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Dynamics simulation on the interaction of intense laser pulses with atomic clusters*

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Under classical particle dynamics, the interaction process between intense femtosecond laser pulses and icosahedral noble-gas atomic clusters was studied. Our calculated results show that ionization proceeds mainly through tunnel ionization in the combined field from ions, electrons and laser, rather than the electron-impact ionization. With increasing cluster size, the average and maximum kinetic energy of the product ion increases. According to our calculation, the expansion process of the clusters after laser irradiation is dominated by Coulomb explosion and the expansion scale increases with increasing cluster size. The dependence of average kinetic energy and average charge state of the product ions on laser wavelength is also presented and discussed. The dependence of average kinetic energy on the number of atoms inside the cluster was studied and compared with the experimental data. Our results agree with the experimental results reasonably well.

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

1. Introduction

During the last several years, the interaction of clusters with high-intensity laser pulses has been studied extensively. The distinction between the average and local density of clusters leads to an important difference between clusters and solids, as well as between clusters and individual atoms when they interact with intense femtosecond laser pulses. Some novel phenomena were observed in the experiments, such as the generation of highly charged ions,[1−4] MeV kinetic energy ions,[2,5,6] keV electrons,[6] keV x-ray emission[3] and neutrons from D-D fusion reduced by laser.[7]

Although many theoretical models[8−13] were proposed to explain the experimental phenomena and explore the interaction mechanism, none of them can adequately explain all the experimentally observed features and some of them even conflict with each other. Ditmire *et al*[10,11] suggested that ions were ionized mainly through the impact ionization of hot electrons driven by an intense femtosecond laser pulse while the tunnel ionization is secondary. On the other hand, the ionization ignition model proposed by Rose–Petruck *et al*[13] and the recent work by Last and Jortner[14] assumed that field ionization by the combined field of laser, ions, and electrons plays a dominant role and the electron-impact ionization is of the minor importance. Meanwhile, the dependence of the kinetic energy and charge state of produced ions on the laser wavelength was observed experimentally,[15,16] but to the best of our knowledge, there has been no detailed theoretical work to study it until now. In this paper, based on classic particle dynamics, the process of intense femtosecond laser pulses with noble-gas clusters is carefully simulated to understand the ionization and wavelength effect. Atomic units are used throughout this paper unless otherwise stated.

2. Simulation method

A basic idea of our simulation method is to treat ions and free electrons as classical particles. Bound electrons do not appear explicitly. Ions and free electrons are driven by the combined field of laser, ions, and electrons. The influence of the magnetic field is also taken into account. The initial geometry of

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the clusters is chosen to be a closed-shell icosahedral structure[17] with an atom spacing of 0.37 nm[11] for Ar clusters.

In an intense laser field, the cluster atoms may be ionized by tunnel ionization (field ionization). This rate can be calculated by using the following formula[18−20]

$$W_{\rm tun} = \sum_{m=-l}^{l} \frac{(l+|m|)!}{2^{|m|}\,|m|!(l-|m|)!} \left(\frac{2e}{n^*}\right)^{2n^*} \times \frac{I_P}{2\pi n^*} \left(\frac{2(2I_P)^{3/2}}{E}\right)^{2n^*-|m|-1} \times \exp\left(-\frac{2(2I_P)^{3/2}}{3E}\right)$$

and

$$n^* = (Z+1)(2I_P)^{-1/2}. \quad (1)$$

Here, l and m are the angular momentum quantum numbers, I_P is the ionization potential, E is the total electric field seen by the ion, n^* is the effective principal quantum number, and Z is the charge state of the ion. The probability of ionization during a time step is given by $W_{\rm tun}\Delta t$. If a random number $p \in [0,1]$ is smaller than the tunnel ionization probability, the tunnel ionization immediately takes place and a new free electron with zero velocity appears near the parent ion in the direction of the ionizing field.

