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DISTRIBUTION AND EVOLUTION OF ELECTRONS IN A CLUSTER PLASMA CREATED BY A LASER PULSE

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We analyze the properties and the character of evolution of the electron subsystem of a large cluster (with the number of atoms $n \sim 10^4$–10^6) interacting with a short laser pulse of high intensity (10^{17}–10^{19} W/cm^2). As a result of ionization in a strong laser field, cluster atoms are converted into multicharged ions, a part of forming electrons leaves the cluster, and the other electrons move in a self-consistent field of the charged cluster and the laser wave. It is shown that electron–electron collisions are inessential both during the cluster irradiation by the laser pulse and in the course of cluster expansion; the electron distribution in the cluster does not therefore transform into the Maxwell one even during the cluster expansion. During the cluster expansion, the Coulomb field of a cluster charge acts on cluster ions stronger than the pressure resulting from electron–ion collisions. In addition, bound electrons remain inside the cluster in the course of its expansion, and cluster expansion does not therefore lead to additional cluster ionization.

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1. INTRODUCTION

We consider the character of equilibrium in the plasma formed by irradiation of a cluster beam by a strong laser pulse of the intensity more than 10^{17} W/cm^2, which is studied experimentally [1–3]. This plasma is used both as a source of neutrons produced with a beam of deuterium clusters [4–6] and for generation of X-rays [7–10]. Under typical experimental conditions, the hierarchy of times of the cluster plasma evolution under consideration is as follows. The typical time of the laser pulse duration ($\tau_1 = 30$–100 fs) is small compared to the cluster lifetime with respect to Coulomb explosion ($\tau_{exp} \sim 100$–1000 fs), which is in turn smaller than the typical time of expansion of the forming uniform plasma, which can reach 1 ns. This hierarchy of times determines the behavior of this plasma.

Along with this time hierarchy, the character of laser energy absorption is important for the yield parameters of the plasma. Excitation of clusters by a laser pulse under consideration proceeds through the electron component of this plasma, but this interaction has a specific character due to parameters of the laser radiation. First, these processes occur in strong fields, because the radiation intensity exceeds the atomic field strength, which is $3 \cdot 10^{16}$ W/cm^2, and we consider higher fields that are available under contemporary experimental conditions [11]. Second, the interaction time is very small, and although it exceeds a typical atomic value, light propagates over the distance 10μm within 30 fs. Therefore, in contrast to the classical character of interaction between radiation fields and atomic systems [12–14], other interaction mechanisms are realized in this case [15–19]. Hence, short-time processes occur because of a short time of the laser–cluster interaction, and over-barrier ionization is the main process of absorption of a strong electromagnetic wave. As a result, the laser radiation energy is consumed by ionization of cluster atoms and is transmitted to the electron component of clusters. Below, we consider the character of development of the electron subsystem of clusters under the action of a strong laser pulse.

In the first stage of the ionization process, free electrons in clusters result from over-barrier ionization of cluster atoms [15]. Electrons of zero kinetic energy are formed in this process, and atoms are transformed into multicharged ions that are found in the ground state. Forming electrons move in the laser and cluster fields

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and receive energy from these fields. When electrons go outside the cluster, a positive cluster charge arises, and interaction between the cluster and laser fields leads to the subsequent electron liberation. In addition, collisions of electrons can cause redistribution of the electron energy, and we analyze the role of such processes.

Because the cluster acquires a positive charge under the action of a laser pulse, it expands as a result of the interaction of the cluster field and its ions. In the course of expansion, the electron subsystem can influence the expansion process. In addition, collisions of electrons with multicharged ions that result from ionization of cluster atoms lead to formation of excited multicharged ions; radiation processes involving multicharged ions are responsible for X-ray radiation of this cluster plasma.

Taken together, processes involving the electron subsystem of clusters determine both the cluster ionization rate and the character of energy redistribution of electrons that affects the processes responsible for X-ray emission of the cluster plasma. The goal of this paper is to analyze the character of electron equilibrium in the cluster plasma and the processes involving electrons.

2. IONIZATION OF CLUSTER ATOMS AND CLUSTERS BY A LASER PULSE

2.1. Formation of multicharged ions inside clusters

Under typical conditions of the process under consideration, the photon energy is smaller than the ionization potentials of cluster atoms and ions, and ionization of cluster atoms results from over-barrier transitions of initially bound electrons in the laser wave field. Because of a high electric strength of the electromagnetic wave, this field decreases the barrier from the Coulomb field of the nucleus, and an initially bound electron can freely leave this Coulomb field.

