Cluster-size dependence of proton kinetic energy from hydrogen-cluster explosion driven by laser pulse

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A simplified Coulomb explosion model is presented for the analysis of the explosion dynamics of hydrogen clusters driven by an ultrashort intense laser pulse. The scaling of the proton kinetic energy with cluster size has been studied in detail based on this model. It is found that the maximum kinetic energy the protons acquire in the laser-cluster interaction rises to a peak and then decreases slightly as the cluster size increases, which can be explained very well by investigating the temporal evolution of outer ionization rate of different-size clusters. It is also indicated that there exists an optimum cluster size to maximize the proton energy for given laser parameters. Moreover, taking the cluster-size distribution as a log-normal function distribution into account, the maximum proton energy increases sharply with the cluster size and then levels off before beginning to fall slowly. The inclusion of a cluster-size distribution into the simulations considerably improves the fit with experimental data. These discussions are useful for the optimum-match determination of laser-cluster parameters to obtain the maximum proton energy in experiments.

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Over the past few years, the interaction of high-intensity laser with clusters has become an active research frontier due to its extensive potential applications. Combining the optimal properties of isolated molecules and condensed phase, clusters have proven to be a unique target that can absorb laser energy very efficiently[1,2], and produce energetic ions with highly charged state[3,4] as well as strong X-ray emission[5−7]. The laser-irradiated clusters containing deuterium generate multi-keV deuterium ions capable of efficiently overcoming the Coulomb repulsive barrier and driving nuclear fusion[8−12]. This will open the potential of developing a compact, easily implemented neutron source.

The interaction of ultrafast intense lasers with clusters mainly contains three sub-processes: ionization of atoms, absorption of laser energy, and expansion of clusters. Whether hydrodynamic pressures or Coulomb repulsive forces prevail in the interaction depends on the laser-cluster parameters. In order to investigate these fantastic processes and explain the observed experimental phenomena, quite a few models have been put forward such as nanoplasma model[13], ionization-ignition model[14], collective oscillation model[15], molecular dynamic model[4,10], and particle-in-cell (PIC) code model[2,16]. The laser-cluster parameter dependence of the ion energy obtained in the expansion process has also been investigated[3,8,10]. For example, neutron yield from femtosecond laser-driven explosions of homonuclear deuterium clusters and heteronuclear clusters containing deuterium is measured and calculated as a function of cluster size[8,12], laser pulse width[8], and laser energy[11,17]. Besides, the effects of the cluster size distribution as well as the laser profile in the interaction area are being realized by some researchers[18−21]. Although correlative researches have been made not only theoretically but also experimentally, so far no complete agreement has been reached on the exact mechanisms as to how these processes happen and evolve.

In this paper, by use of a simplified Coulomb explosion model[21] which avoids the time-consuming calculations of molecular dynamic model and PIC model, we investigate the cluster-size dependence of proton kinetic energy generated from hydrogen clusters irradiated by an intense femtosecond laser pulse. The inherent relationship of outer ionization rate and proton energy has been analyzed in detail. Moreover, it is proven that the inclusion of cluster size distribution in the numerical simulations improves the fit with the experimental result. Deuterium is an isotope of hydrogen, and therefore the study on the interaction of intense femtosecond laser pulses with hydrogen clusters is very helpful for the laser driven neutron fusion research.

For hydrogen clusters irradiated by a high-intensity laser pulse, the interaction process can be simplified as three ordinal sub-processes like optical field ionization (OFI), outer ionization and Coulomb explosion. The atoms inside clusters will be ionized by OFI at the leading edge of the laser pulse. The process that the electrons are removed from their host atoms is called inner ionization. These unbound electrons will be heated and gain more and more kinetic energy from the laser field via laser driven collisional process. Some heated electrons which have higher energies and reside on the outer layer of the cluster will first escape 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. As a consequence of the outer ionization, protons inside the cluster repulse each other by the Coulomb repulsive forces and the explosion of the cluster initiates.

A much more simplified model can be employed to calculate the expansion process of hydrogen clusters without losing the main physical results. It is assumed that no free electrons exist in the cluster until the laser field rises