Free electrons may also appear through electron-impact ionization. If an electron approaches an ion closer than a certain value R_a, an electron impact is assumed to occur. The electron-impact ionization cross section σ is calculated by the following Lotz formula[21]

$$\sigma = \sum_{i=1}^{N} a_i q_i \frac{\ln(E/P_i)}{EP_i} \times \{1 - b_i \exp[-c_i(E/P_i - 1)]\}\,; E \geq P_i, \quad (2)$$

where E is the kinetic energy of the impact electron, P_i is the binding energy of electrons in the i-th subshell, P_1 is the ionization potential, q_i is the number of equivalent electrons in the i-th subshell, and a_i, b_i, and c_i are individual constants. N in Eq.(2) is equal to 1 for hydrogen-like and helium-like ions, while it is equal to 2 for lithium-like through neon-like ions, and equal to 3 for sodium-like through calcium-like ions.

The probability of ionization per electron impact is estimated as[13]

$$P_s = \frac{\sigma}{\pi R_a^2}. \quad (3)$$

If a random number $p \in [0,1]$ is smaller than P_s, the electron-impact ionization takes place and a new free electron is placed with a certain velocity near the parent ion.

In our calculation, the free electrons could be recombined with ions, which was treated in the following way: the free electron is captured by the ion and the charge state of the ion changes from Q to $Q-1$ if the distance between the ion and electron is smaller than a certain value R_a and the following relation is satisfied:[19]

$$\sum_j [Q_j/|\boldsymbol{X} - \boldsymbol{X}_j| - U_{Q_j}(|\boldsymbol{x} - \boldsymbol{X}_j|)] - \sum_i \left(U_Q(\boldsymbol{X} - \boldsymbol{x}_i) - U_{Q-1}(|\boldsymbol{X} - \boldsymbol{x}_i|) - \frac{1}{|\boldsymbol{x} - \boldsymbol{x}_i|}\right) - U_Q(|\boldsymbol{x} - \boldsymbol{X}|) + \boldsymbol{E}\cdot(\boldsymbol{x} - \boldsymbol{X}) + \frac{v^2}{2} < 0. \quad (4)$$

where $\boldsymbol{x}$ is the electron position, $\boldsymbol{X}$ is the ion position, $\boldsymbol{E}$ is the laser field, and v is the electron velocity.

In our classical dynamics simulation, the electric field of electron-electron interaction is modified by a smoothing term in order to avoid a steep increase in forces at very small distances:

$$E(r) = -\frac{1}{r^2 + r_0^2}. \quad (5)$$

The smoothing parameter is taken as $r_0 = 0.38$.

The laser field in the simulation is given by

$$E = E_0 \sin^2\left(\frac{\pi t}{2T}\right)\sin\omega t. \quad (6)$$

3. Results and discussion

The contribution of tunnel ionization and impact ionization to the mean charge state of product ions is shown in Fig.1. From Fig.1 it can be seen that electron-impact ionization plays a minor role for all the clusters studied. The mean charge state obtained for the Ar_{147} cluster in the present work is 4.6, which is close to 4.5 obtained in Ref.[19] under the same condition.

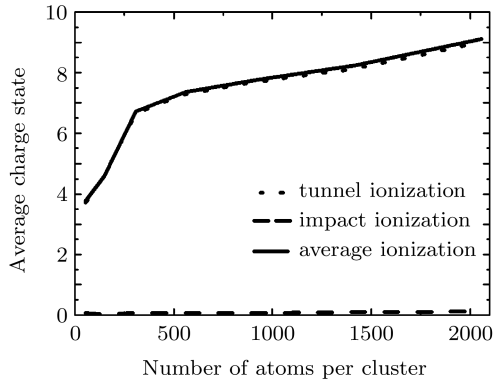


Fig.1. The contribution of tunnel ionization and impact ionization to the average charge state with the irradiation parameters: laser power intensity 1.4×10^{15} W/cm^2, wavelength 780 nm, and pulse width 100 fs.

