Available online at www.sciencedirect.com
SCIENCE DIRECT®
PERGAMON
Radiation Physics and Chemistry 68 (2003) 273–276
Radiation Physics and Chemistry
www.elsevier.com/locate/radphyschem

Cluster explosion in an intense laser pulse

Marian Rusek[a,1,*], Hervé Lagadec[b], Thomas Blenski[b]

[a] Instytut Fizyki, Polska Akademia Nauk, Aleja Lotników 32/46, 02–668 Warszawa, Poland
[b] Commissariat à l'Energie Atomique, DSM/DRECAM/SPAM, Centre d'Etudes de Saclay, 91191 Gif-sur-Yvette, France

Abstract

This manuscript addresses a hot topic in the field of cluster physics: the explosion of rare-gas atomic clusters induced by short, intense laser pulses. Within the Thomas–Fermi model we have developed an numerical approach for an explicitly time-dependent description of small to medium size clusters in 3D. Such an approach, though strongly simplified in comparison to fully quantum-mechanical schemes, is nevertheless expected to yield a qualitatively correct description of the electronic and ionic dynamics of these systems, at a much lower computational cost.

PACS: 36.40.−c; 32.80.Rm; 42.50.Hz; 07.05.Tp

Keywords: Cluster dynamics; Strong-field laser-atom physics

Due to collective many-body effects the laser interaction with the rare-gas atomic clusters (consisting of a few hundred to few thousand atoms) may differ substantially from that of simple atomic and molecular systems. For instance, recent experiments on clusters irradiated by the intense laser pulses have revealed efficient generation of extremely highly charged atomic ions (McPherson et al., 1994a, b; Ditmire et al., 1995; Snyder et al., 1996; Lezius et al., 1997; Ditmire et al., 1997b; Lezius et al., 1998) and generation of electrons and ions with MeV kinetic energies (Lezius et al., 1997; Ditmire et al., 1997a, b; Shao et al., 1996; Ditmire, 1998). In principle the complete description of clusters in intense laser fields requires a solution of the time-dependent Schrödinger equation for many-electron systems. However even for the two-electron systems in strong fields, ab initio wave-function studies require an extremely powerful computer and are only at the initial stage. Thus for a description of cluster explosion an alternative approach should be sought.

Production of highly charged energetic ions by laser irradiation is of an essentially many-electron character and thus requires a theoretical framework within which a large number of electrons could be treated. The time-dependent density functional theory provides such a framework. It may be considered as a nontrivial extension of the steady-state density functional theory to the time domain. Usually this theory is used in the regime of weak fields for which the perturbation theory is applicable. Nevertheless, recently an application of this theory to a study of the response of atomic clusters to an intense laser pulse appeared (Véniard et al., 2001). A simplified one-dimensional model with frozen ion positions and the omission of correlation effects allowed the authors to solve numerically time-dependent Kohn–Sham equations describing electron dynamics in a cluster. Results concerning the initial stage of ionization and high-order harmonic generation in moderate intensity regime were presented.

Several other theoretical models have been proposed to explain the mechanism underlying the production of highly charged energetic ions in interaction of atomic clusters with intense laser pulses. In classical Monte-Carlo simulations of cluster explosion the nuclei and unbound electrons are treated as classical particles obeying Newton's equations of motion (Ishikawa and Blenski, 2000; Last and Jortner, 2000, 2001; Siedschlag and Rost, 2002). The bound electrons are released with a certain probability depending on the electric field strength inside the cluster (Ishikawa and Blenski, 2000;

*Corresponding author.
[1] This work was supported by the Polish Committee for Scientific Research (KBN) under Grant No. 5 P03B 063 20.

