Generation of high-energy protons from the Coulomb explosion of hydrogen clusters by intense femtosecond laser pulses

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The energy distributions of protons emitted from the Coulomb explosion of hydrogen clusters by an intense femtosecond laser have been experimentally obtained. Ten thousand hydrogen clusters were exploded, emitting 8.1 keV protons under laser irradiation of intensity 6×10^{16} W/cm^2. The energy distributions are interpreted well by a spherical uniform cluster analytical model. The maximum energy of the emitted protons can be characterized by cluster size and laser intensity. The laser intensity scale for the maximum proton energy, given by a spherical cluster Coulomb-explosion model, is in fairly good agreement with the experimental results obtained at a laser intensity of 10^{16}–10^{17} W/cm^2 and also when extrapolated with the results of three-dimensional particle simulations at 10^{20}–10^{21} W/cm^2.

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Recent progress in ultraintense femtosecond lasers has enabled the production of ionic radiation energetic enough to induce nuclear reactions, such as fusion, photofission, and electron-positron pair production [1–4]. The generation of high-energy ion radiation by intense femtosecond laser plasma interactions can be effected by two mechanisms. One is by Coulomb explosion in a gas or underdense plasma, and the other is by acceleration in an electrostatic field induced by high-energy electrons driven by a ponderomotive force in an overdense plasma in thin foils [5]. In the present paper we focus on the first of the two mechanisms. Intense femtosecond lasers can expel electrons from molecules or clusters instantaneously without disassociating them, producing highly charged molecular or cluster (parent) ions, a process known as optical field ionization. The molecules or clusters subsequently explode because of the repulsive Coulomb force due to their own highly charged state. This phenomenon is called "Coulomb explosion." During the Coulomb explosion of a parent ion cloud, the elemental ions acquire a high kinetic energy.

The interactions of a femtosecond laser with diatomic molecules have been extensively studied from the point of view of molecular dynamics [6–8]. It has been found that the kinetic energy released from a Coulomb explosion is small, for example, for Cl_2 molecules it is of the order of 10 eV [9]. The formation of highly charged ions and Coulomb-explosion dynamics in intense femtosecond laser fields for larger molecules such as benzene or the fullerene C_{60} have been studied by the present authors [10–12]. For large clusters, Ditmire *et al.* have demonstrated D-D fusion induced by the Coulomb explosion of deuterium clusters in a field of 2×10^{16} W/cm^2 [1,13]. For hydrogen clusters Zweiback *et al.* [14] and Mendham *et al.* [15] have measured the energy distributions of protons exploded from the clusters. However, the distributions do not show the features of cluster Coulomb explosion clearly.

In this paper, the energy distributions of protons emitted by the Coulomb explosion of hydrogen clusters in an intense femtosecond laser field are experimentally and analytically studied. An analytical model is shown to be able to predict the relationship between proton energy and laser intensity for a spherical uniform cluster.

Hydrogen clusters were generated from hydrogen gas (0.5–8.5 MPa of backing pressure) blown into a vacuum chamber (10^{-4} Pa) with a nozzle cryogenically cooled by liquid nitrogen [16]. The mean size of the clusters produced was measured by Rayleigh scattering [16,17]. The gas beam near the output of the gas-jet nozzle was irradiated with He-Ne laser light (wavelength 632 nm). The scattered light in the direction 90° to the laser beam was collected with a lens and imaged onto a photomultiplier tube through a spectrometer. The mean cluster size depends on the backing pressure and varied from 3×10^3 to 2×10^5 atoms/cluster for pressures ranging from 3.5 to 8.5 MPa. To be sure of the mean cluster size we employed the Hagena scaling law [18], using the empirical parameter Γ^*,

$$N=33\{\Gamma^*/1000\}^{2.35},\quad \Gamma^*=k\frac{P\ \ (\mathrm{mbar})}{[T_0(\mathrm{K})^{2.29}]}\left(\frac{\phi\ \ [\mu\mathrm{m}]}{\tan\alpha}\right)^{0.85},$$

where $k=184$ (for H), P is the pressure, $T_0=80$, α is the half opening angle of the jet (15°), and ϕ is the nozzle diameter (200 μm). Then the cluster size scales as $P^{2.35}$, which is in fairly good agreement with the experimental results for P, although the sizes have a discrepancy of more than 50% from each other, due to the measurement uncertainty of the parameters for both the Hagena law and Rayleigh scattering.