It can be understood from the mean free path of electrons inside the cluster. The mean free path of the electron-impact ionization can be calculated via the following formula[19]

$$\lambda = 1/\rho\sigma(E). \tag{7}$$

Here, ρ is the atomic density inside the cluster, E is the incident electron kinetic energy, and $\sigma(E)$ is the impact ionization cross section when the incident electron kinetic energy is E. The mean free path as a function of the incident kinetic electron energy is shown in Fig.2. The diameter of the Ar cluster with the number of atom $N = 923$ is presented as a dashed line. As seen from Fig.2, the mean free path drops quickly to reach its minimum and then slowly increases as the incident electron kinetic energy increases. The minimum mean free path is about 74 times the diameter of the Ar cluster ($N = 923$) when the Ar ion charge state is equal to 1. Although the free path becomes larger with the

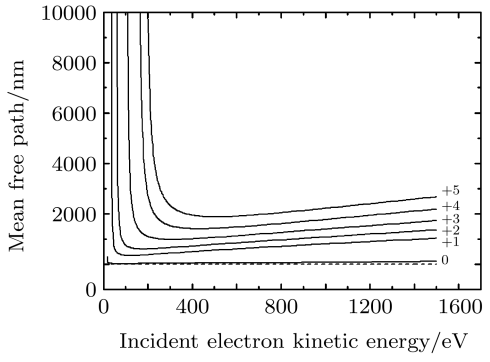


Fig.2. Electron mean free path inside Ar cluster with respect to the electron-impact ionization $Ar^{q+} \rightarrow Ar^{(q+1)+}$ calculated for the incident electron kinetic energy. The diameter of the Ar cluster ($N = 923$) (dash line).

increase of the ion charge state, generally at larger kinetic energy the free mean path is very small and consequently the electron-impact ionization is small too. In Fig.3 the experimental results of Ref.[22] and the present results of the average kinetic energy of ions as a function of the number of atoms inside the cluster are compared. From Fig.3 it can be seen that our results match those of Ref.[22] reasonably well.

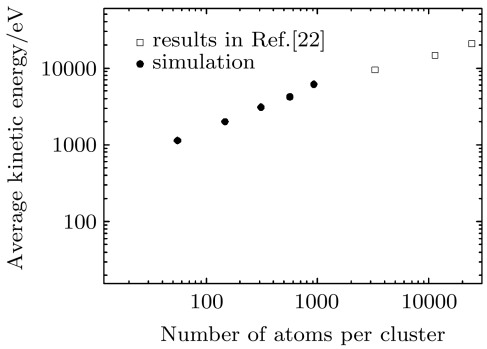


Fig.3. The average kinetic energy of the ions from clusters as a function of the number of atoms inside the cluster with the irradiation parameters: laser power intensity 2×10^{16} W/cm^2, wavelength 780 nm, and pulse width 60 fs.

For a homonuclear cluster, if the cluster expansion mechanism is a Coulomb explosion, the average kinetic energy $\overline{E}_k$ (unit in eV) as a function of the average ion charge state q (unit in e) and the initial cluster radius R_0 (unit in nm) is given by[23,24]

$$\overline{E}_k = \frac{4\pi}{5} B q^2 \rho R_0^2. \tag{8}$$

Here, B is a constant and equal to 1.4385 eV·nm, ρ (unit in nm^{-3}) is the atomic density inside the cluster. The relation between the mean ion energy $\overline{E}_k$ and $q^2R_0^2$ in the present simulation is shown in Fig.4(a). It is revealed that the mean kinetic energy increases nonlinearly with $q^2R_0^2$ and faster than $q^2R_0^2$ does. This is because the charge state of ions is higher at the beginning of the cluster expansion and subsequently becomes lower as a result of recombination. As shown in Fig.4(b), the charge state of the ion in the first half cycle is higher than that in the next half cycle. Simultaneously, figure 4(c) tells us that the mean kinetic energy is roughly linear with R_0^2 and suggests that the expansion process of the clusters after laser irradiation is mainly Coulomb explosion, which is consistent with the conclusion of Refs.[1], [22] and [25].

