Rare gas cluster explosion in an intense laser field

Kenichi Ishikawa and Thomas Blenski

CEA-Saclay, DSM/DRECAM/SPAM, Bât. 522, 91191 Gif-sur-Yvette Cedex, France

ABSTRACT

We study rare gas cluster explosion in an intense, femtosecond laser pulse using Monte Carlo particle dynamics simulations. The obtained dependence of ion energy on ion charge is approximately quadratic for lower charge states and linear for higher ones. This behavior, also observed in experiments, can be entirely explained on the basis of the Coulomb explosion mechanism. Our results also show that electron impact ionization plays only a minor role in ionization.

Keywords: clusters, laser interaction, particle dynamics simulation

1. INTRODUCTION

The intense laser interaction with clusters has been extensively studied for several years.[1–6] Multiply charged energetic ions,[1] high electron temperature,[2] and x-ray emission[3] have been observed in experiments. The laser-cluster interaction involves two processes, i.e., ionization and explosion. Several theories have been developed on the ionization mechanism which leads to the production of high charge states. In a coherent electron motion model by McPherson et al.,[3] multiple ionization arises from impact by coherently moving electrons, behaving like a quasi-particle. Ditmire et al.[2,4] proposed a " nanoplasma" model, in which ions are ionized mainly through impact of hot electrons. Rose-Petruck et al.[5] introduced an "ionization ignition (II)" model, where ionization is driven by the combined field of the laser, the other ions, and the electrons. Concerning the explosion dynamics, related to the high ion energy, the nanoplasma model[2,4] suggests that the cluster expands in a hydrodynamic manner by the pressure of hot electrons confined inside the cluster. Last and Jortner,[6] however, showed that electrons relatively quickly leave even large clusters containing over 2000 atoms and that the genuineness of the nanoplasma is questionable.

In the present study, we investigate the explosion mechanism of rare gas clusters (Ar_{55}, Ar_{147}, Xe_{55} and Xe_{147}) in an intense laser pulse using an improved version of the Monte Carlo particle dynamics simulations used previously by Ditmire.[4] We include ion-electron recombination in addition to tunneling and electron impact ionization, and use real (singular) Coulomb interaction between particles. Our results indicate that electron impact ionization plays only a minor role in ionization and that the experimentally observed charge-energy relation[1] can be entirely explained on the basis of the Coulomb explosion mechanism.

2. MODEL

The nonrelativistic equation of motion of each ion and electron is integrated using the force calculated as the sum of the contributions from all the other particles and the laser electric field. We model the field from an ion of charge state Q as a real Coulomb one from an effective nuclear charge $Q_{\mathrm{eff}}(r)$ of the form,

$$Q_{\mathrm{eff}}(r) = \begin{cases} Z(1 - r/r_a) & \text{for} \quad r < (Z - Q)r_a/Z, \\ Q & \text{for} \quad r \geq (Z - Q)r_a/Z, \end{cases} \tag{1}$$

with Z being the atomic number and r_a the "atomic radius" (1.3 a.u. for Ar and 2.0 a.u. for Xe). This potential is more suitable for the description of ion-electron interaction than a soft Coulomb potential used by Ditmire.[4] To handle an ion-electron close encounter, we resort to Kustaanheimo-Stiefel regularization,[7] widely used in astrophysics.

Free electrons appear through tunnel ionization or elecron impact ionization (EII). We evaluate the tunneling rate via an analytic formula[8] using the total electric field seen by each ion, and calculate the EII cross section using fitting formulas by Lennon et al.[9] for Ar and the Lotz formula[10] for Xe. Upon ionization a new electron is placed

---

near the ion in such a way that the total energy is conserved. Free electrons may be recombined with ions. A pair of an ion of charge Q and an electron is replaced by an ion of charge $Q-1$ if the distance between them is decreasing, if there is no potential barrier between them, and if the total energy of the electron is negative.

The pulse used in our simulations has a field envelope proportional to sine squared with a full width at half maximum of 100 fs and a wavelength of 780 nm. The clusters used in our simulations are Ar_{55}, Ar_{147}, Xe_{55}, and Xe_{147} with a closed-shell icosahedral structure[11] with an atom spacing of 3.76 Å for Ar and 4.32 Å for Xe.[12]

