Explosion of Xenon Clusters Driven by Intense Femtosecond Pulses of Extreme Ultraviolet Light

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The explosions of large xenon clusters irradiated by intense, femtosecond extreme ultraviolet pulses at a wavelength of 38 nm have been studied. Using high harmonic generation from a 35 fs laser, clusters have been irradiated by extreme ultraviolet pulses at intensity approaching 10^{11} W/cm^2. Charge states up to Xe^{8+} are observed, states well above those produced by single atom illumination, indicating that plasma continuum lowering is important. Furthermore, the kinetic energy distribution of the exploding ions is consistent with a quasineutral hydrodynamic expansion, rather than a Coulomb explosion.

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The response of atomic clusters to intense pulses of laser light has been a topic of detailed study in recent years [1–3]. At infrared and optical wavelengths with intensities of 10^{16}–10^{19} W/cm^2, atoms in clusters can absorb large amounts of energy from the laser field, producing x rays, hot electrons, and energetic ions in a process dominated by the large ponderomotive force [1]. At high intensity, the electric field of the laser strips electrons from the cluster by tunnel ionization, and electrons confined to the cluster by space-charge forces further ionize the cluster to high charge states by collisional ionization [1]. If the laser field is strong enough, a condition satisfied if the ponderomotive potential of the laser is greater than the surface binding potential of the ionized cluster, the laser field strips the cluster of its electrons and the charged cluster explodes by Coulomb explosion [4]. On the other hand, if the clusters are large and the ions highly charged, space-charge forces retain electrons within the cluster body, resulting in a ‘‘nanoplasma’’ which explodes by hydrodynamic forces [1].

Recently, the interactions of intense extreme ultraviolet (XUV) pulses (10–100 nm wavelength) with atomic clusters have been explored by a number of groups [5–12]. Such interactions are dramatically different from experiments with intense IR laser pulses. Electrons are easily liberated initially by single photon ionization which can sequentially strip the constituent atoms to high charge states; however, subsequently ionized electrons (including Auger electrons) are not easily removed from the cluster's space-charge forces because of the weak ponderomotive potential of the short wavelength pulse. It is not clear that a nanoplasma will form as the XUV pulse ejects photoionized electrons sequentially, and it is an open question whether XUV irradiated clusters will explode by Coulomb explosion or by hydrodynamic forces.

To date, there have been a number of experiments addressing these issues [5–9]. The most significant experiments on intense XUV irradiated clusters have been performed at the DESY vacuum ultraviolet (VUV) free-electron laser with Xe and Ar clusters of sizes ranging up to $\sim$30 000 atoms/cluster [5–8]. The first surprising result from this machine was that energetic electrons and high ion charge states (Xe^{8+}) are produced when clusters are irradiated with 95 nm light at intensities of $\sim 10^{13}$ W/cm^2 [5] and that the mechanism for ejecting the high Xe charge states from the clusters was by a Coulomb explosion. Later experiments at 95 nm showed electron emission from moderate sized ($\sim$1000 atom) Ar and Xe clusters with a quasi-Maxwellian distribution with a 9 eV electron temperature [7]. More recent experiments at 33 nm in small ($<$100 atom) clusters have also been reported and have found that the electron removal from the cluster was purely sequential, with little evidence for the production of a nanoplasma or a thermal electron distribution [8].

These experiments have bred a number of theoretical studies focused on the reasons for the high charge state production and anomalously high absorption seen in the DESY experiments [10–12]. Santra and Greene have proposed that the large absorption of VUV irradiated clusters and high charge states observed can be explained by a nanoplasma heated through inverse bremsstrahlung (IB) absorption with appropriately corrected collision rates [10]. Siedschlag and Rost have posited that the high charge states are produced by single photon absorption in the cluster facilitated by the distortion of the ionic binding potential by the neighboring ions in the cluster [11]. Finally, Jungreuthmayer *et al.* have performed simulations which indicate that the high charge states result from collisional ionization by an electron cloud heated through a process they refer to as many-body recombination heating [12]. All of these models find that a cluster nanoplasma is formed, a conclusion not completely supported by data [5,8].

