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Femtosecond time-resolved studies of the dynamics of noble-gas cluster explosions

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We have examined the dynamics of noble-gas clusters, heated with high-intensity laser radiation, using pump-probe experiments to temporally resolve the expansion of the clusters. Absorption of the probe radiation is observed to reach a maximum for a particular time delay between pump and probe, dependent on the cluster size. For single-pulse experiments, we find that there is an optimal pulse width to maximize absorption for a given cluster size. Model calculations suggest that these effects are due to resonant heating of the spherical cluster plasma in full support of a hydrodynamic interpretation of cluster interactions.
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High-intensity laser interactions with atomic clusters have been an active area of research in the past few years [1–10]. The high local density of cluster targets greatly increases the coupling of the laser energy to the atoms. In recent years, groups have observed keV electrons [4], MeV ions [5], as well as x rays in the keV energy range [6,7]. Additionally, there have been studies looking at x-ray emission dependence on laser wavelength [8] and absorption of laser energy [9]. More recently, experimental work examining the dynamics of cluster disassembly has been performed [1,10]. Lezius *et al.* [10], for example, showed that the mechanism for cluster expansion in the argon clusters they studied was Coulomb explosion, while in xenon it was a combination of Coulomb explosion and hydrodynamic expansion.

There have been many theories to explain these results. McPherson *et al.* [11] described cluster interactions with a process where collective excitation of the outer-shell electrons produces inner-shell vacancies. Based on previous work by Boyer and Rhodes [12], this theory was used to interpret many experimental results [7,8]. Rose-Petruck *et al.* [13] created an ''ionization ignition model'' where the combined field of the laser and the closely spaced cluster ions cause rapid ionization of the cluster. In contrast to these theories, Ditmire *et al.* [3] proposed a hydrodynamic model of the cluster excitation dominated by collisional ionization and inverse bremsstrahlung heating. This model treats a cluster as a small plasma. In order for this to be valid the clusters must be large enough that the charge on the cluster, from electrons that free-stream away, confines the majority of electrons to the cluster region [11]. We expect to see resonance effects when the natural frequency of the plasma equals the laser frequency, similar to resonance absorption in a bulk plasma. In a bulk plasma this will occur when the electron density equals the critical density, $n_e/n_{\mathrm{crit}}=1$ ($n_{\mathrm{crit}}=m_e\omega^2/4\pi e^2$). The resonance condition for a spherical cluster will not occur when $n_e/n_{\mathrm{crit}}=1$, but rather $n_e/n_{\mathrm{crit}}=3$. This follows from making the dipole approximation for the electric field inside of a cluster with appropriate boundary conditions [3,14]. While evidence for the hydrodynamic model has been growing [1,10], there has been no definitive work, and debate continues [6,8]. In this Rapid Communication we report on a series of experiments in which we temporally resolve the expansion of heated clusters on a femtosecond time scale. Our goal was to observe direct evidence of the resonant heating. We find that there is a very clear expansion time for maximum absorption, dependent on cluster size, clearly demonstrating the effects of resonance in support of the hydrodynamic model.

The laser used for these experiments was a Ti:sapphire laser system operating at a wavelength of 810 nm, capable of producing about 50 mJ in a 50-fs pulse [15]. We could vary the laser pulse length by changing the grating spacing in the compressor [16]. The beam could also be directed through a beam splitter, with one beam going into a variable delay leg, to perform pump-probe style experiments. To minimize the accumulated nonlinear phase, we expanded the beam to a diameter of 8 cm in an all-reflective telescope before passing through the fused silica target chamber window. The beam was then focused with an $f/3$ off-axis paraboloid onto a cluster target. Approximately 70% of the energy was contained in a 9-μm Gaussian spot, with the remaining energy in low-intensity wings. The clusters were produced by expansion in a supersonic Laval nozzle backed with up to 200 psi of xenon or 600 psi of argon. From a calculation of the Hagena parameter, confirmed through Mie scattering, we estimate the average number of atoms per cluster N to be up to 6×10^5 for xenon and 5×10^5 for argon [17,18]. The cluster radii ranged from 85 Å to 205 Å for xenon and 110 Å to 165 Å in argon. The average atomic density 1 mm below the nozzle, where the laser was focused, is estimated from our absorption calculations to vary from approximately 1.7×10^{17} to 6.9×10^{17} atoms/cm^3 for xenon and from 1×10^{18} to 2×10^{18} atoms/cm^3 for argon. This is in reasonable agreement with previous density measurements of these jets [19]. The full width of the jet is about 1 mm.