3. IONIZATION

Figure 1 shows the temporal evolution of the mean ion charge state for the case of an Ar_{147} cluster (solid line) and individual Ar atoms (dashed line) irradiated by a laser pulse with a peak intensity of $1.4 \times 10^{15} W/cm^2$. The mean charge state obtained in a cluster gas is considerably higher than that in an atomic gas. Moreover, high charge states up to 8+ were obtained from the cluster gas though it is not explicitly indicated in the figure. In order to examine the importance of EII, we have performed a simulation with EII switched off, whose result is shown as a dotted line in Fig. 1. It can be seen that EII has practically no effect on the mean charge state. Moreover, the highest charge state produced by EII was only 5+. This result is easy to understand from the viewpoint of mean free path. The electron mean free path with respect to EII of Ar^{5+} in an Ar cluster takes the minimum value of 580 Å at an incident energy of 300 eV. This is even larger than the diameter of Ar_{10^6} (410 Å). Thus, the contribution of EII to the production of high charge states is negligible. A similar argument holds true for Xe clusters. On the other hand, once several atoms are ionized, the total electric field strength seen by each ion can be significantly larger than the laser field alone. This leads to high charge states, driving further tunneling ionization, just as in the II model.[5]

4. CHARGE-ENERGY RELATION

In Fig. 2(a) we plot the relation between the mean ion energy $\bar{E}$ and the mean charge state $\bar{Q}$. The relation can be modeled with $\bar{E} \approx \alpha \bar{Q}^2$, where α is a constant. This indicates that ions are accelerated mainly through a Coulomb explosion mechanism. In fact, electrons quit the cluster before they exchange significant energy with ions, which excludes a hydrodynamic scenario. A 147-atom cluster consists of a central atom and three closed atomic shells. The outermost and middle shells are composed of three and two subshells, respectively. In Fig. 2(b) we plot the mean energy $E_s(Q)$ of the ions with a charge of Q in subshell s ($s = 1, \cdots, 6$) of Xe_{147} in the case of a peak intensity $8.8 \times 10^{15} W/cm^2$. We can model the relation with $E_s(Q) \approx \beta_s Q$, where a constant β_s depends on s. In Fig. 2(b) we have also plotted the average $E(Q)$ of $E_s(Q)$ over the entire cluster as filled circles. Again $E(Q)$ is approximately proportional to Q. We have found that this holds more or less also for Ar_{55}, Ar_{147}, and Xe_{55} and for other values of laser intensity. We simulated the pure Coulomb explosion of Xe_{147}, neglecting the electronic fields in the ionic
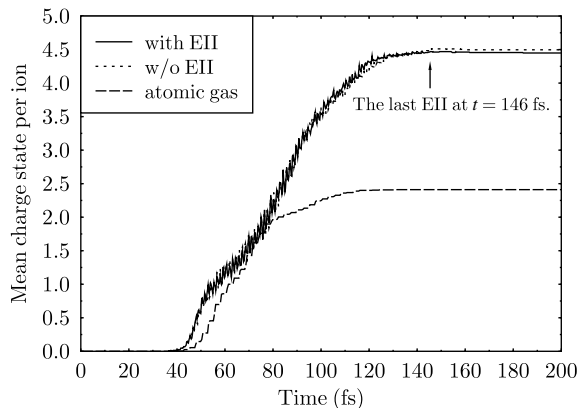

Figure 1. Evolution of the mean charge state per ion in Ar_{147} (solid line) and an atomic gas of Ar (dashed line) irradiated by the pulse with a peak intensity of 1.4×10^{15} W/cm^2. The dotted line is the result for Ar_{147} with electron impact ionization switched off.
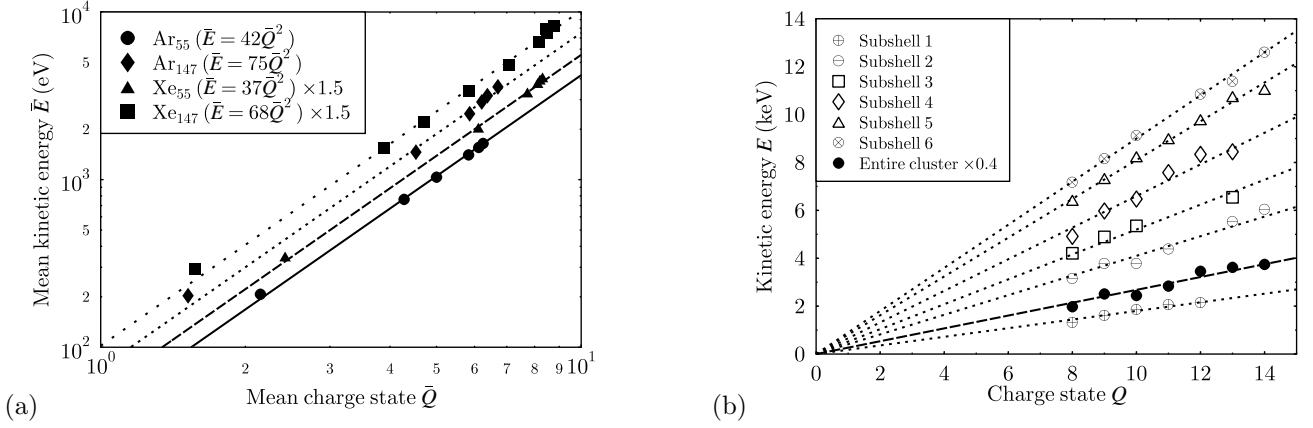
Figure 2. (a) Relation between the mean ion energy $\bar{E}$ and the mean charge state $\bar{Q}$ obtained using different intensities (starting from the lowest value of $\bar{Q}$, 0.35, 1.4, 3.2, 5.6, 8.8, $13 \times 10^{15}\mathrm{W/cm}^2$ for Ar_{55}, Ar_{147}, and Xe_{55}, and 0.35, 0.79, 1.0, 1.4, 1.9, 3.2, 13, 5.6, $8.8 \times 10^{15}\mathrm{W/cm}^2$ for Xe_{147}). The values of $\bar{E}$ for Xe clusters were multiplied by 1.5 for clarity. (b) Mean ion kinetic energy $E(Q)$ as a function of charge state Q for each subshell of Xe_{147} and for the entire cluster (filled circles, multiplied by 0.4 for clarity.) in the case of a peak intensity $8.8 \times 10^{15}\mathrm{W/cm}^2$. The subshells are enumerated outwards starting from the innermost one.