To address these issues we have undertaken a set of experiments using a different source and diagnostic. In this Letter, we present experimental studies of the ion charge state and ion energy spectra of Xe clusters irradiated at $\sim 10^{11}$ W/cm^2 by intense 38 nm pulses produced by high order harmonic generation. We have observed ions with charge states up to 8+ and ions with energy up to 100 eV, a charge state in excess of the single photon ionization threshold of our pulses and an ion energy well above the ponderomotive energy of the XUV pulse. We find that the ion energy spectrum is consistent with a quasineutral hydrodynamic expansion of an 8 eV plasma and inconsistent with a Coulomb explosion, lending strong evidence for the production of a high density nanoplasma in the clusters.

In our experiment, illustrated in Fig. 1, we employed intense pulses of XUV light produced by high harmonic generation (HHG) of an intense femtosecond laser and refocused these pulses into a cluster-producing gas jet. The output of the THOR laser, a Ti:sapphire laser delivering 800 nm, 35 fs pulses of energy up to 500 mJ, was focused into a pulsed jet of argon with a 2 m focal length quartz lens ($f/60$) to an intensity of $\sim 10^{15}$ W/cm^2, and the output of the 21st harmonic at 32.6 eV was optimized. A mask placed 4 m upstream of the lens obscured the central portion of the infrared pulse, and this mask was imaged to an iris 4 m after the focus to remove most of the infrared light; the HHG light is more collimated, and passes through the iris [13]. A 200 nm thick Al filter blocked any residual infrared light. We then selected and focused the 21st harmonic using a spherical Sc/Si multilayer mirror designed to reflect light at 38 ± 5 nm with $f/10$ geometry at near normal incidence [14].

To measure the energy and spatial profile of the focused 21st harmonic, we positioned an XUV sensitive photodiode (AXUV-10, IRD Inc.) after the focus and scanned a knife edge perpendicular to the beam axis. These data are illustrated in Fig. 2. We measured an 8 μm intensity $1/e^2$ half-width and average total energy of 0.5 nJ. Assuming $\sim$15 fs duration for the HHG pulse (which is simply an estimate based on a calculation including harmonic nonlinearity and ionization saturation), this yields a focal intensity of $\sim 5 \times 10^{10}$ W/cm^2.

A second pulsed gas jet nozzle mounted directly above the XUV focus produced a jet of xenon clusters. This jet, with a throat diameter of 500 μm, was backed with a pressure of 13 bar. We used previously published results to ascertain the average size and size distribution of the Xe clusters and estimate that we produced a log-normal distribution of clusters with average size $\langle N \rangle$ ranging up to $\approx 10^4$ atoms [15]. A time-of-flight (TOF) spectrometer with charged grids mounted around the XUV interaction region allowed us to measure the ion charge state distribution by extracting the ions with a field of 50 V/cm. Extracted ions traveled through a field-free region to a multichannel plate. When the extraction grids were grounded, the energetic ions from the explosions of the clusters traveled by their own initial kinetic energy to the multichannel plate detector. In this way, a Xe ion energy spectrum could be directly recovered.

Figure 3 shows the TOF spectrum from the Xe jet produced at various backing pressures when irradiated with the 21st harmonic. High charge states appear at backing pressures where larger clusters are formed (i.e., when $\langle N \rangle \geq 1000$). Multiple XUV photon absorption occurs even in the case of monomer irradiation, resulting in Xe^{3+} production (with some possible evidence for Xe^{4+} probably resulting from the small number of clusters in the jet with $\langle N \rangle = 1$). A broadening of the TOF peaks occurs at large cluster sizes, though we do not believe that this broadening results from kinetic energy of the ions ejected by exploding clusters, which was the explanation assigned to broadening observed in the DESY Xe cluster experiments [8]. We performed Monte Carlo simulations of the TOF spectrum production by modeling the trajectories and resulting TOF signal from an ensemble of ions with various ion energy distributions ejected at the laser focus. We could not reproduce the observed broadening with any reasonable exploding cluster ion energy distribution. Instead, we speculate that this pulse broadening, which manifests itself as a growing feature on the slow side of the peaks from each charge state, results from ions produced by charge exchange between extracted ions and neutral Xe gas sur-

