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F. Greschik, L. Arndt and H.-J. Kull

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Absorption of few-cycle laser pulses by outer ionization of atomic clusters

F. Greschik, L. Arndt and H.-J. Kull
Institute of Theoretical Physics A, Rheinisch Westfälische Technische Hochschule D-52056 Aachen, Germany

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PACS. 36.40.Gk – Plasma and collective effects in clusters.
PACS. 52.38.Dx – Laser light absorption in plasmas (collisional, parametric, etc.).
PACS. 52.65.-y – Plasma simulation.

Abstract. – Laser light absorption is studied by particle simulations for large atomic clusters interacting with intense few-cycle pulses. A significant part of absorbed energy can be attributed to the potential energy required for outer ionization. A simple shell model accounts for the ionization degree and the ionization potential. High absorption fractions are obtained for clustered gases under common experimental conditions.

Over the past decade, the interaction of intense laser pulses with large atomic clusters has received broad attention [1, 2]. As a mesoscopic system between atoms and solids, atomic clusters have unique properties. In particular, strongly enhanced energy absorption has been observed for optical [3] and, more recently for VUV [4, 5] frequencies. Other quite remarkable observations include high-charge states [6, 7], energetic particles [8–10] and keV-photons [6, 11]. Maybe most impressively, high neutron yields have been found from fusion reactions in deuterium clusters [12, 13].

Absorption of laser light by atomic clusters becomes strongly enhanced by the Mie resonance [14]. Collective electron plasma oscillations are resonantly excited by the laser field during the cluster expansion due to the gradual decrease of the electron and ion densities. Detailed experimental studies have confirmed the delay time between the ionization of the dense cluster and the subsequent resonant absorption of the dilute expanding plasma [15]. Some of the absorption mechanisms discussed in this context are inverse bremsstrahlung absorption [14], collisionless resonance absorption [16] and ionization damping [17]. Further heating of outer electrons has been attributed to scattering at the cluster potential [18, 19] and to resonantly enhanced oscillations [20]. There have also been a number of simulations by different approaches which give useful additional insight [21–24]. For metal clusters at lower laser intensities the Mie resonance was studied at a microscopic level on the basis of the time-dependent local-density approximation [25].

The present work is addressed to a different regime of nonresonant absorption of laser pulses whose pulse duration is shorter than the characteristic expansion time of the cluster. Such conditions can be met for few-cycle pulses with pulse durations in the 10 fs regime that have recently become available [26, 27]. These pulses are substantially shorter than those used in most previous laser-cluster experiments and they should offer a unique opportunity to distinguish between resonant and nonresonant absorption processes. Nonresonant absorption at a steep planar surface was originally proposed by Brunel [28]. According to this work, the energy is primarily absorbed by electrons emitted from the surface into the vacuum. Extensions of the model to thin foils and cylindrical columns have also been discussed [29,30]. However, apparently it is not yet well understood how this absorption mechanism works for 3D clusters. In the present work, we have studied nonresonant absorption by outer ionization of clusters on the time scale of few-cycle pulses.

The interaction of a strong laser pulse with an atomic cluster will be treated in the framework of a plasma model. The plasma is assumed to be formed inside the cluster at the beginning of the laser pulse. Typical ionization processes are tunneling ionization and collisional ionization. For simplicity, the ionization step, commonly called inner ionization, will not be explicitly treated. Instead, as a generic initial state, a uniform quasi-neutral plasma sphere with radius R, ion charge Z, ion density n_i and electron density $n_e = Zn_i$ will be assumed. Other initial conditions can be studied in the same manner.

Some basic plasma parameters are the plasma frequency ω_p, the Debye length λ_{De} and the mean particle distance r_s defined by

$$\omega_p^2 = \frac{4\pi q_e^2 n_e}{m_e}, \qquad \lambda_{De}^2 = \frac{T_e}{4\pi q_e^2 n_e}, \qquad \frac{4\pi}{3} n_e r_s^3 = 1, \tag{1}$$

where q_e denotes the electron charge, m_e the electron mass and T_e the electron temperature. When the plasma is heated up to sufficiently high temperatures, the ideal plasma approximation becomes valid. This approximation can be expressed in terms of the coupling parameter

$$\Gamma = \frac{q_e^2}{r_s T_e} = \frac{1}{3}\frac{r_s^2}{\lambda_{De}^2} \; . \tag{2}$$

Here we restrict attention to the ideal plasma regime with $\Gamma \ll 1$.

