# High Energy Ion Explosion of Atomic Clusters: Transition from Molecular to Plasma Behavior

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We have studied the high-intensity femtosecond photoionization of inertially confined noble-gas clusters. The explosion of the resulting highly ionized, high-temperature microplasma ejects ions with substantial kinetic energy. We have observed Xe ions with kinetic energy up to 1 MeV and charge states as high as $40^+$. This ion energy is over three orders of magnitude higher than has previously been observed in the Coulomb explosion of molecules or clusters of any kind and indicates that there is a fundamental shift in the nature of intense laser-matter interactions between molecules and large clusters. [S0031-9007(97)02896-2]

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Research into atomic and molecular clusters is of great interest because such clusters represent a bridge between a molecular and a bulk description of matter [1]. Of fundamental importance to the study of cluster physics is their interaction with light and the nature of cluster photofragmentation. A large number of experiments have studied low-intensity ($<10^{12}$ W/cm$^2$) photoinduced fragmentation [2] and Coulomb fission of clusters [3], in which low energy fragments (0.1–1 eV) are typically observed. Preliminary experiments on high-intensity laser-cluster interactions have suggested that the interaction is much more energetic than that of isolated atoms [4–7], and may be even more energetic than laser-solid interactions [8]. Recent experiments examining the x-ray emission from plasmas formed by the irradiation of noble gas (Ar, Kr, and Xe) clusters have indicated that because of the enhancement of laser-driven collisional excitation within the cluster, efficient production of x rays is possible [4,5]. These and other recent experiments also indicated that highly ionized charge states of the cluster atoms result from the high-intensity laser-cluster interaction [4,5,9,10]. High energy Coulomb explosion of small clusters ionized by femtosecond pulses has been observed as well [9]. The ion energies measured in these experiments ranged from 500 to 1000 eV.

Recent experiments have shown that the electrons in an intensely irradiated cluster undergo rapid collisional heating for the short time ($<1$ ps) before the cluster disassembles in the laser field [6,7]. We previously studied the electron energy spectra produced by the high-intensity ($>10^{16}$ W/cm$^2$) irradiation of large ($>1000$ atoms) Xe clusters with a 150 fs laser pulse [7]. These studies indicated that collisional heating within the cluster can produce electrons with energy of up to 3 keV. A sharp peak near 2.5 keV in the measured electron energy spectrum suggested that the cluster microplasma exhibited a resonance in the heating by the laser pulse similar to the giant resonance seen in metallic clusters [11]. During the laser-cluster interaction, free electrons are produced by photoionization early in the laser pulse. The laser collisionally heats these electrons which further ionize the atoms of the cluster through collisional ionization. A small amount of expansion during the laser pulse lowers the electron density to bring the near infrared laser light into Mie resonance with the free electrons in the cluster. The enhanced laser absorption then rapidly heats the electrons on a fast ($<10$ fs) time scale to a superheated state (with temperature of many keV). The large energy density of these hot electrons will inevitably drive a rapid expansion of the cluster ions.

In this Letter we describe a measurement of the energies of ions ejected from exploding noble gas clusters irradiated by intense femtosecond laser pulses. We find that the ions produced by the exploding clusters are extremely energetic. We have observed highly charged Xe ions with maximum energy up to 1 MeV and charge state up to $40^+$. These measurements indicate that intense short pulse laser interactions with these clusters are fundamentally different than low-intensity photofragmentation of clusters and much more energetic than the Coulomb explosion of small molecules ionized by an intense laser pulse [12]. Instead, the cluster explosion is more closely akin to that of an expanding solid density plasma.

