Two-dimensional effects in the hydrodynamic expansion of xenon clusters under intense laser irradiation

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The inherently two-dimensional nature of the hydrodynamic expansion of large xenon clusters irradiated by intense laser fields is shown in measurements of ion and electron emission as a function of laser pulse duration. These reveal correlation between a resonance in energy absorption by the clusters and asymmetry in cluster expansion. An extension of the hydrodynamic model accounts for the observations; an earlier interpretation of the electron spectrum is found to be incorrect.

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Concomitant with the rapid progress in generation of intense, ultrashort laser pulses, there have been efforts at efficient conversion of laser energy into high-energy particles and radiation. The more notable among these are coherent radiation in the x-ray regime through high-harmonic generation [1], intense incoherent x rays emitted by hot plasmas [2], high-energy electrons obtained by laser wakefield acceleration [3], and, recently, generation of ultrashort pulses of monoenergetic neutrons from nuclear fusion in cluster plasmas [4]. Much recent work has been devoted to the study of cluster-laser interactions due to the surprisingly high efficiency with which large rare-gas clusters absorb laser energy [5] and redistribute it into high-energy ions [6], electrons [7], and photons [8]. However, in spite of potential applications in extreme ultraviolet (EUV) lithography, time-resolved neutron diffraction, and table-top accelerators, proper insight into the dynamics of energy absorption and redistribution remains elusive.

The hydrodynamic model [9] remains the most successful effort at quantitative description of the interaction of large clusters with intense laser fields. After initial ionization by the laser field, the cluster is modeled as a spherically symmetric, uniform density plasma with a negative dielectric constant ε. In this model, it is the hydrodynamic pressure of laser-heated electrons in this plasma that drives the rapid disassembly of the cluster, as opposed to the Coulomb repulsion of highly charged ions. The electric field inside the plasma $[E_{in}(r)=3E_0\cos\theta e^{i\omega t}/(\varepsilon+2)]$ is resonantly enhanced when $\mathrm{Re}(\varepsilon)=-2$. The plasma density (n_e) is initially much larger than the critical density (n_{cr}), but as the cluster expands, the density falls to $3n_{cr}$, and the electric field inside the plasma becomes much larger than the applied field $[\mathrm{Re}(\varepsilon)\approx 1-n_e/n_{cr}]$. This resonance results in enhanced energy absorption in the plasma, and evidence for this phenomenon can be seen in measurements of ion energies as a function of pulse duration—the ion energies are maximum when the peak of the laser intensity coincides with the resonance [10]. The only previously reported electron spectrum (from xenon clusters) [7] comprises a ''warm'' component (0.1–1 keV) emitted predominantly along the laser polarization vector, and a ''hot,'' isotropic component (2–3 keV). It was postulated that the warm component consists of electrons that do not experience the resonantly enhanced field, and therefore undergo few collisions and retain, to some extent, the angular distribution of tunnel ionization, while the hot electrons undergo extensive collisonal heating at resonance, and hence lose all directionality.

In the hydrodynamic model, the in-built assumptions of spherical symmetry (justified largely by the above interpretation of the electron spectrum) and uniform density and temperature automatically imply isotropic expansion of the irradiated cluster. In this article, we demonstrate that, contrary to expectations, asymmetry in ion emission is closely linked to the efficiency of energy absorption in the cluster. A resonance in the latter leads to enhancement of the former. We establish the connection by measuring ion energies both along and perpendicular to the laser polarization vector as a function of pulse duration. The ability to control both the energy of ions and their directionality is obviously critical for applications in which both are important. For instance, deuterium (or deuterium-rich) clusters with significant asymmetry should yield more neutrons as a result of greater probability of head-on collisions. We propose a mechanism that follows directly from the hydrodynamic description of the cluster: the polarization surface charge induced on the cluster by the laser field causes the asymmetry. We also show experimental evidence that the earlier interpretation of the electron spectrum is incorrect—the warm electrons, not the hot ones, are associated with the resonance. The pulse duration dependence of the electron spectrum provides corroboration for the asymmetric resonance mechanism.