The input energy of the laser was measured with a pyroelectric energy meter, measuring transmitted light through one of the mirrors before the target. The laser energy transmitted through the target was collected with an $f/2.3$ lens and measured with a similar energy meter. We checked for scattered laser light (90° and 45° forward) by imaging the interaction region onto a silicon detector with an $f/2.3$ lens. Additionally, we looked for backscattered light by placing a diode behind the dielectric mirror before the entrance of the chamber. We found that only a negligible amount of energy was scattered ($<1\%$ over 4π sr), so the absorbed energy was

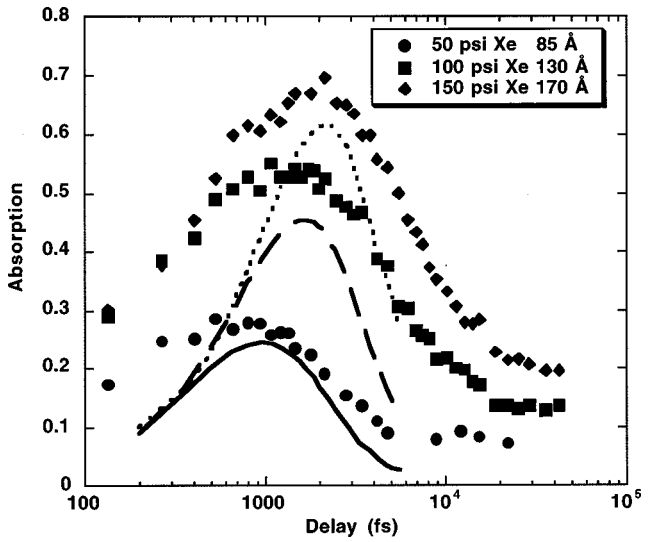

FIG. 1. Absorption measurements for pump-probe experiments. The target is xenon at 50, 100, and 150 psi. The pump pulse was 10% of the 5.4-mJ probe pulse. The peak intensity in vacuum for the pump is 1.6×10^{16} W/cm^2 and for the probe it is 1.6×10^{17} W/cm^2. The pulse length was 50 fs. Average cluster radii are given in the legend. The lines show the results of computer modeling for 50 (solid), 100 (dashed), and 150 (dotted) psi.

taken to be the difference between the input and output energies. The absorption measurements were averaged over 50 shots. We also utilized a variable line-spaced, grazing incidence soft-x-ray spectrometer [20] to look at plasma emission with wavelengths between 170 Å and 300 Å.

In the pump-probe experiments a small (10%) pump pulse was sent into the target to expand the clusters before the probe pulse arrived. This allowed us to probe the disassembly of the clusters as they expanded into a bulk plasma. Both pulses were about 50 fs in length. The probe pulse energy was 5.4 mJ and had a peak intensity in vacuum of 1.6×10^{17} W/cm^2. Figure 1 shows absorption as a function of probe delay for different backing pressures of xenon. The absorption peak in Fig. 1 indicates the presence of resonant heating as the clusters expand. At very small delays, absorption is low; the cluster has little time to expand and the second pulse arrives long before the electron density reaches the resonance condition ($n_e/n_{\rm crit}=3$). However, as we increase the delay, absorption increases. The longer delay allows the electron density to be near the resonance condition when the pulse arrives, greatly enhancing the absorption of laser light. For longer delays, the cluster has continued to expand and the probe pulse is poorly absorbed by the, now underdense, plasma. Eventually, we reach a condition in which the local atomic density within a cluster equals the average atomic density in the jet. At this point the clusters are fully disassembled; the discrete spheres have combined to form an underdense bulk plasma. Hence, there is no enhancement from the presence of clusters; the absorption approaches a constant depending only on the average gas density.

