

SUPERINTENSE LASER FIELDS

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Relativistic effects of the interaction of an intense femtosecond laser pulse with atomic clusters

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***Abstract*. The effects of the interaction of an intense femtosecond laser pulse with large atomic clusters are considered. The pulse intensity is of the order of 10^{18} W cm^{-2}. New effects appear when the magnetic component of the Lorentz force is taken into account. The second harmonic of laser radiation is generated. The second harmonic generation (SHG) efficiency is proportional to the square of the number of atoms in a cluster and the square of the laser radiation intensity. The resonance increase in the SHG efficiency at the Mie frequencies (both at the second-harmonic frequency and fundamental frequency) proved to be insignificant because of a fast passage through the resonance during the cluster expansion. The mechanisms of the expansion and accumulation of energy by electrons and ions in the cluster are discussed in detail. The energy accumulation by electrons mainly occurs due to stimulated inverse bremsstrahlung upon elastic reflection of the electrons from the cluster surface. The equations describing the cluster expansion take into account both the hydrodynamic pressure of heated electrons and the Coulomb explosion of the ionised cluster caused by outer shell ionisation. It is assumed that both inner shell and outer shell ionisation is described by the over-barrier mechanism. It is shown that atomic clusters are more attractive for generation of even harmonics than compared to solid and gas targets.**

Keywords: *femtosecond pulses, superintense radiation, atomic clusters, relativistic interaction, second harmonic.*

1. Introduction

The interaction of an intense laser pulse of frequency ω with gases and plasma of solids is accompanied by the generation of high harmonics. A rarefied atomic gas generates harmonics with frequencies $n\omega$, whose intensity slowly decreases down to the break point at $n\omega = 3.17F^2/(4\omega^2)$ [1], where F is the laser field strength amplitude. We use hereafter the atomic system of units ($m = e = \hbar = 1$). However, the harmonic generation efficiency is low because the gas is rarefied. In the case of dense solid-state plasma, the harmonic generation efficiency is low due to a strong reflection of the incident laser beam.

Atomic clusters are attractive for the generation of harmonics because, on the one hand, they have the density of a solid and, on the other, their size is small compared to the laser radiation wavelength even when a cluster contains millions of atoms. Therefore, a skin layer is not formed inside the cluster, and the electric field is homogeneous. Because of a rapid expansion of the cluster, the cluster plasma in the leading edge of a short laser pulse rapidly becomes subcritical, i.e., transparent for laser radiation. At the same time, the collective properties of the plasma are preserved during a femtosecond laser pulse, and the efficiency of dipole emission of harmonics is proportional to the square of the number of particles in the cluster rather than to its first power, as in a rarefied gas. However, this advantage of clusters is somewhat reduced because the distance between adjacent clusters in a typical cluster beam used in experiments amounts to 5–10 of cluster diameters.

In the nonrelativistic limit, only odd harmonics are emitted. The intensity of the third harmonic emitted by atomic clusters was studied in detail in the model of a nonlinear oscillator for stimulated oscillations of the electronic cloud of the cluster [2]. This intensity increases at the instant when the third-harmonic frequency coincides with the Mie frequency $\omega_{\text{Mie}} = \omega_{\text{p}}/\sqrt{3}$, which is the eigenfrequency of the electronic cloud of a spherical cluster, whose shape does not change during its expansion (surface plasmon).

The even harmonics appear only at intensities above 10^{18} W cm^{-2} due to the relativistic interaction of the field with cluster particles. When solid targets are irradiated, these harmonics can also appear at the intensity above 10^{17} W cm^{-2} due to the plasma density gradient caused by the axial pondermotive force of the laser pulse [3].

Second harmonic generation (SHG) due to quadrupole surface plasma oscillations was discussed in [4]. Its intensity is low because of the additional small parameter $(R/\lambda)^2$ of expansion in multipoles, where R is the cluster radius and λ is the laser radiation wavelength.

