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Ionization dynamics in dense nanoplasmas irradiated by intense laser fields. Pulse shaping

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Abstract
The interaction of nanometer-sized silver clusters with intense laser fields is investigated using a modified nanoplasma model. In particular, the ionization dynamics is considered. The yield of highly charged ionic species can be controlled by pulse shaping. Using a genetic algorithm, optimal pulse shapes for the maximum yield of specific ionic charge states are calculated.

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1. Introduction

The interaction of intense laser radiation with clusters is a field of current interest. Their interaction with electromagnetic fields is very effective because they are objects with initially solid-like atomic density in a nanometer scale size. Thus, absorption of energy is much larger than for the corresponding atomic or bulk systems at the same intensities. Consequently, in laser-cluster experiments the emission of highly charged ions, very energetic electrons, higher harmonics, fast fragments as well as strong x-rays in the multi-keV range is observed. Different theoretical models and simulations indicate that resonant collective absorption plays a central role. The rapid expansion of irradiated clusters is essential as, at a certain time, the cluster reaches the density fulfilling the resonance condition. This can occur during a single pulse. Another method which allows for a better control is the dual-pulse laser excitation with varying time delay between two pulses. Such experiments were performed recently for silver clusters showing a strong dependence of highly charged ion yield as well as of the maximum energy of emitted electrons on the delay time [1]. A further concept to get highly charged ions is the direct control of the laser intensity via pulse shaping [2–4]. In this paper we investigate this concept in the framework of the nanoplasma model [5] and use a genetic algorithm to solve the optimization problem to get a maximum yield of highly charged ions.

2. The nanoplasma model

The nanoplasma model allows us to describe different physical processes like ionization, heating and expansion that occur during the laser–matter interaction on a hydrodynamic level [5]. The clusters are assumed to be initially neutral spheres with uniform temperature and density. The diameter of the spheres should be smaller than the wavelength of the applied laser field. The initial plasma in the cluster is created due to tunnel ionization. Then it will be heated due to the interaction with the laser radiation which is described in the framework of the nanoplasma model by collisional absorption (inverse bremsstrahlung). The heating of the plasma leads to a high pressure in the plasma and consequently to an expansion of the system. For all processes, appropriate approximations have to be found to build up a system of rate equations and hydrodynamic equations in order to simulate the dynamics of the nanoplasma. In the approach presented here, the original model of Ditmire *et al* [5] was modified in some important points. We included the lowering of the ionization energies. This important correlation effect influences essentially the ionization dynamics of the system. It was taken into account using the Stewart–Pyatt shift which interpolates between Debye screening and the ion-sphere model. For further details, see [6, 7].

The heating of the plasma due to collisional absorption can be calculated starting from the balance equation for the electrical current density [8, 9]. For a spherical cluster, it can be written in the following form:

$$\frac{\mathrm{d}}{\mathrm{d}t}\,\mathbf{j}(t) - \frac{\omega_p^2}{4\pi}\,\mathbf{E}^{\mathrm{ext}}(t) + \frac{\omega_p^2}{3}\int_{t_0}^{t} \mathrm{d}\bar{t}\,\mathbf{j}(\bar{t}) = \sum_c \int \frac{\mathrm{d}^3 p_c}{(2\pi\hbar)^3}\,\frac{e_c \mathbf{p}_c}{m_c}\,I_c(\mathbf{p}_c, t) \tag{1}$$

with the plasma frequency $\omega_p^2 = \sum_c \frac{4\pi e_c^2 n_c}{m_c}$ and I_c being the general field-dependent collision integral for species c. The third term on the left-hand side describes the polarization stemming from the mean field contribution. The right-hand side term describes the friction due to collisions. In a Fourier analysis, the corresponding Fourier component can formally be written as a product of a dynamical collision frequency $\nu(\omega)$ and the current $j(\omega)$. Then, the balance equation takes the form

$$-\mathrm{i}\omega\, j(\omega) + \nu(\omega) j(\omega) = \frac{\omega_p^2}{4\pi} E^{\mathrm{int}}(\omega), \tag{2}$$

where $E^{\mathrm{int}}(\omega)$ is the Fourier component of the internal field, which is connected to the external field by

$$E^{\mathrm{int}}(\omega) = \frac{\omega + \mathrm{i}\nu(\omega)}{\left(\omega - \frac{\omega_p^2}{3\omega}\right) + \mathrm{i}\nu(\omega)}\, E^{\mathrm{ext}}(\omega). \tag{3}$$