The lines in Fig. 1 show calculations based on a computer model similar to the one first described in Ref. [3]. In contrast to Ref. [3], however, we calculate the cluster absorption with a Mie scattering code [21], using the previously described Drude model for the cluster index of refraction. For the purposes of laser absorption due to inverse bremsstrahlung heating, Ditmire *et al.* [3] assumed the cluster to be much smaller than the wavelength of light. This is acceptable for a short pulse, since the duration of the pulse is small compared to the expansion time of the cluster. In this work, we are examining that expansion; and after some delay time, this assumption will no longer be reasonable. The Mie code gives us an accurate absorption cross section for any cluster size. The dielectric constant from the Drude model can be used with the Mie code for computing absorption since ionization takes place before the bulk of the laser pulse arrives. In our experiments the Rayleigh range was smaller than the jet length; hence the beam waist is changing through the target. We accounted for this by adjusting the beam waist through the jet in our code, based on a Gaussian beam propagating through focus. The overall model considers a uniform density cluster plasma, taking into account tunnel and collisional ionization, ATI and inverse bremsstrahlung heating, along with the hydrodynamic and Coulomb explosion aspects of the expansion. To account for the distribution of cluster sizes in the jet we ran our code for several cluster sizes and then took a weighted average. While we do not know the exact distribution, previous work in hydrogen [22] shows an asymmetric distribution biased toward larger clusters. Similar to this work we used a superposition of a Gaussian distribution with a full width at half maximum equal to the average cluster size and an exponential distribution producing a tail of large clusters. These results, plotted in Fig. 1, show the trend of larger clusters requiring longer periods of time to expand to resonance. The effect of adding the distribution widened the peaks slightly, but produced little overall effect. The model peaks are somewhat narrower than experiment. This discrepancy may be due to our incomplete knowledge of the exact cluster distribution in the jet. However, the main problem is most likely the limitations of the code itself. The uniform cluster density model does not fully describe the hydrodynamics involved in the expansion process. As gradients develop, the resonance will spread out and not be so clearly peaked. Additional heating mechanisms, not accounted for in the code, might also be adding to the discrepancy. The fluid model does not account for energy deposited when electrons leave the cluster and later reenter the cluster surface. This effect, which is similar to Brunel absorption [23], should be maximum for short pulse lengths (high intensities) and sharply bounded overdense plasmas.

The optimal delay clearly increases with increasing backing pressure. It is well known that the average number of atoms in a cluster N scales as the square of the backing pressure [24]. This leads to the cluster radius r scaling as $r\sim p^{2/3}$. Looking at the expansion of a uniform density sphere with the surface moving at a constant velocity, the time for the cluster to expand to the resonance condition τ_R is simply

$$\tau_R=\frac{r_0}{v}\left[\left(\frac{n_0}{3n_c}\right)^{1/3}-1\right], \tag{1}$$

where r_0 is the initial radius, n_0 the initial density, n_c the critical density, and v is the expansion velocity. From this we would expect τ_R to scale with $p^{2/3}$. Due to the effects of the second pulse the actual scaling should be slightly differ-