Even and odd harmonics generated in the subcritical relativistic plasma due to elastic collisions of electrons with atomic ions in the laser field were considered in [5]. However, their yield decreases with increasing intensity due to a decrease in the frequency of electron–ion collisions. A model describing SHG upon the relativistic interaction of a laser pulse with a subcritical plasma was considered recently in [6]. The second harmonic was studied

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in reflected light. It was shown that the SHG efficiency was saturated at high laser intensities.

In [7], we considered the generation of relativistic harmonics upon irradiation of atomic clusters by a superintense laser pulse, which was caused by elastic electron – ion collisions in the laser field. The generation efficiency of these harmonics is proportional to the number N_e of free electrons inside a cluster because during random collisions with atomic ions each of the ions emits photons irrespective of other electrons.

In this paper, we propose a simple model of the coherent SHG upon the interaction of a moderate intense $[F/(\omega c) < 1]$ linearly polarised laser pulse with large atomic clusters. The second harmonic is induced by the magnetic component of the Lorentz force.

2. Interaction of superintense laser radiation with atomic clusters

Due to the cluster expansion during a laser pulse, the electron concentration n_e, the plasma frequency ω_p, and the Mie frequency $\omega_{\text{Mie}} = \omega_p/\sqrt{3}$ decrease with time. On the other hand, all these quantities increase due to multiple inner shell ionisation of atomic ions by the superintense laser pulse. The Coulomb explosion of the cluster is manly caused by its outer shell ionisation. The mechanism of laser energy absorption depends on the oscillation amplitude of an electron in the laser field. When this amplitude is smaller than or comparable to the cluster size, the laser energy is mainly accumulated due to elastic reflections of electrons from the cluster surface. In the opposite limit, when the dynamics of the electron motion is determined by its thermal rather than the vibrational velocity, the mechanism of inverse bremsstrahlung dominates. In both cases, the collision ionisation of electrons with atomic ions is insignificant compared to field ionisation.

Both the inner- and outer shell ionisation of clusters by the field of a short superintense laser pulse is determined by the over-barrier ionisation. Due to outer shell ionisation, a static Coulomb electric field is produced inside an ionised cluster. Let us find the charge Q accumulated in the cluster by using the Bethe rule (as mentioned above, we use atomic units everywhere)

$$F_{\text{in}} = \frac{I_Q^2}{4Q}. \tag{1}$$

Here, $I_Q = Q/R$ is the ionisation potential of the cluster; F_{in} is the electric field strength inside the cluster (taking into account the Gaussian time envelope of the laser pulse). Thus, due to the outer shell ionisation of the cluster at the leading edge of the laser pulse, the cluster acquires the charge

$$Q = 4F_{\text{in}}R^2. \tag{2}$$

This charge increases with time at the leading edge of the laser pulse and remains constant at the trailing edge because the escaping electrons do not return back.

The inner shell ionisation of atoms in the cluster produced by superintense fields of femtosecond laser pulses also occurs according to the Bethe rule, which in this case has the form

$$F_{\text{in}} = \frac{E_Z^2}{4Z}. \tag{3}$$

Here, E_Z is the ionisation potential of an atomic ion and Z is the degree of ionisation. Let P_Z be a total number of ions with the degree of ionisation Z in the cluster. Then,

$$Q = \sum_Z ZP_Z - N_e. \tag{4}$$

The probability of outer shell ionisation per unit time is described by the expression $W_{\text{fs}} = \delta(t - t_0)$. The parameter t_0 is determined by using the Bethe rule (1). Similarly, the probability of inner shell ionisation can be described by the expression $\Gamma_Z = \delta(t - t_0^Z)$. Here, the quantity t_0^Z is determined from expression (3) for the proper value of Z. Finally, the balance equations have the form

$$\begin{aligned} \frac{\partial N_Z}{\partial t} &= -\Gamma_Z N_Z + \Gamma_{Z-1}N_{Z-1}, \\ \frac{\partial N_e}{\partial t} &= \sum_Z \Gamma_Z N_Z - W_{\text{fs}}N_e, \end{aligned} \tag{5}$$

where N_Z is the number of ions with the degree of ionisation Z.