In considering ionization of individual atoms in a cluster by a laser pulse, we assume its action on cluster atoms to be identical to that of a constant electric field. This assumption is valid at small values of the Keldysh parameter [15]

$$\gamma = \frac{\omega\sqrt{2J_Z}}{F},$$

where F is the electric field strength, J_Z is the ionization potential of a multicharged ion with the charge $Z-1$, and ω is the laser frequency (we use atomic units in this paper). Therefore, this character of ionization of cluster atomic particles is valid for high electric field strengths F of the laser electromagnetic wave. Hence, we use the Bethe formula [20] for the strength F of the electromagnetic wave at which the barrier disappears for an electron with the ionization potential J_Z,

$$F = \frac{J_Z^2}{4Z}, \qquad (1)$$

where Z is the charge of a forming atomic ion. This formula implies that the over-barrier transition leads to liberation of electrons whose binding energy is less than J_Z.

This criterion can be represented in another form using the analysis of dynamics of the electron transition from the Coulomb field of the atomic core. Indeed, near the top of the barrier created by the Coulomb field of the atomic core and the constant electric field, we have the Newton equation

$$\frac{d^2r}{dt^2} \approx 2F\frac{r}{r_0} \qquad (2)$$

for a classical electron, where $r_0 \sim \sqrt{Z/F}$ is the distance of the barrier top from the ion center. From this, we find the typical time

$$\tau_{dep} \sim \sqrt{\frac{r_0}{2F}}$$

for the over-barrier electron transition. The requirement that this time is small compared to the period of the electromagnetic wave gives

$$\tau_{dep}^2\omega^2 \sim \frac{2r_0\omega^2}{F} \sim \frac{Z^{1/2}\omega^2}{F^{3/2}} \ll 1\,. \qquad (3)$$

Because of (1), this criterion is identical to the smallness of the Keldysh parameter. This implies that the mechanism of over-barrier ionization of cluster atoms and ions under the action of a strong electromagnetic wave applies at large intensities of the electromagnetic wave. In particular, the right-hand side of criterion (3) gives the approximate value $3\cdot10^{-3}$ for the laser pulse with $I = 10^{17}$ W/cm^2 and frequency 1.5 eV (Ti:sapphire laser).

Charges Z of ions formed by the over-barrier electron transition are given in Table 1. High values of the charge allow us to use a simple formula for the electron binding energy in this case,

$$J_Z = \frac{Z_{ef}^2}{2(n-\delta_{nl})^2}, \qquad (4)$$

where Z_{ef} is the effective charge that includes shielding of the nucleus charge by atomic electrons, such that $Z_{ef} \geq Z$, and δ_{nl} is the quantum defect for a given

Table 1. The charge of cluster atomic ions resulting from cluster irradiation by electromagnetic waves of different intensities at the cluster of 10^6 atoms. The first value is the ion charge at the cluster center; the value in parentheses is the charge at the boundary

	10^{17} W/cm^2	10^{18} W/cm^2	10^{19} W/cm^2
Kr	12(18)	18(26)	24(27)
Xe	11(24)	19(28)	26(43)
Mo	12(14)	14(24)	22(32)
W	12(38)	22(47)	41(56)

electron shell (usually, $\delta_{nl} < 1$). This formula uses the analogy of multicharged ions with hydrogen-like ions.

We consider one more aspect of this problem. Using the analogy between the action of a strong electromagnetic wave and a constant electric field on an atomic particle, we ignore absorption of the electromagnetic wave as a result of electron release. In reality, this absorption follows from excitation of the electron subsystem of an individual cluster. We assume that as the electric field strength of the laser wave $F \cos \omega t$ varies from zero, it leads to the release of new electron groups in accordance with Bethe formula (1). Hence, as a result of the over-barrier ionization process, free electrons are formed inside a cluster with zero energy.

