

Electron Release of Rare-Gas Atomic Clusters under an Intense Laser Pulse

Christian Siedschlag and Jan M. Rost

Max-Planck-Institute for the Physics of Complex Systems, Nöthnitzer Strasse 38, D-01187 Dresden, Germany

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Calculating the energy absorption of atomic clusters as a function of the laser pulse length T we find a maximum for a critical T^* . We show that T^* can be linked to an optimal cluster radius R^* . The existence of this radius can be attributed to the enhanced ionization mechanism originally discovered for diatomic molecules. Our findings indicate that enhanced ionization should be operative for a wide class of rare-gas clusters. From a simple Coulomb-explosion ansatz, we derive an analytical expression relating the maximum energy release to a suitably scaled expansion time which can be expressed with the pulse length T^* .

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After a basic understanding of the mechanisms governing atoms and molecules subjected to an intense laser pulse [1,2], analogous studies on clusters pioneered by McPherson *et al.* [3] and Ditmire *et al.* [4] have appeared over the last few years with a recent spectacular culmination in the demonstration of deuterium fusion in clusters [5]. Most of these studies do focus on the situation after the laser pulse, namely, on the abundance and kinetic energy spectra of electrons and ions. Some discussion has been devoted to the question if the expansion of the cluster is driven by hydrodynamics or by a Coulomb explosion. Similar studies have been performed in the time domain [6–8]. What has not been investigated in detail is the influence of the laser pulse length on the dynamics. This is surprising since the time scales involved show that the expansion of the nuclei occurs on the same time scale as the pulse lengths which can be chosen, namely, some 10 to 1000 fs, or roughly 10^3 a.u. (which we use hereafter). Apart from the nuclear motion and the pulse length T energy absorption from a laser pulse and subsequent ionization and fragmentation of the cluster involve two additional time scales, the optical cycle $2\pi/\omega = 0.055$ a.u. for the typically used Titan-sapphire laser of 800 nm wavelength, and the period of the bound electrons, which is of the order (hydrogen) of 1 a.u. We work with peak intensities between 10^{14} and 10^{16} W/cm².

In the following we demonstrate that the seemingly complicated process of energy absorption and fragmentation in the laser pulse can be split into three different phases, an atomic phase I, a “molecular” phase II, and a relaxation phase III. Phase I lasts for a time T_0 after the pulse has begun and is characterized by boiling off electrons through multiphoton or tunneling ionization, hence we have termed it the “atomic” phase. We define it to last until every second atom in the cluster has lost one electron, or equivalently until the probability of losing an electron in an atom has reached $p = 1/2$. This probability is calculated from a Krainov tunneling rate [9] where, however, the instant electric field is formed by the laser

and eventually already existing charged particles in the cluster.

Up to T_0 we may assume that the atoms/ions have not moved yet. The second, molecular phase is characterized by Coulomb explosion of the cluster. During this phase, as we show below, the cluster expands to a critical radius R^* which optimizes the energy absorption. Phase III finally, until the end of the laser pulse and beyond, sees a relaxation of the system and the full fragmentation of the cluster proceeding. The existence of these phases follows from a careful analysis of our numerical results. The relevance of the phases is underlined by the time which is spent under phase II. This time turns out to be instrumental for relating the electron release quantitatively to the laser and cluster properties, as shown below.

To simulate the process of energy absorption numerically we have developed a quasiclassical model for small rare-gas clusters. The nuclei are treated completely classically, with the initial configuration defined by minimizing the pairwise Lennard-Jones interactions [10]. Electrons bound to an atom or ion at position \vec{R} are characterized by an effective binding energy $E_b = \epsilon_b + V_{\text{tot}}(\vec{R})$, where the exact atomic binding energy ϵ_b is shifted by V_{tot} , the sum of the potentials from the laser field, and all other charged particles except the atom/ion the electron is bound to. Ionization from such a bound state is accomplished via tunneling along the direction \hat{r} of the instant force at position \vec{R} , $\hat{r} = \vec{\nabla}V(\vec{R})/|\vec{\nabla}V(\vec{R})|$. The time-dependent tunneling action along \hat{r} reads

$$I(t) = \int_{r_1}^{r_2} \sqrt{2[V_{\text{tot}}(r) + V_{\text{atom}}(r) - E_b]} dr \quad (1)$$

with the classical turning points r_i determined by $V_{\text{tot}}(r) + V_{\text{atom}}(r) - E_b = 0$. From $I(t)$ we get the tunneling probability $P(t) = \exp[-2I(t)]$ and finally the tunneling rate $w(t) = P(t)/T_K$ with the classical Kepler period T_K of an orbit with binding energy ϵ_b . For each time step dt , a random number z is compared to the probability $w(t)dt$ for ionization during this time step. If $w(t)dt > z$, the

electron is born as a classical particle and placed at the outer turning point r_2 obeying total energy conservation. From then on, this electron follows Newton's equations, and the next bound electron can be ionized. Hence, strictly sequential ionization is enforced.

