Simulation of the dynamics of laser-cluster interaction

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Abstract. We study the interaction of an intense ($I \gtrsim 10^{15}\text{Wcm}^{-2}$) femtosecond laser pulse with a large ($N > 10000$ atoms) rare-gas cluster. The simulations are based on a mean-field classical transport approach. The electronic dynamics during the interaction with the laser are discussed in more detail. In particular we point out the difference in behavior between the fast electrons and the collectively moving slow electrons and try to shed light on the acceleration mechanisms behind the high energy tail of the electron energy distribution. The benchmark for our simulations is experimental X-ray spectroscopy data. We show a comparison with the experimentally found total X-ray yields and charge-state distributions.

1. Introduction
Ranging in size from a few atoms to several million atoms agglomerated at solid densities, clusters form a link between the gas and the solid phase. One manifestation is the up-conversion of the incident infra-red laser into keV X-ray photons in rare-gas clusters, which takes place with a relatively high efficiency providing large X-ray yields just like in solids, yet is relatively debris-free, a property shared with gas targets [1]. Similar behavior has been observed for the emission of energetic electrons [2] or highly charged ions [3], thus making the interaction of intense short and ultra-short laser pulses with clusters a topic of considerable interest [4; 5].

In a simple picture, the dynamics during the interaction of a strong laser pulse with a cluster can be summarized as follows [6]: the atoms of the cluster are first ionized by the incident laser pulse (inner ionization) and a cold ”nano-plasma” of solid density is formed. The quasi-free electrons take part in a collective oscillation driven by the laser field and, moreover, interact with the field of the surrounding particles. Electron-impact ionization of cluster ions produces additional quasi-free electrons and inner-shell vacancies which are at the origin of characteristic X-ray radiation. As a fraction of the electrons leaves the cluster (outer ionization), a net positive charge is left behind and the cluster begins to expand before disintegrating completely in a Coulomb explosion.

The size of the system and the abundance of mechanisms at play provide a considerable challenge for the theoretical description of the interaction. One approach to the problem is treating the cluster as a ”nanoplasma” submitted to a laser field [7; 8]. The inclusion of the cluster boundaries and the inhomogeneous ionic background during cluster expansion is however problematic. Molecular dynamics simulations on the other hand are limited to clusters of about 1000 atoms [9–11], and a scaling of the results to larger cluster sizes is difficult. Clusters of $\sim 10^4$ atoms have been simulated using a microscopic particle in cell (MPIC) code [12], but typical
runs of such codes take several weeks making the in-depth study of the parameter dependencies tedious and impractical. For large clusters the method of choice thus seems to be a mean-field approach, for example implemented as particle-in-cell (PIC) simulation [13; 14]. In this case, particular attention has to be paid to the correct incorporation of particle-particle effects otherwise neglected or underestimated due to the averaging [15].

From an experimental point of view, the spectroscopy of the emitted ions [3] and electrons [2] gives information on the system a few microseconds after the femtosecond laser-pulse and the cluster disintegration. X-ray spectroscopy [16], on the other hand, presents the advantage of measurements on a much shorter time-scale, down to a few femtoseconds. The inner-shell vacancies in argon responsible for the 3.1 keV characteristic X-ray radiation can not be explained by field ionization unless laser pulses with a peak intensity of the order of \( I \sim 10^{21} \text{Wcm}^{-2} \) are involved. Consequently, the origin of these vacancies must be impact ionization by energetic electrons. The X-ray emission thus probes the high energy tail of the electron energy distribution, thereby providing valuable insights into the electronic dynamics which are the key to a detailed theoretical understanding of laser-cluster interaction. Furthermore, high-resolution X-ray spectroscopy also gives access to the charge-state distribution of the ions emitting the X-ray radiation.

