10-1-1995

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Recommended Citation

T. Ditmire, T.D. Donnelly, R.W. Falcone, M.D. Perry, "Strong X-ray Emission from High-Temperature Plasmas Produced by Intense Irradiation of Clusters," Phys. Rev. Lett. 75, 3122 (1995). doi: 10.1103/PhysRevLett.75.3122

Strong X-Ray Emission from High-Temperature Plasmas Produced by Intense Irradiation of Clusters

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(Received 14 June 1995)

The interaction of an intense laser pulse with large ($\sim$100 Å) clusters present in pulsed gas jets is shown to produce novel plasmas with electron temperatures far in excess of that predicted by above-threshold ionization theory. The enhanced absorption of the laser light by the dense clusters results in the production of high ion charge states via collisional ionization resulting in strong x-ray emission from the hot plasma.

PACS numbers: 52.50.Jm, 36.40.Vz, 52.25.Nr

Much effort has gone into the understanding of the interaction of short ($\leq$1 ps), intense ($>10^{15}$ W/cm^2) laser pulses with matter [1]. Many of these experiments have investigated the production of short wavelength radiation or the generation of energetic electrons. Substantial progress has been made in the generation of bright, coherent soft x rays in the 30 to 100 eV range from underdense plasmas and neutral gases via harmonic generation [2] or x-ray lasers [3]. Incoherent emission extending to the megavolt range has also been observed from the interaction of these intense, short pulses with dense targets [4,5].

High pressure gas jets undergoing rapid expansion produce a unique combination of both gas and solid phase components providing an interesting medium for the study of high intensity laser-matter interactions [6]. Solid density clusters form in the jet, resulting from the cooling associated with the adiabatic expansion of the gas into vacuum. This cooling causes the gas to supersaturate and nucleate. Under appropriate conditions, when the gas jet stagnation pressure exceeds a few atmospheres, the clusters formed in the expanding jet can be quite large ($>10^4$ atoms per cluster) for gases such as Ar, Kr, N_2, and Xe [6]. We have observed anomalous short wavelength emission from these gas jets when irradiated by an intense laser pulse with a duration less than the cluster disassembly time. We find that the production of the anomalous charge states is dominated by collisional heating of large clusters in the laser field during the laser pulse. Time resolved spectroscopy shows that this rapid heating is followed by x-ray emission occurring on a relatively long time scale. In contrast with an earlier report [7], this emission is well described by emission in the underdense plasma that results after the heated clusters expand.

These experiments were motivated by the hypothesis that an adiabatically cooled gas jet might be expected to exhibit enhanced laser absorption relative to a conventional low density gas target due to the presence of solid density clusters. The effect would be significant only if the duration of the laser pulse was less than the expansion time of the cluster. During and after irradiation, the clusters will expand to produce a system with the radiative and kinetic properties of a bulk low density plasma. The presence of clusters has previously been shown to affect laser absorption on solid targets of porous Au black [8] and result in the emission of x rays from charge states in gas targets far beyond those which could be produced by above-threshold ionization (ATI) [9].

Up to an irradiance of approximately 10^{17} W/cm^2, absorption of femtosecond pulses in a low density gas is primarily through strong field ionization. Radiation emitted by the plasma following the laser pulse is well described by conventional radiative and three-body recombination into charge states produced by strong field ionization of individual atoms. The plasma temperature is determined by the single atom ATI energy distribution [10]. Only when the laser intensity approaches 10^{18} W/cm^2 do other heating mechanisms, such as stimulated Raman scattering, become important [11].

The presence of high density clusters makes inverse bremmstrahlung a significant method of absorption and heating due to the high collisionality within the cluster. If the cluster diameter is smaller than a skin depth, this heating occurs nearly uniformly throughout the cluster. Model calculations that include the effects of cluster expansion and electron free streaming during the laser pulse suggest that this mechanism dominates the absorption and heating of clusters with diameters in the 50–150 Å range when irradiated by femtosecond laser pulses. For a 130 fs, 825 nm laser with peak intensity of between 10^{14} and 10^{17} W/cm^2, we find that the electron temperatures reach 1 to 2 keV within the cluster. These hot electrons produce high charge states (e.g., $Z > 8$ for Ar and $Z > 10$ for Kr) in the cluster by collisional ionization before it expands.