We now estimate the excitation time for an individual cluster of n atoms whose electron subsystem acquires the excitation energy roughly equal to $nJ_Z Z$, where Z is a typical charge of forming multicharged ions. Because the incident energy flux of the laser pulse is $cF^2/8\pi$ and the cluster cross section is equal to its geometric cross section

$$\pi R^2 = \pi r_W^2 n^{2/3}$$

(where r_W is Wigner–Seitze radius and n is the number of atoms in the cluster), we find the typical time τ during which all the cluster atoms re converted into multicharged ions of charge Z,

$$\tau = n^{1/3} \frac{8 J_Z Z}{c r_W^2 F^2}. \quad (5)$$

Substituting the electric field strength F from Eq (1), we obtain

$$\tau\omega = n^{1/3} \frac{128}{c} \frac{\omega Z}{J_Z} \left(\frac{Z/r_W}{J_Z} \right)^2. \quad (6)$$

This value is much smaller than the one for cluster sizes under consideration, and it decreases with the increase of the laser pulse intensity because $J_Z \sim Z^2$. Field ionization in clusters can therefore be considered as an instant process. As a result of this analysis, we find that typical experimental intensities of laser pulses provide fast excitation of the electron component of clusters through ionization of their atoms and ions.

2.2. Ionization of a cluster as a whole

Ionization of a cluster under the action of a strong electromagnetic wave is similar to over-barrier ionization of atoms. An electron passes over the barrier and releases. The difference from the case of an atom is that the electron motion in the cluster field is described by classical laws and the cluster size is restricted. The latter allows a released electron to leave the cluster field. The time of electron displacement by a distance of the order of the cluster radius R is given by

$$t_{esc} \sim \sqrt{\frac{R^3}{Q}} \quad (7)$$

(the typical electron velocity is approximately $\sqrt{Q/R}$, where Q is the cluster charge). The criterion $t \ll 1/\omega$ then becomes

$$R^3 \omega^2 \ll Q\ . \quad (8)$$

The cluster charge is determined by the Bethe formula for the over-barrier transition of an electron located in the Coulomb field of a charge Q and in a constant electric field of a strength F. Taking the interaction energy of the electron with the electric field of the cluster to be Q/R at the cluster boundary, we obtain from (1) that the charge Q of this cluster is given by

$$Q = 4FR^2. \quad (9)$$

When criterion (8) is valid, this gives

$$R \ll \frac{4F}{\omega^2} \quad (10)$$

for the cluster size.

We now consider one more aspect of this problem. The electron remains in the region of the cluster Coulomb field under the action of the electromagnetic wave, but can return at another stage of variation of the electromagnetic wave strength if the electron trajectory is determined strongly by the fields of the cluster and of the electromagnetic wave. Of course, a large statistical weight of continuum spectrum states for this electron makes the electron release favorable. It is important that the Coulomb field of the cluster is not constant in time precisely because it is also created by the motion of bound electrons of the cluster. Fluctuations resulting from the motion of internal cluster electrons lead to randomization of the motion of the transferring electron and make its departure from the Coulomb cluster field irreversible. This determines the applicability of simple formulas for ionization of the cluster as a whole.

Thus, as a result of the ionization processes both inside the cluster atoms and for the cluster as a whole, a specific plasma is formed such that multicharged atomic ions of the cluster keep a part of the electrons moving inside the cluster. These electrons are locked inside the cluster, whereas a part of the electrons releases and creates a cluster charge. Later, this system decays as a result of cluster expansion caused by Coulomb forces acting on ions. But the processes of formation of this plasma determine its properties and the character of subsequent cluster expansion.

We consider one more consequence of cluster charging. Because the charge of cluster ions is not compensated by the electron charge, an additional field arises in the cluster. For simplicity, we use the model where the cluster charge is distributed over the cluster uniformly. Then the electric field of the cluster charge with the strength

$$F_{cl} = \frac{Qr}{R^3} = 4F\frac{r}{R} \qquad (11)$$

acts on an ion located at the distance r from the cluster center. This changes the charges of forming multicharged ions inside the cluster. This problem was examined in [21, 22] in detail. Replacing the electric field strength F of the laser wave in Bethe formula (1) with $F + F_{cl}$, we obtain for the ion charge $Z(r)$ at the distance r from the cluster center that

$$F\left(1 + 4\frac{r}{R}\right) = \frac{J_Z^2}{4Z}. \qquad (12)$$

Thus, this charge is larger near the cluster boundary than at its center. The data in Table 1 pertain to the charge of multicharged ions near the cluster center. We also include the charge near the cluster boundary in this table.