Assuming harmonic external fields we get, after cycle averaging, the following expression for the heating rate [6, 7]:

$$\frac{\mathrm{d}U}{\mathrm{d}t} = \frac{\omega_p^2\,\mathrm{Re}\,\nu(\omega)}{8\pi}\,\frac{1}{\left[\omega - \frac{\omega_p^2}{3\omega}\right]^2 + [\mathrm{Re}\,\nu(\omega)]^2}\,|E_0|^2. \tag{4}$$

For the collision frequency, we used improved expressions based on quantum statistical theory [8]. Furthermore, an additional damping term corresponding to electron–surface collisions was included [7].

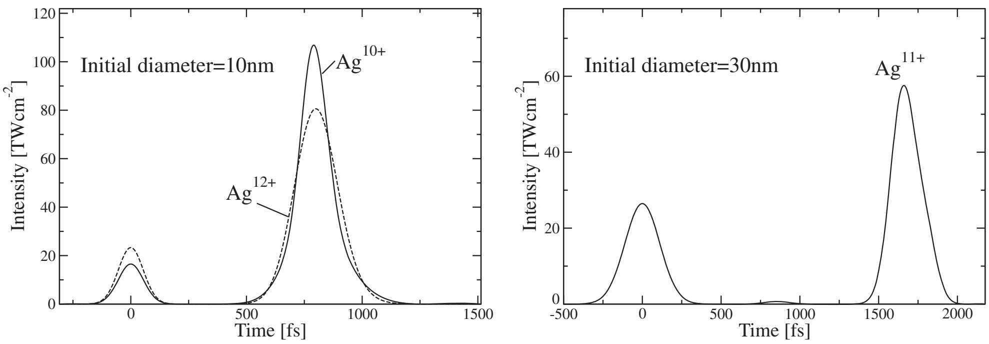

Figure 1. Left: intensity of the laser pulse as a function of time. Solid: optimization for a maximum yield of silver Ag^{10+} ions. Dashed: optimization for Ag^{12+} ions. The initial diameter of the cluster was 10 nm. Right: intensity of the laser pulse as a function of time. Optimization for a maximum yield of Ag^{11+} ions. The initial diameter of the cluster was 30 nm. For both figures, the pulse energy corresponds to a single Gaussian pulse with peak intensity 160 TW cm^{-2} with a duration of 130 fs (FWHM) and $\lambda = 810$ nm.

3. Pulse shaping

Pulse shaping is a modern tool in laser experiments [2–4]. Of special interest is the interaction of femtosecond laser pulses with clusters. With pulse shaping, the dynamics of the system determined by heating, ionization and expansion can be specifically affected. In particular, the yield of highly charged ionic species can be controlled by pulse shaping. For an understanding of the underlying physical processes in the dynamics of laser cluster interaction, a theoretical description using a genetic algorithm and basing on the relatively simple nanoplasma was used. To optimize the yield of a specific ion species or a group of them, we used the following ansatz for the laser pulse intensity:

$$I(t) = \sum_{n=0}^{4} A_n \exp\left[-\frac{4\ln 2(t-\delta t_n)^2}{\sigma_n^2}\right] \tag{5}$$

with σ_n being the full width at half maximum (FWHM) and $\delta t_0 = 0$. Here, one has to account for the constraint

$$\int_{-\infty}^{\infty} I(t)\,\mathrm{d}t = \text{const} \tag{6}$$

for all A_n, σ_n and δt_n to get comparable results. We implemented a genetic algorithm to calculate the parameters for a maximum yield of a specific ionic species. We considered silver clusters with different initial diameters. The wavelength was chosen to be 810 nm. The energy impact, which was constant for all pulse shapes, corresponds to a Gaussian pulse with a width of 130 fs and a peak-intensity of 160 TW cm^{-2}.