The Drude formula for the permittivity of free electrons in classical electrodynamics has the form

$$\varepsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i\nu)}.$$

Here, ν is the frequency of electronic collisions resulting in the decay of electron cloud oscillations. The expression relating the electric field strengths outside and inside the cluster can be easily obtained in the form

$$|F_{\text{in}}| = \frac{\omega^2}{\left[(\omega_{\text{Mie}}^2 - \omega^2)^2 + (\nu\omega)^2\right]^{1/2}}F.$$

Note that this expression uses the electric field strength envelopes as functions of time.

Let us assume that the charge accumulated in the cluster is localised on its surface, i.e., that radius of the ion sphere is larger than the radius of the cloud of free electrons localised inside the cluster. Then, the Coulomb energy accumulated in the ionised cluster is $E_Q = Q^2/(2R)$. The variation of this expression during cluster expansion has the form

$$dE_Q = -\frac{Q^2}{2R^2}dR = -p_Q dV = -p_Q d\left(\frac{4\pi}{3}R^3\right).$$

Here, V is the cluster volume and p_Q is the Coulomb pressure. Therefore, we obtain

$$p_Q = \frac{Q^2}{8\pi R^4} = \frac{2F_{\text{in}}^2}{\pi} = \frac{2\omega^4F^2}{\pi\left[(\omega_{\text{Mie}}^2 - \omega^2)^2 + (\nu\omega)^2\right]}. \tag{6}$$

The equilibrium hydrodynamic pressure p_e of the electron gas is described by the expression

$$p_e = \frac{2E}{3V}, \tag{7}$$

where E is the total energy of the cluster.

The hydrodynamic equation of the cluster surface motion taking into account the Coulomb and hydrodynamic pressures of the electron gas can be written in the form

$$\frac{d^2R}{dt^2} = \frac{3(p_Q + p_e)}{M_i n_i R}.$$

Here, $n_i(t)$ is the concentration of atomic ions inside the cluster and M_i is the ion mass. Finally, we obtain the equation

$$M_i N_a \frac{3}{4\pi R^2}\frac{d^2R}{dt^2} = 3p_e + \frac{6\omega^4 F^2}{\pi\left[\left(\omega^2_{\text{Mie}} - \omega^2\right)^2 + (\nu\omega)^2\right]}, \quad (8)$$

where $N_a = \text{const}$ is the number of atoms in the cluster. This equation can be rewritten in the form

$$\frac{d^2R}{dt^2} = \frac{2E}{M_i N_a R} + \frac{8\pi\omega^4 F^2 R^2}{\pi M_i N_a\left[\left(\omega^2_{\text{Mie}} - \omega^2\right)^2 + (\nu\omega)^2\right]}. \quad (9)$$

3. Heating and cooling of a cluster in the presence of superintense laser radiation

The laser energy is absorbed first of all by electrons inside the cluster. Then, this energy is transformed into the kinetic energy of atomic ions. The cluster expansion is a non-equilibrium process caused by high expansion velocities. Because the increasing pressure of the electron gas is not compensated by the environment, it is impossible to use the known equilibrium statistic expression for the differential $p_e dV$ of the work spent for the cluster expansion for calculating the interaction of radiation with the expanding cluster in this problem. Instead, we will calculate the differential of the kinetic energy of atomic ions in the expanding cluster. We assume, as in the previous section, that ions are uniformly distributed inside the cluster and, therefore, the velocity $\upsilon(r)$ of the radial motion of atomic ions is a linear function of the radius r:

$$\upsilon(r) = \frac{dR}{dt}\frac{r}{R}.$$

The kinetic energy of atomic ions in a spherical layer of thickness dr is described by the expression

$$dE_k = 4\pi r^2 \frac{n_i M_i \upsilon^2(r)}{2}\, dr.$$

By integrating this expression with respect to r, we obtain

$$E_k = \int_0^R 4\pi r^2 \frac{n_i M_i \upsilon^2(r)}{2}\, dr = \frac{3}{10} N_a M_i \left(\frac{dR}{dt}\right)^2.$$