Our experiment used a high power, Ti:sapphire laser [13], which delivers 150 fs pulses at a wavelength of 780 nm. The laser pulses were focused into a time-of-flight (TOF) chamber to a peak intensity of up to $2 \times 10^{16}$ W/cm$^2$. Clusters were produced with a pulsed sonic gas jet, and a skimmer produced a low density Xe or Kr cluster beam which intercepted the laser beam at the laser focus. The average cluster size in the flow from the gas jet was measured through a series of Rayleigh scattering measurements [14]. The energies of the ions produced by the interaction of the laser pulse with the clusters were determined by the flight time of the ions in a field free drift tube of either 38 or 80 cm. A microchannel plate was used to detect the ions in an angular cone of $\sim 3.5°$ from the focus. Three closely spaced grids ($\sim 3$ mm spacing) were placed along the drift tube one third of the distance from the laser focus.

 

The front and back grids were grounded. Charging the middle grid to a voltage $\varphi$ introduced a potential barrier to ions with energy less than $Ze\varphi$, without significantly altering the flight time of higher energy ions. By varying the voltage we were able to measure the charge state distribution of the ions as a function of ion kinetic energy.

Examples of time-of-flight data collected with 55 ± 5 Å (~1500 atom) Xe clusters are shown in Fig. 1. The data exhibit a fast feature near a flight time of zero corresponding to the arrival of fast electrons (energy > 2 keV). The broad feature at flight times >0.5 $\mu$s is the arrival of hot ions ejected from the exploding clusters. Five TOF traces with different angles between the ion detection axis and the laser polarization, varying from 0° (along the laser polarization) to 90° (perpendicular to the laser polarization), are compared. The spectra are virtually identical for all polarization angles, indicating that ions are ejected isotropically with no preference along or perpendicular to the laser polarization. This is in stark contrast to the Coulomb explosion of small, linear molecules ionized at high intensities, in which the ions are preferentially ejected along the laser polarization axis [15].

The TOF trace directly yields an energy distribution function for the ions. The TOF and energy spectra of Xe ions produced from the irradiation of 65 ± 5 Å (~2500 atom) Xe clusters is illustrated in Fig. 2. Most importantly, we observe the production of significant numbers of ions with energies out to 1 MeV. This energy is 4 orders of magnitude higher than has previously been observed in the Coulomb explosion of molecules [12] and about 1000 times higher than the energy of Ar ions ejected in the Coulomb explosion of small Ar clusters irradiated at lower intensity reported by Purnell *et al.* [9]. The average ion energy of the distribution shown in Fig. 2 is defined as $\langle E\rangle = \int Ef(E)\,dE/\int f(E)\,dE$ is 45 ± 5 keV. Thus the average laser energy deposited per ion is substantial.

By varying the gas jet backing pressure we controlled the average size of clusters with which the laser interacts. In general we find that both the maximum and the average ion kinetic energy increase with increasing cluster size, though the energy varies slowly with changing cluster size. The ion energy spectrum resulting from the irradiation of Xe clusters with ~400 atoms (diameter of ~ 35 ± 7 Å) is also shown in Fig. 2(b) for comparison with the spectrum of the larger clusters. Though the ions exhibit somewhat lower energy, the maximum observed ion energy is 250 keV and the average ion energy is 28 keV, there is not a dramatic shift in the shape of the distribution or in the energies of the ions. However, at lower backing pressure, below ~1 bar, we observe no hot ions (with energy >1000 eV). Our Rayleigh scattering measurements indicate that large clusters form only with backing pressure >1 bar (though we expect that small 10–100 atom clusters are still present in the gas flow at lower backing pressure). This transition to hot ion production for large clusters points to an important change in the dynamics of the cluster expansion between small (<100 atom) and large clusters. We have also examined the energies of ions produced from the explosion of Kr clusters with sizes comparable to the Xe clusters studied. The average and maximum energy of the Kr ions tend to be comparable, though slightly lower than the Xe ions under the same conditions. For example, we found that the average ion energy produced from ~1000 atom Kr clusters was 20 keV and