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to a threshold intensity for inner ionization. According to the simple one-dimensional barrier-suppression model[22], the threshold laser intensity which is necessary for the bound electrons to escape without tunneling is calculated as $I_{\mathrm{th}}(\mathrm{W/cm^2}) = 4.00 \times 10^9 E_{\mathrm{ion}}^4(\mathrm{eV})/Z^2$, where E_{ion} is the ionization potential and Z is the charge state. After the inner ionization, the outer ionization takes place. The movement of the protons on the surface of the 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

$$m_{\mathrm{p}}\frac{\mathrm{d}^2R(t)}{\mathrm{d}t^2} = \frac{N_{\mathrm{e}}(t)\,e^2}{4\pi\varepsilon_0 R^2(t)} = \frac{N_{\mathrm{c}}q(t)\,e^2}{4\pi\varepsilon_0 R^2(t)}, \quad (1)$$

where m_{p} is the rest mass of a proton, e is the electron charge, ε_0 is the vacuum permittivity, $R(t)$ is the outer radius of the expanding cluster, and $N_{\mathrm{e}}(t)$ is the amount of electrons removed from the cluster at the instant t by the laser field. Here we define $N_{\mathrm{e}}(t) = q(t)N_{\mathrm{c}}$, while $N_{\mathrm{c}} = 4\pi R_0^3\rho/3$ 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. $N_{\mathrm{e}}(t)$ 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 central wavelength λ, is higher than the Coulomb potential energy U_{c} of the charged cluster[8,19,23], i.e.,

$$U_{\mathrm{p}}\left[=\frac{e^2\lambda^2 I(t)}{8\pi^2c^3\varepsilon_0 m_{\mathrm{e}}} = 933\lambda^2(\mu\mathrm{m})I(t)\,(10^{16}\mathrm{W/cm^2})\mathrm{eV}\right] \geq U_{\mathrm{c}}\left[=\frac{e^2N_{\mathrm{e}}(t)}{4\pi\varepsilon_0 R(t)} = 1.44\frac{N_{\mathrm{e}}(t)}{R(t)\,(\mathrm{nm})}\mathrm{eV}\right], \quad (2)$$

where c is the speed of light, 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. By use of this relationship, the velocities of other protons on the inner layer of cluster can also be obtained since the velocity of a proton on the surface of cluster can be deduced as the time-derivative of its trajectory.

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 (PCE)[21]. The critical laser intensity required for expelling all electrons out of the cluster can be calculated from Eq. (2) as

$$I_{\mathrm{crit}}(\mathrm{W/cm^2}) = 8\pi^2c^3m_{\mathrm{e}}\rho R_0^2/3\lambda^2 \approx 2.73\times10^{15}R_0^2(\mathrm{nm})/\lambda^2(\mu\mathrm{m}). \quad (3)$$

The maximum kinetic energy the protons gain in the interaction is

$$E_{\max} = \frac{1}{4\pi\varepsilon_0}\frac{4\pi}{3}e^2\rho R_0^2 = 0.254R_0^2(\mathrm{nm})\mathrm{keV}, \quad (4)$$

and the average proton energy is calculated as

$$E_{\mathrm{av}} = \frac{3}{5}E_{\max}, \quad (5)$$

where the atomic density of a hydrogen cluster ρ is chosen as 4.22×10^{22} cm^{-3}[17].

If the laser intensity is lower than the critical intensity or its rise time is not so fast, a cluster can only be removed off a part of electrons during the explosion process. In this case, the Coulomb explosion is mixed with some hydrodynamic processes such as inverse bremsstrahlung absorption. As a result, the outer ionization keeps taking place and the amount of net charge inside the cluster changes during the explosion.

Hydrogen clusters with initial local atomic density $\rho=4.22\times10^{22}$ cm^{-3} are supposed to be irradiated by a Gaussian laser pulse with its central wavelength of 800 nm and a pulse duration (full width at half maximum) of 60 fs. The laser intensities are selected from 2×10^{16} to 1×10^{17} W/cm^2 which is located in the adjustable range of focusability of the chirped pulse amplification laser system of 5 TW in our laboratory. The scaling of the maximum proton energy $E_{\max}$ with the square of initial cluster radius R_0 at different laser peak intensities I_{peak} has been calculated as shown in Fig. 1. For lower laser peak intensity, e.g., 2×10^{16} and 4×10^{16} W/cm^2, the maximum proton energy rises to a peak of 2.27 and 4.55 keV when the cluster radius is 5.0 and 7.0 nm respectively, and then falls with the increase of cluster size. The reason is that at a lower intensity the laser field can only outer-ionize smaller clusters completely in a very short time and a PCE occurs, while for large clusters, the outer ionization cannot be finished instantaneously and some electrons reside in the cluster during the expansion process. It can be explained very well by carefully investigating the temporal evolution of cluster outer ionization rate with different cluster sizes. The outer ionization rate is defined as the amount $N_{\mathrm{e}}(t)$ of electrons expelled off the cluster divided by the amount N_{c} of all electrons inside the initial hydrogen cluster.

Figure 2 plots the temporal evolution of cluster outer ionization rate at the laser peak intensity of 4×10^{16} W/cm^2. When the cluster size is relatively small, e.g., 2 nm, the outer ionization rate rises to unit shortly and keeps constant in the later time. This means all electrons can be removed from small clusters before the laser peak comes. Correspondingly the maximum proton energy increases with the enlarged cluster size, as given in Fig. 1.

