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On the importance of damping phenomena in clusters irradiated by intense laser fields

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We study the dynamics of large clusters irradiated by intense and short laser pulses within the framework of the nanoplasma model. A particular attention is paid to the influence of electron surface collisions which have not been considered in previous versions of the model. We show that they dominate Inverse Bremsstrahlung Collisions when plasmon resonance occurs. The dynamics of the cluster changes considerably and the predictions of the model are significantly modified. Moreover, there is no evidence for the presence of highly charged ions and the hydrodynamics pressure is found smaller than the Coulomb one.

I. INTRODUCTION

The interaction between intense laser fields and clusters has received a great deal of attention during the last few years [1–5]. The electromagnetic coupling is so strong that absorption rates of nearly 100% can be achieved [6]. The irradiation of clusters by intense laser pulses may thus lead to exceptionally large energy deposits. This, in turn, can be the source of high energy (keV) electrons [3], highly charged and very energetic ions [2] or fragments [4] as well as X rays in the keV range [7–12]. Even applications to fusion have been reported recently [13]. The basic mechanisms at work in these laser irradiations can be summarized as follows. The laser very quickly strips a sizable number of electrons from their parent atoms. These electrons form a reservoir of quasi free electrons, which can strongly couple to the laser when the electron density matches a critical value characteristic of the dipole eigenfrequency of the system (plasma resonance). The global response of the cluster is characterized by a heating of the electron cloud and electronic emission, in addition to the possibly enhanced dipole oscillations. The net charge and the high excitation energy acquired by the cluster lead to its final explosion. It should be noted that this scenario, at least qualitatively, is, to a large extent, independent of the nature of the irradiated clusters. The difference between metal and rare gas clusters, for example (both of which under experimental investigation), essentially lies in the initial amount of free electrons, which, for high laser intensities, turns out not to be a decisive factor. The general scenario we have just outlined nevertheless does not point out precisely which mechanism is at work, during which phase, and with which importance. As a consequence, although the global scheme is generally accepted, the debate on the relative importance of the competing processes (various ionisation mechanisms inside the cluster, role of the plasma resonance, of electron temperature, of net charge...) remains largely open and various models still compete to explain experimental trends. Since these models are usually schematic, neglecting or adding ingredients may have sizable consequences, which have to be analysed in order to better understand the validity of the various models. It is the aim of this paper to analyze the nanoplasma model [14] in this respect.

Several models have been developed to explain the experimental features observed in the "high" laser intensity regime we consider in this paper. In the "coherent electron motion" model proposed by Boyer *et al.* [15], the electrons behave like a quasi-particle that ionises the atoms in the cluster. In the "ionisation ignition" model [16], the combined field of the laser and the cluster ions produces high charge states, and is responsible for the Coulomb explosion of the cluster. The explosion of small size clusters has been studied with classical molecular dynamics [17,18] and a three-dimensional Thomas-Fermi model [19]. Last *et al.* have invoked the multi-electron dissociative ionisation mechanism (MEDI) to explain the high ionisation efficiency in small and large clusters as well [20,21]. Krainov *et al.* have used a simple Bethe model to explore the dynamics of large Xe clusters and found that the electrons absorb only little energy from the field [22]. The nanoplasma model developed by Ditmire *et al.* [14] offers a complete scenario of the interaction taking into account ionisation, heating and explosion processes simultaneously. Contrary to other models, it takes into account the polarisation field radiated by the charges. The cluster is treated as a spherical plasma where plasmon resonance takes place. In the standard version of the model, large electron temperatures are reached and highly charged ions are produced at resonance. The electron gas exerts a strong hydrodynamic pressure which, combined with the Coulomb one, leads to the cluster explosion. Whether the cluster can be treated as a plasma or not has given rise to a controversy. For example, classical dynamics simulations including the Coulomb field of the ions but neglecting the polarisation field indicate that the electrons are quickly removed at the beginning of the interaction even from large clusters [23]. That makes the existence of a nanoplasma questionable. Until now there have not been any time-resolved measurements at the femtosecond scale to settle the question.