Recent experiments [1; 17] found an unexpectedly low laser intensity threshold for X-ray production. When irradiating large clusters with \( N > 10000 \) argon atoms with infrared (\( \lambda = 800 \text{nm} \)) laser pulses of duration \( \tau = 60 \text{fs} \) at FWHM, characteristic X-ray radiation could be measured for laser peak intensities as low as \( I_{th} \simeq 2.2 \cdot 10^{15} \text{Wcm}^{-2} \). At this intensity, the ponderomotive energy \( U_P = F^2/(4\omega^2) \) (atomic units are used unless otherwise stated) associated with the oscillatory motion of a free electron in a laser field with field strength \( F \) and frequency \( \omega \) is \( U_P \simeq 130 \text{eV} \simeq 5 \text{a.u.} \). This value is more than one order of magnitude below the binding energy \( E_K \simeq 3.1 \text{keV} \) of K-shell electrons in argon. Moreover, the analysis of the high-resolution X-ray spectra show that the mean charge state of the emitting ions can be as high as 13+ when irradiating argon clusters with pulses of an intensity of \( I = 3 \cdot 10^{16} \text{Wcm}^{-2} \).

Both these observations raise questions as to additional heating mechanisms at play in a cluster environment, which allow the electrons to be effectively accelerated well beyond the ponderomotive energy. After a decade of experimental and theoretical studies, the mechanisms causing the efficient heating of electrons in a cluster environment are still a matter of debate [5]. Most prominently, the nanoplasma model [7] predicts the production of fast electrons by inverse bremsstrahlung which is greatly enhanced when the plasma frequency of the electrons in the cluster matches the laser frequency. It has, however, been argued [8; 17] that this resonance should be strongly damped and that the high energy contribution of the electronic energy distribution should not be sufficient to explain the creation of inner-shell vacancies.

In this contribution we try to shed light on the mechanisms behind the production of fast electrons during laser-cluster interactions. We will begin by briefly summarizing our simulation method. This will be followed by a discussion of the electronic dynamics during the laser-cluster interaction, with a particular focus on the behavior of the fast electrons. In the final section, we will compare our simulation results to experimental data from X-ray spectroscopy.

2. Numerical model

As discussed in the introduction, the large size of the clusters (\( N > 10000 \) atoms) calls for some major simplifications in the computational treatment of the system. We therefore use a mean-field approach in which many-particle effects are included via stochastic processes [6; 15]. We also employ a test-particle discretization, i.e. we solve the equations of motion only for a representative fraction of the ensemble of particles, thereby drastically reducing the number of test particles to be followed. The representation fraction \( \alpha \) is limited by computational capabilities.
The test-particle ensemble consisting of \(k = 1, \ldots, \alpha N_e(t)\) electrons and \(j = 1, \ldots, \alpha N\) ions with mass \(M\) and screened charge state \(\tilde{q}_j(t)\) obeys the following Langevin equations:

\[
M\ddot{\mathbf{r}}_j^{(i)} = \tilde{q}_j(t)\mathbf{F}_L(t) + \dot{\mathbf{r}}_j(t)\mathbf{F}_{\text{mean}}(\mathbf{r}_j^{(i)}), \quad \dot{\mathbf{r}}_k^{(e)} = -\mathbf{F}_L(t) - \mathbf{F}_{\text{mean}}(\mathbf{r}_k^{(e)}), t) + \mathbf{F}_{\text{stoc}}(\mathbf{r}_k^{(e)}, \dot{\mathbf{r}}_k^{(e)}), t). \tag{1}
\]

This approach can also be seen as a generalization of classical transport theory (CTT) [18] for a dynamical system open to both particle number variation \(N_e(t)\) and energy exchange with the many-particle reservoir (ions and electrons), as well as with the laser field. In order to take into account that the ions are screened by the surrounding quasi-free electrons, we estimate the screened ionic charge state \(\tilde{q}_j(t)\) from the local number of quasi-free electrons per ion [6].

As the cluster is much smaller than the wavelength of the laser pulse, the laser can be described as a uniform time-dependent external electric field linearly polarized in the \(z\)-direction:

\[
\mathbf{F}_L(t) = F_0 \hat{z} \sin(\omega t) \sin^2\left(\frac{\pi t}{2\tau}\right). \tag{2}
\]

Electron-electron, ion-ion and electron-ion interactions are taken into account by the time-dependent mean field \(\mathbf{F}_{\text{mean}}(\mathbf{r}, t)\), which depends on the positions of all test particles. To evaluate the mean field, the simulation volume is divided in a cylindrical grid of \(64 \times 128\) cells in \((R, z)\), making use of the rotational symmetry around the polarization direction (\(z\)-axis) of the laser field. The grid is equally spaced in \(z\) direction, whereas the step-size in the radial \(R\) direction is chosen such that all cells of the grid have equal volume. At each time step the mean field is obtained by solving the Poisson equation on this grid [19]. Outside the simulation volume, the mean field is assumed to be determined by the net charge and dipole moment inside the simulation volume.