equations of motion but taking account of the ion charge history obtained for the case of Fig. 2(b). The resulting energy-charge relation is very similar to that in Fig. 2(b). This confirms that the approximately linear dependence is due to the mutual Coulomb repulsion between ions.

Lezius *et al.*[1] found a quadratic charge dependence of ion energy for lower charge states and a linear dependence at higher charge states in their experiments with Xe clusters. If we take a spatial profile of the laser intensity into account, we observe a similar behavior in our simulation results. This is illustrated in Fig. 3, which shows the ion charge-energy relation that we obtained by taking average over the simulation results with different values of intensity as in Fig. 2(a). It can be seen from Fig. 3 that the relation is quadratic for lower charges and linear for higher charges. This behavior can be entirely explained as a consequence of Coulomb explosion as follows: for lower charges, the contribution to each charge state Q mainly comes from such a spatial region where the mean charge state equals to Q. As a result, we obtain a quadratic relation, corresponding to the one in Fig. 2(a). On the
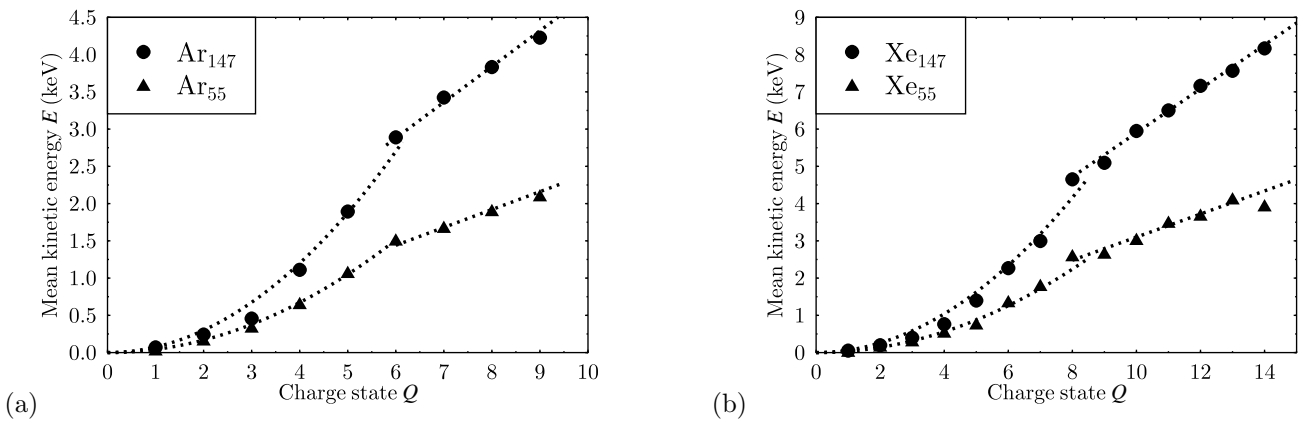

Figure 3. Charge dependence of ion energy of (a) Ar_{55}, Ar_{147} and (b) Xe_{55}, Xe_{147} irradiated by the laser pulse with a peak intensity of $1.3 \times 10^{16}\mathrm{W/cm}^2$, with account of the spatial intensity profile.

