High Intensity Laser Absorption by Gases of Atomic Clusters

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We have measured the energy absorption efficiency of high intensity, picosecond laser pulses in low density gases composed of large atomic clusters. We find that, though the average density of the resulting plasmas is low, the energy absorption can be very high (>95%), indicating that substantial laser energy is deposited per particle in the plasma. Ion energy measurements confirm that this efficient energy deposition results in plasmas with very high (multi-keV) ion temperatures. [S0031-9007(97)03016-0]

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The production of high temperature plasmas with small scale, short pulse, high intensity lasers has been actively pursued during the last ten years. Of particular interest in these studies is the measurement of the energy absorption efficiency of high density plasmas created by intense irradiation of a solid target, and many groups have published work measuring solid target plasma absorption efficiency over a wide range of incident intensities and laser wavelengths on planar [1–5] and microstructured targets [6]. These studies have shown that the plasma typically absorbs a large fraction of the laser energy, between 10% and 80% of the incident energy, depending upon intensity and laser wavelength. Such experiments have shown that a large amount of energy can be deposited per unit area and that high temperatures (>100 eV) are achievable [1,7]. However, rapid heat conduction into the cold, solid substrate beneath the plasma will typically clamp the plasma temperature to a value of <1000 eV [7–9]. Furthermore, most of the deposited energy is contained within the plasma electrons, which cool too rapidly by conduction and hydrodynamic expansion to transfer much of this laser energy to the cold ions.

A gas of large atomic clusters (>1000 atoms/cluster) presents a radically different environment for laser-plasma interaction dynamics [10,11]. In general, low density gases are expected to exhibit very low absorption efficiency (<1%) and the plasmas produced by intense irradiation will generally be quite cold (10–100 eV). The presence of clusters in a gas changes this situation dramatically [11]. Though the average density of a gas containing clusters is low, the local density within the cluster is near solid, and, consequently, will be subject to the rapid heating experienced by a solid target due to collisional inverse bremsstrahlung. Very bright x rays have been observed from gas target plasmas produced by intense femtosecond illumination of clusters [10], indicating that electron temperatures in these plasmas were quite high, far in excess of those expected from a gas composed only of single atoms [11]. These observations suggested that the clusters were very efficient in absorbing laser energy. In this Letter, we report the first laser energy absorption measurements of intense laser pulses in gaseous media containing atomic clusters. We find that cluster gases are at least as efficient as solid targets in absorbing short pulse laser energy, and that, furthermore, much of this energy is deposited in the plasma ions, resulting in plasmas with very hot ion temperatures.

Recent studies of intense laser interactions with individual clusters have confirmed that hot electrons (up to 3 keV) are produced during the laser-cluster interaction [12], and that, furthermore, an even greater energy can be deposited in the ions when these hot, highly ionized clusters explode [13]. These studies indicated that the clusters are rapidly heated by the laser, to a nonequilibrium, superheated state, in large part due to the passage of the free electron density in the cluster through a Mie resonance with the laser field during the cluster expansion [14]. These superheated cluster microplasmas eject electrons with many keV of energy. Soon after the clusters are heated charge separation of the hot electrons causes the clusters to explode, and as a result, much of the energy deposited by the laser in the cluster is converted to ion kinetic energy.

Such studies suggest that plasmas formed from the intense irradiation of gases containing clusters will exhibit large laser absorption. To measure the cluster energy absorption we used a Nd:glass laser based on chirped pulse amplification which produces 2 ps laser pulses with energy up to 0.5 J. These pulses were frequency doubled to 527 nm to eliminate any low intensity laser prepulse, and focused with an $f/12$ lens to a spot of 20 μm ($1/e^2$ diameter) into the output of a pulsed gas jet. The transmitted light was collected with an 8 cm diameter $f/2$ lens to assure that any light refracted by plasma formation at the focus was collected. The energy of the transmitted light was measured with a volume absorbing calorimeter fitted with a 527 nm bandpass filter to pass only laser light. The input laser energy was monitored with a photodiode. Any backscattered light was also monitored with a fast photodiode (none was observed for any of the measurements described in this work). Imaging from the side of the plasma was performed to assure that no significant amount of laser light was side scattered. The onset of cluster formation in the gas jet was determined by monitoring Rayleigh scattering of 532 nm, 10 ns light pulses in the gas jet. This technique allowed us to make estimates for the average cluster sizes in the gas jet [12].