The time derivative of the latter expression gives the work performed per unit time upon expansion:

$$\frac{dE_k}{dt} = \frac{3}{5} N_a M_i \frac{dR}{dt}\frac{d^2R}{dt^2}. \quad (10)$$

The cluster heating is determined from the energy balance equation

$$\frac{dE}{dt} = (N_e - Q)\frac{F^2_{\text{in}}}{2\omega^2}\nu - \frac{3}{5} N_a M_i \frac{dR}{dt}\frac{d^2R}{dt^2}. \quad (11)$$

Here, $E(t)$ is the total energy of electrons inside the cluster as a function of time t.

Thus, we have the system of two equations

$$\begin{aligned} \frac{d^2R}{dt^2} &= \frac{2E}{M_i N_a R} + \frac{8\pi\omega^4 F^2 R^2}{\pi M_i N_a\left[\left(\omega^2_{\text{Mie}} - \omega^2\right)^2 + (\nu\omega)^2\right]}, \\ \frac{dE}{dt} &= \frac{(N_e - Q)\omega^2 F^2 [2E/(N_e - Q)]^{1/2}}{2R\left\{\left(\omega^2_{\text{Mie}} - \omega^2\right)^2 + [2E/(N_e - Q)](\omega/R)^2\right\}} \\ &\quad - \frac{3}{5} N_a M_i \frac{dR}{dt}\frac{d^2R}{dt^2}. \end{aligned} \quad (12)$$

This system should be solved with the initial conditions of the type

$$R(-\infty) = R_0, \ \frac{dR}{dt}(-\infty) = 0,$$

$$E(-\infty) = 0.$$

The Mie frequency is determined by the expression $\omega^2_{\text{Mie}} = (N_e - Q)R^{-3}$, and the field inside the cluster by the expression

$$F_{\text{in}} = \frac{\omega^2}{\left\{\left(\omega^2_{\text{Mie}} - \omega^2\right)^2 + [2E/(N_e - Q)](\omega/R)^2\right\}^{1/2}} F = \frac{E_Z^2}{4Z}. \quad (13)$$

4. Second harmonic generation upon irradiation of clusters by relativistic laser pulses

Stimulated oscillations of free electrons inside the irradiated cluster along the laser pulse propagation (along the z axis) are described by the Newton equation with the magnetic component of the Lorentz force, which for the coordinate z of the centre of mass of a spherical electron cloud has the form

$$\frac{d^2z}{dt^2} + \nu\frac{dz}{dt} + \omega^2_{\text{Mie}} z = -\frac{1}{c}\frac{dx}{dt} H\cos\omega t. \quad (14)$$

Here, $\omega_{\text{Mie}} = \omega_p/\sqrt{3}$; H is the magnetic field strength in a light wave; $\omega_p = (4\pi n_e)^{1/2}$; and $\nu \ll \omega$. We neglect relativistic corrections in the definition of the plasma frequency and assume that a weakly relativistic regime takes place $[F/(\omega c) < 1]$. The amplitude of the external magnetic field is $H = F$, where F is the amplitude of the external linearly polarised electric field directed along the x axis. The right-hand side of Eqn (14) is the magnetic component of the Lorentz force directed along the laser pulse propagation direction.

The electron movement along the electric field (along the x axis) is described by the expression

$$\frac{d^2x}{dt^2} + \nu\frac{dx}{dt} + \omega^2_{\text{Mie}} x = -F\cos\omega t.$$

For the oscillating motion, we have

$$\frac{dx}{dt} = -F\omega\frac{\sin(\omega t + \varphi)}{\left[\left(\omega^2_{\text{Mie}} - \omega^2\right)^2 + (\nu\omega)^2\right]^{1/2}}, \quad (15)$$

where $\tan\varphi = \nu\omega/(\omega^2 - \omega_{\rm Mie}^2)$. A linear approximation of the electron cloud oscillations is valid until $F/\omega_{\rm Mie}^2 \ll R$.