Laser pulses (wavelength 800 nm, pulse duration 130 fs, energy 200 mJ) from a chirped-pulse amplification Ti:sapphire laser were focused onto the hydrogen cluster beam through an $f=850$ mm spherical concave mirror ($f/17$). The intensity profile of the laser focal spot size was measured separately (full width at half maximum 0.2 mm), and the averaged laser intensity at the laser-cluster interaction region,

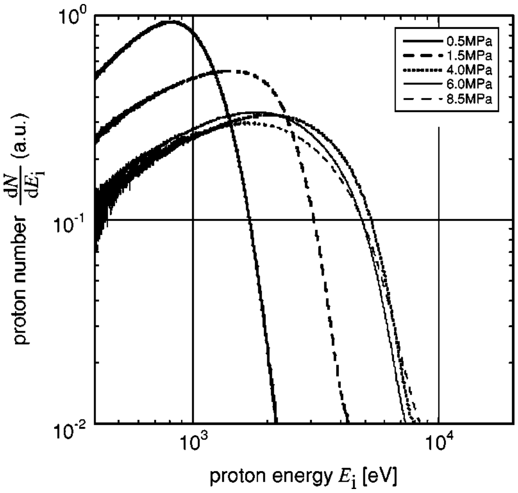

FIG. 1. Energy distributions of protons emitted from hydrogen clusters for varying nozzle backing pressures.

was 6×10^{16} W/cm^2, with an uncertainty of 20%. Protons exploded from the clusters were detected through a flight tube by a microchannel-plate detector located in the direction perpendicular to both the cluster beam and the laser beam propagation. The laser polarization was parallel to the time of flight axis. The flight times of the generated high-energy protons were reduced to an energy distribution.

Figure 1 shows the energy distributions of the protons at different backing pressures, taken from the time-of-flight spectra. As the proton energy increases, the number of protons slowly increases up to a peak energy, and then rapidly decreases as the maximum energy is approached. The dependence of the maximum energy on the backing pressure (corresponding to cluster size) is shown in Fig. 2. For backing pressures smaller than 4 MPa the maximum proton energy increases as the backing pressure increases. For pressures over 4 MPa the maximum energy does not increase and plateaus at 8.1 keV. The reason for the leveling off of the maximum proton energy above 4 MPa is that the laser intensity is insufficient to expel all electrons from the larger clusters produced at these pressures.

We can compare the experimental results with a uniform spherical non-neutral ion cluster model. The model is described in Ref. [19] and is briefly explained here. We consider a spherical cluster with a uniform density n and a radius R and assume equal electron and ion charge densities, with both components having zero temperature and being at rest initially. We first estimate the laser intensity required to expel all the electrons from the cluster. We introduce a normalized laser electric field amplitude $a=eE/m\omega c$, where e is the electron charge, E the electric field, m the electron mass, ω the laser frequency, and c the speed of light. For a laser with a wavelength of λ and an intensity of I, $a=0.85(I/10^{18}\ \mathrm{W/cm^2})^{1/2}(\lambda/\mu\mathrm{m})$. We assume a plasma that is transparent to the laser, i.e., ω is greater than the relativistic plasma frequency $\omega_{pe}=(4\pi e^2 n/m\gamma)^{1/2}$, or the cluster is smaller than the Debye length $\lambda_D=[mc(\gamma-1)/4\pi e^2 n]^{1/2}$, where $\gamma=(1+a^2/2)^{1/2}$ for linearly polarized light, and use the well known relation between the kinetic energy of relativistic electrons, $E_{ek}=(m^2c^4+p_\parallel^2c^2+p_\perp^2c^2)^{1/2}-mc^2$, and the electrostatic potential ϕ, $E_{ek}=p_\parallel c+e\phi$, where the longitudinal and transverse components of momentum are $p_\parallel=mca^2/2$ and $p_\perp=mca$. Here the electromagnetic wave is assumed to be a plane wave. The electrostatic potential appears due to the charge separation. Its value cannot be higher than the potential at the surface of a sphere with radius R: $\phi_{\max}=4\pi ZenR^2/3$, where Z is the ion charge state. If the value $e\phi_{\max}$ is very small compared to the kinetic energy, we obtain $E_{ek}=mca^2/2$. In the case of $E_{ek}\gg e\phi_{\max}$, all the electrons can be blown off by the laser radiation during the time $2R/c$. Thus we can estimate the laser amplitude required for expelling all of electrons from the cluster as

$$a>\left(\frac{8\pi Ze^2n}{3mc^2}\right)^{1/2}\equiv34\left(\frac{Zn}{5\times10^{22}\ \mathrm{cm^{-3}}}\right)^{1/2}\left(\frac{R}{1\ \ \mu\mathrm{m}}\right). \quad (1)$$