In this paper, we study within the framework of the nanoplasma model, the behaviour of the plasma near the plasmon resonance, a key feature of such a model relying on the existence of a quasi free electron gas. This study has been overlooked in previous works. The heating of the cluster at this crucial step has been considered until now as the result of Inverse Bremsstrahlung Collisions (IBC) in which the electrons predominantly absorb the photons from the electromagnetic field when colliding with ions. We show in the first part

that near the resonance, the plasma behaves as a collision-less system when considering the IBC alone. Consequently other damping phenomena have to be considered. These can be electron-electron collisions or the equivalent of Landau fragmentation in finite systems. While rough estimates of the former using static screened cross-sections give a negligible effect, we compute in this paper the latter using a simple approach, modelling this damping by electron surface collisions.

II. NANOPLASMA MODEL AND PLASMON RESONANCE

We review in this section the main features of the standard version of the nanoplasma model and discuss the influence of IBC on plasmon resonance. We consider a cluster in the electric field of an intense linearly polarised laser field $\mathbf{E} = E_{\text{ext}} \cos(\omega t) \mathbf{e}_z$. The dimensions of the cluster are much smaller than the laser wavelength. We assume there are neither temperature nor density gradients and that the electric field inside the cluster is uniform. The cluster quickly turns into a small plasma at the rising edge of the laser pulse as ionisation takes place. The atoms and further produced ions can be ionised by several processes. The laser field first ionises cluster atoms mainly through tunnel ionisation. Once free, these electrons will further ionise atoms/ions through collisions. Due to the high excitations and densities reached inside the cluster, the collisional process is found to be important and produces much higher charge states than optical ionisation alone.

The electromagnetic field inside the cluster \mathbf{E}_{in} is the sum of the incident field and the polarisation field radiated by all the charges in the cluster. It is assumed to be the field inside a dielectric sphere surrounded by a constant field. It then reads $\mathbf{E}' = \mathcal{R}(E_{\text{in}} \exp(i\omega t)) \mathbf{e}_z$ with [11,14]:

$$E_{\text{in}} = \frac{3}{\epsilon + 2} E_{\text{ext}}, \quad (1)$$

where E_{ext} is the amplitude of the external field and ϵ is the plasma dielectric constant. Within the simple Drude model, the latter can be written as:

$$\epsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i\nu)}, \quad (2)$$

where $\omega_p = \sqrt{ne^2/m\epsilon_0}$ is the electronic plasma frequency, n the electron density and ν the electron collision frequency. The polarisation field radiated by the plasma constituents plays an important role in this model. It makes the electric field felt by an atom or an ion in the cluster considerably different from the external field. When the electron density is higher than the critical value $n_c = 3m\epsilon_0\omega^2/e^2$, for which $\omega_p = \sqrt{3}\omega$, the plasma behaves as an electric isolator that prevents (partially) the external field from entering the cluster. Moreover, the electron density is not constant in time inside the cluster. During the time evolution, quasi-free electrons are produced inside the cluster through ionisations and others escape from it. The electrons that escape need to be within one mean free path from the surface of the cluster and their kinetic energy has to exceed the Coulomb potential energy at the cluster surface. At the beginning, electrons leave easily the cluster, but soon a space-charge builds up, retaining the electrons inside the cluster. The electron density increases with time and the electrons absorb energy from the field through collisions. The most energetic electrons can then escape from the cluster and the shielding disappears as the electron density n drops down to n_c . The plasmon resonance occurs for $n = n_c$ for which $|\epsilon + 2|$ passes through a minimum. The electric field is then enhanced and reaches a maximum given by :

$$|E_{\text{in}}(\omega = \omega_p/\sqrt{3})| = \sqrt{1 + \left(\frac{\omega}{\nu}\right)^2} E_{\text{ext}}. \quad (3)$$

