Interaction of intense laser pulses with hydrogen atomic clusters*

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The interaction between intense femtosecond laser pulses and hydrogen atomic clusters is studied by a simplified Coulomb explosion model. The dependences of average proton kinetic energy on cluster size, pulse duration, laser intensity and wavelength are studied respectively. The calculated results indicate that the irradiation of a femtosecond laser of longer wavelength on hydrogen atomic clusters may be a simple, economical way to produce highly kinetic hydrogen ions. The phenomenon suggests that the irradiation of femtosecond laser of longer wavelength on deuterium atomic clusters may be easier than that of shorter wavelength to drive nuclear fusion reactions. The product of the laser intensity and the squared laser wavelength needed to make proton energy saturated as a function of the squared cluster radius is also investigated. The proton energy distribution calculated is also shown and compared with the experimental data. Our results are in agreement with the experimental results fairly well.

Keywords: femtosecond laser pulse, Coulomb explosion, hydrogen atomic cluster
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1. Introduction

Study on the interaction of clusters with intense laser pulses has been a hot field in recent years. Some novel phenomena have been observed in the experiments.[1−6] By intense laser pulse's irradiating deuterium[7] or deuterium rich clusters,[8] the nuclear fusion reactions can take place, which may ultimately lead to the development of a table-top neutron source which could potentially have wide application in materials studies.[7]

Although many theoretical models[9−13] were proposed to explain the experimental phenomena and explore the interaction mechanism, none of them can adequately explain all the experimentally observed features. The energy distribution could not be explained very well, owing to the cluster-size distribution and the laser intensity distribution on the focus unconsidered. Mendham *et al.*[14] gained a rough agreement between numerical and experimental data by considering a log-normal size distribution of cluster sizes. Li *et al.*[15] successfully explained their measured proton energy spectrum and saturation effect of proton energy by considering the effects of cluster-size distribution as well as the laser intensity distribution on the focus spot.

In the present paper, the interaction between intense laser pulses and hydrogen atomic clusters is studied in detail by using a simplified Coulomb explosion model developed by Li *et al.*[15] Our calculated results show that the irradiation of femtosecond laser pulses of longer wavelength on hydrogen atomic clusters may be easier than that of short wavelength to produce highly kinetic hydrogen ions. In view of the similarity between hydrogen and deuterium clusters, the phenomenon suggests that the irradiation of femtosecond laser of longer wavelength on deuterium atomic clusters may be easier than that of short wavelength to drive nuclear fusion reactions. The product of the laser intensity and the squared laser wavelength needed to make proton energy saturated increases linearly with the square of the cluster radius.

2. Simulation method

When clusters are irradiated by an intense laser pulse, the atoms inside the clusters will be ionized by optical field ionization when the laser intensity rises to the threshold intensity and free electrons are gen-

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erated inside the cluster. Subsequently, free electrons are heated and gain kinetic energy from the laser field via laser driven collisional process. A part of free electrons are removed from the cluster by the laser field (outer ionization[15−17]) if their energies are high enough to overcome the positive potential barrier of the cluster ball. The remaining electrons remain inside the cluster. As a result, the cluster acquires the positive charge, the atomic ions inside the cluster repulse each other by the Coulomb repulsive force and the explosion of the cluster takes place.

In the simulation, there are made two elementary assumptions. One is that the cluster is assumed to be a uniform sphere in shape, and the other is that all the atoms and ions inside the cluster are assumed to stay in ground states.