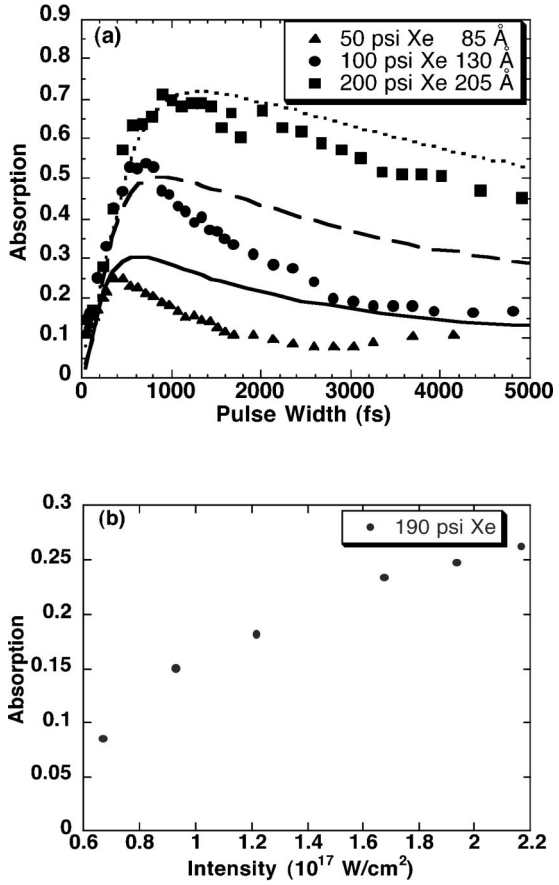

FIG. 2. (a) Absorption measurement for variable pulse length experiments. The target was xenon at 50, 100, and 200 psi. The input energy was ~6.5 mJ, corresponding to a peak intensity in vacuum of 2.3×10^{17} W/cm^2 for a 50-fs pulse. The lines are calculated values for 50 (solid), 100 (dashed), and 200 (dotted) psi. Sizes given in the legend are the average cluster radii. (b) Absorption measurements vs intensity for 190-psi xenon using a 50-fs pulse. In this case the intensity was varied by adjusting the input energy.

ent; our model predicts this scaling to be $p^{0.77}$. From Fig. 1, we see peak absorption occurring at about 800-, 1400- and 2100-fs delays for 50, 100, and 150 psi, respectively. These scale as $p^{0.85}$, which is in reasonable agreement with the predicted value.

Not only do the large clusters reach resonance later, but the resonance peak is broader. If we measure the width of the resonance at 70% of the maximum absorption we find that it is about 2, 3, and 5 ps for 50, 100, 150 psi backing pressures, respectively. In a larger cluster the rate of change in density is less than that for a smaller one. Hence, a larger cluster will be near resonance for a longer period of time, increasing the absorption of laser light. A smaller rate of change in density would also indicate a longer disassembly time. The three curves flatten out, indicating full cluster disassembly, after different delays with larger clusters taking longer periods of time to reach this point. The largest clusters are fully disassembled after 35 ps.

Figure 2(a) shows the results of absorption measurements with a variable pulse width for several pressures. The laser energy is about 6.5 mJ, and a peak intensity is 2.3×10^{17} W/cm^2 in vacuum for a 50-fs pulse. When the pulse is short the absorption is about 15%. At 50 fs, the pulse

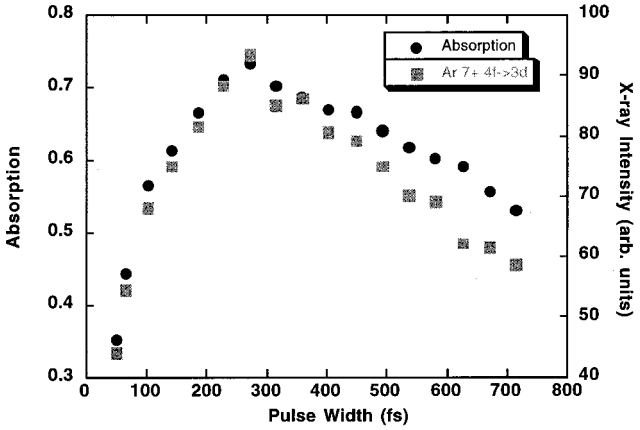

FIG. 3. Measured absorption and x-ray yield for argon, at 300-psi backing pressure, for different pulse widths. Peak intensity in vacuum is 1.9×10^{17} W/cm^2 for a 50-fs pulse.

length is small compared to the hydrodynamic expansion time. An overdense plasma ($\sim75\ n_{crit}$) is formed from each cluster, but by the time it expands to the resonance condition, the laser pulse is already past. As the pulse gets longer the resonance begins to occur during the laser pulse, enhancing the absorption. Eventually, the absorption begins to fall off as the resonance occurs early in the pulse and most of the laser energy interacts with a bulk plasma with relatively low density. The model clearly shows the trend of larger clusters exhibiting optimum absorption at longer laser pulse durations. The general time scales also show reasonable agreement. The limitations of the code are seen at longer pulse lengths, most likely accounted for by bulk plasma and nonlinear propagation effects that the code does not consider.