We note that in the above consideration, we assumed that the laser field penetrates inside the cluster. This is a valid assumption because the skin depth for the laser signal is approximately 100 nm in this case and exceeds the cluster size in the range under consideration (a cluster consisting of 10^6Xe atoms has the radius 25 nm [23]).

3. ELECTRON DISTRIBUTION IN THE CLUSTER PLASMA

3.1. Relaxation of electrons in a cluster

The cluster plasma resulting from a laser pulse is expanding. There are two forces acting on the cluster that cause it to expand during and after the laser pulse. The first is the pressure due to electrons. Cluster electrons collide with ions and push them outside. The second force is determined by the Coulomb force and depends on the charge distribution in the cluster, which is the distribution of ions, and electrons.

The energy distribution of electrons established in the cluster during irradiation is far from the equilibrium one. The relaxation rate of the electron subsystem is determined by electron–electron collisions. The relaxation time can therefore be defined as the time during which a test electron gains the energy Q/R in collisions with other electrons,

$$\tau_{ee} \sim \frac{Q/R}{\epsilon \nu_{ee}}, \qquad (13)$$

where ϵ is the electron energy change and ν_{ee} is the electron–electron collision rate. The electron–electron collision rate is given by [24]

$$\nu_{ee} = N_e\sqrt{2\epsilon}\sigma_t \quad \sigma_t = \frac{4\pi}{\epsilon^2}\ln\Lambda, \qquad (14)$$

where $\ln\Lambda$ is the Coulomb logarithm. Substituting Eqs. (14) in Eq. (13), we obtain the relaxation time

$$\tau_{ee} \sim \frac{Q/R\sqrt{\epsilon}}{4\sqrt{2}\pi N_e \ln\Lambda}. \qquad (15)$$

Substituting Eq. (9) in (15), we finally obtain

$$\tau_{ee} \sim \frac{(Q/R)^{3/2} r_W^3}{3\sqrt{2}Z\ln\Lambda} = \frac{8F^{3/2} r_W^{9/2} n^{1/2}}{3\sqrt{2}Z\ln\Lambda}. \qquad (16)$$

We now compare the relaxation electron time τ_{ee} with the expansion time. The typical expansion time is given by

$$\tau_{exp} = \sqrt{\frac{MR^3}{ZQ}} = \frac{1}{2}\sqrt{\frac{MR}{ZF}}, \qquad (17)$$

where M is the ion mass. The ratio of these times is given by

$$\frac{\tau_{ee}}{\tau_{exp}} = \frac{16}{3\sqrt{2}\ln\Lambda}\frac{F^2 r_W^4}{\sqrt{MZ}} n^{1/3} \gg 1. \qquad (18)$$

The ratios τ_{ee}/τ_{exp} for krypton, xenon, molybdenum, and tungsten clusters under typical laser parameters are given in Table 2. Condition (18) is satisfied for all the parameters considered, as follows from the data in Table 2.

Table 2. The ratio of the relaxation time τ_{ee} for the cluster of 10^6 atoms to the expansion time τ_{exp} for different electromagnetic wave intensities

	10^{17} W/cm^2	10^{18} W/cm^2	10^{19} W/cm^2
Kr	53	359	3100
Xe	55	365	3160
Mo	6.1	57	456
W	4.5	33	241

3.2. Electron subsystem during cluster expansion

We now consider the behavior of electrons in the course of cluster expansion. We use the above fact that the expansion time is large compared to the typical time of equilibrium establishment in the electron subsystem. This implies that in contrast to [25], the distribution function of electrons is not the Maxwell one, and collisions between electrons during cluster expansion can be ignored. We therefore start from the electron distribution by energy that results from laser ionization of atoms and analyze the evolution of electrons located inside the cluster. For simplicity, we use the model where the positive charge is distributed uniformly inside the cluster, and the selfconsistent potential of electrons and multicharged cluster ions is therefore given by

$$U(r) = -\frac{Q}{2R}\left(3 - \frac{r^2}{R^2}\right), \tag{19}$$

where r is the distance from the cluster center, and we consider the interior cluster part $r \le R$. This assumption leads to the electron number density inside the cluster

$$N_e(r) = Z(r)N_i - \frac{3Q}{4\pi R^3} \tag{20}$$

(for the charge $Z(r)$ of an individual atomic ion of the cluster, see Table 1). Because ions are distributed in the cluster uniformly before and at the first stage of its expansion, the charge of an individual ion depends on the distance from the cluster center as

$$Z(r) = Z_0\left(1 + \frac{2Q}{FR^2}\frac{r}{R}\right)^{1/3}, \tag{21}$$

which corresponds to Eq. (12).