4. Numerical results

To calculate the optimal pulse shape, we used a genetic algorithm with about 20 different pools each with a population of 40 species. The following results are related to the pulse shape with the greatest amount of Ag^{10+}–Ag^{12+} ions for different sizes of the irradiated clusters. Figure 1 (left) shows the intensity of the optimized pulse as a function of time for a silver cluster

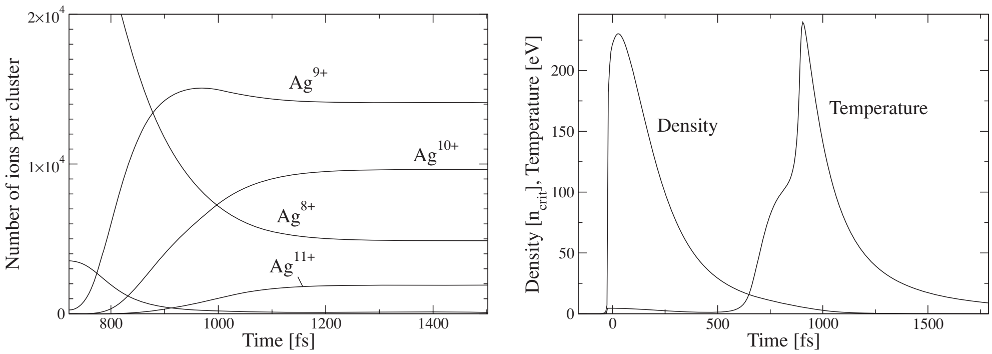

Figure 2. Left: plasma composition of a silver cluster with an initial diameter of 10 nm irradiated by laser pulses optimized to get a maximum yield of Ag^{10+}. Right: electron density and temperature as a function of time. For pulse parameters, see figure 1.

with an initial diameter of 10 nm. The solid line corresponds to an optimization of the Ag^{10+} yield. The dashed line is the result for the Ag^{12+} optimization. Similar to the experimental results [10] there is a double-peak structure with a first pulse of lower intensity followed by the main pulse with higher intensity. Figure 1 (right) shows the laser intensity of an optimized pulse for a bigger cluster with an initial diameter of 30 nm. The shape is similar to the shape for the smaller cluster but the delay time is longer. This can be understood taking into account that the ionization and expansion dynamics in the clusters are functions of the initial size [11]. Smaller clusters expand faster than bigger ones. The resulting plasma composition for the highly charged ions for a 10 nm cluster as a function of time is shown in figure 2 (left). It can be seen that the resulting plasma composition is build up in the trailing edge of the main pulse ($t > 800$ fs). The composition freezes at $t > 1200$ fs to the final composition. For the chosen laser parameters the dominant species are nine- and ten-fold ionized. This strongly peaked distribution of ionization is due to the mono-sized clusters we consider here. In the framework of the nanoplasma model it is possible to calculate the plasma parameters time-resolved. In figure 2 (right) the electron density and temperature of a 10 nm silver cluster irradiated by the optimized laser pulse are shown as a function of time. The first pulse produces a very dense plasma with a density of about 250 times the critical density $n_{\mathrm{crit}} = 1.7 \times 10^{21}$ cm^{-3}. The temperature is about 5 eV. After this first pulse the cluster expands with a rapid decrease of density and temperature. In the vicinity of the Mie resonance [5] ($n_e = 3n_{\mathrm{crit}}$) the second pulse leads to a resonant energy input into the plasma. The temperature rises up to 240 eV. At this stage, the highly charged ions are produced (figure 2 left).

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