Momentum changes of the electrons due to collision processes are taken into account by the stochastic forces \(\mathbf{F}_{\text{stoc}}(\mathbf{r}_k^{(e)}, \dot{\mathbf{r}}_k^{(e)}), t)\). The ionic collision partner \(j\) of the electron is chosen randomly from the same cell. For example, elastic electron-ion scattering is controlled by the probability of each electron to scatter elastically during the time step \(\Delta t\):

\[
P_e = \sigma_e(q_j, E_k^{(e)})(\mathbf{r}_k^{(e)}, t)\dot{\mathbf{r}}_k^{(e)} \Delta t, \tag{3}
\]

which is determined by the energy \((E_k)\) and charge state dependent total elastic scattering cross-section \(\sigma_e\), the local ionic density \(\rho^{(i)}\) and the velocity of the electron. If the electron scatters, the scattering angle \(\theta\) is determined randomly according to the differential cross-section \(d\sigma_e/d\theta\). The scattering cross-sections can be calculated independently by partial wave analysis of parametrized Hartree-Fock potentials for all relevant charge states [15; 20; 21]. The change in momentum associated with the scattering of angle \(\theta\) is one contribution to the stochastic force \(\mathbf{F}_{\text{stoc}}(\mathbf{r}_k^{(e)}, \dot{\mathbf{r}}_k^{(e)}), t)\). Further contributions are, for example, electron-impact ionization of the outer-shell electrons and K-shell ionization. These events are not only marked by a change in momentum, but also by a change in the number of electronic test particles \(\alpha N_e(t)\) i.e. of ionized electrons, as well as in the number of bound electrons \(N_j^{(M)}(t), N_j^{(L)}(t)\) and \(N_j^{(K)}(t)\) in the respective shells of the ionized ion. The cross-section for impact ionization by an electron with kinetic energy \(E\) is evaluated from the Lotz formula [22]. This formula is also used to estimate the contribution of two-step ionization (electron-impact excitation from L-shell to M-shell followed by an impact ionization). Further ionization mechanisms included in the simulation are Auger decay of L-shell vacancies [23], over-barrier field ionization as well as tunnel ionization [6].

Electron-electron scattering is included by randomly pairing up electrons from the same grid cell and picking the relative scattering angle \(\theta\) from a Gaussian distribution of \(\delta = \tan(\theta/2)\) with
variance \[24\]

\[\langle \delta^2 \rangle = \frac{8 \pi \rho^e \ln \Lambda}{v_{\text{rel}}^2} \Delta t \] (4)

where \(\rho^e\) stands for the local electron density and \(v_{\text{rel}}\) for the relative velocity of the two particles. The Coulomb logarithm \(\ln \Lambda\) is approximately 2 for the dense plasmas at hand.

3. Cluster dynamics

To develop insights into the dynamics of the laser-cluster interaction, we focus on the specific case of a cluster with \(N = 3.7 \times 10^4\) argon atoms irradiated by an infrared (\(\lambda = 800\) nm) laser pulse with a pulse duration of \(\tau = 85\) fs and a peak intensity of \(I = 5.3 \times 10^{15}\) Wcm\(^{-2}\). The cluster atoms are initially assumed to form a sphere of solid density (\(\rho(t = 0) = 2.66 \times 10^{22}\) cm\(^{-3}\)) and radius 160 a.u.. The simulation is started at \(t = t_1\) when the laser field reaches the threshold for over-barrier ionization of the neutral argon atoms (\(F_{\text{OBI}}(t_1) \approx 0.08\) a.u., i.e. \(I_{\text{OBI}}(t_1) \simeq 2.2 \times 10^{14}\) Wcm\(^{-2}\)) and the first \(N_e(t_1) = \alpha N\) electrons are released with zero velocity at the position of the atoms. These electrons and ions provide the initial conditions for the propagation of Eq. (1). For this cluster size and pulse duration the representation fraction is chosen to be \(\alpha = 1\) (in the next section where we treat larger clusters \(\alpha = 0.1\)). The number of electrons increases rapidly due to the efficient ionization of the cluster atoms. Electron-impact ionization of the outermost atomic shell is dominating the ionization process, while excitation, Auger decay and ion-proximity effects provide only a minor contribution. At the conclusion of the laser pulse, a mean ionic charge state of Ar\(^{9+}\) is reached. During the pulse the ionic density at the cluster surface begins to drop as argon ions fly away, however, due to the relatively weak and short laser pulse, the radius of the sphere of solid ion density is only reduced by approximately 20% by the end of the pulse.