0969-806X/03/$ - see front matter
doi:10.1016/S0969-806X(03)00299-8

Last and Jortner, 2000, 2001; Siedschlag and Rost, 2002) and collision probability with other electrons and ions (Last and Jortner, 2000, 2001). Multielectron ionization and Coulomb explosion of large Xenon clusters studied using this method revealed that electrons are removed from the cluster mainly when the cluster size is enlarged due to Coulomb explosion (Last and Jortner, 2000). Calculation of the energy absorption of atomic clusters as a function of laser pulse length showed a maximum for a critical length which can be linked to an optimal cluster radius (Siedschlag and Rost, 2002). Molecular dynamics simulations of heteronuclear molecular clusters containing deuterium and heavy atoms indicated a new mechanism for multielectron ionization and Coulomb explosion, providing a source of high-energy deuterons suitable to induce nuclear fusion (Last and Jortner, 2001).

In this paper we further investigate a theoretical approach to explosion of rare-gas atomic clusters in intense laser pulses based on a time-dependent Thomas–Fermi model introduced in our previous article (Rusek et al., 2001). The results of additional simulations are presented and their relation to the conclusions of the recent time-dependent density functional (Véniard et al., 2001) and molecular dynamics models (Last and Jortner, 2000, 2001; Siedschlag and Rost, 2002) is discussed. The Thomas–Fermi model is the earliest and the simplest of the density functional methods. It was introduced by Thomas (1926) and Fermi (1928) in order to describe the self-consistent potential of multielectron atoms. The advantage of this model is its simplicity (atomic number Z is the only parameter) and universality (different atoms differ by scaling units only). The time-dependent version Thomas–Fermi model we use in this paper may be considered as a semiclassical approximation (described by a Bloch-like hydrodynamic model) to the rigorous quantum dynamics of an electron gas. Let us begin by recalling the basic assumptions of the model.

Within the Thomas–Fermi model the ground state structure of an rare-gas atomic (van der Waals) cluster is described by the average electron density $\rho(\vec{r})$ and the positions of the nuclei $\vec{R}_a$. It can be obtained by minimization of the following Thomas–Fermi energy functional (Rusek et al., 2001):

$$\mathscr{E}[\rho(\vec{r}),\{\vec{R}_a\}] = \int \mathrm{d}^3 r \rho(\vec{r}) U(\vec{r}) + \frac{e}{2}\int \mathrm{d}^3 r \left[\sum_{a=1}^{N} Z\delta(\vec{r}-\vec{R}_a) - \rho(\vec{r})\right]\Phi(\vec{r}) - \varepsilon\frac{(Ze)^2}{2}\sum_{a=1}^{N}\sum_{b=1}^{N}\frac{1}{|\vec{R}_a-\vec{R}_b|}, \quad (1)$$

where the relations for the internal (kinetic) energy of an ideal electron gas at temperature $T=0$ are used locally $U(\vec{r}) = c_k[\rho(\vec{r})]^{2/3}$ and the electrons and nuclei interact through the self-consistent Coulomb potential $\Phi(\vec{r})$. In order to stabilize the cluster before the head of the pulse arrives we resort to a very simple approach and artificially diminish the Coulomb interaction between the nuclei (while the electron–electron and electron–nucleus interaction are not changed). This is done by lowering the charge of each nucleus as seen by other nuclei by a small factor $\varepsilon \ll 1$ (in this paper we use the value of $\varepsilon = 10^{-2}$).

To introduce a discretized version of the Thomas–Fermi model suitable for numerical solution we assume that the electron density at any point of space is obtained by summing up the contributions from n smoothed pseudo-particles:

$$\rho(\vec{r}) = \frac{ZN}{n}\sum_{j=1}^{n} w(\vec{r}-\vec{r}_j), \quad w(\vec{r}) = \left(\frac{\alpha}{\sqrt{\pi}}\right)^3 e^{-(\alpha\vec{r})^2}. \quad (2)$$