On the other hand, the heating of the plasma takes place through inverse Bremsstrahlung collisions. In this process, the density of electromagnetic power deposition $\partial U/\partial t$ is given by (U being the electromagnetic energy density inside the cluster) :

$$\frac{\partial U}{\partial t} = \epsilon_0 \mathbf{E}' \cdot \frac{\partial \mathbf{D}'}{\partial t} \quad (4)$$

with $\mathbf{D}' = \mathcal{R}(\epsilon \mathbf{E}')$. When averaging over a laser cycle, only the imaginary part of the dielectric constant gives a non vanishing contribution to the deposited energy. At resonance ($\omega = \omega_p/\sqrt{3}$), the density of power deposition then takes the simple form (Eqs. 1, 2 and 4):

$$\frac{\partial U}{\partial t} = \frac{3\omega^2}{2c} \frac{I_{\text{ext}}}{\nu} \quad (5)$$

in terms of the intensity I_{ext} of the external field $I_{\text{ext}} = c\epsilon_0 E_{\text{ext}}^2/2$. It appears from relations 3 and 5 that the enhancement of the inner field and the plasma heating rate at resonance strongly depend on the value of the electron collision frequency ν . They may become very large if this frequency drops to a small value at resonance. The enhancement of the heating when the collision frequency tends towards zero seems paradoxical since the imaginary part then vanishes. This behaviour can be explained from the fact that, at resonance, according to equation 3, a large flow of electromagnetic energy enters the cluster and even if the collisions are rare, a large amount of photons can be absorbed at each collision, making energy deposition in the cluster important.

In the standard version of the nanoplasma model, the electrons collide only with ions in the plasma. In this case, energy deposition stems from IBC only. During the collisions, photons are mainly absorbed by the electrons because of the large mass difference between electrons and ions. The electron-ion collision frequency ν_{ei} is known to be inversely proportional to the cube of the electron velocity [14,24] except for the slow varying Coulomb logarithm term. It drops to very small values when the electron velocity increases. In the case of a nanoplasma near plasmon resonance, the laser intensity increases and the quiver velocity $\mathbf{v}_q = e\mathbf{E}_{\text{in}}/m\omega$ of the electrons follows. This reduces the collision frequency and the laser intensity increases further. This process can lead to a very small collision frequency and a very high laser intensity inside the cluster.

The explosion of the cluster begins as soon as the resonance occurs. Once heated the electrons expand and pull the cold (heavy) ions outwards with them. They are responsible for a hydrodynamics pressure $P_h = nk_B T_e$ that scales as $1/R^3$ through the electron density n . The other force acting on the cluster stems from the Coulomb repulsion between ions, due to the net cluster charge. The associated pressure reads :

$$P_c = \frac{3}{32\pi^2\epsilon_0} \frac{Q^2 e^2}{R^4} \quad (6)$$

where Qe is the net cluster charge. As it scales as $1/R^4$, the Coulomb pressure is expected to dominate the hydrodynamics pressure only for small size clusters.

III. RESULTS

A. Pure IBC dynamics

In the following, we consider a Xe cluster with 5000 atoms irradiated by a Gaussian laser pulse with wavelength 780 nm, pulse duration 100 fs (FWHM), and peak intensity $10^{16} \text{ W} \cdot \text{cm}^{-2}$. The nanoplasma equations [14,25–28] are integrated numerically using a fifth order Cash-Karp Runge-Kutta method with adaptive step size control. We take into account cluster heating through IBC and cluster expansion due to both hydrodynamics and Coulomb pressures. The considered ionisation mechanisms are direct optical ionisation through tunnel ionisation and electron-ion inelastic collisions due to both electron thermal agitation and laser driven electrons. Electronic temperature and density are adjusted to account for charge conservation and energy loss due to free streaming of electrons from the cluster. Figure 1 shows the variation of the collision frequency ν as a function of time (dashed line). Time $t = 0$ is the time at which the laser reaches the cluster at maximum intensity. The formation of the nanoplasma occurs at $t \simeq -140$ fs when a sufficiently large number of electrons have been stripped from the atoms by tunnel ionisation. From then on, the electron-ion collision frequency ν_{ei} reaches a value close to 1 fs^{-1} and remains stable until $t \simeq 0$, the time at which cluster dynamics starts changing. The electrons that have been progressively heated begin to leave the cluster making the electron density drop. The latter matches the critical density n_c at time $t \simeq 50$ fs and the plasmon resonance occurs. As predicted from the above simple arguments, the electron-ion frequency drops then to a very low value, of the order of 0.1 ps^{-1} in this case.

