X-ray yields from Xe clusters heated by short pulse high intensity lasers

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We have examined the absolute yield of keV x-rays emitted from gaseous plasmas created by the intense irradiation of large Xe clusters. We find that >10 μJ of x rays with photon energies above 1 keV are produced from clustering Xe gas targets when heated by 250 mJ, 2 ps laser pulses at an intensity of $\sim 10^{17}$ W/cm^2. The yields show strong laser intensity dependence and variation with Xe cluster size. © *1997 American Institute of Physics.* [S0003-6951(97)02728-9]

The characteristics of x-ray emission from plasmas produced by high intensity lasers have been extensively studied over the past decade. Short pulse interactions with solid target plasmas have been of great interest as they represent a unique, high brightness source of x rays.[1] Many applications of such compact, high repetition rate sources exist, such as soft x-ray lithography,[2] however, one persistent problem with solid target plasmas has been the production of debris from laser ablation.[3] The x-ray emission properties of gas targets have also been investigated,[4] though gases in general do not emit x rays as strongly as solids.[5]

Very recently, experiments examining the x-ray emission resulting from the intense irradiation of gases composed of atomic clusters of a few hundred to a few thousand atoms per cluster have been reported.[6,7] These interactions appear to be quite different than laser interactions with either solids or monomer gases. Strong (XUV) and soft x-ray emission has been observed from clusters of the heavier noble gases (Ar, Kr, and Xe) and x rays with energies up to 5 keV have been observed from the intense irradiation of Xe clusters.[6] These clusters represent a unique medium; though their average density is low, the density within each cluster is near solid density, and so the collisional processes which lead to efficient laser energy absorption and strong x-ray emission are present.

Though the spectral and temporal characteristics of the x-ray emission from cluster targets has been reported, to date no data exists on the absolute yield of x rays from gases of clusters irradiated by high intensity lasers. Such studies are of interest not only because they illuminate the differences between solids, gases, and clusters as laser targets but also because of the potential application of a gas target as a debris-free source of x rays for proximity lithography. Possibilities for developing an efficient short pulse, high brightness source of x rays also exist, a source of particular interest for experiments requiring time resolution. Xe represents a particularly strong candidate for such studies: it is a high Z element with a strong propensity to cluster. In this letter, we report a study of the absolute yield of keV x rays emitted from gaseous plasmas created by the intense irradiation of Xe clusters.

The experimental set-up for these investigations is shown in Fig. 1(a). The laser used was a Nd:glass laser based on chirped pulse amplification which produced 2 ps full width at half-maximum (FWHM) pulses at 1054 nm with energy up to 0.5 J. These pulses were frequency doubled in a KDP crystal (yielding up to 280 mJ of 527 nm light) and were focused into the output of a xenon gas jet with an $f/12$ lens, yielding a maximum peak intensity of 1.5×10^{17} W/cm^2. The laser was focused 2 mm below the nozzle of a solenoid valve gas jet producing a plasma channel approximately 2.5 mm long. This jet has been fitted with a cooling jacket and can be cryogenically cooled from room temperature down to -170 °C.[8] Xe clusters are formed in the gas jet due to the cooling of the gas associated with its adiabatic expansion into vacuum. The average size of the clusters can be roughly controlled while keeping the average gas density constant by cooling the jet's backing reservoir.

The laser energy absorption efficiency of the Xe cluster jet and the plasma x-ray yield were monitored simultaneously. A detailed account of the absorption measurements is presented in Ref. 9. In brief, the input energy of the laser pulse was monitored with a fast photodiode, which also monitored backscatter of laser energy from the target. (No significant backscatter was observed in these experiments.)

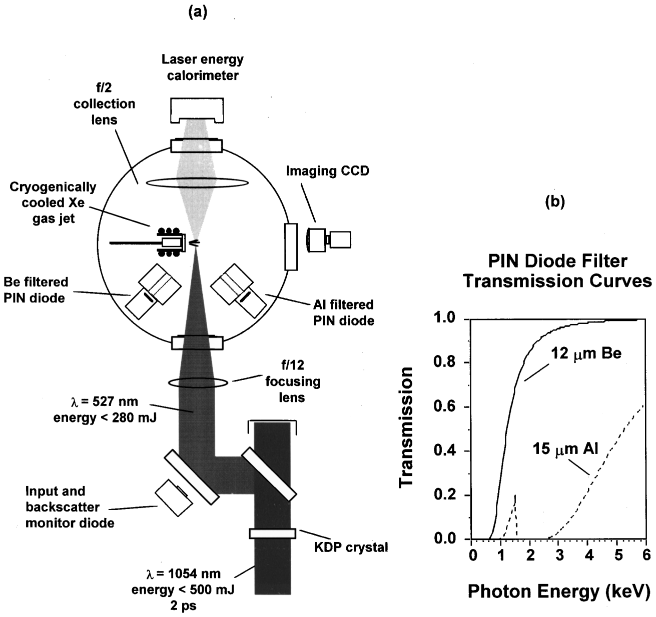

FIG. 1. (a) Experimental layout for measuring x-ray yields from Xe cluster gas jet. (b) Transmission curves for the filters on the two PIN diodes.