We now consider the character of evolution of the cluster during its expansion. Because the selfconsistent cluster potential does not change considerably during the oscillation period of a locked electron, its adiabatic invariant [26, 24]

$$I(t,\epsilon) = \frac{1}{2\pi}\int \left[2m(\epsilon - U(r))\right]^{1/2} dr \tag{22}$$

is conserved. The integration is performed between two turning points of the locked electron with a certain energy ε. This quantity plays the role of an integral of motion for the electron. The distribution function of locked electrons is then a function of the adiabatic invariant,

$$f = f(I(\varepsilon, t)). \tag{23}$$

To obtain information about the cluster evolution, we assume the cluster charge Q to be constant during the cluster expansion and assume the cluster potential for an individual electron to be given by Eq. (19) when the cluster radius R depends on time. The adiabatic invariant for an individual electron is then equal to

$$I = \frac{1}{4\Omega^2}\left[\varepsilon - l\Omega\right], \quad \Omega^2 = \frac{Q}{R^3}, \tag{24}$$

where l is the electron orbital momentum that ranges from 0 to $\Omega^2 R$.

We show that the electron orbital momentum is conserved during the cluster expansion. Indeed, the electron momentum varies in electron collisions with ions and electrons, and because these collisions lead to small scattering angles, the typical momentum variation during cluster expansion is estimated as

$$\Delta l \sim l\frac{\tau_{exp}}{\tau_{ei}}, \tag{25}$$

where τ_{ei} is the typical time of electron–ion collisions. We here take into account that electron–ion collisions are more effective than electron–electron collisions because of the large average charge Z of cluster ions, and the cluster expansion rate is determined by interaction of the cluster charge with each ion. This leads to the following estimates for these parameters:

$$\nu_{ei} \sim \frac{1}{\tau_{ei}} \sim \frac{4\pi N_i Z^2 \ln\Lambda}{3v^3} \sim \frac{nZ^2}{l^3}\ln\Lambda, \qquad \tau_{exp} \sim \sqrt{\frac{MR^3}{ZQ}}. \tag{26}$$

We give typical values of expansion times in Table 3, from which it follows that under typical conditions of cluster evolution,

$$\frac{\Delta l}{l} \sim \tau_{exp}\nu_{ei} \sim \sqrt{\frac{M}{Z}}\frac{nZ^2}{Q^2}\ln\Lambda \ll 1. \tag{27}$$

Table 3. Typical expansion times (fs) at some parameters of the interaction between a laser wave and clusters for different electromagnetic wave intensities and different n

	n	10^{17} W/cm^2	10^{18} W/cm^2	10^{19} W/cm^2
Xe	10^4	92	40	19
	10^5	134	58	28
	10^6	196	84	40
W	10^4	79	33	13
	10^5	116	50	20
	10^6	170	73	30

Thus, the above analysis shows that under typical conditions of cluster evolution, the orbital momentum l of an individual electron is conserved in the course of cluster expansion, and the electron orbital momentum l can therefore be considered as an integral of motion.

We now consider the character of motion of an individual electron in the course of cluster expansion, using conservation of the adiabatic invariant and the angular electron momentum. The motion of locked electrons consists in rotation around the cluster center and oscillations in the radial direction between two turning points. Under cluster expansion, the turning points of the electron also move. Using the expression for the energy of an individual electron

$$\varepsilon = \frac{\Omega^2 r^2}{2} + \frac{l^2}{2r^2} \tag{28}$$

and the relation between Ω and the adiabatic invariant I in Eq. (24), we obtain that the distances $r_{1,2}$ of the turning points from the center are given by

$$\frac{r_{1,2}^2}{R(t)^2} = \frac{4I}{R(t)^2} + \frac{l}{\Omega(t)R(t)^2} \pm \pm 2\left[\frac{4I^2}{R(t)^4} + \frac{2Il}{\Omega(t)R(t)^4}\right]^{1/2}, \tag{29}$$

where $R(t)$ is the current cluster size. Recalling the relation between Ω and cluster size in Eq. (24), we obtain that

$$\frac{r_{1,2}}{R(t)} \approx \frac{l}{(QR(t))^{1/4}} \pm \frac{\sqrt{2I}}{R(t)}. \tag{30}$$