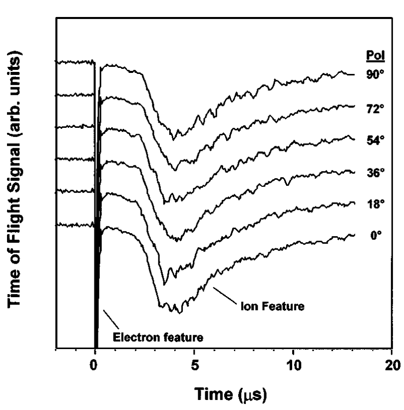


FIG. 1. TOF traces of ions produced by irradiation of 55 ± 5 Å Xe clusters with a peak intensity of $1 \times 10^{16}$ W/cm$^2$. Each plot is an average of 500 laser shots. The drift distance from interaction to detector was 80 cm. The first peak is fast (>2 keV) electrons and the feature at later times is the ions. (Note: Electrons with energy <2 keV do not reach the MCP detector since the front plate is charged to −2000 V). Each curve (offset for clarity) has been taken with a different angle between the laser polarization and the detection axis.

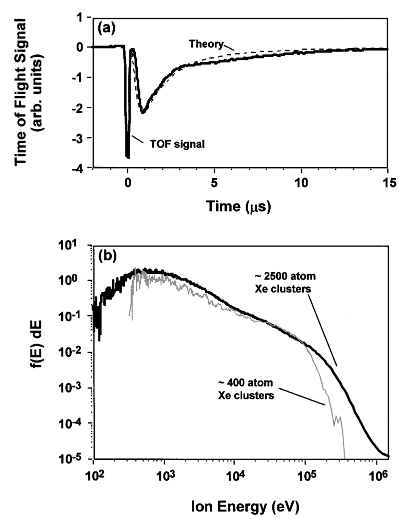


FIG. 2. (a) Time-of-flight spectrum from 65 ± 5 Å (2500 atom) Xe clusters irradiated by a peak intensity of $2 \times 10^{16}$ W/cm$^2$. The drift distance from interaction to detector was 38 cm. The dashed curve shows the calculated TOF trace of an isothermal Xe plasma expansion. (b) Ion energy spectrum from 65 ± 5 Å (2500 atom) Xe clusters [derived from the TOF trace in (a)] and 35 ± 7 Å (400 atom) Xe clusters irradiated with a peak intensity of $2 \times 10^{16}$ W/cm$^2$.

the maximum energy was 350 keV, while 1000 atom Xe clusters exhibited average and maximum ion energies of 38 and 600 keV, respectively.

Another striking feature of the ions produced in the laser cluster interaction is the ionization to high charge states (a phenomenon previously seen in the experiments of Refs. [4,5,10] and theoretically predicted in Refs. [6,16]). Measured charge state distributions of Xe ions produced with a peak intensity of $2 \times 10^{16}$ W/cm$^2$ are shown in Fig. 3 for ion energies of 1, 3, 10, 30, and 100 keV. In general the average charge state of the ions increases as the ion energy increases. For the high energy ions, at 30 and 100 keV the peak of the charge state distribution is around $Z = 20^+ - 25^+$ though a significant number of ions with charge state between $35^+$ and $40^+$ are observed. These are charge states far above the maximum charge state that can be produced by simple field ionization of the Xe atoms [17]. In the cluster the high-temperature electrons created through laser-driven heating rapidly strip the ions by collisional ionization to the high charge states observed [6].