The fraction of laser light absorbed in an argon gas jet as a function of gas jet backing pressure is shown in Fig. 1(a). The peak laser intensity was 7×10^{16} W/cm^2. At low backing pressure the absorption fraction is zero. Rayleigh scattering measurements in the gas jet indicate that clusters begin to form at a backing pressure of >5 bars, a pressure above which the absorption of the laser energy begins to grow. At the highest backing pressure, the absorption is near 80%. At this point the gas plume is composed of Ar clusters with an average size of roughly 80 Å and a cluster density of $\sim 1 \times 10^{15}$ cm^{-3} (corresponding to an average atomic density of $\sim 5 \times 10^{18}$ cm^{-3}). The gas density profile, determined by imaging the visible light emission from the plasma, exhibits a near Gaussian density profile with a $1/e^2$ width of 4.2 mm. For comparison, the measured energy absorption in a Ne gas jet over the same backing pressure range is also shown in Fig. 1(a). Under these conditions, Ne does not nucleate into clusters. As expected, the energy deposition of the laser in the Ne plasma is negligible. Xe also exhibits very high energy absorption at backing pressures which cause cluster formation. Figure 1(b) shows the energy absorption of the laser in a Xe gas jet. The energy absorption begins to grow at lower backing pressure than in Ar. This is due, in some part, to the greater propensity of Xe to cluster than Ar. Xe begins to cluster in our jet at a pressure as low as 1 bar. At the highest pressure measured in Fig. 1(b), the estimated Xe cluster size is 100 Å and the cluster density is $\sim 4 \times 10^{14}$ cm^{-3}.

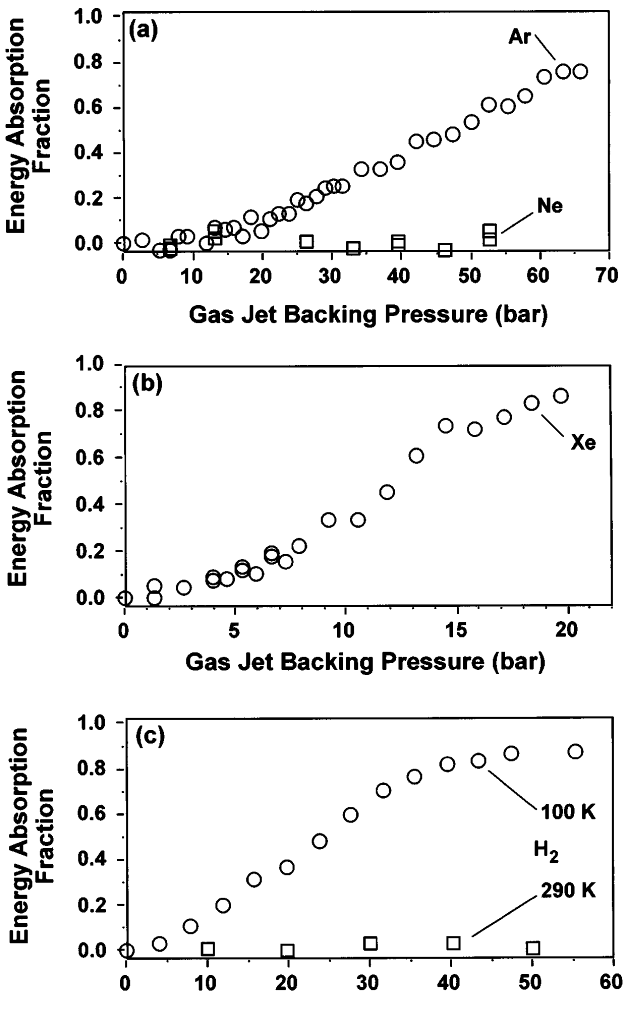

FIG. 1. The fraction of laser light absorbed in a gas jet as a function of gas jet backing pressure with a peak laser intensity of 7×10^{16} W/cm^2. (Each point is an average of three laser shots.) (a) Argon and neon gas jet. (b) Xenon gas jet. (c) Room temperature and cryogenically cooled H_2 gas jet.