The parameter α from Eq. (2) is chosen to minimize the energy Eq. (1) of the Thomas–Fermi atom of given Z. After inserting Eq. (2) into Eq. (1) and treating the Thomas–Fermi energy functional Eq. (1) as the Hamiltonian of the system we arrive at the Hamilton equations of motion for the smoothed pseudo-particles Eq. (2):

$$m\frac{\mathrm{d}\vec{v}_i}{\mathrm{d}t} = -\frac{2}{3}c_k\sum_{j=1}^{n}\left(\frac{1}{\rho(\vec{r}_i)^{1/3}}+\frac{1}{\rho(\vec{r}_i)^{1/3}}\right)\vec{\nabla}_i w(\vec{r}_i-\vec{r}_j) - e\vec{\nabla}_i\Phi(\vec{r}_i) \quad (3)$$

($\mathrm{d}\vec{r}_i/\mathrm{d}t = \vec{v}_i$). Eq. (3) can be understood as a discretized smooth particle hydrodynamics (Lucy, 1977; Gingold and Monaghan, 1977) version of the standard hydrodynamic equations together with the equation of state for an ideal fermion gas which defines the pressure. Therefore, by treating the Thomas–Fermi energy functional as a Hamiltonian of the system we followed (Bloch, 1933) who had viewed the oscillations of the electron cloud in an many-electron atom as a motion of a fluid characterized by a density $\rho(\vec{r},t)$ and a velocity field $\vec{v}(\vec{r},t)$. Eq. (3) for the electron density ρ should be supplemented by the Newton equations of motion for the positions of the nuclei $\vec{R}_a$. The interaction with the laser pulse is treated within the dipole approximation. The linearly polarized wave of a pulse used in the simulations is assumed to have a field envelope proportional to sine squared with a full-width at half-maximum τ and an optical period τ_0.

Let us limit the presentation of the results of our numerical simulations of cluster explosion within the time-dependent Thomas–Fermi model with the discussion of the case of $Z=1$. It is known, that within the static (time-independent) Thomas–Fermi model atoms of different atomic number Z differ by scaling units only. Thus we may expect that simulations performed for $Z=1$ will allow us to gain some qualitative

information about the process of cluster explosion without spending too much computational time. We have prepared an initial state of a $N = 55$ atom cluster in a form of a two shell icosahedron and an atom in the center. In the next step this cluster has been exposed to a strong laser pulse with the peak intensity $I = 1.4 \times 10^{15}$ W/cm^2 (or $\mathscr{F}_0 = 0.2$ [a.u.]). The pulse used in the simulations had a wavelength $\lambda = 800$ nm (or $\tau_0 = 110.425$ [a.u.]) and a temporal full-width at half-maximum $\tau = 106.67$ fs (or $\tau/\tau_0 = 40$). All of the results presented in this paper were obtained using a wave polarized linearly along the x-axis. The only independent parameter left is the mass of the nuclei M which has been set to $M/m = 2000$.

In Fig. 1 we have the kinetic energy $\mathscr{E}_{\mathrm{kin}}$ of the atomic ions fragments coming from the different shells of an $N = 55$ atom cluster versus time t. For convenience the second shell has been splited into two subshells with different initial radiuses. The energy is averaged over the ions coming from a given (sub)shell. Note the stepwise character of the explosion: the ions leaving first being far more energetic than those leaving later. It is seen that the explosion is neither instantaneous nor uniform. It exhibits a layer-like structure in which shells of cluster ions are expelled sequentially. Let us compare these results with kinetic energy of the ions coming from the explosion of an $N = 13$ atom cluster composed of one atomic shell and one atom in the middle. It can be viewed as an $N = 55$ atom cluster without the second shell. The results for this smaller cluster are ploted as an additional curve in Fig. 1. We see a strong increase of energy of the ion fragments (coming from an increase in ionization rate) with the number of atoms in the cluster. This is in agreement with TDFT simulations (Véniard et al., 2001). It is interesting to note that the explosion of an $N = 13$ started faster than the explosion of the first shell in an an $N = 55$ atom cluster as if the outer shell in an $N = 55$ atom the larger cluster prevented the inner shell from moving.