Near plasmon resonance, the collision rate is very small and the internal field reaches very high values ($> 10^{14} \text{ V/m}$). In such intense fields, the electrons become relativistic making thus the treatment of the plasmon resonance problematic, which invalidates the standard nanoplasma equations in this region.

B. IBC plus surface damping dynamics

Due to the small values of the electron-ion collision frequency at resonance, other damping phenomena become more and more relevant. Of particular interest are electron collisions with the surface. These can strongly contribute to the dynamics near the plasmon resonance. When electron velocities increase, the electrons quickly reach the surface and may be reflected many times during the resonance duration. This phenomenon leads to the Landau damping of the plasmon [29] and presents strong analogy with wall dissipation of nuclear physics [30]. To put this phenomenon at a quantitative level and in analogy with the case of large metallic clusters described in the Mie regime [31], we choose for the electron-surface collision frequency the expression $\nu_s = v/R$, where R is the radius of the cluster and $v = \sqrt{\mathbf{v}_{\text{th}}^2 + \mathbf{v}_{\text{q}}^2}$ is the effective electron velocity that takes into account both the thermal velocity $v_{\text{th}} = \sqrt{k_{\text{B}}T_e/m}$ and the quiver velocity of the electron in the field (defined above). Looking at the inner surface of the cluster as a scatterer, the frequency can be interpreted as the inverse of the average time it takes an electron to cross the cluster when reflected by the surface. We have also used more involved expressions for this frequency similar to those derived in Ref. [31] without observing sizable changes in our results. The total collision frequency for the electron is now $\nu = \nu_{\text{ei}} + \nu_s$. Close to the resonance, the electron velocity increases while the electron-ion collision frequency decreases. The surface collisions contribution, which is proportional to the electron velocity, becomes dominant and prevents the total collision frequency to drop and the electric field inside the cluster to diverge. Important changes are then expected for the dynamics of the cluster.

The total collision frequency ν (electron-ion plus electron-surface) is also shown in Figure 1. As expected, the decreasing behaviour of ν exhibited near resonance is suppressed when surface collisions are included. Even when the electron velocity increases, the collision frequency stays around few fs^{-1} , in contrast to the case without surface collisions where it drops to the tenth of the ps^{-1} . Note that at the earlier times, electron velocities are small and the total collision frequency coincides with the electron-ion collision frequency.

We show in Figure 2 the external and internal electric fields for both the pure IBC case

(upper panel) and IBC plus surface collisions (lower panel). During the rising edge of the pulse, electrons are produced from the neutral atoms by field ionisation. The rapid increase of the number of quasi free electrons leads the system through a first modest resonance at $t \simeq -135$ fs. The electric field is then shielded due to the high electron densities reached until $t \simeq 10$ fs when the second important resonance takes place. It occurs much earlier than in the pure IBC case for which the resonance takes place at $t \simeq 50$ fs. The inner field is then only amplified to a modest value ($\sim 3.3 \cdot 10^{11}$ V/m) in comparison with the pure IBC case ($\geq 10^{14}$ V/m). At later times, the electron density drops rapidly due to both electron emission and cluster expansion and the inner electric field becomes identical to the external one.

The modified behaviour of the system at the resonance has significant implications on the dynamics of the cluster. The average ion charge given by $\langle Z \rangle = (\sum_j Z_j N_j)/N$ is of particular interest. Here, N_j is the number of ions with charge Z_j and $N = \sum_j N_j$ is the total number of atoms in the cluster. The average ion charge is found equal to $\sim 10+$, well below the average charge reached when no surface collisions are present ($> 40+$). Before resonance ($t \leq 10$ fs), a rapid increase of the average charge occurs. During this period, the tunnel ionisation and the laser driven ionisation rates are small because of the shielding of the field. Except for the ionisation of the first electrons from the neutral atoms, ionisation by thermal electrons prevails during this phase and allows the cluster to reach an average value of $\sim 9+$ before the occurrence of the plasmon resonance. The latter is only responsible for the increase of the average charge by approximately half a unit. Moreover, the value of the collision frequency at resonance does not allow the field to reach high values that would give higher charge states. It also makes the heating of the cluster and the electronic temperature take weaker values than those obtained in the pure IBC case. For instance, the electronic temperature at resonance is only $T_e = 1$ keV well below the temperature reached when considering IBC alone (> 20 keV). Note that the obtained average charges are in good agreement with the experimental results. For instance, a typical curve is given by figure 7 in Ref. [11]. According to these experimental data, the average value of the ion energy is 45 ± 5 keV. However it is important to notice that most of the ejected ions have an energy