During the Coulomb explosion of a cluster, an ion obtains kinetic energy determined by its initial position in the cluster. Assuming a homogeneous distribution of the ion density n, the charge inside a radius r is given by $Q=4\pi Zer^3n/3$. If the ions peel off from the surface uniformly, an ion at the initial position r acquires an energy $E_i=4\pi Z^2e^2nr^2/3$ at infinity. The maximum ion energy of a cluster with density n and radius R is given by

$$E_{\max}=4\pi Z^2e^2nR^2/3\approx300Z^2\left(\frac{n}{5\times10^{22}\ \mathrm{cm^{-3}}}\right)\times\left(\frac{R}{1\ \ \mu\mathrm{m}}\right)^2\ \mathrm{MeV}. \quad (2)$$

Since the number of ions within a radius r to $r+dr$ is $dN=4\pi nr2dr$, the ion energy distribution function of a simple explosion can be given by

$$\frac{dN}{dE_i}=\frac{3}{4Z^2e^2}\sqrt{\frac{3E_i}{\pi n}}. \quad (3)$$

The ion energy distribution is thus proportional to the square root of the energy. The energy distribution presented by this

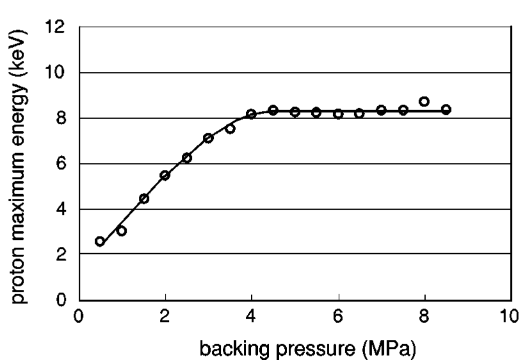

FIG. 2. Maximum proton energy as a function of backing pressure.

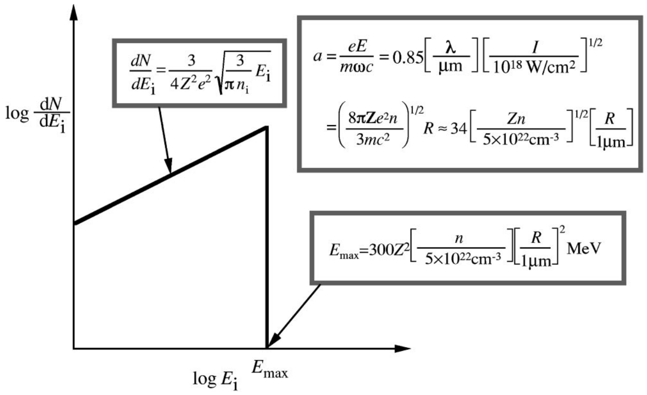

FIG. 3. Energy distribution for a uniform spherical cluster analytical model.

model is shown in Fig. 3. The distribution is examined by three-dimensional particle-in-cell (PIC) simulations in Ref. [19] as given later.

A comparison between Fig. 1 and Fig. 3 shows qualitative agreement, that is, they both show an increase in proton number as the energy increases up to the peak energy. However, in Fig. 1 the proton number does not decrease as rapidly as it does in the model. This is due to the fact that the model considers only a single cluster size, whereas in the experiments there is a distribution of cluster sizes, which all contribute to the observed distribution. Zweiback *et al.* show the proton spectrum in Fig. 5 in Ref. [14]. It does not show dN/dE proportional to $E^{1/2}$ and the truncation at E_{max}. Mendham *et al.* also give the proton energy distribution in Fig. 2 in Ref. [15], and the truncation is not clearly seen.