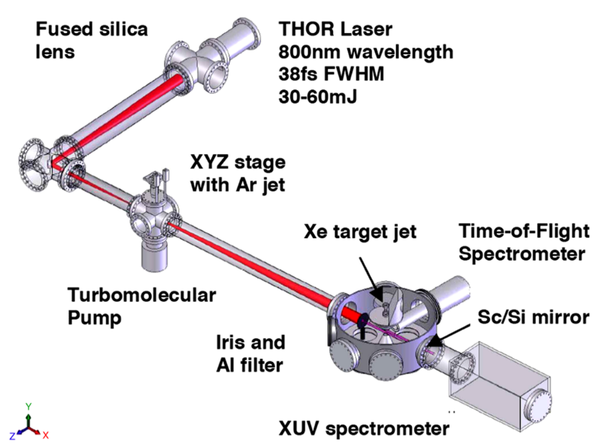

FIG. 1 (color online). Illustration of the high harmonic producing chamber and XUV cluster target chamber. Here pulses of 38 nm light produced by harmonic generation with 35 fs, 800 nm pulses are isolated and refocused in a secondary chamber.

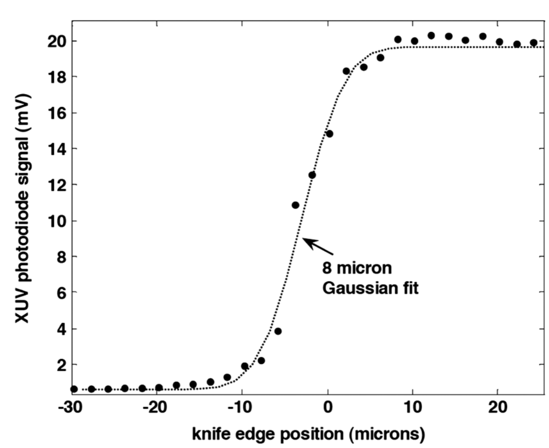

FIG. 2. Measured signal in an XUV sensitive photodiode at the XUV pulse focus passing around a razor blade as a function of blade position in the focus. The solid line is a fit of an 8 μm ($1/e^2$) Gaussian focus.

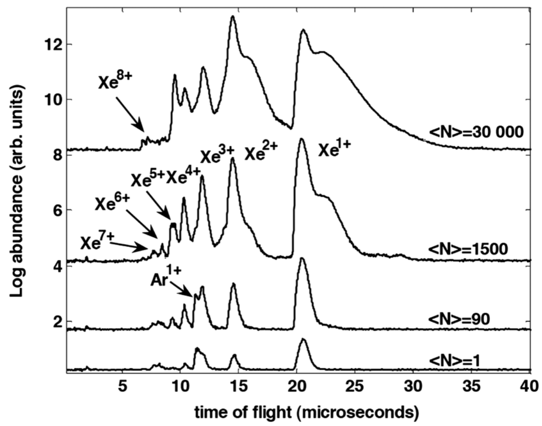

FIG. 3. Ion charge state time-of-flight spectra for various cluster sizes.