The laser field can be described in the framework of the dipole approximation by a time-dependent electric field. It is assumed of the form

$$\boldsymbol{\mathcal{E}} = \boldsymbol{\mathcal{E}}_0 \sin^2\left(\frac{\pi t}{\tau}\right)\sin(\omega t), \tag{3}$$

with a $\sin^2$-envelope. The frequency is chosen to be $\omega = 0.1\omega_p$, the pulse duration $\tau = 8(2\pi/\omega)$. It corresponds to an 8-cycle pulse with wavelength $\lambda = 1.056\,\mu$m for an electron density of $10^{23}\,\text{cm}^{-3}$. The intensity I of the pulse will be defined in terms of the maximum of the pulse envelope as $I = (c/8\pi)\mathcal{E}_0^2$, where c denotes the speed of light. It is noted that the maximum electric field is actually different from $\mathcal{E}_0$. The difference is commonly expressed by the carrier-envelope phase of few-cycle pulses. In the present case this field correction is small and it is therefore neglected.

We have calculated energy absorption with an electrostatic particle code similar to the one described by Pfalzner and Gibbon [31]. Particle simulations of Coulomb systems require a proper treatment of collisions with small impact parameters. Within the ideal plasma approximation ($\Gamma \ll 1$), collisional effects are absent or negligible and the specific form of the force law at small distances therefore does not matter. For convenience, we have chosen a harmonic-core Coulomb force of the form

$$\boldsymbol{F}_{ij} = \frac{Q_i Q_j}{r_0^3}\boldsymbol{r}_{ij} \times \begin{cases} 1 & , \quad r_{ij} < r_0, \\ \left(\frac{r_0}{r_{ij}}\right)^3 & , \quad r_{ij} > r_0. \end{cases} \tag{4}$$

Here $Q_{i,j}$ denote the charges of two simulation particles at the positions $\boldsymbol{r}_{i,j}$ separated by $\boldsymbol{r}_{ij} = \boldsymbol{r}_i - \boldsymbol{r}_j$ and r_0 denotes the cut-off distance below which the Coulomb force is regularized. In practice, the applicability of the ideal plasma approximation can be checked by varying the parameter r_0 to show that it actually has negligible influence on the results under consideration. We also have substituted the force law (4) by the exact force between homogeneously charged spheres of radius r_0, which did not lead to appreciable differences of the results presented. Furthermore, excellent quantitative agreement has been obtained with a relativistic particle-in-cell (PIC) code for two-dimensional systems [30]. Finite-size particles with various shape factors are a basic concept in the simulation of ideal plasmas [32].

In our computations, a cluster with radius $R = 3.2\,\mathrm{nm}$, density $n_e = 10^{23}\,\mathrm{cm}^{-3}$ and charge Q is represented by J pairs of positive and negative spherical charges $\pm Q/J$. The charge-to-mass ratio of the simulation particles is the same as that for ions and electrons, respectively. The number of finite-size particles J can be smaller than the number of real particles. The calculations have been performed with $J = 905$ particles, which was sufficient for obtaining well-defined averages. The radius r_0 can be chosen as large as possible to avoid short-range interactions but small enough to calculate the average force on each particle in good accordance with the Coulomb law. We have chosen $r_0 = R/J^{1/3}$, which corresponds to the mean distance between simulation particles. Initially, the pairs are placed at rest on a regular grid within a sphere of radius R. Energy absorption of the cluster due to the interaction with the laser pulse is calculated by taking the difference of the total energy of the N-particle system before and after the interaction, $W = W_f - W_i$, where W_i denotes the total energy of the initial and W_f the total energy of the final state. Similarly, the change of the potential energy $U = U_f - U_i$ and of the kinetic energy $K = K_f - K_i$ is evaluated.

We first present results of a calculation with rigid ions in fig. 1. Due to the large ion mass, ion motion plays no essential role on the time-scale considered. It can be seen that the absorbed energies are increasing with the laser intensity. Within an intermediate intensity regime between 10^{16} and $10^{18}\,\mathrm{W/cm}^2$ the increase is approximately linear in the intensity. At lower intensities absorption strongly decreases, at higher intensities it grows at a reduced rate. In this intensity regime a significant fraction of electrons is removed from the cluster by the laser field, which is called outer ionization. The behavior of absorption closely correlates with the degree of outer ionization, which will be discussed in more detail below.