The interaction between two particles with charge Q_1 and Q_2 and position vectors \vec{r}_1 and \vec{r}_2 , respectively, is described with a smoothed Coulomb potential

$$V_{\text{softcore}} = \frac{Q_1 Q_2}{\sqrt{(\vec{r}_1 - \vec{r}_2)^2 + a_1(Q_1) + a_2(Q_2)}}, \quad (2)$$

where the a_i are charge-dependent soft-core parameters. For electrons we used $a(-1) = 0.1$, while the ionic a_i are chosen such that the potential minima for each ion always coincide with the quantum mechanical binding energy. This choice prevents artificial classical autoionization.

The model allows us to follow the full time-dependent evolution of the cluster with all interactions for a long time (10^5 a.u.) to investigate the influence of the cluster expansion during the laser pulse on its energy absorption. Although it implies, e.g., for xenon clusters, to propagate up to 200 charged particles, the computation can be handled with moderate resources due to the crucial simplification which arises from treating bound electronic motion not explicitly.

For the systems we have investigated the total electron release from the clusters which does not change any more after the laser pulse is over. Hence, it can serve as a robust observable which changes as a function of the pulse length T as shown in Fig. 1 for a Xe_{16} cluster in comparison with the corresponding electron release (i.e., ionization) of a Xe atom. The energy content of the laser pulse $\mathcal{E} = \int_0^T F(t)^2 dt$ has been kept constant which means that

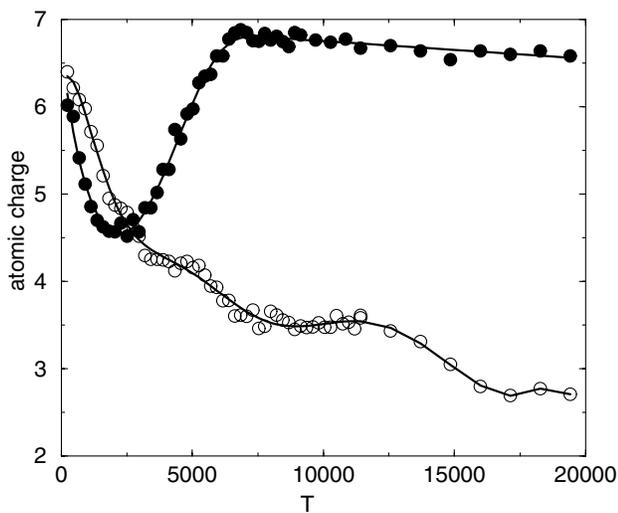


FIG. 1. Average atomic charge after the laser pulse for Xe_{16} (\bullet) and Xe (\circ) as a function of pulse length and for fixed energy of the laser pulse. The lines are to guide the eye.

the peak intensity F_0^2 of the pulse with amplitude $F(t) = F_0 \sin^2(\pi t/T) \cos \omega t$ decreases with increasing pulse length T according to $F_0^2 \propto 1/T$. As a reference for this energy normalization we chose a pulse with $F_0 = 0.16$ a.u. and a pulse length of 20 optical cycles. One sees that the ionization of an atom increases towards shorter pulse lengths T or equivalently, higher peak intensity. Indeed, atomic ionization depends on the peak intensity F_0^2 rather than on the pulse length T which is obvious if the electron yield is dominated by sequential ionization depending exponentially on F_0 via the Krainov rate [9], but only linearly on the pulse length. The oscillations in the single atom case are due to the atomic shell structure.

For the cluster the situation is quite different: for short T (high intensity F_0^2) the electron release/atom follows that of an isolated atom. In fact, it is even slightly lower than in the atomic case, due to the much larger Coulomb field of a multiply ionized cluster which has not significantly expanded. However, the electron release increases again and reaches a maximum for some optimum pulse length T^* at considerably smaller peak intensity. Hence, the cluster *expansion* plays an important role for the energy absorption, in contrast to the atom for which this degree of freedom does not exist. A comparison of time scales shows that indeed the Coulomb explosion of the ions in the cluster happens on the same time scale as the pulse duration (10^3 – 10^4 a.u. or equivalently some 10 to 100 fs). Hence, the dependence of the electron release on T points to the spatial expansion of the cluster which may in turn exhibit a maximum electron release for a certain cluster radius R . We define R in terms of the averaged distance between two ions in the cluster,

$$R(t) = \left(\frac{1}{N} \sum_{i=1}^N \min_{i \neq j} \{ |\vec{R}_i - \vec{R}_j|^2 \} \right). \quad (3)$$