of the order of 0.3-2 keV with charges between 5 and 10, according to figure 13 of the same reference. The higher charge states are obtained for very energetic ions at the far tails of the energy distribution. Altogether, this would give an average ion charge around 5 to 10. This is in good agreement with the predictions in similar situations of our model (with average charge 7-8). When no surface collisions are taken into account, the average value of the ion charge is much higher due to the huge field amplifications, and obviously contradicts the experimental results.

Note that the nanoplasma model is an average model with no temperature/density gradients, and for which only average values can be compared to the experiment. The model as such predicts very narrow charge distributions (with few units around the average value), and the contribution of highly charges is not predicted at all. Other processes have to be taken into account to explain the origin of these small but important contributions (the highest charge states) [16,21]. Including temperature/density gradients in the model might also result in broader ion charge distributions.

We have also calculated the energy spectrum of ejected electrons by summing up the energy distribution of the electrons that leave the cluster during the entire laser pulse (upper part of Figure 3). The corresponding energy spectrum when no surface collisions are taken into account is shown in the lower part of the figure. The total spectrum (shown by a solid line in the upper figure) is the sum of a “warm” and spread peak (dotted line) that results from the electrons leaving the cluster before resonance and a smaller “hot” and sharp peak (dashed line) resulting from the contribution of electrons ejected at and after the resonance. These two peaks are centered around 2 keV and 4 keV, respectively. Even though the maximum temperatures reached in the cluster are relatively low, only $T_e \simeq 0.5$ keV before resonance and 1 keV at resonance, the ejected electrons have larger kinetic energies. This comes from the assumption that the electron distribution in the cluster is Maxwellian and the fact that electrons leaving the cluster should be energetic enough to overcome the surface barrier. This makes the contribution of the Maxwellian tail important and explains why a significant amount of energy is carried away by the emitted electrons. The corresponding energy spectrum when no surface collisions are taken into account is shown in the lower

part of the figure. In this case, the spectrum exhibits a smaller contribution from "warm" electrons and a completely different shape for the "hot" contribution, with a large tail corresponding to a huge electron emission at the second resonance.

C. Hydrodynamics versus Coulomb pressure

We plot in Figure 4 the temporal evolution of the Coulomb and the hydrodynamics pressures responsible for the cluster expansion. The possibility for the hydrodynamics pressure to dominate the Coulomb one in large clusters is one of the most striking consequences of neglecting surface collisions [11,14]. In our case, the hydrodynamics pressure is greater than the Coulomb one only before resonance when the cluster has not yet reached a significant charge. During this phase, the ionisation rate is higher than the streaming rate and both the electron density and the temperature simultaneously increase. The hydrodynamics pressure $P_h = n_e k_B T_e$ rises as a result. At resonance, however, the kinetic energy of electrons is sufficiently high for the electrons to escape the cluster because the cluster net charge is not large enough to confine them. This is found to happen in a very short time scale (~ 15 fs). In the pure IBC case, the important rise of the electronic temperature at resonance compensates for the loss of electrons and thus maintains a competitive hydrodynamics pressure. This is obviously not sufficient in the case when surface collisions are considered. The rise of the hydrodynamics pressure is then very modest and falls down rapidly as both the remaining electrons leave and the cluster begins to expand. The Coulomb pressure is found to dominate the expansion dynamics of the cluster as soon as the resonance is approached. It exceeds by an order of magnitude the hydrodynamics pressure at resonance. When the cluster expands, the Coulomb pressure drops rapidly because of the $1/R^4$ dependence but is always greater than the hydrodynamics contribution although this last one scales as $1/R^3$. This statement is confirmed by considering even larger clusters. We have calculated the Coulomb and hydrodynamics pressure for a Xe cluster of $5 \cdot 10^4$ atoms. The conclusions remain unchanged. The loss of electrons proceeds faster than the expansion of the cluster. As a result the Coulomb pressure prevails even longer after the resonance.