The optimal pulse width is much shorter than the optimal delay for the pump-probe experiments. This is explained by the structure of the laser pulses in the two experiments. In the pump-probe experiments the pump energy is deposited and then the cluster undergoes a free expansion until the probe pulse arrives. The velocity of the expansion is set by the

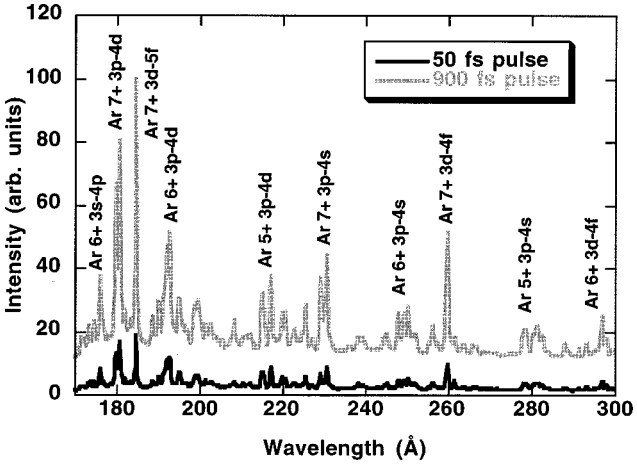

FIG. 4. Spectra for 600-psi argon. Input energy was ~5.4 mJ for both graphs. The 50-fs pulse corresponds to a peak intensity in vacuum of 1.9×10^{17} W/cm^2 and the 900-fs pulse corresponds to 1.1×10^{16} W/cm^2. The spectrum for 900 fs is offset 10 arb. units for clarity.

small pulse, and no acceleration takes place during the delay period. When we use a single pulse, energy is constantly being deposited in the cluster; the atoms continue to accelerate throughout the laser pulse. This leads to higher expansion velocities and the resonance condition is achieved sooner.

Since we are keeping the energy constant in these experiments, the intensity increases as we shorten the pulse. It has been previously observed, using a 2-ps pulse, that as the laser intensity increases, the absorption saturates and eventually decreases [9]. To see if the increasing intensity was responsible for the decrease in absorption at the shortest pulses, we measured the absorption of laser light as a function of intensity, keeping the pulse width constant and varying the input energy. The shortest pulse of 50 fs was used, since this produces the highest intensities and would therefore cause the most saturation. The results of these experiments are shown in Fig. 2(b). Over the intensity range in which these experiments were performed, the absorption is a monotonically increasing function of the laser intensity, clearly indicating that it is the change in pulse width and not the change in intensity that is responsible for the results in Fig. 2(a). This shows that results taken with different lasers that have different pulse widths, but the same intensity [8], cannot be freely compared; energy and pulse width must be considered independently.

Figure 3 shows both soft-x-ray yield for the sodiumlike argon $3d-4f$ transition and laser absorption. The target was argon at 300 psi. The x-ray yield tracks the absorption very closely. Peak absorption (or x-ray yield) occurs for a pulse just under 300 fs. This shows that the absorption that we measure is an accurate indicator of x-ray yield.

The spectra from argon (600 psi) at two different pulse lengths are shown in Fig. 4. The spectra were integrated over 20 shots. The pulse length was 50 fs, corresponding to a peak intensity in vacuum of 1.9×10^{17} W/cm^2 for the upper plot; and the pulse length was 900 fs, corresponding to 1.1×10^{16} W/cm^2 for the lower one. The input energy is the same in both graphs ($\sim$5.4 mJ). The x-ray yield increased almost 400% by making the pulse length 18 times longer. Although the intensity has dropped by over an order of magnitude, the two spectra look similar. Collisional ionization still produces a significant amount of sodiumlike argon at 1.1×10^{16} W/cm^2, while tunnel ionization does not [25].

In conclusion we have demonstrated that spherical resonance absorption plays an important role in cluster interactions with high-intensity lasers, in full support of a hydrodynamic model of cluster interactions. Using a pump-probe setup, we find that absorption is maximum for a certain delay, depending on cluster size. For a given cluster size there is an optimal pulse width to maximize absorption of the laser pulse. This maximum absorption corresponds to maximum soft-x-ray emission, and we can control the x-ray conversion efficiency by temporal control of the laser pulse.

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