Because $R(t)$ increases in time, Eq. (30) implies that the electron trajectory remains inside the cluster in the course of cluster expansion and the relative distance of turning points from the cluster center $r_{1,2}/R(t)$ decreases. Because the second term in Eq. (30) decreases faster than the first one, the motion of a locked electron tends to transform into rotation.

The motion of an electron in the cluster field consists of oscillation in the cluster potential $U(r)$ and rotation around the cluster center characterized by the electron angular momentum l. Equation (30) gives the behavior of turning points of a test electron as the cluster expands. We see that turning points of a test electron move from the cluster center slower than the radius increases. This implies that electrons remain inside the cluster during cluster expansion, and cluster expansion does not therefore lead to an additional cluster ionization.

We now consider this problem from another standpoint, introducing the energy E of a test electron such that the energy on the cluster boundary is zero. Assuming that the electron transition outside the cluster boundary means the electron release, we rewrite Eq. (24) for the adiabatic invariant as

$$E = I\Omega^2 + l\Omega - W, \tag{31}$$

where $W = 3\Omega^2 R^2/2$ is the cluster well depth. We introduce the reduced electron energy

$$\widetilde{E} = \frac{E}{l\Omega} = \left[\frac{I}{R^2} - \frac{3}{2}\right] R^2\Omega + 1. \tag{32}$$

If $\widetilde{E}$ becomes positive, the electron releases. But because $R^2\Omega = \sqrt{QR}$ increases as a result of cluster expansion, an initially negative value $\widetilde{E}$ remains negative, i.e., cluster ionization does not occur as a result of cluster expansion in the absence of collisions involving electrons.

We next consider the character of cluster expansion that proceeds under the action of the Coulomb field of the cluster charge and under the action of electron collisions. The force acting on a test ion from the cluster charge is given by

$$\mathcal{F}_{cl} = \frac{QZ}{R^2}. \tag{33}$$

The force acting on the test ion in collisions with electrons is

$$\mathcal{F}_{el} \sim \nu_{ei} p \sim \frac{4\pi N_i Z^2}{3v^2} \ln\Lambda, \tag{34}$$

where p is the typical electron momentum. Substituting the minimal electron velocity $v = \sqrt{Zn^{1/3}/R}$ in this formula, we obtain an estimate for the force acting on the cluster boundary due to collisions with electrons,

$$\mathcal{F}_{el} \le \frac{3n^{2/3} Z \ln\Lambda}{R^2}. \tag{35}$$

Comparing the above forces, we have

$$\frac{\mathcal{F}_{cl}}{\mathcal{F}_{el}} = \frac{4}{3}\frac{Fr_W^2}{\ln\Lambda} \gg 1. \quad (36)$$

The cluster expansion rate is therefore determined by the Coulomb force that acts on ions from the cluster charge. Hence, the equation for the evolution of the cluster boundary has the form

$$M\frac{d^2R}{dt^2} = \frac{ZQ}{R^2} \quad (37)$$

and its approximate solution is

$$R \approx \sqrt{2\frac{ZQ}{R_0 M}}\,t. \quad (38)$$

A typical time T of the cluster expansion is

$$T = \sqrt{\frac{MR_0^3}{ZQ}}. \quad (39)$$

Typical cluster expansion times are given in Table 3. We see that the cluster expansion time becomes comparable to the pulse duration under the laser intensities of the order 10^{19} W/cm^2. It may affect cluster evolution during irradiation and after it.

4. CONCLUSION

Thus, the analysis of the behavior of an individual cluster in the field of a strong electromagnetic wave shows that electrons locked in the cluster cannot leave it after the laser pulse. Neither electron–electron collisions nor the cluster expansion lead to a considerable additional release of electrons. The pressure created by electron–ion collisions gives a small contribution to the cluster expansion rate. In addition, this analysis demonstrates that under large intensities of the laser pulse, expansion during the irradiation can affect the character of interaction between the laser pulse and the cluster.

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