IV. CONCLUSIONS

In conclusion, we have investigated in this paper the dynamics of large clusters irradiated by intense and short laser pulses within the framework of the nanoplasma model. In particular, we were interested in the cluster behaviour at the plasmon resonance. The role of plasmon resonance is shown to be overestimated when only IBC is considered. A very strong enhancement of the laser field inside the cluster is obtained and the cluster behaves as a collision-less system. We show that surface collisions are important and must be incorporated in the model to describe correctly the dynamics. This leads to considerable changes in the predictions of the nanoplasma model. Taking into account surface collisions, we find that the field enhancement at resonance is not sufficient to create highly charged ions. The hydrodynamics pressure is found to play a negligible role in the cluster expansion that explodes mainly by Coulomb repulsion even for large clusters ($> 10^4$ atoms). However, the electron spectrum still exhibits the double peak structure as observed in experiments, even though the second "hot" peak is much smaller.

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Figures

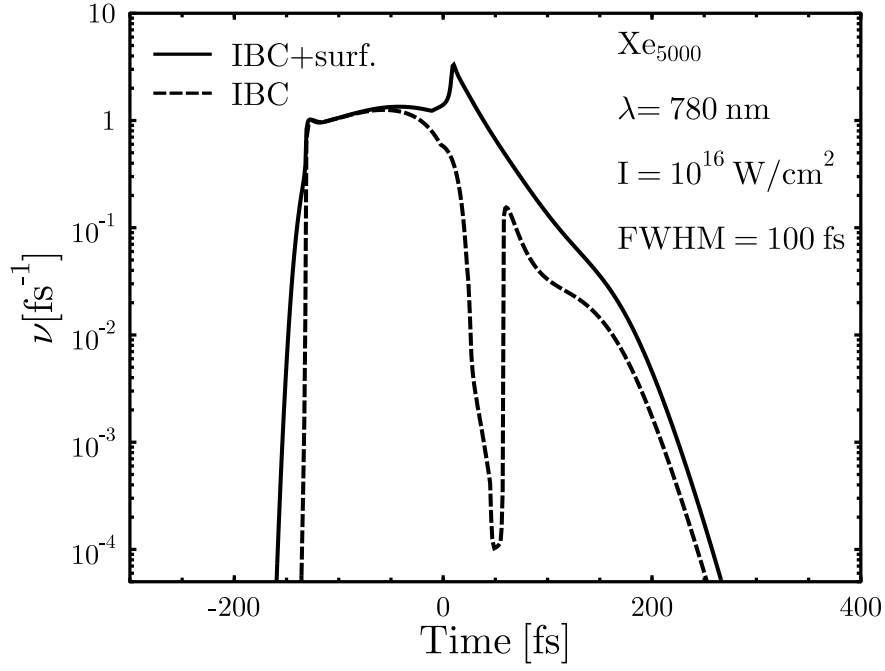


FIG. 1. Time evolution of the electron collision frequency for a 5000 atoms Xenon cluster irradiated by a 100 fs, 780 nm pulse with peak intensity (at $t = 0$) of $10^{16} \text{ W} \cdot \text{cm}^{-2}$. The dashed line shows the results with IBC alone and the solid line the results with IBC plus surface collisions.