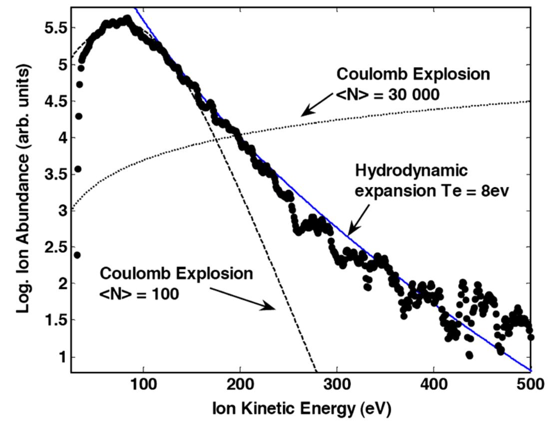

FIG. 4 (color online). Xe ion kinetic energy spectrum from field-free time-of-flight of the exploding cluster ion. The spectrum is fit to a model of Coulomb explosion of large clusters (dotted line) and small clusters (dashed line), as well as the spectrum of a quasineutral expansion of a spherical cluster with an initial temperature of 8 eV (solid line).

rounding the interaction region [16]. In Xe clusters the charge states we observe are high, with a significant number of Xe ions with charge up to 5+ when the cluster size distribution had average size of $\geq$ 1500 atoms per cluster. We also observe a small number of ions with charge states between 6+ and 8+ at the largest cluster sizes. These charge states are remarkably similar to those observed in the DESY experiments at pulse intensity 2 orders of magnitude higher.

The next question that we wished to address was whether these highly charged ions in the cluster exploded by a Coulomb explosion, as has been surmised in the DESY results, both at 100 and 30 nm. To do this we examined the energy spectrum from the exploding Xe clusters by field-free TOF by the ejected ions. Figure 4 shows the energy spectrum of Xe ions ejected from the exploding clusters when the average cluster size is 30 000 atoms. This ion distribution peaks at around 100 eV and has a tail that extends out toward 600 eV.

We calculated the spectrum expected from Coulomb exploding clusters by convolving the spectrum of a single Coulomb exploding cluster with a log-normal cluster size distribution using the procedure described in Ref. [4]; this has been shown to model accurately the explosions of deuterium clusters irradiated by high intensity 800 nm laser pulses [4]. The calculated spectrum for Coulomb exploding clusters with $\langle N \rangle = 30\,000$ (and $Z \sim 1$) is illustrated in Fig. 4. This distribution is very different than the observed spectrum, peaking well above 2 keV. For comparison, the spectrum from a best fit distribution with $\langle N \rangle = 100$ is also illustrated in this figure. Such a Coulomb explosion distribution accurately matches the shape of the ion spectrum peak at 100 eV but does not accurately match the tail of the energy distribution. On the other hand, we also fitted our observed ion spectrum to that of a hydrodynamically expanding quasineutral cluster nanoplasma using the model for a spherical quasineutral plasma detailed in Ref. [17]. This best fit is shown in Fig. 4 and passes through the data over the entire energy range from 10 to 600 eV. This spectrum is consistent with an initial electron temperature in the Xe cluster plasmas of 8 eV, and the fit to our data is excellent over most of the entire ion energy range. (Incidentally, this electron temperature is very close to that of the electron spectrum observed in Ref. [7] from XUV free-electron laser irradiation of Xe clusters.)

Our observed ion energy spectrum is strong evidence that near quasineutral nanoplasmas are formed in our XUV pulse irradiation of the Xe clusters and that the explosion mechanism is not by Coulomb explosion but instead by hydrodynamic expansion. Our measured electron temperature of 8 eV is consistent with the amount of heating we expect by IB in the XUV field, which we have estimated by calculating the electron heating using IB formulas for a plasma with finite electron temperature. (We use the bremsstrahlung absorption rate derived in Ref. [18].) More enigmatic is the production of high charge states under these conditions. The models of both Jungreuthmayer *et al.* [12] and Santra and Greene [10] propose that the high charge states observed in high intensity XUV irradiation of clusters result from electron collisional ionization of ions in the transient nanoplasma that is formed in the cluster (a process that has been well documented in IR irradiated clusters [1]). Such a model has been further explored by Ziaja *et al.* [19]. However, this picture is inconsistent with our data. The equilibrium average charge state $\bar{Z}$ found by numerical solution of the Saha equations for the inner cluster density and 8 eV electron temperature is $\bar{Z} \approx 2.4$. The calculated charge state distribution has no significant number of ions with charge greater than $3+$. We conclude that collisional ionization alone cannot explain our observations.