By substituting Eqn (15) into (14), we obtain

$$\frac{{\rm d}^2 z}{{\rm d}t^2} + \nu\frac{{\rm d}z}{{\rm d}t} + \omega_{\rm Mie}^2 z = \frac{F^2\omega}{2c}\,\frac{\sin(2\omega t + \varphi)}{\left[\left(\omega_{\rm Mie}^2 - \omega^2\right)^2 + (\nu\omega)^2\right]^{1/2}}. \quad (16)$$

The right-hand side of Eqn (16) is the effective electric field directed along the laser pulse propagation direction. One can see that the frequency of this field is 2ω. Here, we neglect the constant component of the electric field, which induces the constant dipole moment. Equation (16) can be written in the form

$$z = -\frac{F^2\omega}{2c}\times\frac{\sin(2\omega t + \varphi + \chi)}{\left\{\left[\left(\omega_{\rm Mie}^2 - 4\omega^2\right)^2 + (2\nu\omega)^2\right]\left[\left(\omega_{\rm Mie}^2 - \omega^2\right)^2 + (\nu\omega)^2\right]\right\}^{1/2}}. \quad (17)$$

Here, $\tan\chi = 2\nu\omega/(4\omega^2 - \omega_{\rm Mie}^2)$. The linear approximation is valid when the condition $z \ll R$ is fulfilled. This condition is weaker than the condition $x \ll R$.

The dipole moment along the laser beam direction is $d_\parallel = -N_{\rm e} z$, where

$$N_{\rm e} = n_{\rm e}\frac{4\pi R^3}{3} = \omega_{\rm Mie}^2 R^3. \quad (18)$$

The second harmonic power emitted by one cluster integrated over angles is

$$P_2 = \frac{4\langle|\ddot{d}_\parallel|\rangle}{3c^3}. \quad (19)$$

By substituting expression (17) into (19), we obtain

$$P_2 = \frac{8R^6F^4\omega^6}{3c^5}\times\frac{\omega_{\rm Mie}^4}{\left[\left(\omega_{\rm Mie}^2 - 4\omega^2\right)^2 + (2\nu\omega)^2\right]\left[\left(\omega_{\rm Mie}^2 - \omega^2\right)^2 + (\nu\omega)^2\right]}. \quad (20)$$

One can see that P_2 is a function of time because $F = F(t)$ and $n_{\rm e} = n_{\rm e}(t)$.

The second harmonic is mainly emitted perpendicular to the laser beam direction. The dipole power emitted by a cluster in the fundamental (first) harmonic integrated over all angles is

$$P_1 = \frac{2F^2R^6}{3c^3}\,\frac{\omega^4\omega_{\rm Mie}^4}{\left(\omega_{\rm Mie}^2 - \omega^2\right)^2 + (\nu\omega)^2}. \quad (21)$$

The power ratio of the second and first harmonics for the Mie resonance ($\omega_{\rm Mie} = \omega$) is proportional to the dimensionless relativistic parameter

$$\frac{P_2}{P_1} = \left(\frac{2F}{3\omega_{\rm Mie}c}\right)^2. \quad (22)$$

5. Decay mechanisms

Let us now elucidate the meaning of the collision frequency ν. The first of the possible decay mechanisms is inverse bremsstrahlung. In this case, ν is the electron–ion collision frequency. However, because this frequency is proportional to F^{-3} in the case of superintense laser pulses, the relativistic electrons inside a cluster do not virtually collide with ions.

In the relativistic case, the decay appears due to elastic reflections of electrons from the cluster walls. In the presence of a laser field, an electron absorbs in each collision the energy equal to $F_{\rm in}^2/(2\omega^2)$. Because the average collision frequency is determined by the electron motion inside the cluster, we obtain the estimate

$$\nu \sim \frac{(2E_{\rm e})^{1/2}}{R} = \frac{1}{R}\left(\frac{2E}{N_{\rm e} - Q}\right)^{1/2},$$

where $E_{\rm e}$ is the average energy of electrons inside the ionised cluster.

