Vol 13 No 2, February 2004
1009-1963/2004/13(02)/0196-05

Chinese Physics

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Three-dimensional simulation on explosions of hydrogen atomic clusters irradiated by an intense femtosecond laser pulse*

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(Received 20 May 2003; revised manuscript received 4 August 2003)

Using classic particle dynamics simulations, the interaction process between an intense femtosecond laser pulse and icosahedral hydrogen atomic clusters H_{13}, H_{55} and H_{147} has been studied. It is revealed that with increasing number of atoms in the cluster, the kinetic energy of ions generated in the Coulomb explosion of the ionized hydrogen clusters increases. The expansion process of the clusters after laser irradiation has also been examined, showing that the expansion scale decreases with increasing cluster size.

Keywords: intense femtosecond laser pulse, cluster, icosahedron, Coulomb explosion
PACC: 5250J, 3640J

1. Introduction

Researches on the interaction of intense femtosecond laser pulses with clusters have recently been a hot field.[1−18] Cluster media are different from monoatomic gaseous media and solid media when they are irradiated by intense femtosecond laser beams. For the gaseous monoatomic media the absorption of laser energy is rather weak and the application of such media to the effective conversion of the laser energy into x-ray emission is limited and the kinetic energies of ions and electrons produced in the interaction are low. On the other hand, for solid media the absorption of laser energy is usually high, but the drawback is that the solid targets are not as easily penetrated by laser beams as the gaseous media, and the rapid heat conduction into the cold substrate will restrict the temperature of the plasmas created. Clusters are considered to be the bridge systems between isolated molecules and the condensed matter. Atomic clusters formed in supersonic expansion of a high-pressure gas into vacuum have been proposed recently as an alternative solution combining the advantages of both gaseous and solid targets.[1] For instance, recent experiments on clusters irradiated by intense laser pulses have revealed several extremely high-energetic phenomena, that are not encountered in previous experiments restricted to atoms and small molecules: efficient generation of highly charged atomic ions,[2−8] generation of keV electrons and very energetic ions with MeV kinetic energies,[6,7,9−11] and the emission of intense x-rays.[2,4,12,13] Based on the production of high-ion-temperature plasmas, table-top nuclear fusion is realized[14] by the explosions of deuterium clusters heated by an intense femtosecond laser.

Several theoretical models have been proposed to explain the mechanism underlying the production of highly charged energetic ions in the interaction of atomic clusters with intense laser pulses. In the coherent electron motion model,[15] high charged states arise from impact ionization by coherently moving electrons. In the microplasma model[1,7] a cluster is treated as a spherical plasma ball and its dynamic process in a laser field can be described by hydrodynamic equations. The Coulomb expansion of this plasma ball is effectively boosted by heating through collisions between the laser-driven electrons and the ions (or inverse bremsstrahlung). In this model, the

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collisional ionization and tunnelling ionization determine the charged states of the ions. In Monte Carlo simulations of the cluster explosion driven by intense laser,[16] an ionization ignition mechanism has been proposed to account for the enhanced ionization. According to the ignition mechanism the enhanced rate of over-the-barrier ionization stems from the combined fields of the laser and the Coulomb attraction from neighbouring ions. When the intensity of the laser beam becomes sufficiently large, electrons are rapidly stripped from ions. Classical dynamics simulations performed in the frozen geometry approximation for fixed ionic charges predict the quasiresonance mechanism.[17]

In this paper, we report a three-dimensional simulation used for the study on the interaction between hydrogen atomic clusters and an intense femtosecond laser pulse. The kinetic energy distributions of ions and electrons generated in the explosion of clusters are presented. The expansion process of the clusters is also studied.

2. Theoretical model

The usual theoretical approaches are used to describe van der Waals bonded atomic clusters as the systems of particles interacting via a short-range potential such as the Lennard-Jones potential. It is well known that one of the particularly stable geometric configurations of such clusters takes the form of a closed-shell icosahedron. Multilayer icosahedral clusters possess complete K outer layers. A K shell icosahedron must contain a well defined total number of atoms [19]

$$N_K = (10K^3 + 15K^2 + 11K + 3)/3. \qquad (1)$$

In this paper H_{13}, H_{55} and H_{147} are chosen as the laser targets and are assumed to be icosahedral for simplicity. A real cluster plume has a wide cluster size distribution, and for a particular size usually there are plenty of isomers. The icosahedral assumption of these clusters in this work aims at making the calculations easier to draw the main conclusions: for example, the cluster size determines the expansion scale and energy distribution of ions and electrons, while the results may not deviate much from the reality.

