INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS D: APPLIED PHYSICS
J. Phys. D: Appl. Phys. **35** (2002) 2935–2938 PII: S0022-3727(02)39837-1

Plume splitting and sharpening in laser-produced aluminium plasma

S S Harilal, C V Bindhu, M S Tillack, F Najmabadi and A C Gaeris

Center for Energy Research, University of California San Diego, 9500 Gilman Drive, La Jolla, CA 92093-0417, USA

E-mail: Harilal@fusion.ucsd.edu

Received 23 July 2002
Published 4 November 2002
Online at stacks.iop.org/JPhysD/35/2935

Abstract
Plume splitting and sharpening were observed in laser-produced aluminium plasma created using 532 nm, 8 ns pulses from a frequency doubled Nd : YAG laser. Measurements were made using 2 ns gated fast photography as well as space and time resolved optical emission spectroscopy. The motion of the leading edge of the plume was studied with several background air pressures and the expansion of the plume front was compared with various expansion models. Combining imaging together with time resolved emission diagnostics, a triple structure of the plume was observed.

1. Introduction

The interaction of pulsed laser ablation plumes with a background gas has received increased attention recently due to its importance in laser deposition [1], nanoparticle formation and growth [2], cluster production [3], etc. Ablation into a background gas results in shock waves as expansion fronts propagate through the gas. In the moderate intensity range (1–10 GW cm^{-2}), gas dynamic effects are thought to play a leading role in determining spatial and velocity distributions of the vaporized material. Laser-produced plasma is transient in nature with characteristic parameters that evolve quickly and are strongly dependent on irradiation conditions such as incident laser intensity, laser wavelength, irradiation spot size, ambient gas composition and ambient pressure [4–6]. In spite of the extensive literature on this subject, expansion dynamics of laser-produced plasma remains incompletely understood. The different processes involved, including laser absorption, evaporation, transient gas dynamics, radiation transport, condensation, ionization and recombination, are rather complex and require further investigation. In particular, the experimentally observed splitting of the plume into an energetic component travelling at near vacuum speed and a component slowed down by the ambient gas has been difficult to understand and to describe theoretically [7].

In this paper, we report 2 ns gated fast photography and time and space resolved spectral emission studies in a laser-produced aluminium plasma created by 532 nm, 8 ns pulses from a Nd : YAG laser at moderate background air pressures. Fast side-on views of the plume expansion are made by recording overall visible emission from the plasma plume. This is useful for understanding the hydrodynamics of plume propagation [8, 9]. We particularly emphasize the plume sharpening and splitting observed with background air pressure of 150 mTorr during later stages of the plasma expansion. The results obtained with fast imaging are compared with the observation of a double peak temporal emission structure for the excited ionic species in the aluminium plasma.

2. Experimental details

Focused pulses from a frequency doubled Nd : YAG laser were used to create aluminium plasma in a stainless steel vacuum chamber. To avoid errors due to local heating and drilling, the target was rotated about an axis parallel to the laser beam. The laser beam was attenuated by a combination of wave plate and cube beam splitter and focused onto the target surface at normal incidence using an antireflection-coated plano-convex lens. The beam energy was monitored using an energy meter (Ophir).

The plume imaging was accomplished using an intensified CCD camera (PI MAX, Model 512 RB, 512 × 512 pixels, pixel size = 19 μm × 19 μm, 1 : 1 aspect ratio) placed orthogonal to the plasma expansion direction. A Nikon lens (f/4) was used to image the plume region onto the camera to form a two-dimensional image of the plume intensity. The visible radiation from the plasma was recorded integrally in the wavelength range 350–900 nm. In order to eliminate 532 nm stray photons reaching the camera, a magenta subtractive filter was used. A programmable timing generator was used to control the delay time between the laser pulse and the imaging system with overall temporal resolution of 1 ns.