First we assess the influence of the size of the cluster on the electron release under the reference pulse of 20 field cycles. The size of the cluster is varied preserving its shape by scaling the ionic positions $\vec{R}_i^\lambda = \lambda \vec{R}_i^0$ with a factor λ compared to the equilibrium positions \vec{R}_i^0 . As can be seen from Fig. 2, a critical value of the mean interionic distance, $R^* = \lambda^* R_0$, exists, where the ionization yield shows a maximum. The position of R^* hardly changes upon variation of the laser frequency. The ionization yield, however, increases with increasing frequency: this is due to the smaller ponderomotive oscillation amplitude at higher frequencies, which leads to increased interaction between quasi-free electrons driven by the laser field and those still well in reach of the cluster ions.

The mechanism responsible for the existence of R^* was first identified in diatomic molecules under the name CREI or enhanced ionization (ENIO) [11,12] with the (linear) laser polarization parallel to the molecular axis.

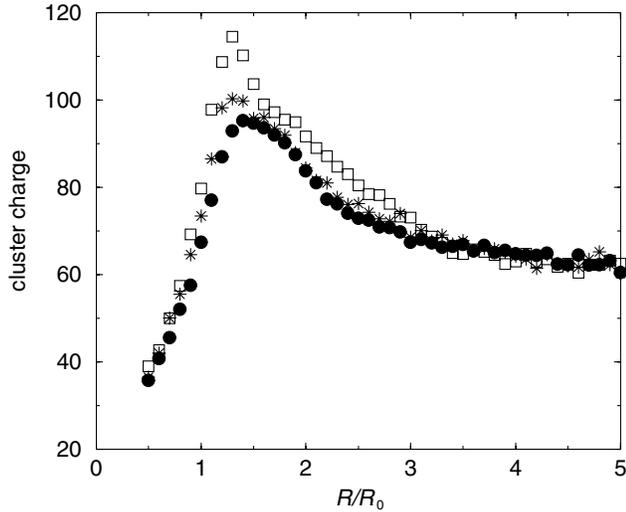


FIG. 2. Electron release of Xe_{16} after the laser pulse for fixed nuclei. At fixed pulse energy the pulse length was $T = 55$ fs and the frequencies $\omega = 0.055$ a.u. (\bullet), $\omega = 0.075$ a.u. (\star), and $\omega = 0.11$ a.u. (\square).

It might seem astonishing that the cluster also exhibits ENIO although there is no preferred axis which could align with the polarization axis. This is even more surprising since no enhancement was found for diatomic molecules if the polarization is perpendicular to the molecular axis or if the laser is circularly polarized. However, a distinct feature of ENIO is the insensitivity to changes in the laser frequency which we also find in the cluster (Fig. 2). This fact, together with the relation of T^* to the critical radius R^* as presented below, provides sufficient evidence that intense laser field dynamics of clusters is structured by ENIO as is the corresponding dynamics of molecules. For clusters, ENIO is even more general since there is no restriction with respect to the polarization of the laser: The direction of the axis for linear polarization does not matter and ENIO also occurs for circular polarization as shown in Fig. 3.

Our findings exemplified here for Xe_{16} have been confirmed by extensive calculations for a number of clusters of 8 to 30 atoms for the elements Ne, Ar, Kr, and Xe. These calculations clearly demonstrate that ENIO plays an important role for small rare-gas clusters under intense laser fields with quantitative consequences as we see next.

One could think that the relation of T^* and R^* is directly given by the Coulomb-explosion mechanism. The latter indeed links R^* to a certain time interval τ , but not to the entire pulse length T^* . The reason lies in the existence of the different phases as described in the beginning.

Only phase II drives the relevant Coulomb explosion, namely, the onset of the cluster expansion. For the maximum electron release this time interval ends if the critical radius R^* is reached at half the pulse length $T^*/2$ when the