In the case when an intense laser pulse interacts with the inert gas clusters, such as Ar_n, Kr_n and Xe_n, early in the pulse, the ionization of atoms in the cluster is dominated by tunnelling ionization. After a number of electrons have escaped from the cluster, they are driven by the laser field and can continue to ionize the clusters later in the laser pulse by collisional ionization. On the other hand, after the initial optical ionization, inelastic collisions between electrons and ions within the cluster become an important ionization mechanism.[1] In this paper, the classical Newton equations of motion for all of the electrons and nuclei are then integrated over the duration of the laser pulse. This approach admits the optical ionization and collision ionization, and ignores the tunnelling ionization, but it should provide qualitative insight into the ionization dynamics and the dependence on, e.g., the cluster size.

The Newton equations of motion can be established as follows:

$$m_i \frac{\mathrm{d}\boldsymbol{v}_i(\boldsymbol{r}_i,t)}{\mathrm{d}t} = \sum_{j,j\neq i} \frac{q_i q_j}{4\pi\varepsilon_0|\boldsymbol{r}_i-\boldsymbol{r}_j|^2}\frac{\boldsymbol{r}_i-\boldsymbol{r}_j}{|\boldsymbol{r}_i-\boldsymbol{r}_j|} + e\nabla\phi(\boldsymbol{r}_i,t), \quad (2)$$

where $\boldsymbol{r}_i$ and $\boldsymbol{r}_j$ denote the displacements of the particles (including ions and electrons), q_i and q_j are the charge of the particles, m_i is the mass of the particle, ε_0 is the dielectric constant, and ϕ is the potential induced by the laser field.

The interaction of particles with the laser pulse is treated within the dipole approximation by

$$\phi(\boldsymbol{r},t) = \boldsymbol{r}\cdot\boldsymbol{F}(t), \qquad (3)$$

where $\boldsymbol{F}(t)$ is the electric field of the incoming wave. The linearly polarized wave of the laser pulse used in the simulations is assumed to be a field envelope proportional to the sine function and the Gaussian profile,

$$\boldsymbol{F}(t) = \boldsymbol{F}_0\sin(\omega t)\exp\left(-(2\ln 2)\frac{t^2}{\Delta t^2}\right), \qquad (4)$$

where F_0 and ω are the amplitude and frequency of the laser oscillation respectively. In order to avoid the singularity of the Coulomb field, a soft-core model is used. Equation (2) can be changed to

$$m_i \frac{\mathrm{d}\boldsymbol{v}_i(\boldsymbol{r}_i,t)}{\mathrm{d}t} = \sum_{j,j\neq i} \frac{q_i q_j(\boldsymbol{r_i}-\boldsymbol{r}_j)}{4\pi\varepsilon_0(|\boldsymbol{r}_i-\boldsymbol{r}_j|^2+a)^{3/2}} + e\nabla\phi(\boldsymbol{r}_i,t), \quad (5)$$

where a is a non-zero parameter. In our work, a is supposed to be $0.0005\,\mathrm{nm}^2$. The Runge–Kutta law can be used to solve the above equation matrix. In order to improve the calculation precision, the sixth order Runge–Kutta law has been used in this paper.

The hydrogen atomic cluster is stable initially, and the electrons are circling around the nuclei. When a laser pulse irradiates the cluster, the atoms are ionized by the laser field at the early stage of the laser irradiation. Because of their smaller mass, electrons gain higher acceleration than the nuclei. So at the beginning of the ionization, the nuclei in the cluster are inertially confined while the ionized electrons are substantially heated by inverse bremsstrahlung and are quickly removed out of the cluster by the laser field. Then eventually the Coulomb explosion of the cluster will happen due to the strong repulsion between nuclei. Consequently, the potential energy of the fully ionized cluster will convert into the kinetic energy of ions.

3. Results and discussion

Icosahedral hydrogen atomic clusters composed of 13, 55 and 147 atoms are respectively interacted with an 814 nm laser pulse with a peak power intensity of $1.4 \times 10^{15}\,\mathrm{W/cm^2}$ and a temporal full width at half maximum of 25 fs. The distance of the neighbouring atoms in the cluster is taken to be 0.44 nm in the present calculations. The clusters explode due to the Coulomb repulsion forces after the laser irradiation. Figure 1 shows four snapshots of a 55-atom hydrogen cluster at different times in the laser field. (Ions are represented by the large dark spots and the electrons are represented by the small diamond spots.) At a time t=12.5 fs before the peak of the laser pulse, the cluster begins to expand. As is clear in this image, many of the electrons are ionized and are circled around the body of the cluster. The cluster itself is expanding isotropically. At the peak of the laser pulse (t=0), many of the electrons fly away from the cluster and the electron density in the vicinity of the cluster falls and the cluster has expanded essentially. 12.5 fs later the cluster continues to expand via Coulomb forces.