To demonstrate the applicability of the rigid ion model, ion motion has been included in a few calculations for typical rare-gas clusters. Choosing Ar- and Xe-ions with a relatively large charge state of $Z = 10$ yields the results shown in fig. 2. The main effect of ion motion is to slightly increase the radius of the ion sphere within the interaction time, thereby lowering the effective Coulomb fields. However, the influence of ion motion on absorption is seen to be still small for Ar-ions and nearly absent for Xe-ions. As may be expected, the largest differences occur in the regime of partial outer ionization at intermediate intensities. In these calculations, the initial conditions have also been varied. The particles have been randomly placed within the cluster sphere. The effect of the initial conditions leads to minor changes at small intensities, where outer ionization is no longer the dominant absorption mechanism.

The present scaling of the absorbed energy with the laser intensity cannot be simply explained in terms of the well-known Brunel model of absorption at a plane surface [28]. In this model, the number of emitted electrons N^{out} and the energy gained by these electrons K^{out} is estimated by

$$N^{out} \propto n_e \xi \propto \mathcal{E}_0, \qquad K^{out} \propto N^{out} U_P \propto \mathcal{E}_0^3 \propto I^{3/2}, \tag{5}$$

where

$$\xi = \frac{q_e \mathcal{E}_0}{m_e \omega_p^2}, \qquad U_P = \frac{q_e^2 \mathcal{E}_0^2}{4 m_e \omega^2}.$$

Here ξ denotes the displacement of an electron inside the solid and U_P the ponderomotive energy of an electron in the laser field. According to this estimate the absorbed energy is

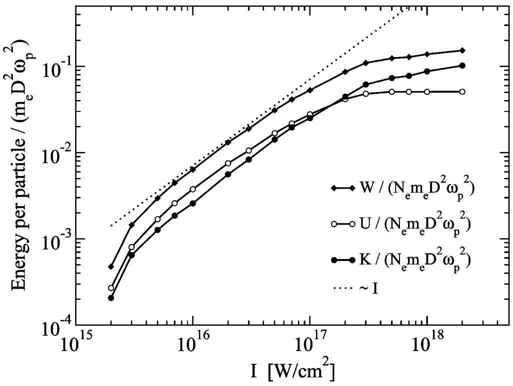

Fig. 1

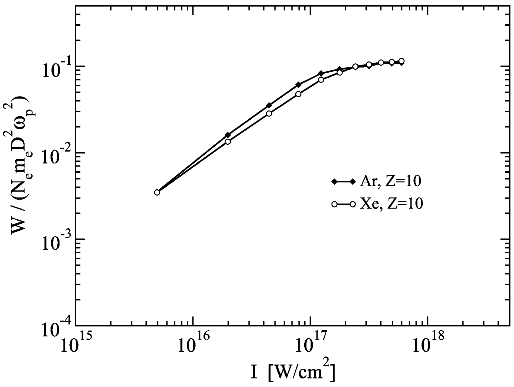

Fig. 2

Fig. 1 – Energy absorbed by a cluster with rigid ions due to the interaction with laser pulses of different intensities I. In the regime of outer ionization, the absorbed energy increases approximately linearly as shown by the dashed line. (W: total energy, U: potential energy, K: kinetic energy, D: cluster diameter, N_e: number of electrons).

Fig. 2 – Total energy per particle absorbed by clusters of mobile Ar- and Xe-ions with charge state $Z = 10$. Due to their smaller mass, Ar-ions reach larger expansion velocities, leading to somewhat enhanced absorption at intermediate intensities.

expected to scale as $I^{3/2}$. The intensity scaling shown in fig. 1 is basically modified by the spherical geometry due to the saturation of outer ionization.

We now present a simple model that allows one to analytically predict the degree of outer ionization and the potential energy gain of the ionized cluster. The ions are described by a rigid homogeneously charged sphere with radius R, density n_i and an average charge state Z. The electrons are assumed to relax to a quasistatic equilibrium state. For simplicity, this equilibrium is modelled by another negatively charged sphere with density $n_e = Zn_i$ and radius $R_e < R$. After the laser pulse, the electron and ion spheres form a neutral core surrounded by a positively charged concentric shell. The basic problem is to estimate the radius of the final electron sphere as a function of the laser intensity.