Note the interaction of laser radiation with clusters was described similarly in paper [8], but for lower laser radiation intensities and, hence, for different ionisation and decay mechanisms.

6. Numerical simulation

Let us select the parameters of a numerical experiment. We will assume that the intensity of laser radiation is 10^{18} W cm^{-2}. Clusters consist of krypton atoms. The ionisation potentials of krypton ions as functions of the electric field inside the cluster are presented in Fig. 1.

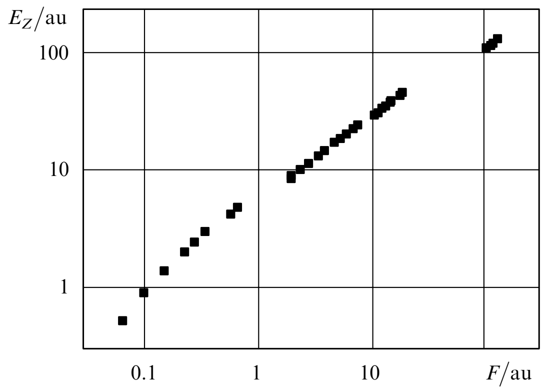

Figure 1. Inner-shell ionisation potentials of atomic krypton ions in a cluster for a specified electric field strength F according to the Bethe rule.

The laser photon energy was taken to be 0.057 au ($\hbar\omega = 1.5$ eV). An external electromagnetic pulse of duration $\tau = 60$ fs has a Gaussian shape. We assume for simplicity of calculations that the initial electron energy is zero. Indeed, the ionisation of atoms by a light wave with the superatomic strength occurs according the Bethe rule (see section 2). Therefore, an electron leaves an atom (or a multiply charge ion) virtually immediately after the electric field inside the cluster achieves a certain value. For this reason, the escaping electron will have a minimal energy. We will assume that ions at the initial instant are singly ionised. This simplification eliminates the difficulties involved in the simulation of a prepulse.

Figures 2–6 present the results of calculations performed for the initial radii $R_0 = 5$, 10, 30, and 50 nm. Figure 2 shows the time dependence of the cluster radius.

We considered above in our model only one cluster. However, in fact the expansion of clusters in laser fields should be studied taking into account the presence of other clusters. When the cluster radius increases by several times, it is necessary to consider other clusters as well. One can see from Fig. 2 that clusters with small initial radii expand faster. Therefore, the region of applicability of our model for them narrows down. Note also that for clusters with initial radii 5 and 10 nm, our model becomes invalid before the maximum laser radiation intensity is achieved.

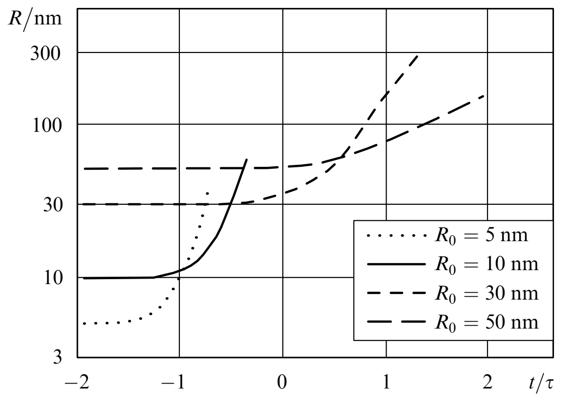

Figure 2. Time dependences of the cluster radius R for different initial radii R_0.

Figure 3 shows the time dependence of the total energy of cluster particles. The total energy increases with time at the leading edge of a light wave. This means that laser heating of the cluster occurs. However, as the cluster radius increases, losses of the cluster energy appear, which is also demonstrated in Fig. 3. The time dependence of the energy also characterises the applicability of the model; however, this estimate is quite rough. A strong decrease in the energy due to cluster expansion was obtained by neglecting the interaction with adjacent clusters. Roughly speaking, this estimate allows one to determine the instant when the interaction between charged particles in the cluster can be neglected. This gives the upper bound of the applicability of our model. Under the conditions of applicability of the model (Fig. 3), the energies for different initial radii are of the same order of magnitude.