We have recently shown [11] that ion emission from Ar clusters has two components that can be distinguished by their angular distributions. The low-energy component is isotropic, while the high-energy component is emitted preferentially along the laser polarization vector. It was postulated that the asymmetry arises due to two related yet different forces. First, the charge state distribution on the cluster is asymmetric due to the preferential removal of electrons from the poles of the cluster (the axis being defined by the direction of the linearly polarized laser field [12]). Subsequent Coulomb explosion of the cluster is then asymmetric. Second, the net field experienced by the ions at the poles is significantly different over two consecutive laser half cycles due to the contribution of the radial Coulomb field. This results in a net cycle-averaged force on these ions along the laser polarization direction [13]. Taken together, these two

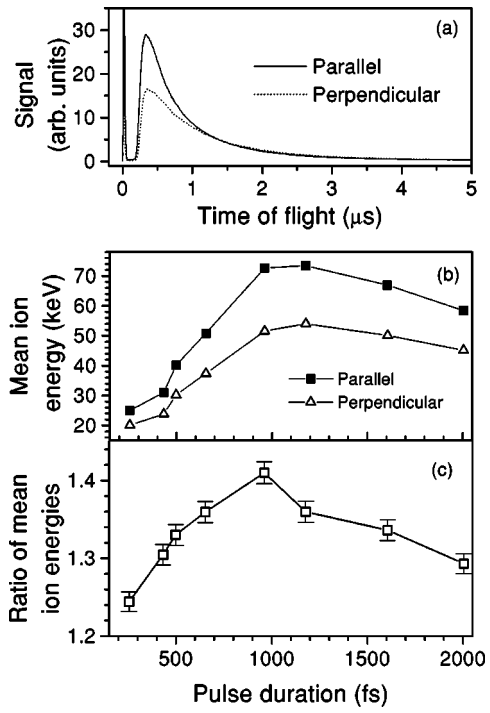

FIG. 1. (a) TOF spectra of Xe ions. The pulsewidth was 1.2 ps, for which the mean ion energy was maximum for this cluster size and laser energy. The mean energy along the laser polarization is 73 keV; in the perpendicular direction it is 54 keV. The sharp spike near $t=0$ is due to electrons. (b) Mean ion energies as a function of pulse duration. Laser intensity was 1×10^{16} W cm^{-2} at 200 fs pulsewidth. (c) The ratio of the mean ion energies shown in (b).

effects were found to explain the important features of the observed angular dependence. The connection between this scenario and the mechanism proposed here is discussed later.

Our experimental setup is similar to the one described recently [11], with some modifications. The clusters were produced using a solenoid-driven pulsed nozzle with a 500 μm throat diameter, and the centerline beam was transfered to a high vacuum chamber via a 55° nickel skimmer with a 250 μm diameter aperture. A dual microchannel plate (MCP) assembly was used at the end of a 19 cm time-of-flight (TOF) spectrometer to detect the ions and the electrons. Ion energies were determined by converting the field-free ion TOF to energy. Electron energies were measured by scanning the voltage on a retarding potential analyzer from zero to -5 kV, integrating the signal near $t=0$ over time, and then differentiating the yield thus obtained with respect to the voltage. The MCP signal was recorded using a 500 MHz, 1 GS/s digital oscilloscope. The Ti:sapphire laser used can generate 55 mJ per pulse, with 100 fs pulsewidth, but only up to 12 mJ was used for this series of experiments. The pulse duration of the laser was varied by changing the grating separation in the compressor. Xenon cluster sizes were estimated using Hagena's scaling law [14]; the mean cluster size in the present experiments was 1.5×10^5 atoms per cluster at 6 bar stagnation pressure.

