Rare Gas Clusters in Intense Laser Fields

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General Problem Description

Laser pulses with an intensity up to several times $10^{20} \text{ W/cm}^2$ can be nowadays produced in several laboratories worldwide. Already at $3.5 \times 10^{16} \text{ W/cm}^2$ the electric field of the laser equals the one “seen” by the electron in the hydrogen atom on its first Bohr orbit. As a result, electrons are quickly freed when such an intense laser pulse impinges on matter, and a plasma is formed. Since the power in these laser pulses is so enormous, the pulse durations must be small. The shortest pulses achieved consist of only two laser cycles, corresponding to ~5 femtoseconds ($10^{-15}$ seconds). Such short times are hard to imagine, but a femtosecond is to a minute what a minute is to the age of the universe.

One goal in intense laser-matter interaction is to dump as much laser energy as possible into the target. In doing so, very energetic (keV) electrons and ions (MeV) can be produced. When two fast, light ions collide, nuclear fusion may occur. This has been demonstrated in experiments with targets containing deuterium. The high charge states present in the laser-generated plasma give rise to bright X-ray emission with great potential for applications in nanolithography, for instance.

Clusters, being aggregates of 10 up to $10^6$ atoms that are held together by either the van der Waal-force (rare gas clusters) or by the delocalized electrons (metallic clusters), bridge the gap between bulk material and single atoms. It turned out that clusters absorb laser energy particularly well. Contrary to bulk material, no thin, overcritical plasma layer is formed at the target’s front surface which acts like a mirror and reflects the remainder of the laser pulse. This is because the cluster radii are small compared to both laser wavelength (typically 800 nm) and the so-called skin depth. Moreover, in clusters the fast electrons cannot escape into the cold bulk material but are exposed to the laser field throughout the entire pulse.

Numerous experiments and theoretical investigations related to the interaction of intense laser fields with rare gas clusters have been performed (see Refs. [1,2] for reviews). Our work in 2003 mainly focused on the ionization dynamics of small rare gas clusters.

Problem Details and Work Done in the Reporting Period

*Time-dependent density functional (TDDFT) studies: Dynamical ionization ignition.* Time-dependent density functional theory was used to study the ionization dynamics of clusters. To this end, the spin-degenerate time-dependent Kohn-Sham equation (TSDKSE) was solved. However, solving the TSDKSE in full dimensionality for rare gas clusters in laser fields is very demanding since, contrary to metal clusters and fullerenes, the so-called jellium approximation where the ion background is

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smeared is a priori not applicable. We therefore studied a 1D model of a cluster where the laser polarization direction is the sole degree of freedom. We solved the TDKSE for the electrons (atomic units (a.u.) are used unless noted otherwise),

\[
\frac{i}{\hbar} \frac{\partial \psi_j(x,t)}{\partial t} = (-\frac{1}{2} \nabla^2 + U + V_{\text{xc}} + V_{\text{ion}} + V_{\text{laser}}) \psi_j(x,t),
\]

where \( U, V_{\text{xc}}, V_{\text{ion}}, V_{\text{laser}} \) are the Hartree potential, the exchange-correlation potential, the potential created by the ions, and the dipole interaction with the laser field, respectively. Soft-core Coulomb potentials were used for the particle interactions, and the exchange-only local density approximation was employed for \( V_{\text{xc}} \) (see Ref. [3] for details). The ions were propagated according to their classical equations of motion.

Fig. 1 shows the electron dynamics of a 1D model cluster consisting of 9 ions with 4 active electrons each, submitted to a laser pulse of intensity \( 1.3 \times 10^{14} \text{W/cm}^2 \) at 800nm wavelength. The laser pulse was ramped up over 3 cycles and held constant thereafter.

![Figure 1: Logarithmically scaled electron density vs space and time for a cluster consisting of 9 ions and 4 active electrons per ion. The laser intensity was \( 1.3 \times 10^{14} \text{W/cm}^2 \), the laser wavelength was 800nm. The laser field was ramped up over 3 cycles and afterwards held constant. See text for discussion.](image)

Initially, the electron density is located close to the ion positions (yellow maxima). Then, as the laser is ramped up, electrons leave the cluster ("outer ionization") and oscillate in the laser field (green and red). However, as electrons leave the cluster the positive ion background gives rise to a potential that hinders further electrons from escaping straightaway.

Moreover, the positively charged ions repel each other so that the entire cluster Coulomb-explodes (diverging yellow stripes of high electron density in Fig.1). Some electrons are just removed from their parent atoms ("inner ionization") but remain bound with respect to the cluster as a whole. The most interesting aspect of the electron dynamics in Fig. 1 is the wave packet bouncing from one side of the cluster to the other. This collective electron dynamics affects both the energy absorption from the laser and further inner ionization, as it is shown in Fig 2.