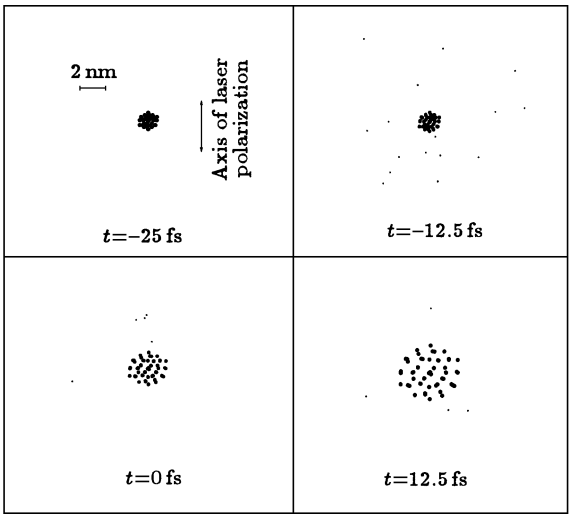


Fig.1. Four snapshots of a 55-atom hydrogen cluster at different times in the laser field with a peak power intensity of $1.4 \times 10^{15}\,\mathrm{W/cm^2}$. Ions are represented by the large dark spots, and the electrons are represented by the small diamond spots.

The final kinetic energy distributions of the produced ions and electrons calculated through the Newton equations (2) are shown in Fig.2. It can be seen from Fig.2(a) that with the increase of the number of atoms in the cluster, the ionic kinetic energy distribution after Coulomb explosion moves to a higher energy region, and the maximum ionic kinetic energy also becomes higher. For H_{13} cluster, E_{max}=14.9 eV; for H_{55} cluster, E_{max}=49.0 eV; and for H_{147} cluster, E_{max}=107.0 eV. These results can be interpreted by recognizing that with the increase of the number of atoms in the cluster, after ionization the repulsion forces of ions augment at the same time because the number of ions increases. We can also see from

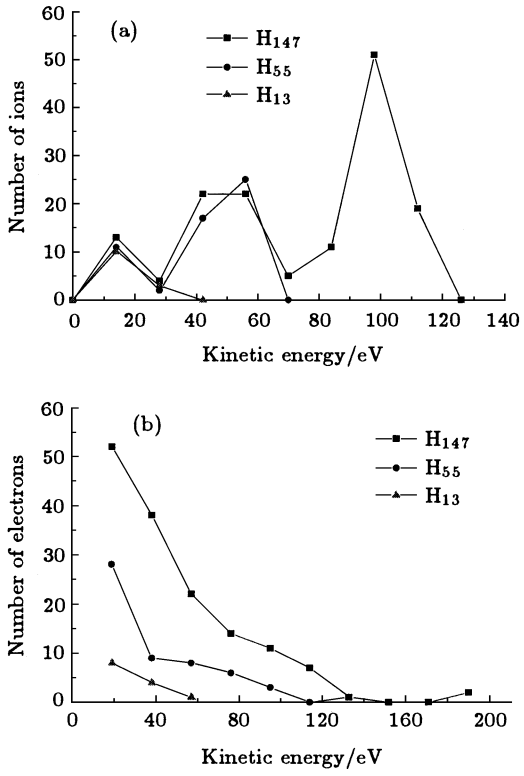


Fig.2. Kinetic energy distributions of the product ions and electrons. (a) Kinetic energy distributions of ions, (b) kinetic energy distributions of electrons. Irradiation parameters: laser power intensity $1.4 \times 10^{15}\,\mathrm{W/cm^2}$, wavelength 814 nm, pulse width Δt=25 fs.

Fig.2(a) that the energy-yield plot of the ions fluctuates strongly, especially for H_{55} and H_{147}, with many ions having almost the same energy. The origin of this clearly associates with the symmetry of these clusters. The energy spectrum of the electrons produced during the interaction is shown in Fig.2(b). The most remarkable aspect, if any, of this energy distribution is that the vast majority of the electrons exhibit energies below 120 eV.

The expanding process of the clusters during and after the laser irradiation is already examined in Fig.1, and the quantitative demonstration concerning the cluster expansion is shown in Fig.3. The effective distance R_{eff} of the cluster is defined as the mean-square root of the atom distribution

$$R_{\text{eff}} = \sqrt{\frac{1}{n}\sum_{i=1}^{n} R_i^2}, \tag{6}$$

where R_i is the atomic coordinate (the centre of mass of the cluster before explosion is supposed to be the grid origin). R_0 denotes the initial radius of the cluster before expansion. Figure 3 demonstrates that during the initial stage of the Coulomb explosion the clusters expand rapidly. After about 40 fs the clusters already extensively expand and the distances between ions are so large that the repulsion forces between ions are too weak to be considered. Thus the ions travel with a steady speed. Figure 3 also shows that with the increase of the number of atoms in the cluster, the scale of expansion decreases. At a certain value of $R(t)/R(0)$ the cluster can be defined as disassembly. A suitable laser pulse width should be chosen so that a cluster before disassembly can absorb as much energy as possible from the laser pulse.