For space and time resolved spectroscopy, an optical system was used to image the plasma plume onto the entrance slit of the monochromator/Spectrograph (Acton, Spectra-Pro 500i, f/6.5), so as to have one to one correspondence with the sampled area of the plume and the image. The optical system was translated to monitor different parts of the plume. Spatial resolution provided by our optical system was better than 0.5 mm. The monochromator was equipped with three gratings: 150, 600 and 2400 g mm^{-1}. One of the exits port of the spectrograph was coupled to an intensified CCD camera. The other exit port of the monochromator was coupled to a photomultiplier tube (PMT). For spatially resolved studies, different regions of the plasma plume were focused onto the monochromator slit. The characteristic lines were selected by the monochromator and the PMT output was coupled to a 1 GHz digital phosphor oscilloscope (Tektronix, TDS 5014). This set-up provides the ability to measure the arrival time as well as decay of the emission from constituent species at a specific point within the plasma, which are extremely important parameters related to the evolution of laser ablated material.

3. Results and discussion

Two-dimensional images of laser-produced plasma provide an orthogonal view of the expansion with respect to the aluminium target plane. Typical ICCD images of the expanding aluminium plume at different times after the onset

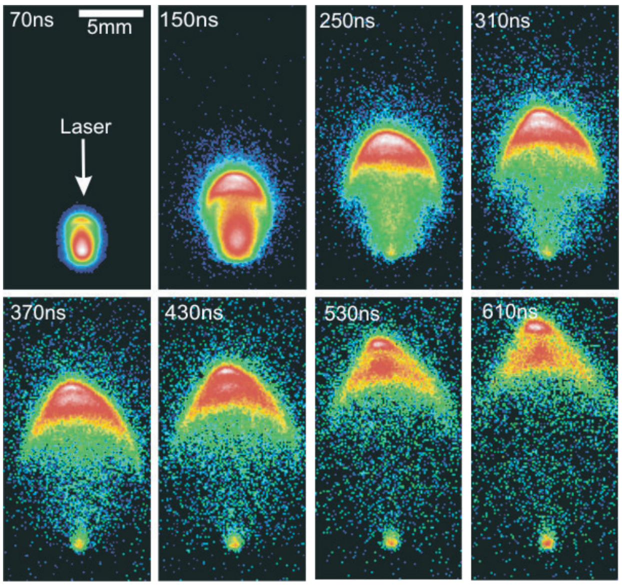

Figure 1. The evolution of visible emission from an aluminium plume recorded using an ICCD camera (gating time 2 ns). The laser power density used was 3 GW cm^{-2} and background pressure was 150 mTorr. All the images are normalized to their maximum intensity.

of plasma are given in figure 1. The images were obtained at a background pressure of 150 mTorr and at a power irradiance of 3 GW cm^{-2}. The gating time is 2 ns and each image is obtained from a single laser pulse. Timing jitter is less than 1 ns. Since the laser intensity exceeds the ablation threshold of the target, the laser beam evaporates and ionizes material, creating a plasma plume above the material surface. Initially, the atoms, molecules and ions undergo collisions in the high-density region near the target forming the so-called Knudsen layer, to create a highly directional expansion perpendicular to the target. Ions in the expanding plasma may be accelerated as high as a few hundred electronvolt. Generally at high fluences, the fast evaporative process produces interaction of the laser pulse with the rapidly generated plasma, further increasing the ion production.

Plasma emission begins on the target surface soon after the laser photons reach the surface and as time evolves it separates into two components. The light emission from the plume very close to the target surface exists even after 500 ns. This stationary emission region close to the target surface may result from gas collisions between the plume ejecta in the high pressure region of the initial expansion, resulting in a Knudsen layer with stopped and/or backward moving material [10, 11]. The other component expands very rapidly with time in a highly forward-directed pattern.