Here we discuss the energy dependence for a cluster size distribution. Figure 3 is valid only for a single cluster size, but actually the sizes will be widely distributed, although in the present experiment the cluster size distribution could not be measured. We assume clusters are distributed as

$$\frac{dN_c}{dN_a}=f(N_a)=\exp\left[-\frac{\ln^2 N_a}{2w^2}\right], \tag{4}$$

where N_a is the cluster size normalized by the model size and w is proportional to the full width at half maximum of the distribution [20]. Figure 4(a) shows the distributions for $w=0.5$, 1.0, and 1.2. The energy distribution in Fig. 3 can be described as $dN/dE=E^{1/2}u(E_{max}-E)$, where $u(x)$ is 0 for $x<0$ and 1 for $x>0$. When R_0 is the maximum radius of clusters from which all electrons are expelled for a given laser intensity, E_{max} is proportional to the product of the density of atoms in a cluster and the square of the radius. Here e is the ion energy E normalized by $E_{max}(R_0)$ and r is the radius R normalized by R_0. For $R<R_0$ $(r<1)$ $e_{max}=E_{max}(R)/E_{max}(R_0)=R^2/R_0^2=r^2$. For $R>R_0$ $(r>1)$ the cluster is partially ionized, and here we assume that the density of ions in the cluster is reduced, and then $e_{max}=E_{max}(R)/E_{max}(R_0)=(R/R_0)^3nR^2/nR_0^2=R_0/R=r^{-1}$. Equation (4) can be rewritten as the radius distribution $g(r)=dN_c/dr$ with N_a proportional to $R^3(r^3)$. The total energy distribution is given by

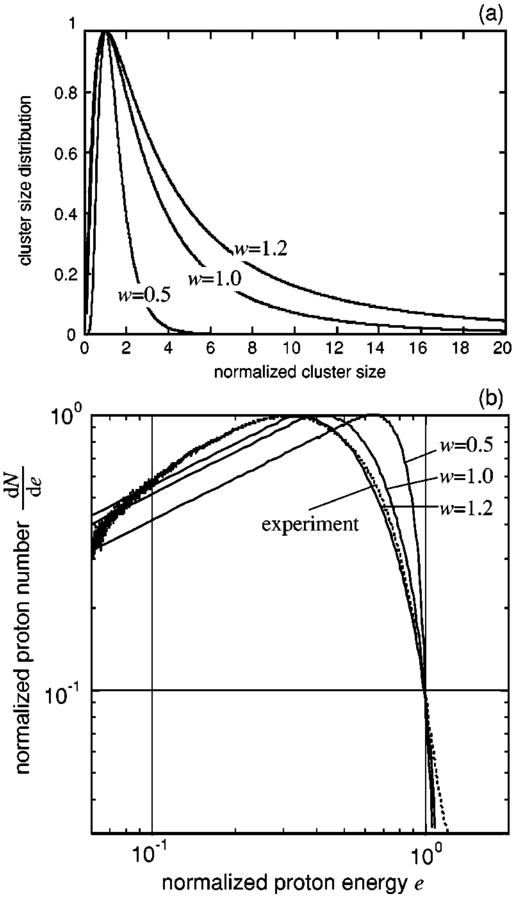

FIG. 4. (a) Log-normal cluster size distributions. The most abundant cluster size is normalized to unity. (b) Energy distributions of protons calculated with the spherical cluster Coulomb-explosion model and log-normal cluster size distributions, compared with the experimental result for 4 MPa backing pressure.

$$\frac{dN}{de}=\int_0^1 g(r)e^{1/2}u(r^2-e)dr+\int_1^\infty g(r)e^{1/2}u(r^{-1}-e)dr. \tag{5}$$

Figure 4(b) shows the energy distributions for $w=0.5$, 1.0, and 1.2 with the experimental result for 4 MPa backing pressure. All distributions are normalized at maximum and also at the energy giving 1/10 of the maximum ion distribution. The energy distribution can be fairly reproduced by Eq. (5) with $w=1.2$. We think in the present experiment the cluster size is broadly distributed.