Figure 1(a) shows typical TOF spectra from Xe clusters for parallel and perpendicular polarizations. The TOF spectra were converted to energy distributions, using $f(E)=f(t)\times(dE/dt)^{-1}$; the mean ion energy and the total ion yield were computed from the distribution function. The mean energy, the maximum energy, and the total ion yield all maximize along the direction of polarization. As noted above, the hydrodynamic model demands an optimum pulse duration for most efficient energy absorption from the laser pulse. We have observed the pulsewidth dependence in the ion energies, with the mean ion energy reaching a maximum at a pulse duration of 1.2 ps. Thus, unlike argon clusters studied in [11], these xenon clusters undergo hydrodynamic expansion. Also shown in Fig. 1 are the mean energies along both directions [Fig. 1(b)], and the ratio of the two [Fig. 1(c)], as functions of laser pulse duration. The laser energy, and not the intensity, was kept constant for these measurements. From the hydrodynamic model, and from the measurements of Zweiback *et al.*, the maximum in the ion energies was expected, but the corresponding variation in the asymmetry of expansion that we observed was surprising.

The asymmetry shown in Fig. 1 shows that the mechanism that causes asymmetry in the ion emission is closely related to the resonant enhancement of the electric field in the expanding cluster plasma. But, as noted above, the hydrodynamic model [9] assumes spherical symmetry and, hence, does not provide any information on mechanisms that might be responsible for the observed asymmetry or its evolution. The scenario proposed in [11] also does not provide any clue about the pulse duration dependence of the asymmetry, and in any case relies on the Coulomb field of the charged cluster, which plays a subordinate role in clusters near resonance. A closer examination of the phenomenon responsible for the resonance shows that the hydrodynamic description does, indeed, contain the physics that might drive an asymmetric expansion. The hydrodynamic model accounts for the enhancement of electric field due to the polarization of the medium, but fails to consider the polarization surface charge distribution,

$$\sigma_{pol}=\frac{3}{4\pi}\left(\frac{\varepsilon-1}{\varepsilon+2}\right)E_0\cos\theta e^{i\omega t}, \tag{1}$$

and the action of the laser field on this charge. The laser field exerts a cycle-averaged pressure

$$P_{pol}=\frac{1}{2}\mathrm{Re}(\vec{\sigma}\cdot\vec{E}^*_{av})=\frac{9}{16\pi}\frac{|\varepsilon|^2-1}{|\varepsilon+2|^2}E_0^2\cos^2\theta, \tag{2}$$

where $E_{av}=1/2(E_{in}+E_{out})$ and $E_{out}=\varepsilon E_{in}$, on the surface of the cluster. Thus, the resonance in the electric field is inextricably linked to a resonance in this ($\cos^2\theta$)-dependent force. The hydrodynamic model does not take this pressure into account, but it is clear that if the force is comparable to the hydrodynamic or Coulomb pressures, its consideration is mandatory. Figure 2 shows the dependence of E_{in} and P_{pol} on $\mathrm{Re}(\varepsilon)$, assuming the laser intensity to be 1×10^{15} W cm^{-2}, and a fivefold enhancement of the field at resonance. We find that the polarization pressure is as large as 8×10^{11} N m^{-2} at the peak. Since the hydrodynamic pressure, for a plasma with an electron density of 5.4

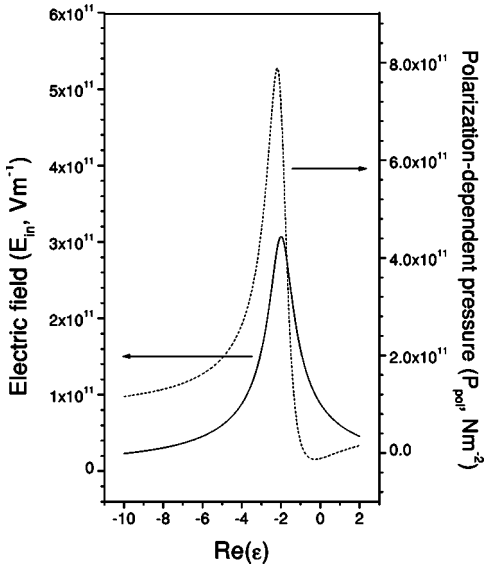

FIG. 2. The electric field amplitude and the polarization-dependent force [Eq. (2)]. The value of ε was chosen so as to produce a fivefold enhancement of the electric field at resonance. Im(ε) was held constant. The laser intensity was taken to be 1×10^{15} W cm^{-2}.