At early times, the plume front is spherical in nature, but as time evolves the plume front becomes sharpened. Along with sharpening, the expanding plume front splits into a fast and slow moving cloud indicating plume splitting. These peculiar plume splitting and sharpening phenomena are observed only in a particular pressure range (around 150 mTorr); we have not observed such phenomena at lower or higher pressures. Coincidentally, this pressure falls within the transition from collisional to collisionless interaction of the plume species with the gas.

Observation of a multiple peak distribution in a laser-produced plasma has been reported previously [12, 13]. Bulgakov and Bulgakova [14] reported the observation of a double layer in a laser created graphite plasma during charged-collector probe measurements and they explained the formation of double peak structure in ion time-of-flight (TOF) distribution as due to the formation of an ambipolar electric field in the expanding plume. Plume sharpening behaviour suggests that higher kinetic energy particles are emitted closer to the target surface normal. It has been reported that ions of highest ionization state dominate in the direction normal to the target, and that their concentration falls sharply away from the normal [15]. Puretzky *et al* [16] observed a peculiar sharpening of the biomolecule plume during gated LIF (laser-induced fluorescence) imaging studies due to the small concentration of heavy biomolecules propagating within a narrow angular distribution, which continues to sharpen over extended times after laser irradiation.

Chen *et al* [17] reported that partial ionization of the vapour due to temperature increase near the shock wave front also can result in an acceleration of the flow. The addition of kinetic energy to the laser-induced flow through absorption of incident laser energy will result in a more moderate deceleration of the shockwave velocity than the predicted blast-wave theory [18]. Simultaneously, the part of the ablated species that collide with background gas will lose their kinetic energy. Thus, the plume itself might be split as indicated in the experimental observation. When the background gas pressure is high enough, the plume is continuous and no such plume splitting is observed.

In order to obtain a better understanding of the plasma expansion and evolution, we used the imaging data to create position-time plots of the luminous front at several background air pressures (150 mTorr, 1.3 and 10 Torr), as shown in figure 2. The symbols in the figure represent experimental data points and the curves represent different expansion models. Plume expansion in the early stage (<40 ns) is linear irrespective of the background gas pressure (the straight-line fit in the graph corresponds to $R \propto t$). The plume expansion at 1.3 Torr is represented by a shock model given by $R = \xi_0(E_0/\rho_0)^{1/5}t^{2/5}$ where ξ_0 is a constant which depends on γ, the specific heat capacity of the expanding gas. The shock model describes the explosive release of energy, E_0 through a background gas of density ρ_0. At later stages the plume expansion is described by a drag model which is given by $R = R_0(1 - \exp(-\beta t))$ where R_0 is the stopping distance of the plume and β is the slowing coefficient ($R_0\beta = v_0$). The plume front position of the plasma at 150 mTorr showed a $t^{0.445}$ dependence.

To further elucidate underlying mechanism, we performed time resolved spectral emission studies of different species in the expanding plasma. Measurements of the temporal locations of the maximum intensity as a function of distance give an estimate of the velocity component perpendicular to the target surface. Figure 3 shows the typical TOF profiles of Al^+ species (358.6 nm) recorded at different distances from the target surface. The time evolution of the spectral emission profiles obtained in this work clearly reveals that the species ejected by the Al plume exhibit twin peaks for the TOF distribution. The twin-peak distribution is observed only beyond a certain distance from the target. The estimated expansion velocity of the faster peak and slower peaks in the initial stages are 7.6×10^6 cm s^{-1} and 2.5×10^6 cm s^{-1}, which correspond to a kinetic energy of 800 eV and 88 eV, respectively. The faster peak moves with a velocity very close to the free expansion velocity (straight-line fit in the image R–T plot). We observed twin peak distribution in the emission profiles of neutral aluminium species also.