The hot cluster microplasmas will undergo rapid hydrodynamic expansion into the surrounding underdense plasma during and after the laser pulse. The time for the density within the cluster to drop from the solid density present initially n_0 to the surrounding ambient density n_s is a measure of the expansion time of the cluster and can

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be estimated by assuming a sonic expansion. The resulting cluster expansion time is approximately

$$\tau_{\mathrm{ex}} \approx r_0 \sqrt{\frac{m_i}{ZkT_e}} \left(\frac{n_0}{n_s}\right)^{1/3}, \qquad (1)$$

where r_0 is the initial cluster radius, kT_e is the cluster electron temperature, and m_i is the ion mass. For an argon cluster (initial lattice spacing ~3.8 Å) with an initial radius of 50 Å, an initial electron temperature of 1 keV, a $Z \approx 8$, and a surrounding bulk plasma with a density of 10^{18} atoms/cm^3, the expansion time is approximately 1 ps. After the cluster expands, the plasma dynamics are determined by the underdense uniform plasma.

Our experiments were performed with a Cr:LiSAF laser that produces 0.5 J pulses at 825 nm with 130 fs pulse duration [12]. The supersonic gas jet used in these experiments is a Mach 8 Laval nozzle that produces atom densities of $(1-5) \times 10^{18}$ atoms/cm^3 for backing pressures of 100 to 700 psi [13]. For all experiments the laser was focused ~1 mm from the nozzle output, which is 1.4 cm from the jet throat. We can estimate the extent of the atomic clustering for this jet from the empirical gas jet scaling parameter of Hagena [14,15], which is given by

$$\Gamma^* = k \frac{(d/\tan\alpha)^{0.85} p_0}{T_0^{2.29}}, \qquad (2)$$

where d is the jet throat diameter in μm (150 μm for our jet), α is the jet expansion half angle (~5° for our jet), p_0 is the backing pressure in mbar, T_0 is the initial gas temperature (~298 K for our experiments), and k is an empirical constant ($k \approx 2900$ for Kr, 1700 for Ar, 180 for Ne, and 4 for He [15]). Clustering begins when this parameter exceeds ~300 [6,14,16] and large clusters ($>10^4$ atoms/cluster) predominate when $\Gamma^* > 5 \times 10^4$ [7,15,16]. This parameter varied from $\sim 1 \times 10^4$ to 1×10^5 for Ar and $\sim 2 \times 10^4$ to 2×10^5 for Kr backing our jet with 100 to 600 psi, respectively. It never exceeded 5000 for Ne and He in our experiments.

We confirmed the presence of large clusters in this gas jet under standard operating conditions by a series of Mie scattering measurements. The gas at the output of the jet nozzle was irradiated with approximately 1 μJ of light from the frequency doubled LiSAF laser at 412 nm. The centerline of the approximately 1 mm diameter flow was probed with a beam of approximately 400 μm in diameter. The 90° Mie scattered light was collected with a lens and imaged onto the face of a photomultiplier tube. Figure 1 shows the scattered light signal as a function of backing pressure for He, Ne, Ar, and Kr. No significant light scattering above the noise level is observed from either Ne or He over the range of backing pressures studied. The scattered light signal from the expansion of Ar and Kr, however, exhibits nonlinear growth with backing pressure, rising above the noise with as little as 150 psi backing the gas jet. Assuming 100% condensation of the atoms into clusters [17], the observed scattered signal levels at the highest backing pressures suggest that the mean cluster size is $(2-10) \times 10^4$ atoms for Ar and $(1-5) \times 10^5$ atoms for Kr, in good agreement with that expected from the clustering parameter, Γ.