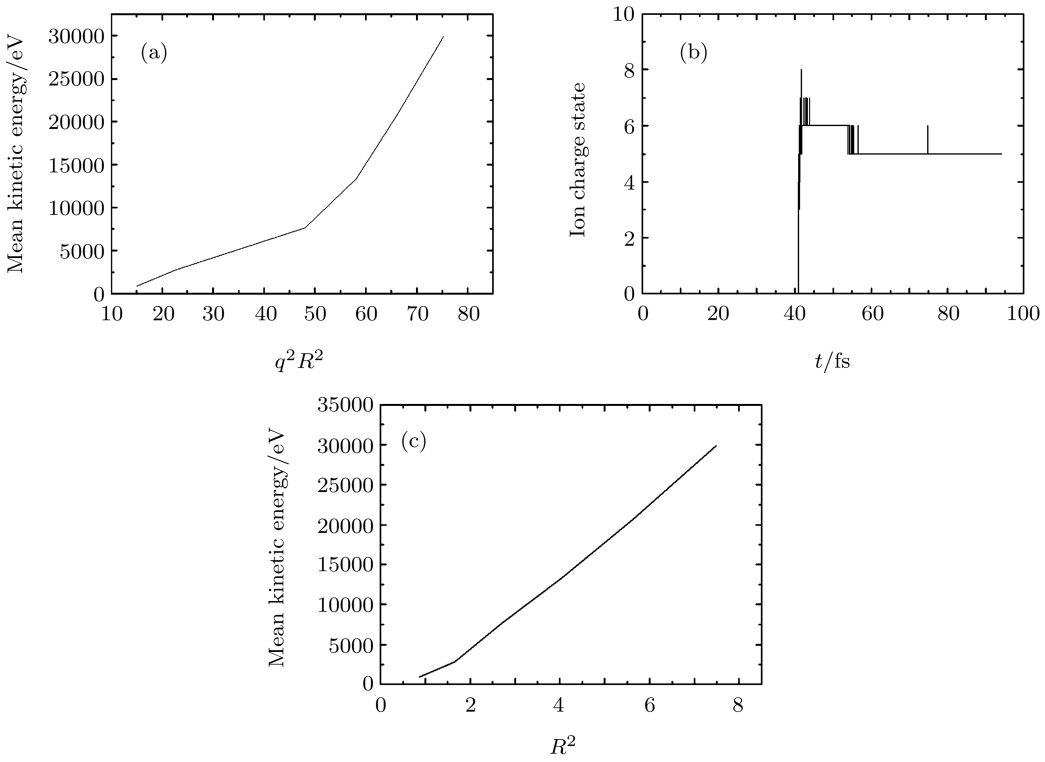


Fig.4. (a) The average energy of the ions from clusters as a function of $q^2R_0^2$; (b) The charge state of an ion inside the Ar_{147} cluster as a function of time t; (c) The average energy of the ions from clusters as a function of R_0^2. The parameters are the same as those in Fig.1.

In the past study, the kinetic energy of ions as a function of laser intensity and laser pulse width was extensively investigated. There has been no detailed theoretical work on the dependence of the ion kinetic energy on the laser wavelength. In the present simulation, the wavelength effect is studied and shown in Fig.5. As seen from Fig.5(a), the average kinetic energy of the product ions increases with the increasing laser wavelength. It is understandable if you pay attention to the fact that the mean charge state of product ions increases with the laser wavelength as shown in Fig.5(b). The average energy absorbed by the electron in a single collision can be given by the following formula[26,27]

$$\begin{aligned} \Delta E &\approx 2U_P \\ &= 2 \times \frac{e^2 I \lambda^2}{8\pi^2 \varepsilon_0 m_e C^3} \\ &= 1.86 \times 10^{-13} I \lambda^2 (\text{eV}). \end{aligned} \tag{9}$$