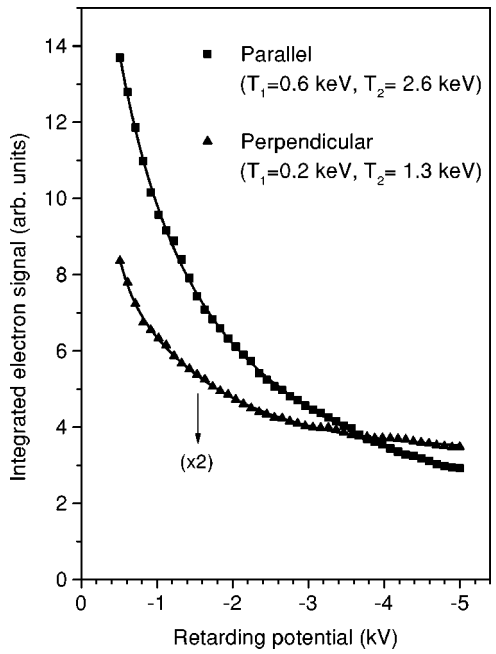

FIG. 3. Integrated electron yield as a function of repeller voltage. The lines are exponential fits, with two temperatures in both cases. The signal in the perpendicular direction is multiplied by a factor of 2.

$\times10^{21}$ cm^{-3} ($=3n_{cr}$ for 800 nm) and a temperature of 1 keV, is 3×10^{11} N m^{-2}, we conclude that the cluster expands under the combined influence of both the pressures. The asymmetric force is manifested in the directionality of the emitted ions. From Fig. 2, we also expect the asymmetry in ion emission to peak at a shorter pulse duration than the mean ion energy, and then fall off at a faster rate as the pulse duration becomes too long for efficient energy absorption. Our experimental data (Fig. 1) are consistent with both these expectations.

In [11], asymmetric ion emission from Coulomb-exploding argon clusters was attributed to the generation of higher charge states at the poles of the cluster and a cycle-averaged force on the polar ions due to rapid changes in the charge state within a laser cycle. The mechanism proposed here is a macroscopic description of the same phenomenon. The most important difference between the two is the role played by the Coulomb field, which was critical for the production of higher-charge states at the poles in conjunction with the laser field. But the pressure due to the polarization of the cluster does not depend on the residual charge on the cluster; it would be present even in a neutral cluster. This pressure should affect the electrons as well as the ions. According to the model, an electron can leave the cluster provided it has enough energy to overcome the Coulomb barrier, and if its mean free path is longer than the distance to the surface of the cluster. The polarization pressure effectively causes the barrier to be lowered asymmetrically and should, therefore, cause an asymmetry in the electron distribution, influencing in particular the component that is produced by the resonance. This conclusion is opposite to the one reached in [7], where resonance-heated electrons were claimed to be isotropic.

In order to verify this claim, we have measured electron energies under similar conditions as the ions, and Fig. 3 shows the integrated electron signal as a function of repeller voltage. Each data point is an average of 50 shots. First, we note the absence of the hot electron peak reported in [7]. In fact, the thermal fits show good agreement with the data, although two temperatures were necessary for the fitting. The hot electron peak, if any, must lie above 5 keV, the maximum repeller voltage used for these measurements. Indeed, the signal from the MCP does not go to zero when the repeller voltage is -5 kV, implying either a significant photon component in the signal, or electrons with energies greater than this limit. In [7], the hot and the warm electrons were distinguished by the temporal separation between the two; we have also observed two such peaks in the MCP signal, and found the earlier peak to be isotropic, but it remains on application of -5 kV on the repeller. Second, the warm electron peak itself extends to 5 keV, an energy value that is considerably higher than reported in [7]. And finally, although the warm electron yield is much smaller along the direction perpendicular to the laser polarization, the spectrum is qualitatively similar to the parallel component. The differences between our data and the data reported in [7] might be due to the difference in the cluster sizes since our clusters are estimated to be nearly five times larger.