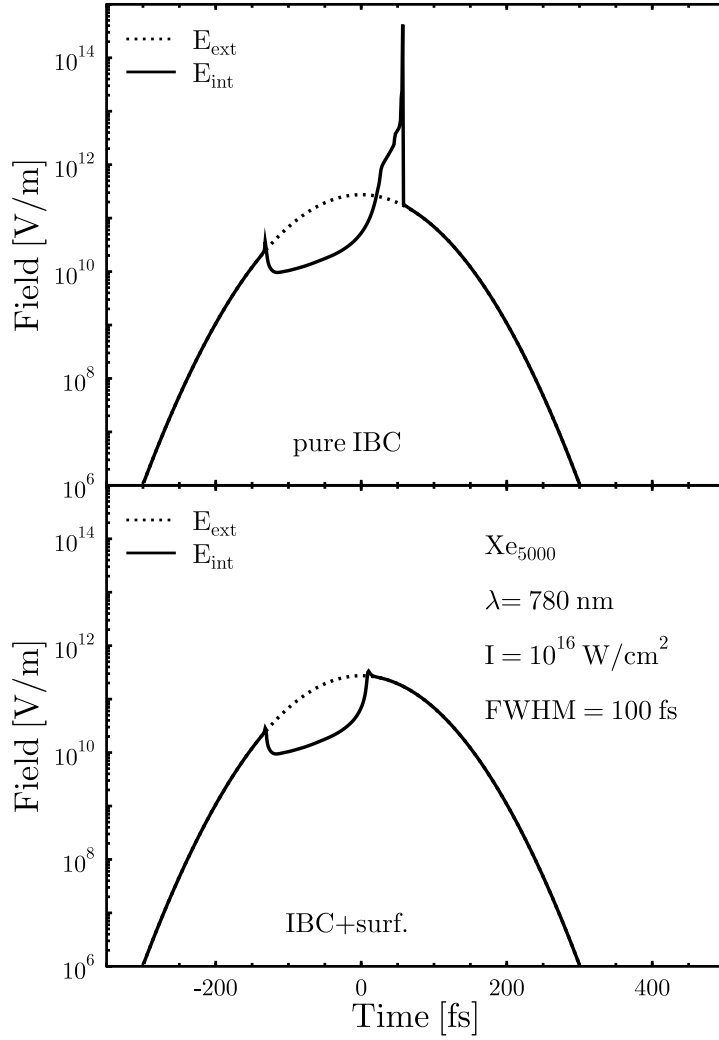


FIG. 2. Time variation of the amplitudes of the internal and external electric fields (Eq. 1) for the pure IBC case (upper panel) and IBC plus surface collisions (lower panel). The laser and cluster parameters are the same as those in Figure 1. After a first modest resonance, the inner field is shielded until the second plasmon resonance takes place.