To investigate the role of the electron dynamics in the cluster explosion process we have also performed additional simulations of the Coulomb explosion of a cluster. This time the ions were treated as point-like particles of given charges $Q_a(t)$ as in the classical Monte-Carlo simulations (Ishikawa and Blenski, 2000; Last and Jortner, 2000, 2001; Siedschlag and Rost, 2002). In the first case the charges of each ion at a given instant of time were chosen equal to the charges of the ions calculated in the full Thomas–Fermi simulation, i.e., $Q_a(t) = Q_a^{\mathrm{TF}}(t)$. A plot illustrating the energies of the ions obtained in this case is presented in Fig. 2. The kinetic energy of the ions coming from different shells also looks qualitatively similar to Fig. 1 but their numerical values are higher. Notice, that this not a mere constant energy shift, the difference depends on the shell of the cluster. This confirms the fact that the space-charge distribution due to electrons present inside the cluster plays an important role in the explosion process. In out second simulation the charges of all ions were identical and equal to the average charge from the TF simulation, i.e., $Q_a(t) = 1/N \sum_{b=1}^{N} Q_b^{\mathrm{TF}}(t)$. In this case the results are different: the energies of the ions coming from different shells are almost the same (except the central atom). This shows the dependence of atomic ionization rates on the original position of an atom in the cluster and is in agreement with results of the with 1D TDFT simulations (Véniard et al., 2001) (however in our case the most ionized atoms come from the outer shells of the cluster) (Fig. 3).

In summary, the dynamics of small rare-gas atomic clusters ionized by a high-intensity femtosecond laser pulse have been studied qualitatively using a three-dimensional refinement of the time-dependent Thomas–Fermi model. It was confirmed that the explosion is

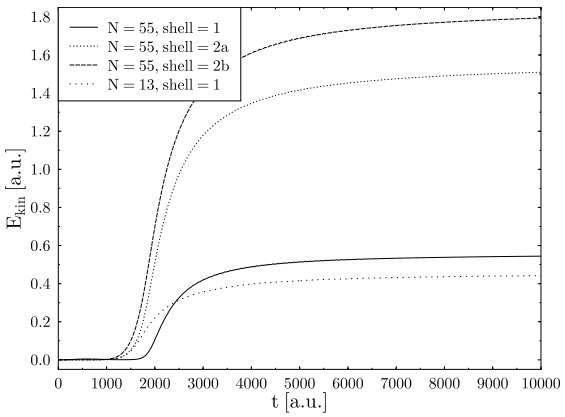

Fig. 1. Average kinetic energy $\mathscr{E}_{\mathrm{kin}}$ of the Thomas–Fermi ions coming from different (sub)shells of $N = 55$ and 13 atom clusters ploted versus time t.

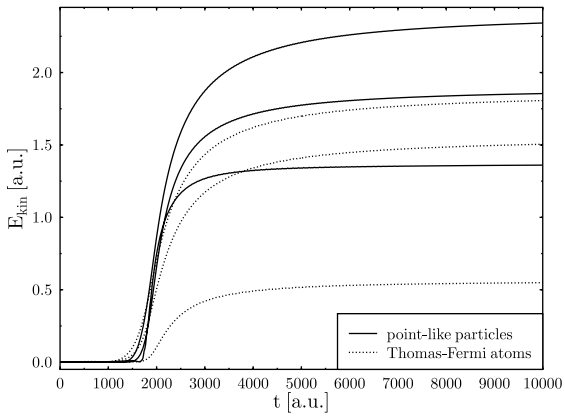

Fig. 2. Approximate average kinetic energy $\mathscr{E}_{\mathrm{kin}}$ of the point-like ions coming from different (sub)shells of $N = 55$ atom clusters ploted versus time t.