The laser intensity in the simulation is given by

$$I = I_{\rm peak}\sin^2(\omega t)\exp\left(-(4\ln 2)\frac{(t-\Delta t)^2}{\Delta t^2}\right). \quad (1)$$

There are no free electrons inside the cluster initially. The bound electrons of atoms will be set to be free when the laser intensity rises to a threshold intensity. According to the simple one-dimensional Barrier-suppression model,[15,18,19] the threshold intensity can be calculated by using the following formula:

$$I_{\rm th}({\rm W\ /\ cm^2}) = \frac{cE_{\rm ion}^4({\rm eV})}{128\pi e^6 q^2} = 4.00\times 10^9\frac{E_{\rm ion}^4({\rm eV})}{q^2}, \quad (2)$$

where $E_{\rm ion}$ is the ionization potential, q is the charge state of ion and the charge state q is one for hydrogen ions, c is the speed of light and e is the electron charge.

According to Newton's law, if the cluster is assumed to be a uniform sphere in shape, the movement of the protons on the surface of the cluster with a radius of R_0 can be given via the following formula:

$$m_{\rm p}\frac{{\rm d}^2R(t)}{{\rm d}t^2} = \frac{N_{\rm e}(t)e^2}{4\pi\varepsilon_0 R^2(t)}, \quad (3)$$

where $m_{\rm p}$ is the rest mass of a proton, e is the electron charge, $R(t)$ is the outer radius of the expanding cluster, and $N_{\rm e}(t)$ is the number of electrons removed from the cluster at the time t.

When the energies of free electrons are high enough to overcome the positive potential barrier of the cluster ball, they can escape from the cluster. Consequently, $N_{\rm e}(t)$ can be obtained by assuming that the electrons can be expelled from the cluster if the ponderomotive energy $U_{\rm p}$ that the electrons acquire in the laser field with intensity I and central wavelength λ is larger than the Coulomb potential energy $U_{\rm c}$ of the charged cluster, i.e.[15,19,20]

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

where ε_0 is the vacuum permittivity, c is the speed of light, $m_{\rm e}$ is the rest mass of electron. Because every hydrogen atom has only one bound electron, the number of free electrons is equal to that of the atoms inside the cluster after the hydrogen atom inside the cluster has been ionized by optical field ionization. Therefore, the number of electrons $N_{\rm e}(t)$ removed from the cluster at the time t may not be larger than that of hydrogen atoms inside the cluster. Consequently, the following condition must be met for hydrogen cluster:

$$N_{\rm e}(t) \leq 4\pi\rho R_0^3/3, \quad (5)$$

where ρ is the atomic density of hydrogen cluster and R_0 is the initial radius of hydrogen cluster.

The movement of protons on the surface of cluster can be calculated by the following means: when the laser intensity I is larger than the threshold intensity $I_{\rm th}$, the hydrogen atoms inside the cluster will be ionized by optical field ionization and part of the electrons which have higher energies will escape out of the cluster. And the electron number $N_{\rm e}(t)$ removed from the cluster at time t can be gained from Eq. (4) and the Eq. (5) must be satisfied at the same time. The movement of protons on the surface of cluster can be gained by solving the differential equation Eq. (3). The energies gained by protons on the surface of the cluster correspond to the maximum kinetic energy $E_{\rm max}$. Under the consideration of uniform expansion, the radial expanding velocity of a proton inside the cluster increases linearly with its radial position increasing. Therefore, the kinetic energies of protons at the initial radial position r inside the cluster can be deduced via the following formula:

$$E(r) = \frac{r^2}{R_0^2}E_{\rm max}. \quad (6)$$

Under the assumption of an initially uniform spherical cluster, the number of protons within a radial range, $r \to r + {\rm d}r$, is

$${\rm d}N = 4\pi\rho r^2{\rm d}r. \quad (7)$$

So the average proton energy can be given as

$$E_{\rm av} = \frac{1}{N}\int_0^{R_0} E(r){\rm d}N = \frac{3}{5}E_{\rm max}. \quad (8)$$

The initial atomic density of hydrogen clusters is assumed to be $3\times10^{22}/\mathrm{cm}^3$ [21] through this paper.

3. Results and discussion

Combining the motion equation (3) and Eq. (8), we can study the explosion processes of hydrogen clusters. The average kinetic energies that the protons can gain in the Coulomb explosion at different cluster sizes for different laser parameters can also be studied.