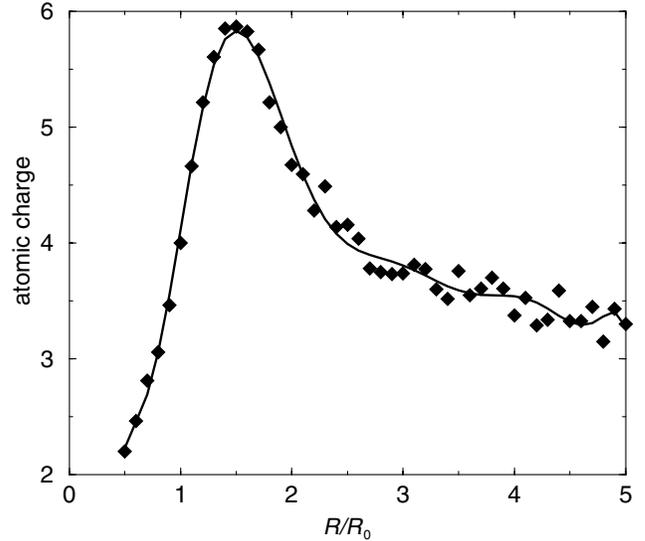


FIG. 3. Electron release of Xe_{16} at $\omega = 0.055$ a.u. as in Fig. 2 but for circular polarization.

laser pulse intensity has its maximum. The interval begins, however, only at T_0 when each atom in the cluster has on average a 50% probability of being ionized. At this time the cluster still has its equilibrium radius R_0 . Hence, we get $\tau = T^*/2 - T_0^*$ as the relevant time during which the cluster expands from R_0 to R^* .

Having identified phase II and its time interval τ as the one which controls the maximum number Q^* of released electrons, we can use the dynamics of Coulomb explosion to derive a quantitative relation between Q^* and the optimum pulse length T^* . To this end we assume that the ionic motion in τ can be described by an averaged ionic charge which is proportional to the averaged final charge of each atom in the cluster, i.e.,

$$Q_i = \alpha Q^*/N, \quad (4)$$

where N is the number of atoms in the cluster. Furthermore, we assume that in phase II the main kinetic energy goes into the expansion of the cluster without changing its shape. Using the same parametrization as before, we write now for the time-dependent position $\vec{R}_i(t)$ of an atom or ion $\vec{R}_i(t) = \lambda(t)\vec{R}_i^0$, where \vec{R}_i^0 is the equilibrium position of the atom before expansion, i.e., at time $t = T_0$. The kinetic energy $K = M/2 \sum_i [d\vec{R}_i(t)/dt]^2$ reads then

$$K = \left(\frac{d\lambda}{dt}\right)^2 \frac{M}{2} \sum_i^N (\vec{R}_i^0)^2 \equiv \left(\frac{d\lambda}{dt}\right)^2 K_0. \quad (5)$$

K_0 has form and units of a moment of inertia and represents the influence of the shape of the cluster on its kinetic energy during the expansion. The potential energy $V = \sum_{i>j} Q_i Q_j / |\vec{R}_i(t) - \vec{R}_j(t)|$ simplifies to

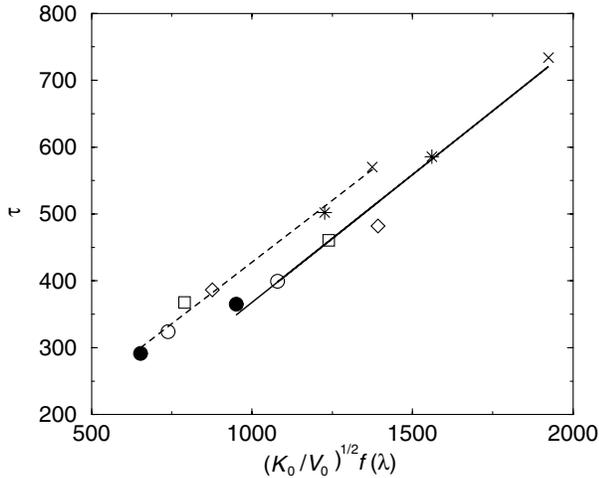


FIG. 4. Expansion time (numerical data) as a function of $(K_0/V_0)^{1/2}f(\lambda)$ and linear fits (see text). Two different energy normalizations were used: $F_0 = 0.16$ a.u. (solid line) and $F_0 = 0.25$ a.u. (dashed line), both at a frequency of $\omega = 0.055$ a.u. and a pulse length of $T = 55$ fs. \bullet : Ar₁₆, \circ : Ar₂₀, \square : Ar₂₅, \diamond : Ar₃₀, \star : Kr₁₆, and \times : Xe₁₆.

$$V = \lambda^{-1}(\alpha Q^*/N)^2 \sum_{i>j=1}^N |\vec{R}_i^0 - \vec{R}_j^0| \equiv \alpha^2 V_0/\lambda. \quad (6)$$