ΔE denotes the average energy absorbed by the electron in a single collision, I is the laser intensity (unit in W/cm^2), λ is the laser wavelength (unit in μm). It can be seen from Eq.(9) that ΔE is proportional to λ^2. As a result, with increasing laser wavelength, the electrons produced inside the cluster can gain more kinetic energy to escape from the cluster, and consequently, the electric field inside the cluster becomes stronger and the tunnel ionization rate becomes larger. Although these results are in conflict with the simulation results obtained in Ref.[14], they generally agree with the experimental results.[15,16] In Refs.[15] and [16], the clusters were radiated by the intensive nanosecond laser and the phenomenon of higher ion kinetic energy and charge state obtained with a longer wavelength laser, was observed. Although the results in Ref.[28] are under different conditions and it is impossible to make a quantitative comparison between ours and theirs, it should be noticed that in Ref.[28] the mean ion energy increases and the average charge state per atom stays the same with increasing the laser wavelength. This is because when the average charge state and the ionization potential of the ion are very high, it is very difficult to get a higher charge state, consequently, the average charge state will not change significantly with the laser wavelength.

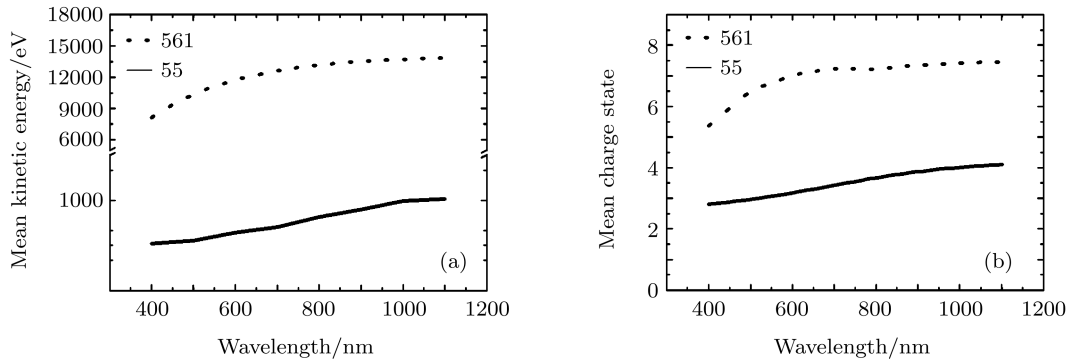


Fig.5. The average ion energy (a) and the mean charge state (b) calculated for different wavelengths with laser intensity 1.4×10^{15} W/cm^2 and the pulse width 100 fs.

Figure 6 shows the ion energy spectrum of the Ar clusters with different sizes. It can be seen that with increasing cluster size the ionic kinetic energy distribution and its maximum moves to a higher energy region. This is in agreement with the experimental result of Ref.[2]. The ion distribution exhibits pronounced oscillations which result from the initial shells and subshells of the clusters. With the increase of the number of atoms inside the cluster, the influence of the initial subshells of the cluster to the ion energy spectrum is larger. Similar simulation results are also obtained by Isidore Last[14] and Xia.[29]

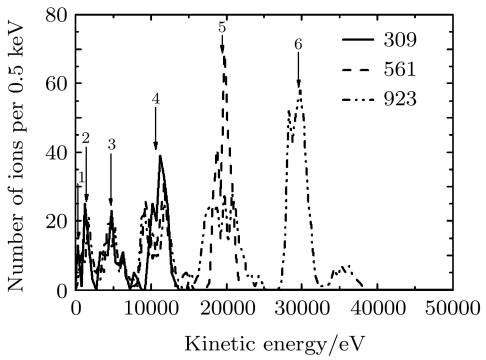


Fig.6. Kinetic energy distribution of produced ions. The numbers indicate the main shell of the clusters. The parameters are the same as those in Fig.1.