The relationships between average proton energy and squared mean cluster radius at different laser wavelengths are studied and shown in Fig. 1. The laser intensity and the pulse duration used to obtain the results in Fig. 1 are 2×10^{16} W/cm^2 and 60 fs, respectively. From Fig. 1 it can be seen that the average proton energy increases to a maximum and then falls with the increase of cluster size. This is because at lower intensities the laser field can only outer-ionize smaller clusters completely in a very short time according to Eq. (4), while for larger clusters the outer ionization cannot be finished immediately and part of the free electrons still reside in the cluster during its expansion. At the same time, it can also be seen that the average proton energy increases with the increase of laser wavelength. The longer the laser wavelength is, the larger the mean cluster radius to maximize average proton energy will be. According to Eq. (4) it can be easily understood that the number of electrons removed from the cluster becomes larger with laser wavelength increasing. Therefore, the laser field with longer wavelength can outer-ionize larger clusters completely. It suggests that the irradiation of femtosecond laser of longer wavelength on hydrogen atomic clusters may be easier than that of shorter wavelength to produce high kinetic hydrogen ions. In view of the similarity between hydrogen and deuterium clusters, the phenomenon suggests that the irradiation of femtosecond laser of longer wavelength on deuterium atomic clusters may be easier than that of shorter wavelength to drive nuclear fusion reactions.

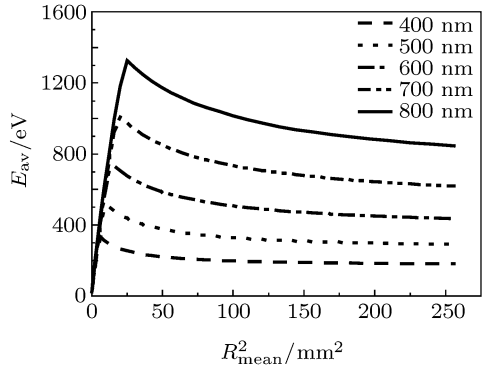


Fig. 1. Curves for average proton energy E_{av} versus squared mean cluster radius, R^2_{mean}, for different laser wavelengths. The laser intensity and the pulse duration are 2×10^{16} W/cm^2 and 60 fs, respectively.

The curves for average proton energy as a function of laser intensity for different laser wavelengths are shown in Fig. 2. The mean cluster radius R_{mean} and laser duration are set as 6.0 nm and 60 fs, respectively. As seen from Fig. 2, the average proton energy increases quickly with the increase of laser intensity when the laser intensity is low and becomes saturated after the laser intensity has reached a certain value, which is in qualitative agreement with the results obtained from dynamics simulation.[22] They also found that the average proton energy increased quickly with laser intensity increasing when the laser intensity is low and turns saturated after the laser intensity has reached a certain value. The reason is that the outer-ionization of free electrons, which strongly influences the average proton energy, increases with laser intensity increasing according to Eq. (4). Since the number of electrons for the cluster with a given size is definite and the total electrons are expelled completely when the laser intensity has increased to a certain value, there exists a minimum peak intensity for average proton energy to become saturated at a certain laser wavelength for a hydrogen cluster with a given size.

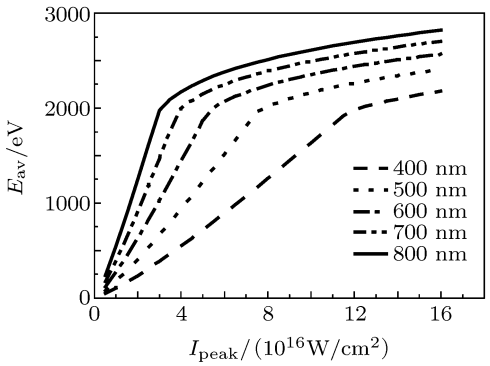


Fig. 2. Curves for average proton energy E_{av} versus laser peak intensity I_{peak} for different laser wavelengths, with mean cluster radius R_{mean} and laser duration being 6.0 nm and 60fs, respectively.