In the beginning of the interaction, the cluster behaves like a polarizable sphere driven by the laser field [15]: the electron cloud oscillates with respect to the ionic background, and the resulting net positive and negative charges at the cluster poles induce a dipole field. As, after reaching the first ionization threshold, the electron density \(\rho^{(e)}(t > t_1)\) is so high that the associated plasma frequency \(\omega_p = \sqrt{4 \pi \rho^{(e)}(t)/3}\) exceeds the driving frequency of the laser, the dipole field screens the laser field inside the cluster. At the cluster poles, on the other hand, the electric field is enhanced. The collective displacement of the electron cloud can still be observed near the pulse maximum (fig. 1). However, as a fraction of the quasi-free electrons
have left the cluster, an additional overall net positive charge is built up, which produces a strong asymmetry in the potential (fig. 2): at one cluster pole this positive charge is just compensated by the electron cloud, thus flattening the potential, while at the opposite pole, the potential is steepened due to the additional charge. These polarization effects lead to a reduction of the laser field inside the cluster by nearly a factor 10 throughout the whole pulse, while enhancing the laser field by a factor 2-3 at the cluster poles.

![Figure 3.](image)

**Figure 3.** (Colour online) Kinetic energy distribution of the quasi-free electrons inside the initial cluster boundaries at different times during the laser pulse: $t_1 = 35$ fs (red), $t_2 = 70$ fs (green) and $t_3 = 100$ fs (blue). The dashed lines represent Maxwell-Boltzmann energy distributions with $T(t_1) = 0.2$ a.u. (red), $T(t_2) = 1.2$ a.u. (green) and $T(t_3) = 2.8$ a.u. (blue).

Due to the small effective field inside the cluster and the ongoing production of slow electrons by ionization, the electrons inside the cluster have on average energies well below the ponderomotive energy $U_P = 12$ a.u. corresponding to the intensity of $I = 5.3 \times 10^{15}$ W cm$^{-2}$ of the free laser field (see fig. 3). While Maxwell-Boltzmann distributions with time-dependent temperatures ranging up to $T \approx 3$ a.u. represent the low-energy portion of the electron spectrum (inside the cluster) quite well, they grossly underestimate the high energy tail of the electron distribution.

For a better understanding of the behavior of the fast electrons, we look for the subset of fast electrons within the ensemble depicted in fig. 1: while the high density electron cloud is shifted to positive $z$ values, thus producing there a flattened potential, a small fraction of electrons (yellow) occupies the space left of the cluster (negative $z$ values), where the steep potential accelerates the electrons toward the cluster. Examining the spatial dependence of the mean kinetic energy distribution (fig. 2) identifies the electrons to the left of the cluster as fast electrons. This subensemble of fast electrons transits the cluster from one pole to the other during approximately half a laser cycle, before being turned back by the enhanced laser field at the electron-depleted pole. Due to the mismatch in arrival times of the electrons and the strongly time and coordinate dependent field, the fast electron bunch is dissolved when exiting the cluster and then rebuild on reentering the cluster (fig. 4 and animation). A similar behavior was also observed for a 2D cluster simulation in Ref. [13].

