Coulomb explosion of the clusters is guaranteed.

# References


[1] Ditmire T, Smith R A, Tisch J W G and Hutchinson M H R 1997 *Phys. Rev. Lett.* **78** 3121

[2] Ditmire T, Tisch J W G, Springate E, Mason M B, Hay N, Smith R A, Marangos J and Hutchinson M H R 1997 *Nature* **386** 54

[3] Ditmire T, Zweiback J, Yanovsky V P, Cowan T E, Hays G and Wharton K B 1999 *Nature* **398** 489

[4] Ditmire T, Zweiback J, Yanovsky V P, Cowan T E, Hays G and Wharton K B 2000 *Phys. Plasma* **7** 1993

[5] Zweiback J, Smith R A, Cowan T E, Hays G, Wharton K B, Yanovsky V P and Ditmire T 2000 *Phys. Rev. Lett.* **84** 2634

[6] Ditmire T, Donnelly T, Rubenchik A M, Falcon R W and Perry M D 1996 *Phys. Rev.* A **53** 3379

[7] Rose–Petruck C, Schafer K J, Wilson K R and Barty C P J 1997 *Phys. Rev.* A **55** 1182

[8] Parks P B, Cowan T E, Stephens R B and Campbell E M 2001 *Phys. Rev.* A **63** 063203

[9] Sakabe S, Shimizu S, Hashida M, Sato F, Tsuyukushi T, Nishihara K, Okihara S, Kagawa T, Izawa Y, Imasaki K and Iida T 2004 *Phys. Rev.* A **69** 023203

[10] Mendham K J, Tisch J W G, Mason W B, Hay N, Smith R A and Marangos J P 2002 *J. Phys.* B **35** 663

[11] Li S H, Liu B C, Ni G Q and Xu Z Z 2003 *Chin. Phys.* **19** 856

[12] Lei A L, Li Z, Ni G Q and Xu Z Z 2000 *Chin. Phys.* **9** 432

[13] Smith R A, Ditmire T and Tisch J W G 1998 *Rev. Sci. Instrum.* **69** 3798

[14] Madison K W, Patel P K, Price D, Edens A, Allen M, Cowan T E, Zweiback J and Ditmire T 2004 *Phys. Plasma* **11** 270

[15] Lewerenz M, Schilling B and Toennies J P 1993 *Chem. Phys. Lett.* **206** 381