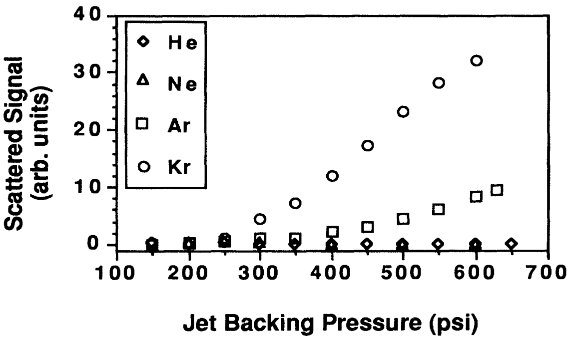

FIG. 1. Mie scattered light signal as a function of backing pressure for He, Ne, Ar, and Kr.

Enhancement of the absorption of laser light by the presence of clusters was investigated by a series of experiments in which a small fraction (0.01–0.1) of a gas known to cluster is mixed with He and passed through the gas jet. It is well known that the use of He as a carrier gas will significantly enhance the formation of clusters of heavier atoms in a gas expansion [17], while the He does not itself undergo clustering. The gas mixture was illuminated by the 130 fs pulse focused by an $f/25$ lens producing a peak intensity of up to 10^{17} W/cm^2. A grazing incidence extreme ultraviolet (XUV) spectrometer was used on the laser axis to disperse the resulting plasma emission. Time integrated XUV data were recorded with a microchannel plate detector and time resolved spectra were taken with an x-ray streak camera capable of 10 ps time resolution.

Since the He does not itself cluster, observation of the intensity of the Ly-α transition in He$^+$ at 304 Å allowed us to study the dynamics of the bulk plasma without concern for intracluster effects. The presence of the clusters served to absorb laser energy, which resulted in a thermal plasma that can collisionally excite the He levels. Figure 2 shows the measured, time integrated intensity of the Ly-α line as a function of focused laser intensity for a variety of gas mixture conditions. The intensity required to optically ionize He to He^{2+} is approximately 1.5×10^{16} W/cm^2 with a 130 fs laser pulse. Above this intensity Ly-α light will be emitted due to the recombination of electrons into the upper levels of the doubly ionized He. As shown in Fig. 2, when a plasma is formed from 100% pure helium we observe a small amount of Ly-α light at peak intensities above the threshold for tunneling ionization to He^{2+}. Addition of a small fraction of Ne does not significantly change the observed Ly-α signal. This is consistent with the fact that large clusters are absent in the He and He/Ne expansions and the small observed signal is due only to direct strong-field ionization by the laser.

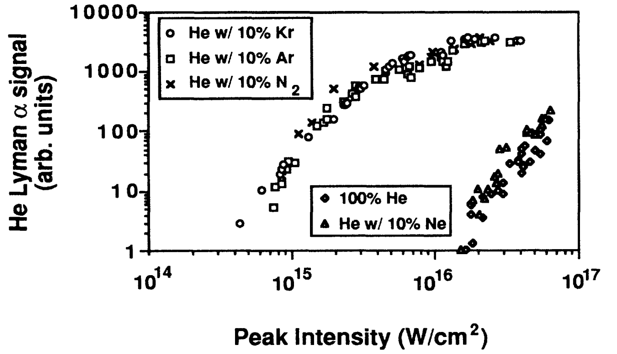

FIG. 2. Time integrated intensity of the He Ly-α line as a function of focused laser intensity for a variety of gas mixtures (350 psi gas jet backing pressure).

When a small amount of Ar, Kr, or N_2 is mixed with the helium, all gases with a strong propensity for forming large clusters, the magnitude of the He Ly-α signal is significantly enhanced exceeding the signal of the pure helium by nearly a factor of 100 at the highest intensities. Furthermore, the Ly-α signal appears at an intensity that is 20 times lower than the threshold for the production of He^{2+} predicted by tunneling ionization. This large enhancement in Ly-α radiation can be explained by the creation of a hot plasma produced by efficient laser energy absorption into the dopant gas clusters. Time resolved data of the He Ly-α line (radiative lifetime = 133 ps) indicate that the emission from the pure helium plasma decays after about 1.5 ns, consistent with the relatively cold ($\sim$40 eV) optically ionized plasma predicted by tunnel ionization [10]. The emission from the plasma formed by irradiation of the gas mixtures lasts for approximately 150 ns. This long-lived emission is evidence for a hot ($>$100 eV) thermal plasma that is slowly cooling by hydrodynamic expansion.