The weak dependence of the ion charge state with ion kinetic energy and the lack of significant variation of ion energy with cluster size suggests that the traditional mechanism of strong field molecular fragmentation, Coulomb explosion, is not the primary mechanism causing the high energy explosion of the atomic clusters. The large attractive charge of the ionized cluster prevents electrons from escaping until late in the laser pulse [6]. As a consequence the force propelling the ion explosion is dominated by the hydrodynamic pressure associated with the hot electrons within the cluster and the Coulomb forces are less important. The pressure associated with the hot electrons within the cluster is merely $P_{\text{hyd}} = n_e k_B T_e$, on the other hand, the "pressure" driving the Coulomb explosion of a spherically charged cluster is approximately [6] $P_{\text{Coul}} \approx Q^2 e^2 / 8\pi r^4$ where $Q$ is the positive charge on the cluster resulting from escaped electrons. Not only do large clusters prevent a buildup of $Q$ due to their strong space charge, the hydrodynamic pressure scales as $1/r^3$ (through the electron density) while the Coulomb pressure scales as $1/r^4$, indicating that for larger clusters, the "plasma" hydrodynamic pressure will dominate over any Coulomb expansion force.

To further explore this aspect of the cluster explosion we have conducted numerical modeling of the cluster explosion during the irradiation by an intense, femtosecond laser pulse. The details of this model have been presented in Ref. [6]. In brief, the model treats the cluster as a spherical microplasma, subject to the standard processes of a laser-heated plasma such as collisional heating, as well as collisional and tunnel ionization. The cluster expansion is treated as an expanding sphere with uniform density. The rate of electrons free streaming from the cluster is calculated, accounting for the space-charge and electron collisional mean free path in the cluster. Both hydrodynamic and Coulomb contributions to the cluster expansion are included.

The results of one such calculation are shown in Fig. 4 in which a 65 Å Xe is irradiated by a 150 fs, 780 nm pulse with a peak intensity of $2 \times 10^{16}$ W/cm$^2$. In Fig. 4(a) the temporal evolution of the cluster radius and radial velocity are shown. The cluster very rapidly expands during the pulse once heating of the electrons in the cluster has begun. The maximum radial ion energy is 255 keV, consistent with the very high ion energies observed in our experiment. The relative contributions

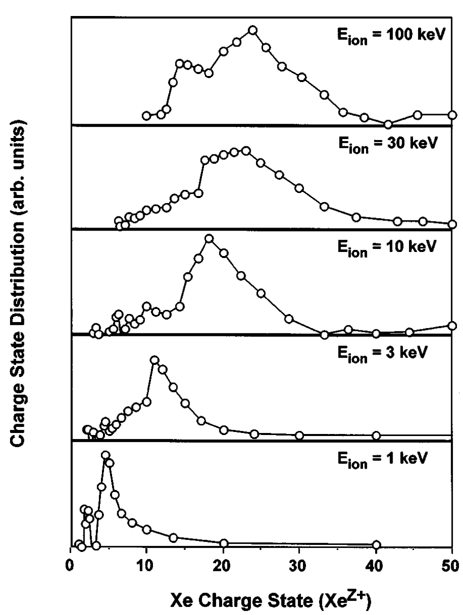


FIG. 3. Measured charge state distribution of 65 Å Xe clusters irradiated by a peak intensity of $2 \times 10^{16}$ W/cm$^2$ for five different ion kinetic energies. (Each data point represents the number of ions between $Z$ and $Z + \Delta Z$.)

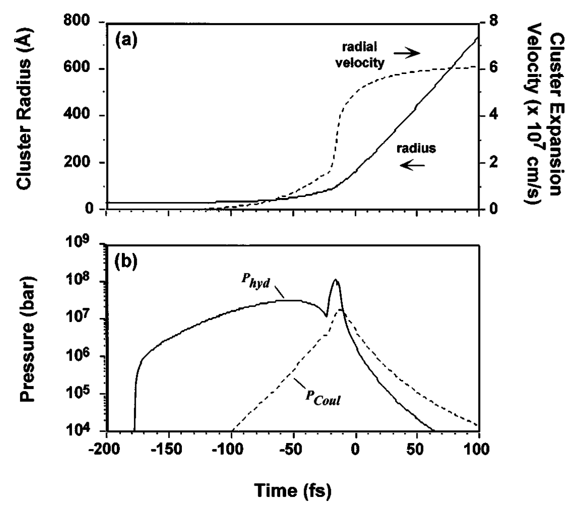


FIG. 4. (a) Calculated time history of the radius and maximum radial velocity of the ions in a 65 Å Xe cluster irradiated by a 150 fs, 780 nm pulse (centered at $t = 0$) with a peak intensity of $2 \times 10^{16}$ W/cm$^2$. (b) Calculated hydrodynamic and Coulomb pressure driving the cluster expansion.