It is also found that the laser wavelength has a great effect on the minimum laser intensity as well as the saturated average energy. The longer the laser wavelength is, the lower laser intensity required to reach a greater saturation value of proton energy will

be. It can be well explained from Fig. 3. By calculating the values of average energy of protons for different cluster sizes, for different laser peak intensities which are larger than the threshold intensity $I_{\rm th}$ and for different laser wavelengths when the laser pulse width is 60 fs, we find that the product $I_{\rm peak}\lambda^2$ has a critical value to make proton energy saturated for a given cluster size and the proton energy would turn saturated if the product $I_{\rm peak}\lambda^2$ is larger than the critical value. Figure 3 shows the critical product $I_{\rm peak}\lambda^2$ as a function of the squared mean cluster radius, $R^2_{\rm mean}$. It can be seen that the product of the laser intensity and the squared laser wavelength needed to make proton energy saturated increases linearly as the squared cluster radius increases. When $I_{\rm peak}\lambda^2$ is bigger than the critical value, the average proton energy increases slightly. The reason is that the saturated average proton energy is reached when the total free electrons are removed completely from the cluster in a very short time and the cluster does not expand obviously. At this time, the outer radius of the expanding cluster, $R(t)$, is approximately equal to R_0 and the number of electrons $N_{\rm e}(t)$ is $4\pi\rho R_0^3/3$. According to Eq. (4), the expression $I_{\rm peak}(10^{16}\ {\rm W}\ /\ {\rm cm}^2)\lambda^2(\mu{\rm m}) \approx 5.14\times10^{-4}\times4\pi\rho R_0^2$ can be obtained.

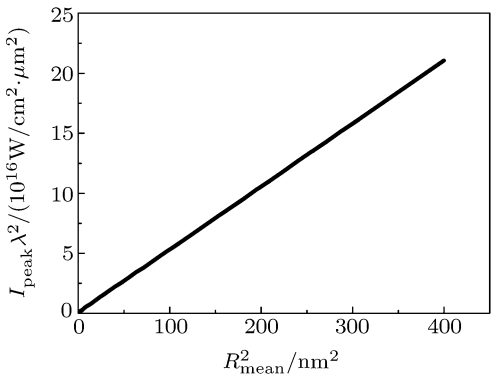


Fig. 3. Critical product $I_{\rm peak}\lambda^2$ of laser intensity and squared laser wavelength to make proton energy saturated as a function of squared mean cluster radius, $R^2_{\rm mean}$, with laser pulse width being 60 fs.

The saturated average proton energy $E_{\rm sav}$ as a function of squared mean cluster radius is shown in Fig. 4. As seen from Fig. 4, the saturated average proton energy $E_{\rm sav}$ also increases linearly as squared cluster radius increases. The result is in qualitative accordance with the theoretical prediction of pure Coulomb explosion.[23,24] In pure Coulomb explosion model, free electrons can be expelled completely from the cluster immediately after the atoms have been ionized and the average proton energy increases linearly with the squared cluster radius. In our calculation, the saturated average proton energy is reached when the total free electrons have been removed completely from the cluster in a very short time. In this case, it can be thought that a quasi pure Coulomb explosion takes place.

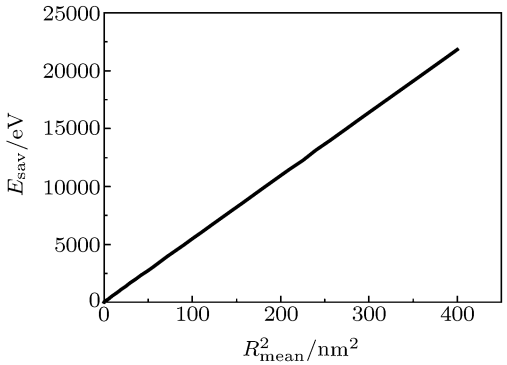


Fig. 4. Saturated average proton energy $E_{\rm sav}$ as function of the squared mean cluster radius $R^2_{\rm mean}$, with laser pulse width being 60 fs.