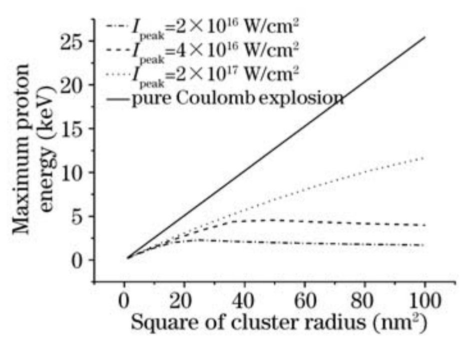

Fig. 1. Scaling of the maximum proton energy with the square of cluster radius for different laser peak intensities I_{peak}.

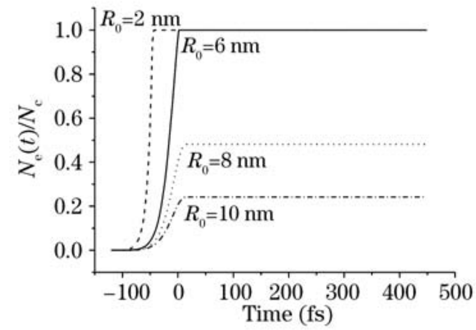

Fig. 2. Temporal evolution of cluster outer ionization rate $N_e(t)/N_c$ with different cluster sizes R_0. The laser peak intensity I_{peak} is $\sim 4\times10^{16}$ W/cm^2 and the pulse duration is 60 fs. The zero time marks the peak of the laser pulse.

For the cluster size larger than 6 nm, the outer ionization rate keeps a certain value which is less than unit after the laser peak. The reason why the rate does not vary any more after the laser peak is that the falling edge of laser pulse cannot provide electrons with enough ponderomotive energy for departing from the cluster and some electrons have to reside in the cluster. Moreover, the final value of outer ionization rate begins to decrease with the enlarged cluster size. Therefore the maximum energy the protons acquire will saturate and slightly decrease as the cluster size increases. The maximum Coulomb potential of the cluster plasma produced by the laser field is no longer proportional to the square of the cluster radius but determined by the laser intensity. This saturation effect of proton energy as a function of cluster size is in agreement with our experiment[24] as well as the reported observations[3,19]. The saturation point shown in Fig. 1 can be seen as a rough judgment when the cluster expansion begins to transit from a PCE to a mixed expansion driven by Coulomb pressure and hydrodynamic factors. It also indicates that there exists an optimum cluster size to maximize the average proton energy for given laser parameters. However, if we calculate the maximum cluster radii for a PCE at different laser intensities by using the formula R_0 (nm)$=1.572\times10^{-8}\sqrt{I\ (\text{W/cm}^2)}$ given by Eq. (3), the cluster radii at different laser intensities of 2×10^{16} and 4×10^{16} W/cm^2 are calculated as 2.22 and 3.14 nm respectively, which are a little less than half of the cluster radii corresponding to the proton peak energies as mentioned above. It means that this calculated cluster radius doesn't correspond to the cluster radius for the saturation of proton energy. For higher peak intensity, e.g., 1×10^{17} W/cm^2, the maximum cluster radius for a PCE is calculated as 4.97 nm while our simulation indicates that the required radius for the saturation of proton energy can be as large as 10 nm. Therefore, under this laser intensity the maximum proton energy increases all the time with cluster size as shown in Fig. 1 and the maximum energy is proportional to the square of the cluster radius. For PCE, the maximum proton energy increases linearly to the square of the cluster radius, and this increasing trend is faster than those under the laser intensities considered above because in the latter cases the effect of the pulse duration is included.

In experimental reality, the formation of clusters does produce a distribution of sizes. Some researches[18−20] indicate that the cluster size distribution can be assumed as a log-normal distribution function

$$f\left(N_c\right) \propto \exp\left[-\ln^2\left(N_c/N_0\right)/2w^2\right], \qquad (6)$$

where N_0 is the modal cluster size and w is proportional to the FWHM of the distribution. The width of the distribution is assumed approximately equal to N_0 and thus w is 0.4087, accordingly. The mean cluster size $\bar{N}$ relates to N_0 by $N_0 = \bar{N}/1.29$. Taking the cluster size distribution into account, we calculated the mean cluster-size dependence of the maximum and the average kinetic energy which the protons acquire when the laser peak intensity is 4×10^{16} W/cm^2 and the pulse width is 60 fs, shown in Fig. 3. The mean cluster radius R_{mean} is defined by $\bar{N} = 4\pi R_{mean}^3\rho/3$.