This efficient laser energy absorption, however, is not limited to high atomic number species. We have also measured the absorption of laser energy in gases of hydrogen clusters. We produce H_2 clusters by cryogenically cooling the backing reservoir of our gas jet. Figure 1(c) shows the energy absorption in a hydrogen gas jet as a function of pressure for two different reservoir temperatures. At a temperature of 290 K, a temperature at which hydrogen does not exhibit any clustering, the energy absorption is essentially zero. However, with a jet temperature of 100 K, a temperature at which we observe strong clustering of the hydrogen, the energy absorption rapidly rises with backing pressure. At a pressure of 55 bars ($\sim$100 Å clusters), nearly 90% of the laser energy is deposited in the hydrogen plasma.

This efficient energy absorption occurs over a very wide range of laser intensities. The absorption fraction of the laser energy in an argon jet with 40 bars of pressure ($\sim$80 Å clusters) as a function of the peak laser intensity is shown in Fig. 2(a). The absorption rises from zero at an intensity of 2×10^{13} W/cm^2 and slowly increases with intensity to 70% at an intensity of 5×10^{15} W/cm^2 after which the absorption begins to drop with increasing intensity. Xe absorption, shown in Fig. 2(b) at a backing pressure of 20 bars, also exhibits this trend. Like argon, the absorption rises at a low intensity, saturates, and drops at the highest intensity.

There are two striking features of this data. The first is the very low intensity at which the gas jet begins to absorb laser energy. In the Xe jet the absorption begins to rise at an intensity of 10^{12} W/cm^2. This is an intensity nearly 1 order of magnitude lower than that at which atomic Xe is ionized to any significant degree by multiphoton processes in a picosecond pulse of this wavelength. (The estimated error in determining the laser intensity is less than a factor of 2.) This fact seems to indicate that the production of a very small number of electrons by multiphoton ionization in the cluster is sufficient to seed an avalanche breakdown within the cluster, which is then followed by collisional heating and the bulk of the laser absorption. This is a

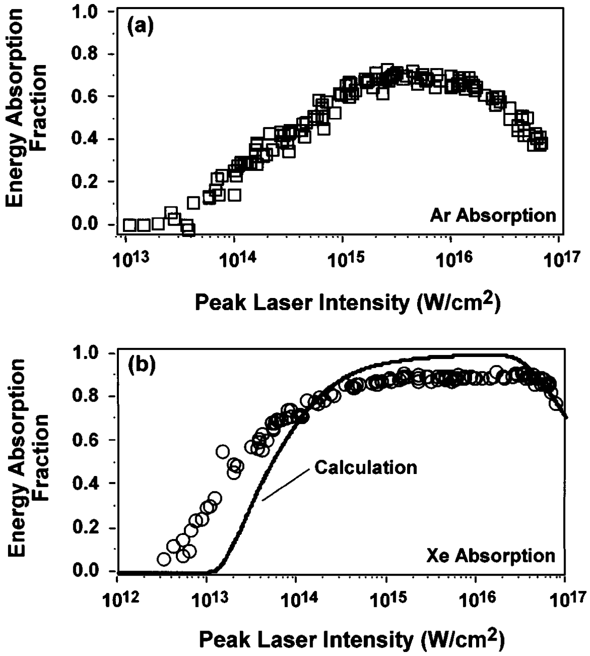

FIG. 2. The absorption fraction of the laser energy in a jet as a function of the peak laser intensity. (Each point is a single laser shot.) (a) Argon jet with 40 bars of backing pressure (~80 Å clusters). (b) Xenon jet with 20 bars of backing pressure (~100 Å clusters). The solid line is the calculation of the cluster absorption model.

situation similar to the breakdown of dielectric solids by short pulse radiation [15].