Figure 5 shows the curves for average proton energy $E_{\rm av}$ versus laser pulse width (FWHM, which stands for full width at half maximum) for different laser wavelengths. The hydrogen cluster radius is set as 6.0 nm and the laser peak intensity $I_{\rm peak}$ is 2×10^{16} W/cm^2. It can be seen that the average proton energy increases until it reaches a peak value, then decreases as the laser width increases, which indicates that an optimum pulse width exists for a given laser intensity and given cluster size at a certain laser wavelength. Synchronously, it is also found that the optimal pulse width decreases with the increase of wavelength. This is because for longer wavelength laser pulse it takes shorter time to outer-ionize the cluster completely

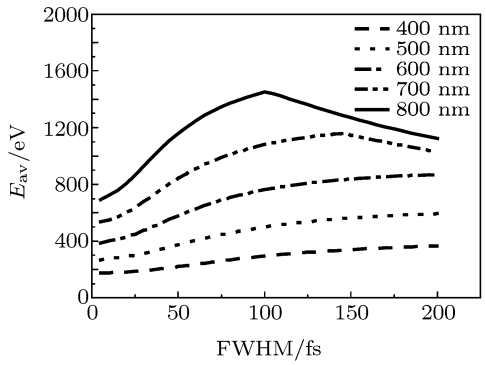


Fig. 5. Curves for average proton energy $E_{\rm av}$ versus laser pulse width (FWHM) for different laser wavelengths, with laser peak intensity $I_{\rm peak}$ and cluster radius being 2×10^{16} W/cm^2 and 6.0 nm, respectively.

and thereby more Coulomb potential energy can be accumulated in the cluster, which will be converted into the proton energy. Therefore, the optimal pulse width decreases with the increase of wavelength and the higher average proton energy will be gained.

In Fig. 6, the curves for average proton energy $E_{\rm av}$ versus laser wavelength for different laser intensities are shown. The laser pulse width and cluster radius are 60 fs and 6.0 nm, respectively. It is revealed that the average proton energy increases with the increase of laser wavelength which is in qualitative agreement with the results of our present dynamics simulation[25] and Petrov and Davis.[26] The reason is that the number of electrons removed from the cluster becomes larger with laser wavelength increasing and thereby more Coulomb potential energy can be accumulated in the cluster, which will be converted into the proton energy. Therefore, the average proton energy increases with the increase of laser wavelength. When the peak intensities are 5×10^{17} and 1×10^{18} W/cm^2, the average proton energy increases slowly as laser wavelength increases. The reason is that at the higher intensity the laser field can outer-ionize the cluster with a given size completely and the average proton energy becomes saturated. When the laser intensity is low, the average proton energy increases very quickly at the beginning, subsequently, the increasing trend of the average proton energy becomes slow. It can be understood easily that at lower intensities the laser field can only partly outer-ionize the cluster. And the outer-ionization degree of cluster increases very quickly with the increase of laser wavelength according to Eq. (4). However, the number of electrons for the cluster with a given size is definite and the total electrons are expelled completely when the laser wavelength reaches a certain value. It can also be seen that lower laser intensity requires longer laser wavelength so as to make the average proton energy saturated. For the lower laser intensity, it can be seen that free electrons can be removed completely from the cluster when the laser wavelength is very long (see Fig. 3).