Equation (2) gives the relationship between maximum energy and cluster size. For $E_{max}=8.1$ keV, as shown in Fig. 2, Eq. (2) give a cluster radius of 7.7 nm, which is in only fair agreement with the 4.8 nm measured by Rayleigh scattering. However, if we take the peak energy of the distribution, about 2.2 keV, to be the maximum energy for an average-size cluster, the cluster radius given by Eq. (2) is 4.0 nm, which is in good agreement with the 4.8 nm Rayleigh scattering measurement. From Eqs. (1) and (2) the laser intensity for full ionization can be related to the maximum energy, $a=1.97E_{max}^{1/2}$. For $E_{max}=8.1$ keV, the laser intensity is $I=6.7\times10^{16}$ W/cm^2, which is very close to the intensity of

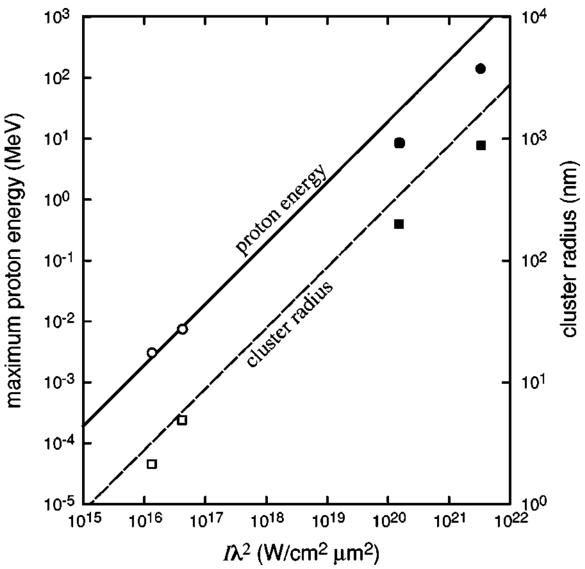

FIG. 5. Maximum proton energy (solid line) and cluster size (broken line) for the laser intensity given by a uniform spherical cluster analytical model. The horizontal axis shows the product of the laser intensity and the square of the laser wavelength. The circles and squares are maximum energy and cluster size, respectively. The open and closed symbols are the experimental results and the three-dimensional PIC simulation results of Ref. [19], respectively.

6×10^{16} W/cm^2 estimated from the focal spot size and laser energy. Note that the maximum energy is directly proportional to the laser intensity. We can conclude that the simple spherical cluster model can be used to determine the characteristics of high-energy ions emitted from a cluster exploded by an intense laser.

In the spherical cluster model, the exploded proton energy is proportional to the laser intensity, as shown in Fig. 5. Both the experimental results and predictions for higher intensities made using the three-dimensional PIC simulation are also given in Fig. 5. The PIC simulation is described in Ref. [19], but here is briefly mentioned. A flat top laser pulse with a rise time of $(3\omega)^{-1}$ (ω is the laser frequency) irradiates a spherical cluster of radius R_0. The number of particles (electrons and protons) is 1 071 580 in total. The three-dimensional simulation boxes are approximately (12λ, 5λ, 5λ) for a cluster of $R_0=0.2$ μm and (31 λ, 20 λ, 20 λ) for $R_0=0.8$ μm, where λ is the laser wavelength. The number of meshes is (300, 128, 128) and (400, 256, 256), respectively. The simulation results show that the energy distributions of protons are proportional to the square of the energy and truncated at the maximum energy, similar to the prediction of the spherical cluster model. For the clusters (the laser intensity) of $R_0=0.2$ μm ($a=10$) and $R_0=0.8$ μm ($a=50$), the maximum energies are 9 MeV and 150 MeV, respectively. Although the laser intensity used in our studies was insufficient to explode clusters larger than 10 nm, we can predict higher-energy results up to MeV for 100-nm-size clusters and a laser intensity of 10^{18} W/cm^2.

Usually protons generated in thin foil plasmas are distributed in a Boltzmann distribution with the high-energy protons located only at the tail end of the distribution. Coulomb-explosion protons, on the other hand, have a narrow energy band, which is a significant advantage. The former generates protons in a beamlike fashion, whereas the latter is isotropic. Although Coulomb explosions can be induced only in underdense gas by intense femtosecond lasers, they can be an efficient way to generate high-energy bursts of ions if a target material with a local density high enough for Coulomb explosion and average density high enough for efficient emission, yet low enough for laser propagation, is available, such as foam structured targets [21]. From the point of view of feasibility of laser produced ion sources, the details of both approaches, e.g., ion intensity, efficiency, and energy spectrum narrowing, must be further studied. The present experimental results have shown the validity of the model, which will be useful for future experiments.

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