3

other hand, ions of higher charges come from the same region where the intensity is highest. This leads to a linear relation, corresponding to the one in Fig. 2(b). In Ref. 1 the relation was reported to be quadratic in the entire range of $1 \leq Q \leq 8$ in the case of Ar. This is probably because the authors of Ref. 1 used sufficiently high intensity $(5 \times 10^{17} \text{W/cm}^2)$ to produce Ar^{8+} while the yield of Ar^{Q+} $(Q \geq 9)$, for which a linear relation should be expected, was very low since the ionization potential of Ar^{8+} (422 eV[13]) is much higher than that of Ar^{7+} (143 eV[13]).

The preceding discussion has an important impact on the interpretation of the experimental results by Lezius *et al.*.[1] These authors interpreted the linear relation found for higher charge states as a sign of hydrodynamic expansion. Our results, however, indicate that this is not necessarily correct.

5. CONCLUSIONS

We have studied the explosion of rare gas clusters containing up to 147 atoms in an intense laser field via Monte Carlo particle dynamics simulations. Electron impact ionization plays only a minor role in ionization. The entire charge dependence of ion kinetic energy, including a linear part at higher charge states, formerly attributed to hydrodynamic expansion, can be understood on the basis of Coulomb explosion.

It is true that our simulation results cannot directly exclude the possibility of hydrodynamic expansion in larger clusters containing over 1000 atoms. However, it is striking that the overall feature of the charge-energy relation in Fig. 3 is similar to the one obtained by Lezius *et al.*[1] Last and Jortner[6] showed that electrons quickly leave even large clusters containing 2097 atoms. Moreover, according to our results, the mean energy of ions is 10-20% higher when they are emitted along the direction of laser polarization than perpendicular to it, which agrees with a recent experimental finding.[14] These agreements strongly indicate that the cluster explosion may be essentially governed by Coulomb explosion also in large clusters.

REFERENCES

1. M. Lezius, S. Dobosz, D. Normand, and M. Schmidt, "Explosion dynamics of rare gas clusters in strong laser fields," *Phys. Rev. Lett.* **80**, pp. 261–264, 1998.
2. T. Ditmire, T. Donnelly, A. M. Rubenchik, R. W. Falcone, and M. D. Perry, "Interaction of intense laser pulses with atomic clusters," *Phys. Rev. A* **53**, pp. 3379–3402, 1996.
3. B. D. Thompson, A. McPherson, K. Boyer, and C. K. Rhodes, "Multi-electron ejection of inner-shell electrons through multiphoton excitation of clusters," *J. Phys. B* **27**, pp. 4391–4400, 1994.
4. T. Ditmire, "Simulation of exploding clusters ionized by high-intensity femtosecond laser pulses," *Phys. Rev. A* **57**, pp. R4094–R4097, 1998.
5. C. Rose-Petruck, K. J. Schafer, K. R. Wilson, and C. P. J. Barty, "Ultrafast electron dynamics and inner-shell ionization in laser driven clusters," *Phys. Rev. A* **55**, pp. 1182–1190, 1997.
6. I. Last and J. Jortner, "Quasiresonance ionization of large multicharged clusters in a strong laser field," *Phys. Rev. A* **60**, pp. 2215–2221, 1999.
7. S. J. Aarseth, "Direct methods for N-body simulations," in *Multiple Time Scales*, J. U. Brackbill and B. I. Cohen, eds., pp. 377–418, Academic Press, Orlando, 1985.
8. A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, "Ionization of atoms in an alternating electric field," *Sov. Phys. JETP* **23**, pp. 924–934, 1966.
9. M. A. Lennon, K. L. Bell, H. B. Gilbody, J. G. Hughes, A. E. Kingston, M. J. Murray, and F. J. Smith, "Recommended data on the electron impact ionization of atoms and ions: Fluorine to nichel," *J. Phys. Chem. Ref. Data* **17**, pp. 1285–1363, 1988.
10. W. Lotz, "Electron-impact ionization cross-sections and ionization rate coefficients for atoms and ions from hydrogen to calcium," *Z. Phys.* **216**, pp. 241–247, 1968.
11. J. A. Northby, "Structure and binding of Lennard-Jones clusters: $13 \leq N \leq 147$," *J. Chem. Phys.* **87**, pp. 6166–6177, 1987.
12. A. Bondi, "van der Waals volumes and radii," *J. Phys. Chem.* **68**, pp. 441–451, 1964.
13. R. D. Cowan, *the Theory of Atomic Structure and Spectra*, Univ. of California Press, Berkeley, 1981.
14. E. Springate, N. Hay, J. W. G. Tisch, M. B. Mason, T. Ditmire, M. H. R. Hutchinson, and J. P. Marangos, "Explosion of atomic clusters irradiated by high-intensity laser pulses: Scaling of ion energies with cluster and laser parameters," *Phys. Rev. A* **61**, p. 063201, 2000.