The second important feature of the data in Fig. 2 is the saturation in the absorption fraction followed by a drop in absorption with a further rise in laser intensity. To explore these dynamics we have conducted modeling of the absorption of the laser light in an extended medium of clusters. To do this, we utilized the plasma model of cluster heating as detailed in Ref. [14]. This model calculates the energy deposited in an expanding cluster microplasma. It accounts for inverse bremsstrahlung heating of the electrons, tunnel and collisional ionization, and both hydrodynamic and Coulomb explosion contributions to the cluster expansion. To account for the effects of multiphoton ionization at low intensities on the avalanche breakdown in the cluster we use the multiphoton ionization cross section for neutral Xe measured in Ref. [16] for 586 nm pulses. This model was then solved for a 2 ps pulse propagating through a series of spatial cells, in which the cluster absorption dynamics were solved. The measured density profile of our gas jet was used in the code. The absorption through the gas jet was calculated as a function of intensity and integrated over a Gaussian focal spot intensity distribution. Any effects of refraction on the pulse during its propagation through the gas jet plasma were ignored. The results of this calculation are shown for Xe as the solid line in Fig. 2(b). The general behavior of the absorption with increasing laser intensity is well reproduced by the model, including the drop in absorption at high laser intensity.

The modeling indicates that this feature in the data appears to be the result of the limited time over which the clusters absorb laser energy in the laser pulse. The rapid expansion of the clusters once they begin to be heated by the laser causes them to disassemble on a 100–200 fs time scale, a time scale faster than our 2 ps pulse. As a result, the leading edge of the pulse initially experiences the highest absorption as it propagates into the cluster medium. This leads to an effective pulse shortening as the pulse propagates deeper into the medium. When the peak intensity is high enough, the pulse "burns through" the cluster medium, and the trailing edge of the pulse propagates through the medium without any significant absorption.

This effect is illustrated in Fig. 3 in which the calculated temporal pulse profile after propagation through the Xe cluster gas jet is compared with the initial pulse profile at two initial peak intensities. At a peak intensity of 4×10^{16} W/cm^2 [Fig. 3(a)] most of the pulse is absorbed and only a small amount of light near the end of the pulse succeeds in exiting the medium. At a peak intensity of 1×10^{17} W/cm^2 [Fig. 3(b)] a much larger fraction of the trailing edge of the pulse is transmitted through the gas, due to the rapid disassembly of the clusters in the rising edge of the pulse. The close agreement of the code with the observed rollover of the absorption in the data of Fig. 2(b) at the highest intensities seems to confirm that the clusters explode rapidly, and, therefore, only absorb laser energy on a subpicosecond time scale. In fact, the calculated trend of the absorption is remarkably similar to that of the data over the entire range of intensity.

The very high absorption observed in these measurements indicates that a large amount of laser energy can be deposited in a small volume in these clustering gas jet targets. In our experiments, the laser energy is deposited in a volume of $\sim\pi\omega_0^2 l \sim 1 \times 10^{-6}$ cm^3 (where ω_0 is the laser focal spot radius and l is the plasma length). At the highest intensity, over 0.25 J of energy is deposited in this small volume. Thus, with an average atomic density of 5×10^{18} cm^{-3} in the gas jet, this implies that up to 300 keV of energy is deposited per atom. However, unlike a solid target, in which the deposited laser energy is conducted away from the hot plasma on a time scale comparable to the laser pulse width, thus clamping the plasma

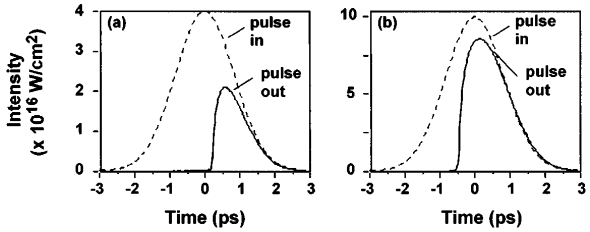

FIG. 3. Calculated pulse profile after propagation through the Xe cluster gas jet (solid lines) compared with the initial pulse profile (dashed lines). (a) Peak intensity of 4×10^{16} W/cm^2. (b) Peak intensity of 1×10^{17} W/cm^2.

temperature [7,9], heat conduction is very slow in the low density gases used in these experiments [14]. As a result, the plasmas created are extremely hot.