At the time where the wave packet is close to the left cluster boundary (Fig. 2a,b), the laser field is zero. However, the internal field due to the space charge is maximum. A quarter of a laser cycle later (Fig. 2c,d) the wave packet is in the cluster center so that the field \( F_{\text{int}} \) due to the space charge is low but the laser field has its maximum then. Hence, throughout the laser cycle there is always an electric field inside the cluster. Therefore inner ionization is more efficient as if there was the laser field alone.
Figure 2: Snapshots of the electron density and the total potential (a,c) at the two time instants indicated by the horizontal lines in the relative electron density contour plots (b, d). Red and yellow color mean accumulation of electrons, blue and black a lack of electrons. The bar at the bottom of the density plots indicate the center of mass of the electrons inside the cluster. Arrows, +, and – in the potential plots illustrate the forces $F_{\text{laser}}$, $F_{\text{int}}$ exerted by the laser field and the space charge, respectively. The laser intensity was $3.8 \times 10^{13} \text{ W/cm}^2$, the wavelength 800nm. See text for discussion.

We named this effect “dynamical ionization ignition” [3] since it is the generalization of the well-known (static) ionization ignition effect [4].

**Semi-classical molecular dynamics (SCMD) simulations.** Full 3D laser-cluster calculations were performed using molecular dynamics with the inner-atomic dynamics treated explicitly. Since classical many-electron atoms are unstable (i.e., autoionizing) we followed an approach introduced by Kirschbaum and Wilets [5] where an uncertainty relation $rp \gg \xi$ is implemented by introducing a momentum-dependent potential $V_H$ (“Heisenberg potential”) of the form

$$V_H = \frac{\xi^2}{4\alpha r^2 \mu} \exp \left( \alpha \left[ 1 - \left( \frac{rp}{\xi} \right)^4 \right] \right),$$

with $\xi, \alpha$ are parameters (to be adjusted) and $r, p$ are the distance between electron and ion and the relative momentum, respectively (see [6,7] for details). This Heisenberg potential prevents electrons from falling into the “black hole” of the nucleus and thereby stabilizes the classical many-electron atom.

Our working Hamiltonian describing $N_a$ atoms at the positions $\vec{R}_i$ and $Z$ active electrons per ion at the positions $\vec{r}_j$ reads

$$H = \sum_{i=1}^{N_a} \frac{P_i^2}{2M} + \sum_{j=1}^{Z N_a} \frac{p_j^2}{2} + \vec{E}(t) \cdot \left( \sum_{j=1}^{Z N_a} \vec{r}_j - Z \sum_{i=1}^{N_a} \vec{R}_i \right)$$

$$+ \sum_{i=1}^{N_a} \sum_{j=1}^{Z N_a} \left( V_H - \frac{Z}{|\vec{R}_i - \vec{r}_j|} \right) + \sum_{i=1}^{N_a} \sum_{k=1}^{N_a} \frac{Z^2}{|\vec{R}_i - \vec{R}_k|} + \sum_{j=1}^{Z N_a} \sum_{l=1}^{Z N_a} \frac{1}{|\vec{r}_j - \vec{r}_l|},$$

where $\vec{P}_i, \vec{p}_j$ are ion and electron momenta, respectively, $M$ is the ion mass, and $\vec{E}$ is the electric field of the laser. We studied the dynamics of a model xenon cluster consisting of 54 atoms with all 6 electrons in the 5p shell active [7]. Particular attention was given to the ionization and energy absorption mechanisms and their
wavelength dependence. In this contribution, due to the limited space, we only present the results for the relative inner and outer cluster ionization.

Figure 3: Relative ionization (i.e., normalized to the single, isolated atom ionization shown in the inset) per cluster atom as a function of the peak laser intensity for the two laser wavelengths 800nm (red) and 100nm (blue). Solid curves: inner ionization, dotted: outer ionization. Multiply the laser intensity in a.u. with $3.51 \times 10^{16}$ in order to obtain the common units W/cm$^2$. See text for discussion.

Here, “relative” means that the result was normalized to the single, isolated Xe atom ionization in the same laser field so that one can immediately infer whether the cluster ionizes more or less efficiently. Fig. 3 shows the relative ionization per cluster atom for the two laser wavelengths 800 (red) and 100nm (blue). In both cases the laser pulse was ramped up over 8 femtoseconds (fs), held constant for 26.5fs, and ramped down again over 8fs. The single atom result is shown in the inset. It is seen that at low laser peak intensities relative cluster ionization is high for both wavelengths. This is due to (dynamical) ionization ignition: the first freed electrons escape from the cluster and the electric field of the ions enhances further inner ionization. However, as later electrons are trapped by the cluster potential, relative ionization drops. In the short wavelength case it drops even below unity, indicating that the single atom actually ionizes more efficiently than atoms inside the cluster. This is due to the tiny excursions of the electrons in high-frequency laser fields (note that a free electron oscillates with an amplitude $\frac{E}{\omega^2}$ where $\omega$ is the laser frequency), leading to inefficient transport of the electrons away from the cluster. In fact, it was found that electron emission at short wavelengths is isotropic, as it is expected from thermionic emission, while at long wavelengths it is strongly peaked in laser polarization direction, indicating the dominance of the quiver motion in the laser field.

Resource Usage at PC²
The Siemens hpcline was used. Both TDDFT and semi-classical molecular dynamics code are not truly parallel. The computing cluster was used to perform parameter studies in an efficient way, e.g., running the program for a fixed cluster configuration but, say, 16 different laser peak intensities.

References