Our measurements also show that the higher the ionization stage, the greater the kinetic temperature. This is probably due to enhanced absorption of laser light by the plasma. After the laser action ceases the charge composition of the plasma is governed by three-body recombination, which dominates over radiative and/or dielectronic recombination due to the rapid drop in electron temperature as the plasma expands. The ions located at the front of the plasma acquire the largest energy during hydrodynamic acceleration and the interaction time for recombination is very much reduced. As observed with ion TOF studies, these ions propagate with high kinetic energies close to the free expansion velocity. This small group of high-energy ions transports a significant fraction of absorbed energy. These ions interpenetrate into background gas without much attenuation in kinetic energy. The ions located in the inner plume layers are accelerated much less due to hydrodynamic expansion. They remain much longer in the denser state, which is being subjected to strong recombination, which in turn enhances the emission from these regions.

Surprisingly, the position of the plume front recorded using ICCD imaging at 150 mTorr moves much slower than the free expansion model. The slow moving component in the TOF species matches well with the plume front position recorded using ICCD imaging. However, we see almost no signal corresponding to the faster profile in the TOF species using ICCD imaging studies. The TOF profiles indicate that the yield or concentration of the higher kinetic energy peak is rather low compared to the slow moving component, which leads us to believe that the dynamic range of the ICCD is too low to observe these photons. Since we observe plume splitting

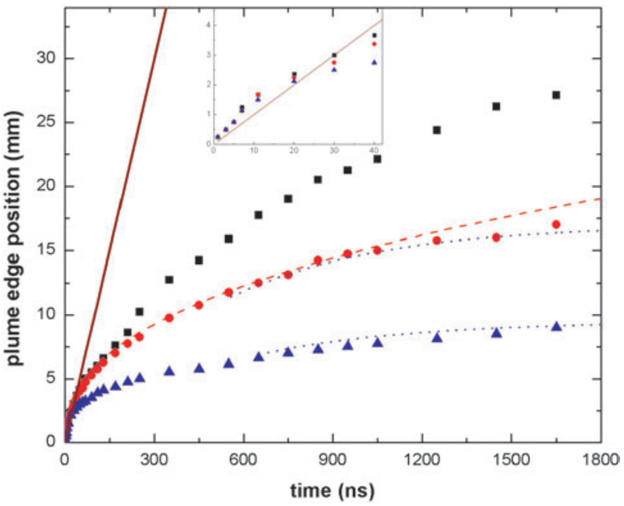

Figure 2. Position-time plots of the luminous front of the aluminium plume produced in background air pressure of 150 mTorr (■), 1.3 Torr (●) and 10 Torr (▲) measured from gated ICCD plume images. The symbols in the figure represent experimental data points and curves represent different expansion models. The dashed curve represents the shock wave model and the dotted line shows the drag model fit. Fitting parameters are $\beta = 0.002$ ns^{-1} and $R_0 = 17$ mm (1.3 Torr) and $R_0 = 9.5$ mm (10 Torr). The data below 40 ns is blown up in the inset.

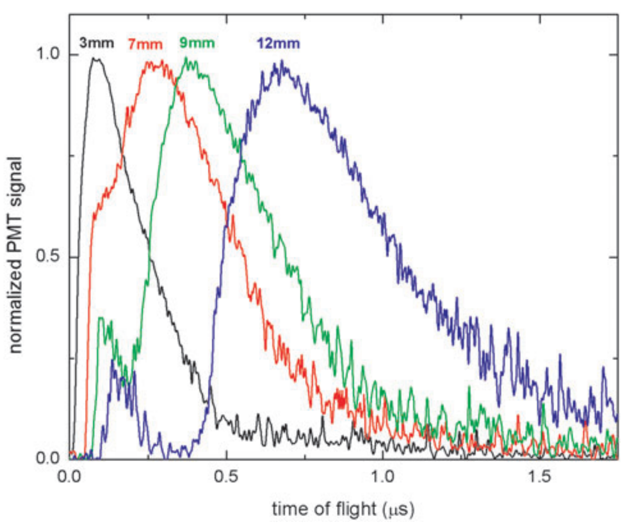

Figure 3. Intensity variation of spectral emission with time for Al^+ species (358.6 nm) recorded at different distance from the target surface. The laser power density was 3 GW cm^{-2} and the background air pressure was 150 mTorr for these measurements.