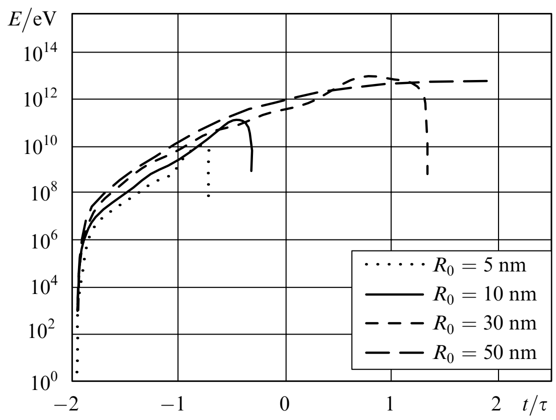

Figure 3. Time dependences of the total energy E of electrons in a cluster for different R_0.

Consider the time dependence of the second harmonic power (Fig. 4). This power increases with increasing the initial cluster radius. A complex jump-wise structure of this dependence is explained by the cluster ionisation. The ionisation is confirmed by the time dependences of the number of electrons in the cluster presented in Fig. 5. Jumps in Figs 4 and 5 occur simultaneously. The final distributions of ions over the degree of ionisation are presented in Fig. 6. One can see that, as the initial radius increases, the average degree of ionisation also increases (recall that the initial condition for all clusters is single ionisation). Such a result does not contradict to a common sense because clusters with small initial radii fly apart before the laser field intensity achieves its maximum.

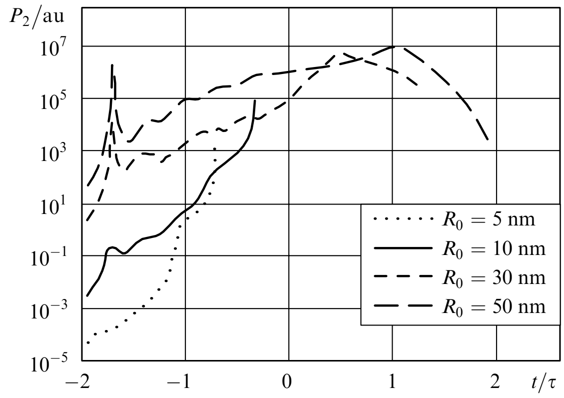

Figure 4. Time dependences of the second harmonic power for different R_0.

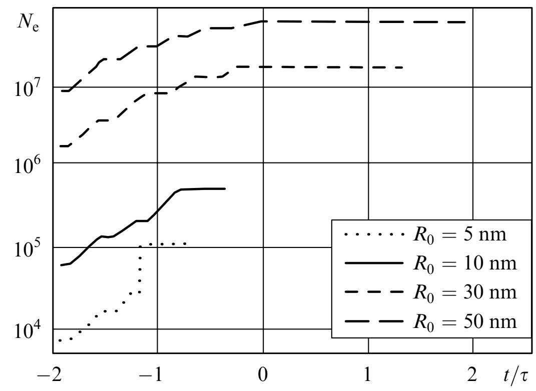

Figure 5. Time dependences of the total number of electrons inside a cluster for different R_0.

Consider now Figs 7 and 8. The dependences presented in them demonstrate the contribution of pressures p_Q and p_e to the results of calculations. The pressure p_Q, i.e., the Coulomb pressure plays the main role. The electron gas pressure p_e only introduces a substantial correction to the calculation. In particular, it is the pressure p_e that causes a drastic decrease in the energy (Figs 3 and 8). Therefore, the contributions from both pressures should be taken into account in calculations.