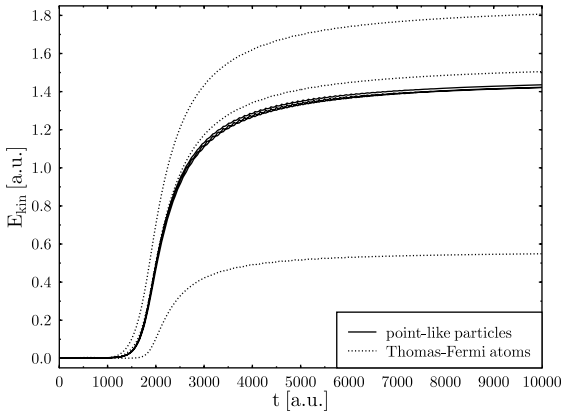

Fig. 3. Approximate average kinetic energy $\mathscr{E}_{kin}$ of the point-like ions coming from different (sub)shells of $N = 55$ atom clusters ploted versus time t.

neither instantaneous nor uniform. It exhibits a layer-like structure in which shells of cluster ions are expelled sequentially. It seems that the inner shells of the cluster start to expand first and "push" the outer shells. Stepwise character of the explosion was seen also in the kinetic energy of the outgoing atomic ion fragments: the ions leaving first were far more energetic than those leaving later. The role of electron dynamics in the cluster explosion at the initial stages has been also investigated.

References

Bloch, F., 1933. Z. Phys. 81, 363–376.
Ditmire, T., 1998. Phys. Rev. A 57 (6), R4094–R4097.
Ditmire, T., Donnelly, T., Falcone, R., Perry, M., 1995. Phys. Rev. Lett. 75, 3122.
Ditmire, T., Tisch, J.W.G., Springate, E., Mason, M.B., Hay, N., Marangos, J.P., Hutchinson, M.H.R., 1997a. Phys. Rev. Lett. 78, 2732.
Ditmire, T., Tisch, J.W.G., Springate, E., Mason, M.B., Hay, N., Smith, R.A., Marangos, J., Hutchinson, M.H.R., 1997b. Nature 386, 54.
Fermi, E., 1928. Z. Phys. 48, 73–79.
Gingold, R., Monaghan, J., 1977. Mon. Not. R. Astron. Soc. 181, 375–389.
Ishikawa, K., Blenski, T., 2000. Phys. Rev. A 62, 063204.
Last, I., Jortner, J., 2000. Phys. Rev. A 62, 013201.
Last, I., Jortner, J., 2001. Phys. Rev. Lett. 87, 033401.
Lezius, M., Dobosz, S., Normand, D., Schmidt, M., 1997. J. Phys. B 30, L251.
Lezius, M., Dobosz, S., Normand, D., Schmidt, M., 1998. Phys. Rev. Lett. 80 (2), 261–264.
Lucy, L.B., 1977. Astron. J. 82 (12), 1013–1924.
McPherson, A., Luk, T.S., Thompson, B.D., Borisov, A.B., Shiryaev, O.B., Chen, X., Boyer, K., Rhodes, C.K., 1994a. Phys. Rev. Lett. 72 (12), 1810–1813.
McPherson, A., Thompson, B.D., Borisov, A.B., Boyer, K., Rhodes, C.K., 1994b. Nature 370, 631.
Rusek, M., Lagadec, H., Blenski, T., 2001. Phys. Rev. A 63, 013203.
Shao, Y.L., Tisch, T.D.J.W.G., Springate, E., Marangos, J.P., Hutchinson, M.H.R., 1996. Phys. Rev. Lett. 77 (16), 3343–3346.
Siedschlag, C., Rost, J.M., 2002. Phys. Rev. Lett. 89, 173401.
Snyder, E.M., Buzza, S.A., Castleman Jr., A.W., 1996. Phys. Rev. Lett. 77 (16), 3347–3350.
Thomas, L.H., 1926. Proc. Cambridge Philos. Soc. 23, 542–548.
Véniard, V., Taïeb, R., Maquet, A., 2001. Phys. Rev. A 65, 013202.