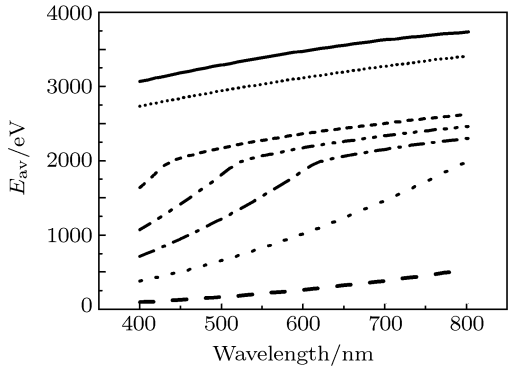


Fig. 6. Curves for average proton energy $E_{\rm av}$ versus laser wavelength for different laser intensities. The laser intensities are 1, 3, 5, 7, 10, 50, 100($\times10^{16}$ W/cm^2) in bottom-to-top sequence. The laser pulse width (FWHM) and cluster radius are 60 fs and 6.0 nm, respectively.

The curves for average proton energy $E_{\rm av}$ versus laser wavelength for different cluster sizes are shown in Fig. 7. The laser pulse width is set as 60 fs and the laser peak intensity $I_{\rm peak}$ is 2×10^{16} W/cm^2. When the cluster radius is small, the average proton energy increases very quickly with the increase of laser wavelength at the beginning and becomes saturated finally. When the cluster radius is large, the average proton energy increases quickly as the laser wavelength increases. For a certain laser intensity, it is easy to conclude that the larger the cluster size is, the longer the laser wavelength needed to make the average proton energy saturated will be. Since free electrons can be easily removed from the clusters with the increase of laser wavelength, the average proton energy increases with the increase of laser wavelength. But, because the number of free electrons is quite small for the small clusters, free electrons can be removed completely when the laser wavelength reaches a certain value. Then the average proton energy becomes saturated for the small clusters after the laser wavelength has reached a certain value. For large clusters, the laser of longer wavelength is required to make the average proton energy saturated for a certain laser intensity according to Fig. 3.

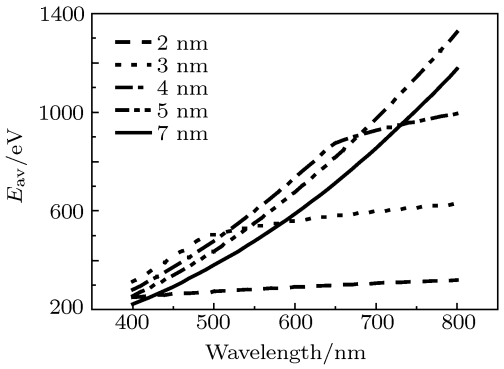


Fig. 7. Curves for average proton energy $E_{\rm av}$ versus laser wavelength for different cluster sizes. The laser intensity and laser pulse width are 2×10^{16} W/cm^2 and 60 fs, respectively.

Combining Eq. (3) and Eq. (7), the energy spectra of protons at different laser parameters can be inves-

tigated. In Fig. 8, the energy spectrum of protons is compared with the experimental date[15] in the same condition. The laser intensity is 1.4×10^{17} W/cm^2, the laser wavelength and laser pulse width (FWHM) are 800 nm and 60 fs respectively. In the calculation, the cluster size distribution is considered and assumed to be a lognormal distribution function[14]

$$f(N_{\rm c}) \propto \exp[-\ln^2(N_{\rm c}/N_0)/(2\omega^2)], \qquad (9)$$

where N_0 is the modal cluster size and ω is proportional to the FWHM of the distribution. The FWHM is approximately equal to N_0, and ω is 0.4087. The mean cluster size $\overline{N}$ is related to N_0 by $N_0 = \overline{N}/1.29$. The mean cluster size $\overline{N}$ is related to $R_{\rm mean}$ by $\overline{N} = 4\pi R_{\rm mean}^3\rho/3$. The calculated proton energy spectrum is in good agreement with the experimental results except in the high energy range. The difference in high energy range probably results from the attenuation of ultra intense laser in an assembly of clusters and the assumed uniform laser intensity distribution approximation on the focus spot, which needs to be studied in more detail.