7. Conclusions

The model of macroscopic collective (coherent) oscillations of the electron cloud along the laser beam direction

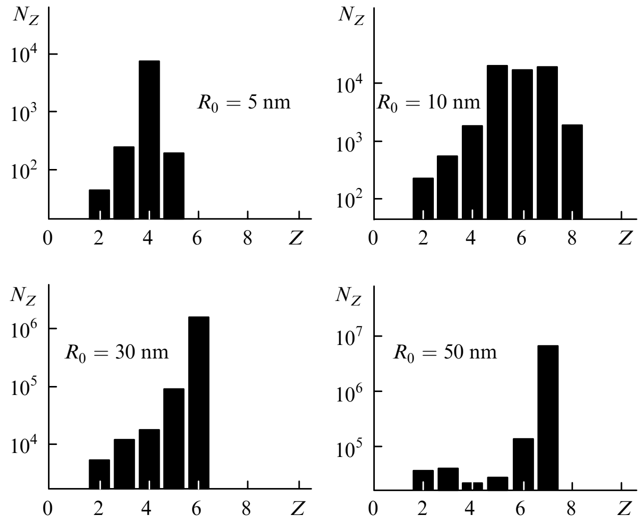

Figure 6. Histograms of the distribution of the number N_Z of multiply charged atomic ions over the degree of ionisation at the end of calculations for different R_0.

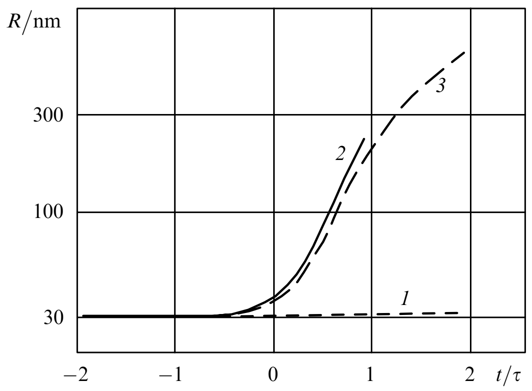

Figure 7. Comparison of the time dependences of the cluster radius R calculated taking into account only p_Q (*1*), p_Q and p_e (*2*), and only p_e (*3*) for $R_0 = 30$ nm.

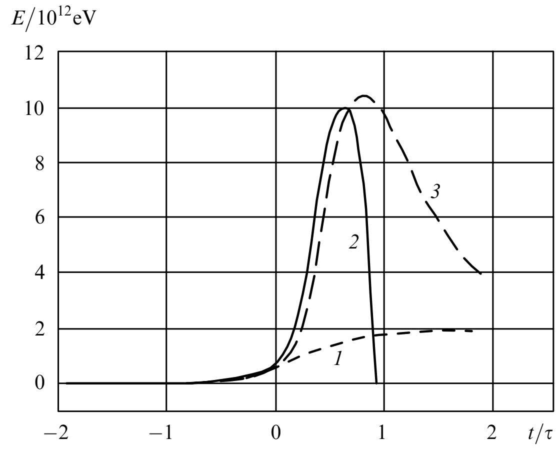

Figure 8. Comparison of the time dependences of the total energy E calculated taking into account only p_Q (*1*), p_Q and p_e (*2*), and only p_e (*3*) for $R_0 = 30$ nm.

proposed in the paper predicts that the harmonic generation efficiency is proportional to N_e^2. The angular distribution of the second harmonic dipole emission is determined by the quantity $\sin^2\theta$, where θ is the angle between the laser pulse direction and the propagation direction of the second harmonic photons.

The generation of even harmonics in a skin layer of the supercritical plasma upon the interaction of laser radiation with dense targets was considered in our paper [9]. The harmonic generation efficiency was determined by the frequency of collisions of electrons with atomic ions inside plasma. This efficiency decreases in a strong laser field with increasing the field intensity [10]. In the case considered in this paper, the second harmonic intensity increases with increasing the laser pulse intensity. However, the role of the Mie resonances at the fundamental and second harmonic frequency proves to be insignificant because of the fast passage through these resonances during cluster expansion.

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