We also measured the electron peak simultaneously with the ion spectrum as a function of the pulse duration, and these are shown in Figs. 4(a) and 4(b), respectively. The spectra were recorded with the laser polarization along the TOF axis. There is some ringing in the MCP output, so that the electron yield seems to become negative near $t=30$ ns, but this experimental artifact does not affect our conclusions. The correlation between the warm electron component and

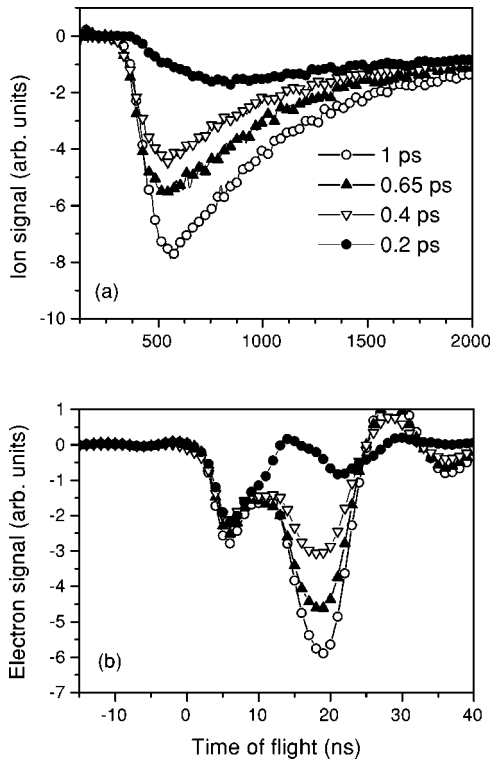

FIG. 4. MCP signals from ions (a) and electrons (b) for four different pulse durations. The cluster size was 1.5×10^5, and the laser intensity at 200 fs was 8×10^{15} W cm^{-2}. Laser polarization was along the TOF axis.

the ion energy spectrum is apparent, and offers strong evidence of a common mechanism for the generation of both. We also note the much weaker dependence of the first peak (at $\sim$5 ns), which might be due to hot electrons, on the pulse duration. In terms of the analysis in [7], these results are unexpected, since the fraction of hot electrons should have increased near resonance, while the warm electron component should have become less important. Instead, if the polarization charge is taken into account, the observed behavior of the warm electrons and the close correspondence with ion energies is expected. Any electrons with significantly higher energies would, on the other hand, have large enough mean free paths to free-stream directly from the interior of the cluster, and should not be much influenced by the polarization charge. Note that the association of the warm electrons with the resonance raises a question about the source of the hot electrons, for which we have no answer at present.

In summary, we have discovered a relationship between energy absorption and asymmetry in cluster disassembly in the hydrodynamic regime. An extension of the hydrodynamic model is proposed, wherein the polarization charge induced on the surface of the cluster gives rise to the asymmetry. Our electron energy measurements show that the asymmetric warm electrons are produced by the resonance, offering strong support for this mechanism. Clearly, a two-dimensional model with self-consistent field and charge distributions is important. Milchberg *et al.* [15] recently reported a self-consistent model, but it is one dimensional. Our results indicate the possibility of simultaneous maximization of ion energy and directionality by control of resonance in energy absorption. The improved understanding of the electron energy spectrum and its dependence on the resonance has implications for designing more efficient sources of EUV radiation for lithography, using either clusters or liquid drops.

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