Our previous studies of single cluster dynamics indicate that the majority of the laser energy absorbed by a cluster is transferred to the kinetic energy of the radially exploding ions [13]. This cluster explosion is driven by the ambipolar potential created by the expansion of the hot electrons, in a situation similar to the expansion of a hot solid target plasma into vacuum [17]. With the high deposited energy density in our experiments, we expect the ion temperature of the plasma to be quite large. To explore this we have examined ion energies through ion time-of-flight measurements from the plasma. These measurements were made with an isolated Faraday cup charge collector located 40 cm from the plasma. The ions were detected at an angle of 20° from the laser axis, nearly along the axis of the cylindrical plasma formed by the focused laser pulse in the gas jet. A Xe ion time-of-flight spectrum is shown in Fig. 4. A fast spike coinciding with the laser creation of the plasma is observed at $t = 0$. This spike is caused by photoelectron ejection from the charge collector by UV radiation emitted by the plasma. This fast spike is expanded in the inset of Fig. 4. Superimposed on the photoelectron signal is a negative going spike resulting from the arrival of fast electrons from the plasma.

A long time scale feature is then seen after this initial spike, corresponding to the arrival of the Xe ions. The corresponding ion kinetic energy is shown across the top of this plot. As can be seen from Fig. 4, the Xe ions exhibit very large kinetic energy. We observe ions with energy up to 400 keV (faster ions are obscured by the presence of the photoelectron spike). No fast ions of any sort are observed in gases that do not exhibit clustering in the gas jet. These high Xe ion energies are consistent with the ion energies observed from exploding Xe clusters heated by a 140 fs, 780 nm pulse [13]. Clearly, the ion temperature of the cluster plasma is large, significantly greater than that achieved in solid target interactions.

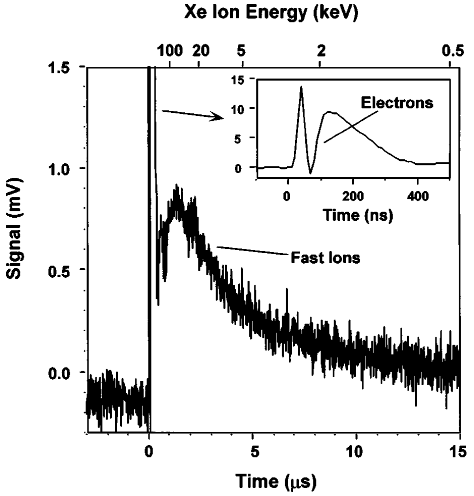

FIG. 4. Xe ion time-of-flight spectrum with a gas jet backing pressure of 20 bars and a laser peak intensity of 8×10^{16} W/cm^2. (Inset) Same data expanded to show fast time scale signal.

In conclusion, we have measured the energy absorption efficiency of high intensity laser pulses in a gaseous medium of clusters. We have examined a number of cluster species and we find that the absorption efficiency of these media is much higher than that of a gas of single atoms and is comparable to solid target plasmas. The high absorption efficiency is the result of collisionally driven heating of the clusters which ultimately explode after this heating. As a result, a plasma with a very high ion temperature is produced. The remarkably high efficiency with which the cluster medium absorbs intense laser radiation makes possible the creation of moderate density plasmas with hot ion temperatures using small scale, table-top lasers delivering modest energy. Such interactions may ultimately make feasible table-top scale experiments on fusion and high temperature astrophysical plasmas.

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