The maximum kinetic energy of electrons (i.e. the cut-off energy of the energy distribution in fig.3) inside or in proximity of the cluster can be used to study the contributions to the electron heating of the monopole field, resulting from the charging of the cluster, and of the laser field
Figure 4. (Colour online) Snap shots of the phase space $z, v_z$ at times $t = 73$ fs (left) and $t = 74$ fs (right). Only electrons which belonged at some previous time to the high energy tail of the energy distribution and are within $50 \, \text{a.u.}$ of the $z$-axis are represented ($\diamond$). The dots mark the positions at the 4 previous time steps. The upper graph shows the current field (green line) along the $z$-axis. An animated series of such pictures spanning a whole laser cycle is provided with this paper.

Figure 5. (Colour online) Red: Maximum electron kinetic energy in the simulation volume as a function of time. The rapid oscillations in the energy reflect the acceleration and deceleration of the electron bunch when turning around at the cluster poles. Green: Electrostatic potential at the center of the cluster.

enhanced by the cluster polarization (fig. 5). The maximum kinetic energy an electron can get from the acceleration due to the space charge of the charged-up cluster can be estimated from the value of the potential at the origin (center of the cluster). While this contribution of the monopole field nicely mimics the time dependence of the maximum energy, it underestimates its absolute value. This is explained by the fact, that in addition to the field of the charged cluster, the fast electrons are pulled back into the cluster by the laser field which is enhanced by the polarization of the cluster.
4. Comparison with experimental results from X-ray spectroscopy

The simulation gives access to the number of K-shell vacancies in cluster ions produced by electron-impact ionization. This allows to compare simulation results with experimental data from X-ray spectroscopy [1; 17]. For this comparison, the spatial Gaussian intensity profile of the laser beam [25] has to be taken into account, resulting in an averaging over several laser intensities for each data point. For laser intensities of the order of $10^{16}$ W cm$^{-2}$, the results for the charge state distribution of the X-ray emitting ions show a good agreement, even though the production of Ar$^{16+}$ is still underestimated (fig. 6). However at low intensities near the threshold for X-ray production ($\sim 10^{15}$ W cm$^{-2}$), the simulations give only a mean charge state of 9+ to 10+, while the experiments appear to show no significant decrease of the mean charge state $\bar{q} \approx 12.5+$ with decreasing laser intensities. The origin of this discrepancy is as of now unknown.

Figure 6. (Colour online) Charge state distribution of the ions with K-shell vacancies. The argon cluster with $\langle N \rangle = 5 \times 10^5$ atoms is irradiated by a laser pulse of duration 60 fs (FWHM in intensity) and intensity $3.5 \times 10^{16}$ W cm$^{-2}$. The simulation (purple) and the experiments (yellow) are normalized at 14+.

Figure 7. (Colour online) Intensity dependence of the absolute X-ray yield from argon clusters ($\langle N \rangle = 3.7 \times 10^4$ atoms) irradiated by laser pulses with durations (FWHM in intensity) of 60 fs (squares), 150 fs (circles) and 610 fs (triangles). Simulation results (full symbols) and experimental data (open symbols) are compared assuming a clustering ratio of 5%.

The X-ray yield per cluster is determined by the number of K-shell vacancies created, corrected by the mean fluorescence yield $\eta \approx 0.12$ [23]. To extract the absolute X-ray yield, one would also need the clustering ratio of the gas in order to know the number of clusters formed in the interaction volume. Unfortunately, while the mean atomic density of the argon gas is known, the clustering ratio is not yet accessible experimentally and remains a free parameter, which we chose to be 5%. With this choice kept fixed, we get very good quantitative agreement between simulations and experiments for the X-ray yield at different intensities and pulse lengths (fig. 7).

5. Conclusion

In summary, we have developed a mean-field approach to the complex dynamics of laser-cluster interaction. We find X-ray yields and charge state distributions in satisfying quantitative agreement with the experimental results. We have shown that, while the large majority of the electrons follows a collective motion driven by the laser field and gains only little energy, the
electrons of the high energy tail of the kinetic energy distribution behave very differently and get heated due to the combined effects of the monopole field of the charged cluster and the laser field enhanced by the polarization of the cluster. The dependence of these heating mechanisms on parameters such as cluster size, laser pulse length or laser wavelength remains, however, to be studied more closely, which also calls for a more careful analysis of the ionic dynamics during cluster explosion.

Acknowledgments
This work is supported by FWF SFB-16 (Austria).

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