of the hydrodynamic pressure and the Coulomb pressure are shown in Fig. 4(b). During the majority of the ion acceleration seen in Fig. 4(a), the dominant force is the hydrodynamic force, with very little contribution from the Coulomb explosion force. Our calculation also predicts the appearance of the high ion charge states observed (>20+ for Xe ions). We find that rapid collisional ionization by the hot electrons within the cluster can strip the ions to very high charge states (up to $Xe^{40+}$ in our calculations), a mechanism discussed at length in Ref. [6]. We have also performed calculations for Kr clusters of similar size under our experimental conditions. We find that, in general, the dynamics are very similar to those of the exploding Xe clusters, with hydrodynamic forces dominant in driving the explosion, though the ion energies tend to be lower than produced from the Xe clusters (~130 keV from 1200 atom Kr clusters), a trend consistent with our experimental findings.

The numerical analysis suggests that the explosion of the cluster ions is very much like that observed in the hot electron driven expansion of the laser-heated solid target plasma, which typically produces ion energies of > 100 keV [18]. These high ion energies are the result of fast electron-driven hydrodynamic expansion of the quasineutral plasma into vacuum [19]. To reinforce this similarity, we have calculated the well known self-similar solution of ion energies [19] resulting from the isothermal expansion of an Xe plasma with $Z = 20$ and an electron temperature of 2.5 keV, the measured hot electron temperature of the Xe clusters under these conditions [7]. (Note that the actual expansion is not isothermal; the actual temperature is a complicated function of time. However, this calculation is intended only as a qualitative comparison and the temperature variation will not dramatically alter the shape of the calculated ion TOF trace [19].) This solution is compared with the measured ion TOF trace of Fig. 2(b). The measured TOF trace bears a striking similarity to that of the calculated hot electron driven hydrodynamic expansion.

This close similarity indicates that, for clusters even as small as a few hundred atoms, the dynamics of the expansion appear to approximate that of solid density, hot electron driven plasma expansion, and that the Coulomb explosion model for the fragmentation is no longer valid. The hot electrons produced by collisional heating in the cluster produce an ambipolar potential which accelerates the ions. In this model the average ion energy will be of the order of $E_{\rm ion} \sim \overline{Z} k_B T_e$ [18] (where $\overline{Z}$ is the average charge state of the cluster microplasma). This implies that if $k_B T_e = 2.5$ keV, a cluster with $\overline{Z} \sim 20^+$ will exhibit an average kinetic energy of ~50 keV, in good agreement with our observed average ion energies. The average charge states observed in Kr tend to be somewhat lower, $15^+ - 20^+$, explaining the lower kinetic energies observed. Lower ionization states in Kr were found in our numerical modeling as well. This resulted in a lower cluster electron density and a lower plasma pressure, explaining the lower ion energies observed from Kr clusters.

The efficiency with which the cluster converts laser energy to particle energy is remarkable. Ultimately the intense laser interaction with clusters of ~1000 atoms results in very efficient coupling of laser energy to the ions through rapid heating of the cluster microplasma before it disassembles. The high ion energies and charge states observed in the explosion of intensely irradiated clusters is very much like the ions observed in the expansion of a laser-heated solid density plasma into vacuum and is unlike the Coulomb explosion of smaller molecules in strong laser fields. Thus clusters of greater than a few hundred atoms represent an important transition in the dynamics of intensity laser-matter interaction from molecules to solids.

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