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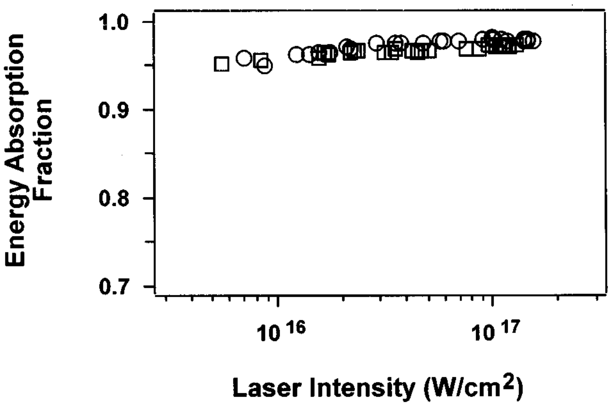

FIG. 2. Measured absorption efficiency of the laser in the Xe gas jet backed with 35 bar as a function of laser intensity. Gas jet reservoir temperature was 20 °C (squares), -85 °C (circles).

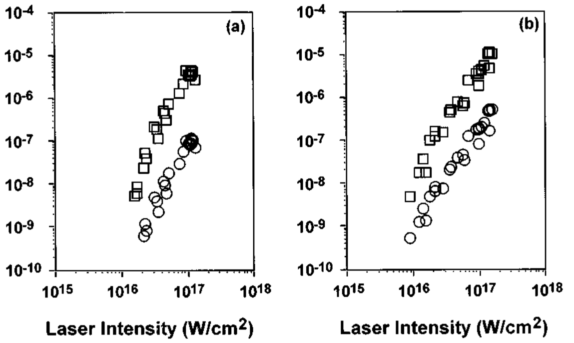

FIG. 3. Integrated x-ray yield into 4π from the Xe jet backed with 35 bar at a temperature of 20 °C (a) and -85 °C (b). Squares are the yield measured by the Be filtered diode; circles are the yield measured by the Al filtered diode.

The laser energy transmitted through the Xe gas jet was collected with an $f/2$ lens (to collect all refracted light) and measured with a large aperture energy calorimeter.

The time integrated x-ray yield of the Xe cluster plasma was measured with two silicon PIN photodiodes (UDT XUV-100), optimized for absolute measurement of x-ray yield from laser produced plasmas. Each diode was located 45° from the laser axis and both were placed 14 cm from the plasma. Any hot electrons produced in the plasma were deflected from the front of the detectors with a 0.1 T magnetic field located 6 cm in front of the diodes. A 12-μm-thick Be filter placed in front of the first diode blocked radiation with photon energy below 1 keV, and a 15-μm-thick Al filter placed in front of the second diode filtered most photons with energy below 3 keV. The calculated transmission curves of the Be and Al filters are shown in Fig. 1(b).

We examined x-ray yields and absorption by the Xe gas jet over a range of backing reservoir pressures and temperatures. The average size of the Xe clusters in the gas jet can be simply estimated using published scaling laws for cluster formation.[10] We have also previously conducted Rayleigh scattering measurements on the clusters produced in the gas jet which allow an additional check on the average cluster size produced.[11] For the data presented in this work, the jet was operated with a backing pressure of 35 bar, yielding an estimated average atom density of $\sim 1\times10^{19}$ atoms/cm^{-3}. Under these conditions, Xe forms large clusters even with the jet at room temperature, and all Xe atoms have condensed into clusters.[10] At room temperature (20 °C), we estimate that the average Xe cluster diameter was approximately 200 Å. When the jet reservoir was cooled to -85 °C we estimate that the average Xe cluster size grew to 400 Å.

We find that over the range of intensities studied the Xe cluster jet is very efficient at absorbing laser energy. Figure 2 shows the faction of energy absorbed by the Xe cluster jet with 35 bar backing pressure and two different jet temperatures as a function of laser intensity. We find that in both cases nearly 100% of the incident laser energy is absorbed in the gas jet over the intensity range from 5×10^{15} to 1.5×10^{17} W/cm^2. This very high absorption in a clustering medium is in sharp contrast to that of a gas of single atoms alone. It is a result of the large collisional inverse bremstrahllung absorption by the electrons in the high density clusters. This large absorption results in a very high temperature plasma.[7]

Consequently, the resulting x-ray yield from this plasma can be quite high. The measured yield of x rays emitted into 4π steradians is illustrated in Fig. 3. Figure 3(a) shows the integrated yield of x rays above 1 keV and above 3 keV as a function of laser intensity from Xe plasmas produced in the jet backed with 35 bar of pressure and the jet at 20 °C. We observe up to 5 μJ of x-ray energy emitted above 1 keV from the Xe plasma. The yield above 3 keV is substantially lower, <0.2 μJ. Figure 3(b) shows the same data taken when the gas jet was cooled to -85 °C. In this case, the maximum yield above 1 keV rises to 12 μJ at the highest laser intensity and the yield above 3 keV increases to 0.5 μJ.