For this purpose, it is helpful to follow the electron dynamics from the final equilibrium state backward in time. In the falling part of the laser pulse there is no outer ionization. The electron sphere therefore just oscillates around the center of the ion sphere without crossing it. In a quasistatic equilibrium the center-of-mass position ξ of the inner electrons is linearly related to the laser field $\mathcal{E}$ by

$$\frac{\xi}{R} = \frac{\mathcal{E}}{E_C}, \tag{6}$$

where $E_C = Q/R^2$ is the Coulomb field at the surface of the ion sphere with total charge Q. At the maximum field $\mathcal{E}_{max}$ of the laser pulse, the electron sphere touches the surface of the ion sphere and the corresponding maximum displacement ξ_{max} satisfies

$$R_e + \xi_{max} = R. \tag{7}$$

Combining (6) and (7), defining $I_C = cE_C^2/(8\pi)$ and setting $\mathcal{E}_{max} \approx \mathcal{E}_0$, one obtains the

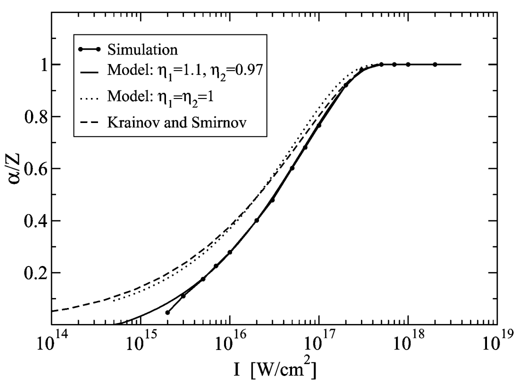

Fig. 3

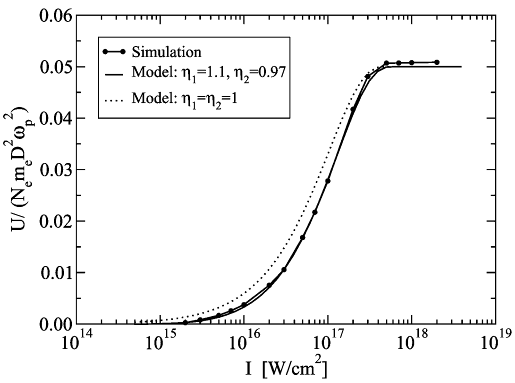

Fig. 4

Fig. 3 – Degree of outer ionization α of a cluster with charge state Z as a function of the laser intensity I. Predictions of the present shell model with parameters η_1 and η_2 are compared with the simulation results and previous analytic work [2].

Fig. 4 – Potential energy of the cluster after the interaction with a laser pulse as a function of the laser intensity. Predictions of the shell model with parameters η_1 and η_2 are compared with the simulation results.

inverse aspect ratio $\varrho = R_e/R$ of the two shells as a function of the laser intensity,

$$\varrho = 1 - \sqrt{\frac{I}{I_C}}. \qquad (8)$$

At $I = I_C$ the cluster becomes completely ionized up to the given charge state Z. The ionization degree α can be defined as the ratio of the total number of outer electrons over the total number of ions and atoms in the cluster. Setting $\alpha = Z(V - V_e)/V$, where V_e and V are the volumes of the electron and ion spheres, respectively, and using (8) yields

$$\frac{\alpha}{Z} = 1 - \left(1 - \sqrt{\frac{I}{I_C}}\right)^3. \qquad (9)$$

As an example, fig. 3 shows the ionization degree as a function of the laser intensity for a cluster with the parameters $R = 3.2\,\mathrm{nm}$ and $n_e = 10^{23}\,\mathrm{cm}^{-3}$. For these parameters $I_C = 4.9 \cdot 10^{17}\,\mathrm{W/cm}^2$, which is about 3 orders of magnitude larger than typical atomic ionization thresholds. The model proves in good agreement with our simulations and previous analytic work [2]. An even better quantitative agreement can be obtained by slightly generalizing eqs. (6) and (7) in the form $\eta_1 \xi_{max}/R = \mathcal{E}_0/E_C$ and $\eta_2 R_e + \xi_{max} = R$ with fitting parameters η_1 and η_2. The parameter η_1 adjusts the effective displacement ξ_{max}, the parameter η_2 the effective radius R_e. The present simulation results can be accurately fitted by setting $\eta_1 = 1.1$ and $\eta_2 = 0.97$.