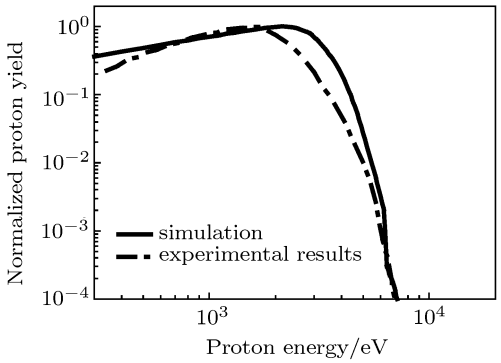


Fig. 8. Calculated energy distribution of protons compared with the experimental results[15] in the same condition. The laser intensity is 1.4×10^{17} W/cm^2, the laser wavelength and laser pulse width (FWHM) are 800 nm and 60 fs respectively.

The curves for the scaling of average proton energy $E_{\rm av}$ with squared mean cluster radius $R^2_{\rm mean}$ for different laser peak intensities $I_{\rm peak}$ are studied in Fig. 9 with the cluster size distribution included. The pulse duration is 60 fs. The wavelength is 800 nm. As shown in Fig. 9, for laser intensities $I_{\rm peak}$, e.g. 1×10^{16}, 2×10^{16} and 5×10^{16} W/cm^2, the average proton energy increases very steeply as the squared mean cluster radius increases at the beginning. And $E_{\rm av}$ versus $R^2_{\rm mean}$ exhibits near saturation as the squared mean cluster radius increases to a certain value for laser intensity $I_{\rm peak} = 1\times10^{16}$ W/cm^2. The saturation effect of proton energy as a function of cluster size is in accordance with those measured in experiments.[27–29] For laser intensity $I_{\rm peak} = 5\times10^{17}$ W/cm^2, the average proton energy increases linearly with the increase of squared mean cluster radius. It implies that a pure Coulomb explosion takes place. It can be easily understood that at lower intensities the laser field can only outer-ionize smaller clusters completely in a very short time and a pure Coulomb explosion can occur, while for large clusters, the outer ionization cannot be finished instantaneously and part of the free electrons reside in the cluster during its expansion. However, for higher intensities the laser field can only outer-ionize larger clusters completely in a very short time and a pure Coulomb explosion can occur.

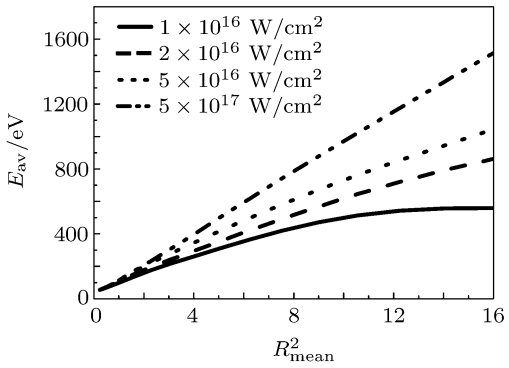


Fig. 9. Curves for scaling of average proton energy $E_{\rm av}$ with squared mean cluster radius $R^2_{\rm mean}$ for different values of laser peak intensity $I_{\rm peak}$ with the cluster size distribution included. The pulse duration is 60 fs. The wavelength is 800 nm.

4. Conclusions

We study the interaction between hydrogen atomic clusters and intense femtosecond laser pulses by using a simplified Coulomb explosion model. We find that the irradiation of a femtosecond laser of longer wavelength on hydrogen atomic clusters may be a simple, economical way to produce highly kinetic hydrogen ions. The phenomenon suggests that the irradiation of a femtosecond laser of longer wavelength on deuterium atomic clusters may be easier than that of a shorter wavelength to drive nuclear fusion reactions. We also find that the product of the laser intensity and the squared laser wavelength needed to make proton energy saturated increases linearly as the squared cluster radius increases.

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