The model of Siedschlag and Rost, however, suggests that the high charge states result from the distortion of the ionic binding potential by neighboring charged ions [11], an effect not present in a quasineutral cluster plasma because the neighboring ion potentials are screened by electrons. Rost's group does point out in a more recent paper that electron screening is important in determining the extent of the lowering of the ionization potential of ions in the cluster nanoplasma [20]. We believe that our data, which appear to arise from explosions of nearly quasineutral clusters, can be explained within the scope of this picture using a simple ionization potential lowering model. In a dense plasma the presence of an electron cloud around an ion will lower the ionization potential (even when the plasma is effectively neutral) because of the reduced energy needed to liberate an electron into the sea of electrons around the plasma's ions—an effect commonly termed continuum lowering. In our strongly coupled cluster nanoplasmas (where the strong coupling parameter $\Gamma \approx 0.7$), the usual Debye-Hückel model is not appropriate [21,22], but we can use the ion-sphere model [21] to calculate the magnitude of this continuum lowering in our Xe nanoplasmas. In this regime the ionization potential will be lowered by

$$\Delta I_p = \frac{9}{10}(2\bar{Z} - 1)e^2 n_i^{1/3}, \tag{1}$$

where n_i is the ion density in the cluster. This predicts that at the ion density of the unexpanded cluster, which has been ionized to $Z \sim 4$, the ionization potential will be lowered by about 24 eV. This would be sufficient to reduce the ionization potential of Xe^{4+} sufficiently (where $I_p \approx 54$ eV) to allow direct photo-ionization up to Xe^{5+} (as well as the lower charge states up to this point). An estimate of the photoionization cross section of the lowered potential of Xe^{4+} indicates that a high degree of photoionization is possible given the photon fluence of our XUV pulse. This plasma continuum lowering effect will be at work in a completely quasineutral cluster plasma, so it is consistent with the observation of an ion energy spectrum that appears to arise from the hydrodynamic expansion of quasineutral cluster nanoplasmas.

The presence of a small number of charge states with $Z = 6$–$8+$ can then be explained by nonequilibrium collisional ionization of the Xe^{5+} ions produced rapidly ($\sim$20 fs) by photoionization during the XUV pulse by the hot tail of the 8 eV electron distribution. Using the usual Lotz formula for the collisional ionization cross sections [23] with ionization potentials corrected by continuum lowering and integrating this cross section over an 8 eV Maxwellian, we calculate that roughly 5% of Xe^{5+} ions can be collisionally stripped up to Xe^{8+} during $\sim$1.5 ps, the time for our 15 nm, 8 eV clusters to expand hydrodynamically to roughly twice their initial radius. So the presence of small numbers of Xe^{6+}–Xe^{8+} in our data can be explained by electron collisional ionization from the photoionized Xe^{5+}. With the charge states subsequently frozen in place by the subsequent expansion of the cluster, an effect well known in cluster explosion experiments driven by IR radiation [2,3]. We have also estimated the probability of two-photon ionization of Xe^{5+} to these higher charge states and have concluded that the intensity of our XUV pulse is well below that needed to lead to significant ionization to Xe^{6+}–Xe^{8+}.

In summary, we have experimentally studied the explosions of large Xe clusters when irradiated by intense, femtosecond pulses of XUV radiation produced by high order harmonic conversion of a Ti:sapphire laser. We find that the ion spectrum of the explosion is consistent with that of the expansion of a quasineutral plasma with electron temperature of 8 eV and inconsistent with a Coulomb explosion. Charge states up to 5+ observed in these explosions are produced by photoionization of ions whose ionization potentials have been depressed by plasma continuum lowering and a small fraction of ions are further stripped by nonequilibrium electron collisional ionization.

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