The dynamics of the cluster expansion are shown in Fig.7. The effective radius R_{eff} is defined as

$$R_{\text{eff}} = \sqrt{\frac{1}{n}\sum_{i=1}^{n} R_i^2}. \tag{10}$$

Here, R_i is the atomic distance from the center of mass. At the timescale of 40–53 fs, the scale of expansion decreases with the number of atoms inside the cluster, whereas at the timescale of $t > 53$ fs, the scale of expansion increases with the increase of the number of atoms inside the cluster. It is different from the result of Ref.[14] in which it was suggested that the scale of expansion decreases with the increase of the number of atoms inside the cluster during and after the laser pulse. Since free electrons can escape more easily from small clusters, the expansion of small clusters is faster than that of large clusters at the beginning of expansion. But, because the charge state of an ion from large clusters is higher, the ions from a larger cluster can reach a higher velocity than ions from a smaller cluster. That is why the phenomenon, that the ionic energy increases as the number of atoms inside the cluster increases, was observed in experiments of Refs.[2] and [25] and the present simulation.

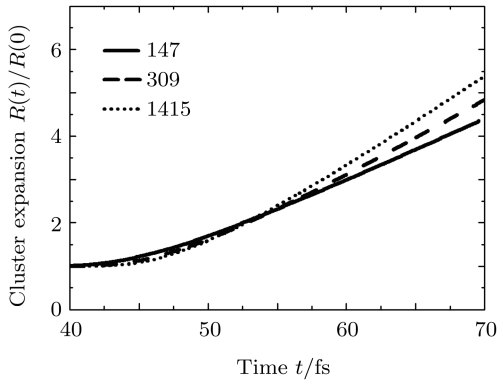


Fig.7. The expanding process of clusters. The parameters are the same as those in Fig.1.

The general picture of the ionization is displayed in Fig.8. The outer ionization event is recorded when the distance R between an electron and a cluster center is large enough to ignore the interaction of this

electron with other electrons and ions. This condition is given as follows: $R > 20R_{\rm eff}$. Since the number of electrons inside the clusters increases very steeply and reaches the maximum to 2 fs, the ionization ignition mechanism exists in the ionization process of atoms inside the cluster. The number of electrons outside the cluster increases subsequently and reaches the saturation.

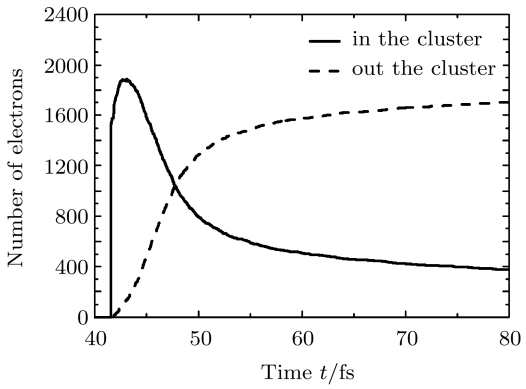


Fig.8. Ionization dynamics inside the Ar_{309} cluster. The solid line is the number of electrons inside the cluster. The dotted line is the number of electrons outside the cluster. The parameters are the same as in Fig.1.

4. Conclusions

We have studied the interaction between rare-gas clusters and intense femtosecond laser pulses by using the classical particle-dynamics simulation. We found that the ionization proceeds mainly through tunnel ionization by the combined field from ions, electrons and laser, while the contribution of impact ionization is secondary. We also found that a higher charge state could be reached in the front half cycle, whereas in the next half cycle, the field inside the cluster becomes small owing to cluster expansion and the recombination led to a lower charge state. In our calculation, the mean kinetic energy of ions generated in the interaction is roughly linear with R_0^2, which confirms that the expansion process of the clusters after laser irradiation is mainly Coulomb explosion. Further, the present simulation shows that the increase of the laser wavelength results in the enhancement of the charge state and energy of the produced ion. This suggests a simple, economical way to produce highly charged atomic ions.

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