with imaging studies for the low kinetic energy component in the TOF profile, we conclude that there exists a triple plume structure with one component travelling near vacuum speeds and two slower components. The estimated temporal separation at a specific spatial point in plasma between the two slow layers observed with the imaging studies is ~50 ns. The TOF kinetic energy distribution of the slow component is rather broadened, and hence we do not clearly see the splitting of the delayed TOF distribution as observed with the imaging studies. Only through the combination of imaging and TOF spectroscopy do we determine the full plume structure.

4. Conclusions

The expansion dynamics of laser-produced Al plasma has been studied using fast photography and time resolved emission diagnostic techniques. The behaviour of the plume is found to be strongly influenced by the background gas pressure. From these studies, three temporal stages can be distinguished. In the earlier stage (<40 ns) the expansion is almost linear with time irrespective of the background gas pressure used. As time evolves the plume propagation is well characterized by a spherical shock wave model. For time >500 ns, the plume is decelerated rapidly and comes to rest due to numerous collisions with the background gas molecules and the drag model is a good approximation in this regime. We observed a peculiar plume splitting and sharpening at 150 mTorr. The results obtained with TOF emission studies along a particular line of sight also show plume splitting. Combining both diagnostic tools, a triple structure of the plume is deduced. Highly energetic ions moving with the free expansion velocity are not observed with fast imaging studies, may be overlooked because of bright plume luminescence of slow component.

Acknowledgment

This work was supported by the US Department of Energy under the grant number DE-FG03-99ER-54547.

References

[1] Chrisey D B and Hubler G K 1994 *Pulsed Laser Deposition of Thin Films* (New York: Wiley)
[2] Puretzky A A, Geohegan D B, Fan X and Pennycook S J 2000 *Appl. Phys. Lett.* **76** 182
[3] Kroto H W, Heath J R, O'Brien S C, Curl R F and Smalley R E 1985 *Nature* **318** 162
[4] Amoruso S, Bruzzese R, Spinelli N and Velotta R 1999 *J. Phys.* B **32** R131
[5] Gordillo-Vazquez F J, Perea A, Chaos J A, Gonzalo J and Afonso C N 2001 *Appl. Phys. Lett.* **78** 7
[6] Harilal S S, Bindhu C V, Nampoori V P N and Vallabhan C P G 1998 *Appl. Phys. Lett.* **72** 167
[7] Chen K R, Leboeuf J N, Wood R F, Geohegan D B, Donato J M, Liu C L and Puretzky A A 1996 *J. Vac. Sci. Technol.* A **14** 1111
[8] Geohegan D B and Puretzky A A 1995 *Appl. Phys. Lett.* **67** 197
[9] Harilal S S, Bindhu C V and Kunze H J 2001 *J. Phys.* D **34** 560
[10] Kelly R 1990 *J. Chem. Phys.* **92** 5047
[11] Pietsch W 1996 *J. Appl. Phys.* **79** 1250
[12] Laube S J P and Voevodin A A 1998 *Surf. Coat. Technol.* **105** 125
[13] Harilal S S 2001 *Appl. Surf. Sci.* **172** 103
[14] Bulgakov A V and Bulgakova N M 1995 *J. Phys. D: Appl. Phys.* **28** 1710
[15] Thum-Jaeger A, Sinha B K and Rohr K P 2001 *Phys. Rev.* E **63** 016405
[16] Puretzky A A, Geohegan D B, Hurst G B, Buchanan M V and Luk'yanchuk B S 1999 *Phys. Rev. Lett.* **83** 444
[17] Chen K R, Leboeuf J N, Wood R F, Geohegan D B, Donato J M, Liu C L and Puretzky A A 1995 *Phys. Rev. Lett.* **75** 4706
[18] Finke B R and Simon G 1990 *J. Phys.* D **23** 67