The enhanced absorption and heating by clusters should lead to high charge states and short wavelength ($<$100 Å) x rays being produced at relatively modest laser intensity as described previously. A second grazing incidence spectrometer was used to measure x-ray emission above 100 eV emitted 90° to the laser propagation axis. Fig. 3(a) shows the time integrated spectrum between 40 and 100 Å produced by the laser focused to an irradiance of approximately 1.5×10^{16} W/cm^2 into the gas jet backed by 500 psi of pure Kr. The spectrum exhibits strong emission from the $4p$-$3d$ and $4s$-$3p$ arrays of Kr^{10+}, Kr^{11+}, Kr^{12+}, and Kr^{13+} [18]. Tunnel ionization predicts that intensities of 3×10^{17}, 4×10^{17}, 6×10^{17}, and 8×10^{17} W/cm^2, respectively, are required to produce these states by optical ionization, nearly an order of magnitude in excess of the actual laser irradiance present.

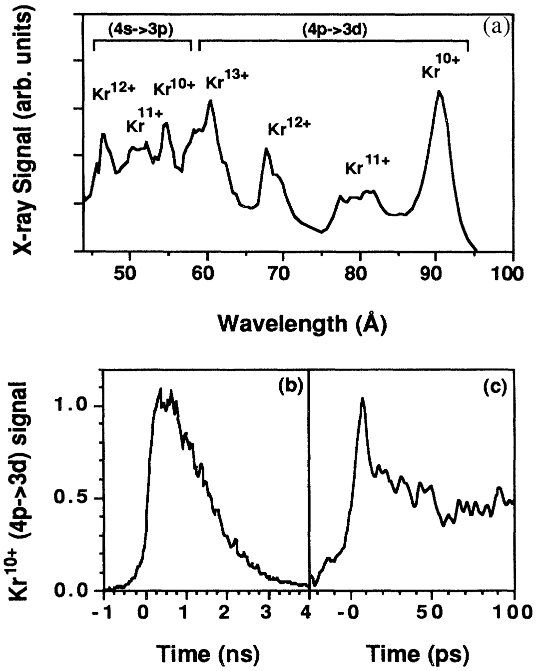

FIG. 3. (a) Time integrated spectrum produced in Kr (500 psi gas jet backing pressure) with an intensity of 1.5×10^{16} W/cm^2. (b),(c) Measured time history of the Kr^{10+} $4p$-$3d$ line.

Anomalous lines from Kr^{10+} in this wavelength range were observed previously by McPherson *et al.* under similar conditions in a time integrated experiment using a 200 fs KrF, 248 nm laser to produce a plasma in a Kr gas jet [7]. They attributed these lines to laser induced inner-shell excitation induced by excitation of small (few atom) clusters. This mechanism would produce radiation emitted on the time scale of the laser pulse ($<$1000 fs). We find in our experiment that large clusters ($>10^4$ atoms) are present in the gas jet and that the emission from these lines is dominated by relatively long time scale processes (expansion, cooling, and recombination). The time history of the Kr^{10+} $4p$-$3d$ line is shown in Fig. 3(b) with a streak camera resolution of approximately 280 ps. The radiation from this line is emitted for nearly 3 ns after the passage of the laser through the gas jet. This lifetime is consistent with the long-lived emission of a hot, underdense bulk plasma.

Further evidence for the interaction of the laser with large solid density clusters is found when the time history of the Kr emission is observed over the first 100 ps. Figure 3(c) shows the time history of the Kr^{10+} 90 Å line for the first 100 ps after illumination by the laser. The time resolution here is $\sim$10 ps. We observe an initial spike in the emission of the Kr^{10+} line, faster than the time resolution of the streak camera, followed by the long-lived emission demonstrated in Fig. 3(b). A similar spike is seen on all lines observed in Fig. 3(a). This initial spike is indicative of intense x-ray emission by the dense cluster microplasma immediately after heating by the laser. This short time scale emission can be explained by enhanced collisional excitation within the hot plasma during the brief time in which the local density is high within the cluster. The fast (<1 ps) expansion of the cluster is then followed by lower intensity emission by the lower density bulk plasma on a long (3 ns) time scale. We should emphasize that these data indicate that the vast majority of the x-ray emission comes not from the cluster but from the underdense plasma. The initial spike in the emission contains less than 1% of the total x-ray signal emitted from the plasma.