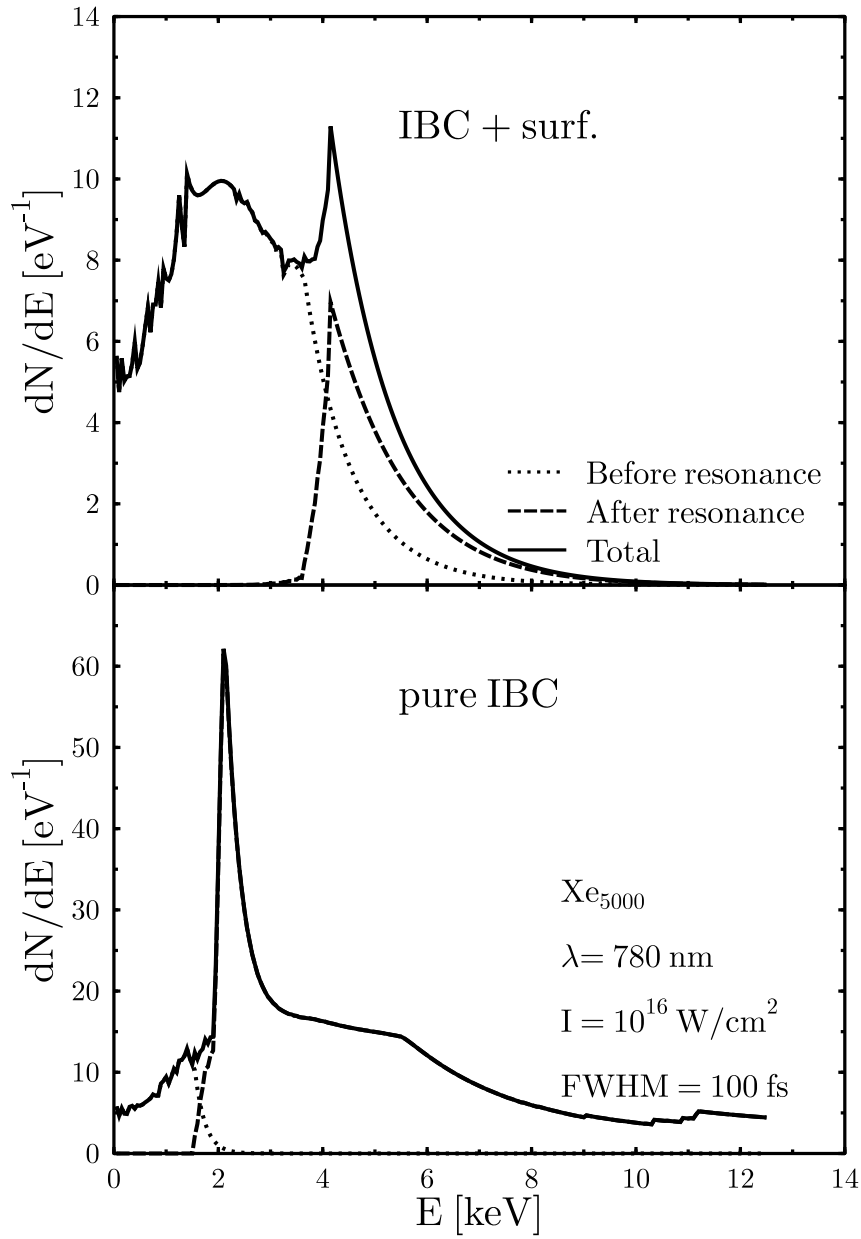


FIG. 3. Energy spectrum of ejected electrons during the entire laser pulse. The solid line shows the total spectrum while the dotted and dashed lines show the spectrum before and after the resonance, respectively.

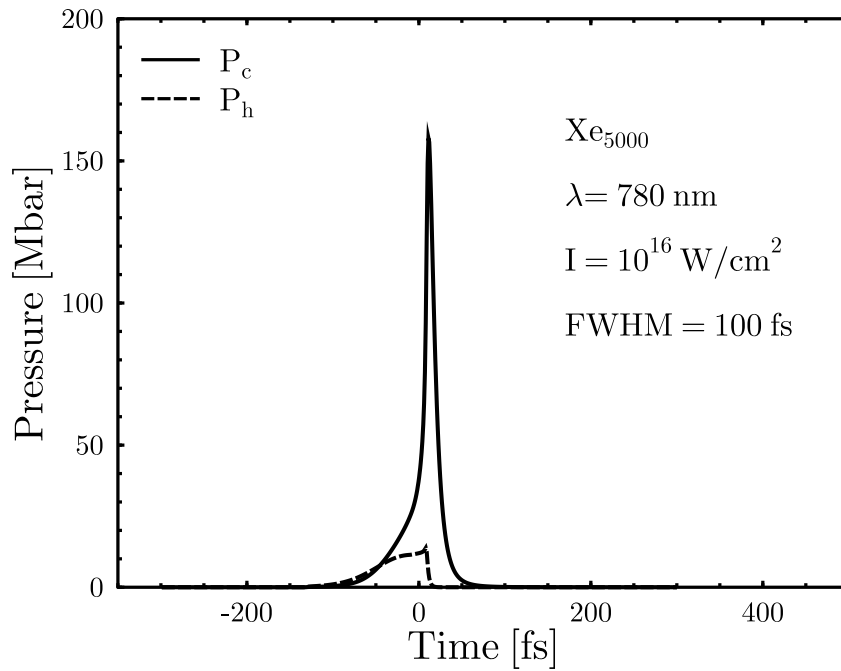


FIG. 4. Variation of the Coulomb and the hydrodynamics pressures in the cluster. The laser and cluster parameters are the same as those in Figure 1. The maximum is reached at resonance for both pressures. The Coulomb pressure is dominant for $t > -50 \text{ fs}$.