The x-ray yield data in both cases follow a well defined power law dependence with changing laser intensity. The laser intensity was varied by changing the amount of laser energy focused into the jet. The yield above 1 keV in the room temperature Xe jet scales quite strongly with laser intensity. It varies as yield(Be)$\sim I^{3.0}$. The yield through the Al filter nearly follows this scaling law as well, [with yield(Al)$\sim I^{2.8}$]. The yield from the cooled jet exhibits a slightly slower yield dependence with yield(Be)$\sim I^{2.4}$ and yield(Al)$\sim I^{2.3}$. The strong dependence with laser intensity is particularly remarkable since the lack of variation in the laser energy absorption data indicates that the energy deposited into the plasma is rising only linearly with a linear increase in intensity. This favorable scaling with laser intensity indicates that higher conversion efficiency into x rays may be possible with modest increase in laser intensity. The yield will ultimately saturate with increase in intensity, however, we expect that the yield (and conversion efficiency) can be substantially increased since the observed x-ray energy is still a very small fraction of the deposited laser energy in the plasma.

From the great difference between the yields from the two diodes, it is clear that the majority of the observed x-ray emission rests in the 1–3 keV region for both jet temperatures. We attributed this to the strong M-shell emission bands in Xe in the 8–15 Å range (0.8–1.6 keV).[5] Strong emission in this wavelength range has been previously observed in Xe cluster targets heated with a high intensity, 300 fs KrF laser.[12] This is the wavelength range of particular interest for proximity lithography applications.

The most striking aspect of this data is that the measured x-ray yields are comparable to yields observed from high intensity short pulse illumination of solid targets. For example, Gordon *et al.* have reported x-ray yields from planar aluminum and gold targets irradiated by 120 fs pulses with energies up to 200 mJ and intensity up to 10^{18} W/cm^2.[13] These measurements found that integrated x-ray yields above 1 keV were about 2 μJ from Al targets and just above 10 μJ from Au targets for input laser energies similar to ours. Though our measured yields are not as high as those reported in Ref. 13 from microstructured targets, the yields are comparable to those found from planar solids.

This result is in itself quite remarkable since yields from gas targets are typically expected to be substantially lower than from solids due to the gas's much lower average atomic density. For example, Celliers *et al.* found that x-ray yields from plasmas produced by a 30-J-long pulse (13 ns) Nd:glass laser in the vicinity of 1 keV were much lower in Xe gas jets than from planar solid targets.[5] The difference in behavior we observe for short pulse irradiation can be attributed to the unique nature of the laser interaction with the clusters in the gas. Though Celliers *et al.* did not consider the possibility of clusters in their Xe gas plume, the presence of clusters in those long pulse laser experiments is expected to be insignificant. Any clusters in the jet will disassemble on a picosecond time scale.[7] Consequently, a ns laser pulse will interact predominantly with a low density plasma without any clusters. A short pulse laser, on the other hand, interacts with inertially confined clusters of high local density. Thus, because of the high collisionality inside the cluster, the laser absorption can be quite large, a phenomenon manifested in Fig. 2. To a short pulse laser, the cluster medium approximates a solid in many respects, largely explaining the similarity between x-ray yields in our Xe cluster gas with those from high Z planar targets like Au.

Despite some similarities with solids, important differences in the cluster interactions do exists. In general, the specific absorption and energy transport characteristics of the cluster target are quite different from those of solids. One important experimental difference can be seen in the power law dependence of the x-ray yields from the clustering gas jet which are faster than those found from the solid targets. In the experiments of Gordon *et al.*, the yield scaled as $I^{1.7}$ for Au targets and $I^{2.1}$ for the Al targets.[13] This may be due to the nonlinear nature of the heating in the expanding cluster. For example, the cluster microplasmas can exhibit collective electron absorption, a mechanism absent in solids.[14]

Finally, we see that there is some dependence of the x-ray yield on cluster size. The yield from the larger 400 Å Xe clusters is roughly a factor of two higher than that from the 200 Å clusters. This may be a result of the fact that larger clusters disassemble more slowly than smaller clusters in the laser field.[7] The larger clusters maintain their solidlike character longer, a fact which has a number of consequences for the nature of the x-ray emission. Nonetheless, this scaling suggests that an optimum cluster size may exist for maximum conversion efficiency to x rays.

In summary, we have reported a measurement of the absolute energy yields of x rays produced by the intense irradiation of a gas of Xe clusters with a 2 ps laser. We find that x-ray yields of over 10 μJ in the x-ray range above 1 keV are possible with under 300 mJ of laser energy. The Xe clusters formed in the gas jet are very efficient at absorbing short pulse laser light and, consequently, result in x-ray emission that is close to that previously observed from solid target plasmas. Furthermore, the yields appear to exhibit some dependence with cluster size, suggesting that it may be possible to optimize the output of the x-ray conversion efficiency for practical applications.

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