It can be seen from Fig. 3 that the numerical simulation without cluster-size distribution included clearly predicts an optimum cluster radius ~7 nm for the maximum proton energy under the laser pulse condition. In contrast, when the cluster-size distribution with FWHM of the distribution equal to the modal cluster size N_0 is included, the numerical data show a broader peak. The maximum proton energy increases sharply with the cluster size and then levels off before a slow fall. Moreover, the proton energy corresponding to a given mean cluster size is higher than that in the former case. As Ref. [18] reported, the inclusion of a cluster-size distribution into the simulations considerably improves the fit with experimental data. This broadening effect on the peak results from the fact that the optimum-size cluster of 7 nm is present in the distribution when the mean cluster radius is between 5 and 10 nm and the maximum proton energy resulting from the expansion of the optimal cluster will be observed when the mean cluster size is between these values. Besides, around the optimum cluster size, the inclusion of size distribution makes the average proton energy less than that in the case with no cluster-size distribution included. It is also because the wide size distribution allows some clusters to contribute very low energy and then to drag off the average effect.

Another property that should be paid more attention is, when the cluster-size distribution is excluded, the

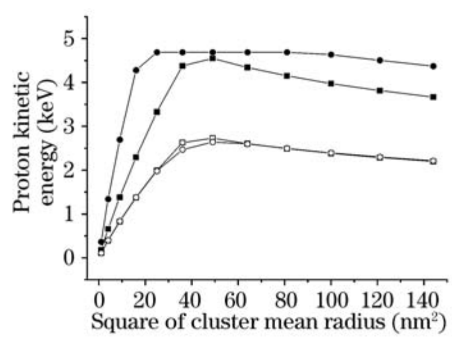

Fig. 3. Scaling of the proton kinetic energy with the square of cluster mean radius R_{mean}. The laser peak intensity is $\sim 4\times10^{16}$ W/cm^2 and the pulse duration is 60 fs. The solid squares and circles represent the maximum proton energy values without cluster size distribution included and with FWHM of the distribution equal to the modal cluster size N_0, respectively. The open data points represent the average proton energy in the above two cases respectively.

proton energy of very small cluster approaches to zero. However, when the size distribution is included, the maximum proton energy is not zero even if the mean cluster size is small. The explanation can also be obtained from the wide cluster-size distribution. Although the mean size is small, some relatively large cluster in the size distribution can still be outer-ionized completely and provide higher energy than the energy corresponding to a single cluster whose size equals to the mean cluster size. At the same time, the percentage of larger clusters in the size distribution is high enough to make its contribution outstand for the observation of maximum energy.

One of our relative experimental researches[25] has reported that the maximum proton energy, which increases from 1.5 to 4.5 keV, is detected when the mean cluster radius varies between 1 and 3 nm. In the experiment, the maximum laser intensity is estimated to be $\sim 2 \times 10^{16}$ W/cm^2. However at this laser intensity the maximum proton energy in our simulation is only 2.25 keV even if the cluster size changes a lot in a wide region, as given in Fig. 1. The great difference between the experiments and simulations may primarily result from the rough estimation of laser intensity. The real maximum laser intensity in the experiments may be higher than that estimated, which allows larger clusters to be outer-ionized completely and to acquire higher proton energy. As shown in Fig. 3, when the laser intensity rises to 4×10^{16} W/cm^2 in the numerical simulation, the proton energy can reach 4.7 keV, which is close to the measured maximum energy. Besides, our simulation indicates that corresponding to the same proton energy region as the measured, the required cluster radius ranges from 2 to 4 nm. The cluster radius range is 1 nm bigger than the radius range estimated in the above experiment. Therefore, we think that the relation of maximum proton energy and cluster radius obtained in our simulation is helpful for the estimation of cluster mean size in experiments. This issue will be subjected to a further study in future.

In conclusion, the scaling of the proton kinetic energy with hydrogen cluster size has been studied in detail based on a simplified Coulomb explosion model. It is found the maximum kinetic energy the protons acquire rises to a peak and then decreases slightly as the cluster size increases. It also indicates that there exists an optimum cluster size to maximize the proton energy for given laser parameters. This scaling can be explained by investigating the temporal evolution of outer ionization rate of hydrogen clusters with different sizes. The inclusion of a cluster-size distribution into the simulations considerably improves the consistency between theoretical fit and experimental data. These discussions are useful for the optimum-match determination of laser-cluster parameters to obtain maximum proton energy in experiments. In view of the similarity between hydrogen and deuterium clusters, the investigation of the Coulomb explosion of hydrogen clusters can be useful for the study of laser-deuterium cluster interaction to obtain ideal neutron sources.

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