Due to the lack of, to the best of our knowledge, experimental data for comparison, the simulation data concerning the hydrogen ion energies obtained in this study are much lower than those given by Ditmire *et al*[11] in their classical particle dynamics simulations on argon clusters. The reason for these differences is clear since in their work the argon ions possess high charge states (7^+–10^+) that make the Coulomb explosion of the cluster much stronger. Taking that into account, it is believed that the data obtained in the present simulations are reasonable. Very recently, molecular dynamics simulations were performed by Last and Jortner[20] on D_n and $(D_2O)_n$ clusters in order to investigate the laser driven nuclear fusion. For D_n clusters, they reported the maximum ion energy for n=959 is about 500 eV. Compared with this value, the results we obtained in this work are practical.

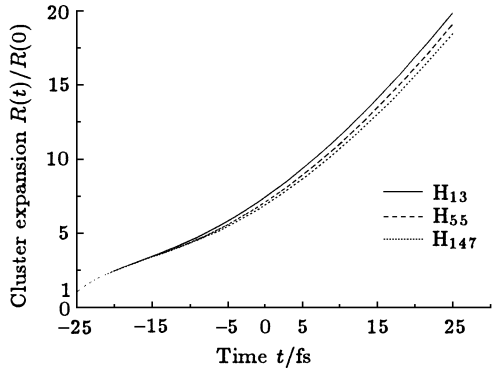


Fig.3. The expanding process of clusters during and after the laser pulse. Irradiation parameters: laser power intensity 1.4×10^{15} W/cm^2, wavelength 814 nm, pulse width Δt=25 fs.

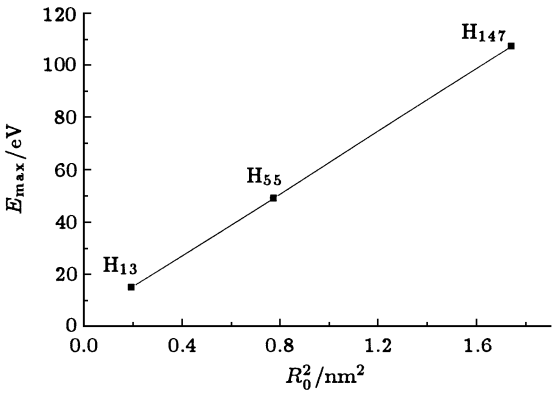


Fig.4. The calculated maximum kinetic energy $E_{\max}$ of the ions from Coulomb explosion of H_n clusters as a function of the square of the cluster size.

For a homonuclear cluster with identical charges (q in e units) on each atom and uniform atomic density ρ (unit in 10^{-3} nm^{-3}), the kinetic energy E (per ion in eV) is maximum for ions at the cluster surface at the initial cluster radius R_0 (unit in 10^{-1} nm),[21]

$$E(R_0) = (4\pi/3)Bq^2\rho R_0^2, \tag{7}$$

where B is a constant. Equation (7) provides a unique case of a cluster size equation (CSE),[22] in the scaling law $E \propto R_0^2$. Figure 4 illustrates the calculated maximum kinetic energy E in the presence of ions from Coulomb explosion of H_n clusters as a function of the square of the cluster size. The ion energy increases roughly linearly as the cluster size increases. The result is in accord with the theoretical prediction of Eq.(7). The present simulations indicate that to obtain higher ion energies, larger hydrogen clusters and intense laser pulse with a suitable width are required.

4. Conclusion

Using Newton equations of motion, we have simulated the interaction between the icosahedral hydrogen clusters and an intense femtosecond laser pulse. We find that with the increase of the number of atoms in the clusters, the kinetic energy of ions produced in the interaction increases at the same time. It is also found that the calculated maximum kinetic energy $E_{\max}$ of the produced ions from Coulomb explosion of H_n clusters is roughly proportional to the square of the cluster size. Because of the symmetrical structure of the icosahedron, the distribution of the ionic kinetic energy features some pinnacles. The energy distribution of electrons produced in the interaction is obtained and shows that the vast majority of electrons exhibit energies below 120 eV for H_{13}, H_{55}, and H_{147}. The expansion process of the cluster has also been examined, indicating that with the increase of cluster size, the expansion scale decreases. The present simulations indicate that higher ion energy can be generated when larger hydrogen clusters are used.

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