A significant fraction of the energy absorbed by the cluster can be attributed to the change of the potential energy $U = U_f - U_i$. The initial energy $-U_i$ is the ionization energy for inner ionization. It scales linearly with the number N_e of electrons. The final energy U_f is the ionization energy for outer ionization. It consists of a sum over the Coulomb energies of about N_e^2 particle pairs at an average distance of the order of $N_e^{1/3}$ and scales approximately as $N_e^{5/3}$. Since $U_f/U_i \propto N_e^{2/3}$, the ionization energy for outer ionization becomes the dominant contribution for sufficiently large clusters. Neglecting the potential energy of the distant outer electrons it reduces to the potential energy of the ion shell. Ultimately, this electrostatic energy will be transformed into ion kinetic energy by Coulomb explosion. Calculating the field energy due to a uniformly charged shell $R_e < r < R$ one obtains the result

$$\frac{U}{N_e m_e \omega_p^2 D^2} = \frac{1}{40}\left[2 - 5\varrho^3 + 3\varrho^5\right], \tag{10}$$

where $D = 2R$ is the diameter of the ion sphere and N_e the total number of electrons. Combining eqs. (8) and (10), one obtains the potential energy of the ionized cluster as a function of the laser intensity

$$\frac{U}{N_e m_e \omega_p^2 D^2} = \left\{ \begin{array}{l} \frac{1}{40}\frac{I}{I_C}\left[15\left(1 + \frac{I}{I_C}\right)\right. \\ \left. -\sqrt{\frac{I}{I_C}}\left(25 + 3\frac{I}{I_C}\right)\right] \end{array} \right. . \tag{11}$$

This shell model and a corresponding extension with parameters $\eta_1 = 1.1$ and $\eta_2 = 0.97$ proves in excellent agreement with particle simulations. A comparison of results is shown in fig. 4 for the same parameters used in fig. 3. The potential energy in the simulations includes the binding energy $-U_i$. It therefore approaches a slightly larger value at large intensities.

For weakly ionized clusters, $I \ll I_C$, the potential energy is proportional to the energy of the laser field inside the cluster volume $U \to 27\ (I/c)V$. For a completely ionized cluster, $I = I_C$, it reduces to the well-known potential energy of a sphere, $U \to (3/5)\ Q^2/R$. It is also instructive to compare the potential energy per particle with the ponderomotive energy U_p of (5). From the limiting forms one obtains $U/N_eU_p \to 54\ \omega^2/\omega_p^2$ for $I \ll I_C$ and $U/N_eU_p \to (36/5)\ \omega^2/\omega_p^2$ for $I = I_C$. In both limits this fraction is proportional to ω^2/ω_p^2.

From the present study one can draw the following conclusions. Absorption by outer ionization closely correlates with the potential energy gain of the cluster. To a good approximation one can set $W = \gamma U$, where $\gamma \approx 1$–3 is of order unity and U can be simply predicted by the shell model as shown in fig. 3. Outer ionization can account for significant absorption in common cluster gases. The absorption coefficient a can be estimated from the power density $P = aI = \omega n_{Cl} W/(2\pi l)$ absorbed by a cluster gas of density n_{Cl}, where l denotes the number of laser periods required for outer ionization of a single cluster. At the saturation intensity $I = I_C$ the absorption coefficient becomes

$$a = \frac{24\pi}{5}\frac{\gamma}{l}\frac{n_{Cl}R^3}{\lambda}. \tag{12}$$

It basically scales with the wave number and the volume fraction occupied by the clusters in the gas. Assuming values from a previous experiment [3], $n_{Cl} = 4 \cdot 10^{14}\,\mathrm{cm}^{-3}$, $R = 5\,\mathrm{nm}$, $\lambda = 0.5\,\mu\mathrm{m}$ and setting $\gamma = 2$, $l = 4$, one arrives at an absorption length of the order of $L = a^{-1} \approx 1\,\mathrm{mm}$. It will be interesting to demonstrate absorption by outer ionization with intense few-cycle pulses under similar experimental conditions.

In summary, we have studied absorption by outer ionization of clusters. A shell model for inner electrons has been proposed to estimate the ionization degree and the absorbed energy. The model is found in good agreement with particle simulations. An estimate of the absorption coefficient indicates significant absorption for intense few-cycle pulses in clustered gases under realistic experimental conditions.

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