The differential equation for the expansion in terms of $\lambda(t)$ is obtained via the energy balance $K(t) + V(t) = E \equiv V(T_0)$, where at time T_0 before the expansion the kinetic energy of the atoms is zero. With the help of Eqs. (5) and (6) it can be written in the form

$$\frac{d\lambda}{dt} = \alpha[(1 - \lambda^{-1})V_0/K_0]^{1/2}. \quad (7)$$

Equation (7) can be solved analytically by separation of variables to yield

$$t(\lambda) - T_0 = \left(\frac{K_0}{V_0\alpha^2}\right)^{1/2} f(\lambda), \quad (8)$$

where we have set $\lambda = 1$ for $t = T_0$ and $f(\lambda) := \sqrt{\lambda(\lambda - 1)} + \ln(\sqrt{\lambda - 1} + \sqrt{\lambda})$. For the maximum energy release the critical radius $R^* = \lambda^* R_0$ should be reached after time $t(\lambda^*) = T^*/2$. This is the desired relation between the static ENIO mechanism at R^* and its dynamical effect during the cluster expansion in the time interval $\tau = T^*/2 - T_0^*$.

The proportionality factor in Eq. (4) determines the fraction of the final charge by which the expansion from R_0 to R^* during phase II is effectively driven. If phase II is indeed the crucial dynamical time span which universally controls the electron release we expect α to be the same for all types of clusters we consider, independent of the atomic element or cluster size. Under this assumption, we predict from Eq. (8) a linear relation between the expansion time τ and $(K_0/V_0)^{1/2}f(\lambda)$. In Fig. 4 we can see that

this prediction is actually very good: shown are the expansion times τ as a function of the cluster-dependent values of $(K_0/V_0)^{1/2}f(\lambda)$ for different clusters. A linear fit to the data yields $\alpha = 0.38$ and $\alpha = 0.37$ for energy normalized pulses at $F_0 = 0.16$ and $F_0 = 0.25$, respectively. The correlation coefficient is in both cases higher than 0.99. Hence, α is the same for different clusters, and it is almost the same for different energy normalizations of the laser pulse.

To summarize, we have shown that the enhanced ionization mechanism is operative for small rare-gas clusters over a wide range of parameters. Moreover, from a careful analysis of the Coulomb-explosion process, we conclude that energy absorption and subsequent ionization of the cluster proceeds in a very similar way for different clusters, irrespectively of the number and sort of atoms in the cluster. It is only for large rare-gas clusters, with $N \sim 10^3$ or more, that we expect a transition to a nanoplasma behavior, as has been found in hydrodynamical simulations of such systems [4,13]. Where and how this transition happens will be the subject of further studies, as well as the connection with enhanced energy absorption recently reported for small *metal* clusters [14].

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- [1] C.J. Joachain, M. Doerr, and N. Kylstra, *Adv. At. Mol. Opt. Phys.* **42**, 225 (2000).
 - [2] J. F. McCann and J. H. Posthumus, *Philos. Trans. R. Soc. London A* **357**, 1309 (1999).
 - [3] A. McPherson, T. S. Luk, B. D. Thompson, A. B. Borisov, O. B. Shiryaev, X. Chen, K. Boyer, and C. K. Rhodes, *Phys. Rev. Lett.* **72**, 1810 (1994).
 - [4] T. Ditmire *et al.*, *Phys. Rev. A* **53**, 3379 (1996).
 - [5] T. Ditmire *et al.*, *Nature (London)* **386**, 54 (1997).
 - [6] C. Rose-Petruck, K. J. Schafer, K. R. Wilson, and C. P. J. Barty, *Phys. Rev. A* **55**, 1182 (1997).
 - [7] I. Last and J. Jortner, *Phys. Rev. A* **62**, 013201 (2000).
 - [8] K. Ishikawa and T. Blenski, *Phys. Rev. A* **62**, 063204 (2000); M. Rusek, H. Lagarde, and T. Blenski, *Phys. Rev. A* **63**, 013203 (2001).
 - [9] M.V. Ammosov, N. B. Delone, and V.P. Krainov, *Sov. Phys. JETP* **64**, 1191 (1986).
 - [10] See <http://brain.ch.cam.ac.uk/jon/structures/LJ/> for a compilation of potential minima.
 - [11] T. Seideman, M. Yu. Ivanov, and P. B. Corkum, *Phys. Rev. Lett.* **75**, 2819 (1995).
 - [12] T. Zuo and A. D. Bandrauk, *Phys. Rev. A* **52**, R2511 (1995).
 - [13] J. Zweiback, T. Ditmire, and M. D. Perry, *Phys. Rev. A* **59**, R3166 (1999).
 - [14] L. Köller, M. Schumacher, J. Köhn, S. Teuber, J. Tiggesbäumker, and K. H. Meiwes-Broer, *Phys. Rev. Lett.* **82**, 3783 (1999).