We have observed similar enhanced absorption and heating resulting in the production of high charge states and long-lived x-ray emission in all species which undergo significant clustering. Figure 4(a) shows the time integrated spectrum of Ar heated by a pulse focused to 8×10^{15} W/cm^2. Strong emission from lines below 50 Å produce in neonlike Ar (Ar^{8+}) are readily observed. To produce this charge state by tunnel ionization would require a focused intensity of $>1.5 \times 10^{18}$ W/cm^2, more than 2 orders of magnitude higher than that used in the experiment. These 250–400 eV photons result from the formation of a hot plasma ($T_e > 250$ eV). The measured photon yield on these lines indicates that up to (2–5)% of the laser energy is converted to x rays in this wavelength range, yields that are comparable to solid targets [8]. Time resolved data on these lines exhibit similar behavior to that of the Kr lines, with the Ar^{8+} 49 Å $3s$-$2p$ line, for example, lasting over 2 ns [Fig. 4(b)].

The mechanism of enhanced absorption in large, solid density clusters producing hot microplasmas and x-ray emission on a time scale determined by hydrodynamic expansion and cooling is supported by a one-dimensional hydrodynamic model and kinetics calculations of the low density plasma. We assume the cooling is determined by adiabatic expansion of a cylindrical plasma with corrections for conductive and free steaming cooling, and we use the kinetics code FLY [19] to determine the time history of various charge states. FLY solves rate equations to determine the time-dependent population of energy levels in the different ion species found in the plasma. We find that the observed charge states and resulting line time histories imply an initial plasma temperature of ~1000 eV in the argon cluster. The calculated time history of emission on the Ar^{8+} 49 Å line in a cylindrical plasma, with an initial electron temperature of 1000 eV and a final ion density of 1×10^{18} cm^{-3}, is compared to the measured 49 Å emission in Fig. 4. The calculated decay time is in reasonable agreement with the measured time history, predicting a long (>1 ns) decay consistent with the data.

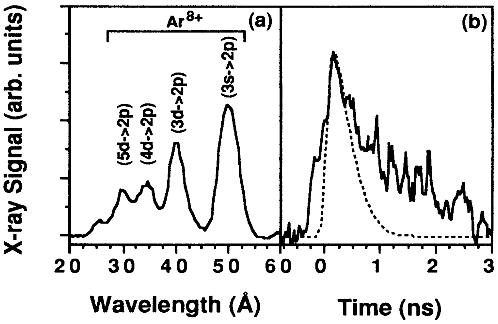

FIG. 4. (a) Time integrated spectrum produced in Ar (500 psi gas jet backing pressure) with an intensity of 8×10^{15} W/cm^2. (b) Measured (solid) and calculated (dashed) time history of the Ar^{8+} 49 Å line.

In conclusion, we have shown that large clusters produced in expanding gas jets can be used to produce hot, moderate density plasmas with intense, short-pulse lasers. Enhanced absorption is observed when the pulse duration of the laser is less than the expansion time of the cluster. Plasmas containing charge states far beyond that predicted by optical field ionization of individual atoms are produced by the illumination of clusters by femtosecond pulses of 10^{16} to 10^{17} W/cm^2. The resulting strong x-ray emission occurs on a time scale determined by hydrodynamic expansion and cooling of the plasma. Furthermore, the x-ray yields are comparable to those that can be achieved with solid targets. These cluster heated plasmas have the potential for providing a source of strong, x-ray radiation with the modest irradiance (10^{15} to 10^{17} W/cm^2) produced by small-scale short-pulse lasers through a unique combination of the advantages inherent to both solid and gas targets.

We would like to acknowledge helpful discussions with R. A. Smith and the technical assistance of R. Jones. This work was supported by the AFOSR and the DOE under Contract No. W-7405-Eng-48.

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