Laser-ablation-assisted-plasma discharges of aluminum in a transverse-magnetic field

J. S. Lash, R. M. Gilgenbach,[a] and C. H. Ching
Intense Energy Beam Interaction Laboratory, Nuclear Engineering Department, The University of Michigan, Ann Arbor, Michigan 48109-2104

(Received 9 February 1994; accepted for publication 23 May 1994)

Laser-ablation-assisted-plasma discharges (LAAPD) have been used to enhance the ionization of laser ablated aluminum metal. Ablation is accomplished by focusing a KrF excimer laser (248 nm, 40 ns, $\leqslant$0.4 J) on a solid aluminum target with a fluence of 4 J/cm^2. Peak plasma discharge voltage is 1–4 kV and peak plasma current is 0.2–1 kA, while peak power is 0.1–1 MW. Gated emission spectroscopy is used to determine the charge states and the electronic temperatures within the plasma discharge. With unmagnetized discharge parameters of 3 kV and 760 A, the observed light emission is dominated by transitions from Al^{2+} ions indicating nearly complete ionization of the plume. From the emission spectra intensities, an Al^{2+} electronic temperature of 3.3 eV is determined. Emission spectra from unmagnetized LAAPD of 1.2 kV and 280 A show no visible Al^{2+} ion transitions indicating cooler plasma and a lower ionization state. Introducing a 620 G transverse magnetic field (at 1.2 kV, 280 A) enhances the ionization due to the increased electron confinement and leads to the observance of the Al^{2+} lines as seen with discharges of 3 kV and 760 A.

Laser ablation has found important applications over the past decade in deposition of thin films of metals and ceramics.[1] Excimer lasers have been utilized extensively for deposition of metal-oxide, high-temperature superconductors, and metal-nitride electronics materials.[1–3] Several investigators have observed that the ions in laser ablation plumes play a significant role in the deposition process.[4,5] Recent neutral-metal-atom density measurements suggest that the fractional ionization is relatively low in laser ablation plumes from metals after the end of the laser pulse (due to recombination).[6–8] However, laser ablation research on ceramics has shown singly and doubly ionized species are ejected.[9,10] Thus, a technique for adjusting the fractional ionization after the laser pulse in laser ablated plumes from metals could be useful in film deposition studies. Another advantage of ions (over neutral atoms) is their ability to be controlled directionally by electric fields (e.g., for deposition in microelectronic trenches). Recently, Dyer's group has used magnetic fields to enhance the ionization in MgO laser ablation.[10] A number of researchers have used unmagnetized plasma discharges to stimulate the production of and deposition of carbon/amorphic diamond films.[11,12] Previous research in these authors' lab has employed laser-guided discharges (30 kV, 10–20 kA) in atmospheric-pressure gases for localized melting of metals.[13]

In this article, we describe experiments in which laser-ablation-assisted-plasma discharges (LAAPD) have been utilized with high voltage (1–4 kV) and high current (0.2–1 kA) in vacuum to enhance the ionization in laser-ablation plumes. The large electrical power (~MW) supplied by a pulsed capacitor bank is sufficient to generate a nearly fully ionized plume of metal ions from a laser ablated aluminum target. Spectroscopic measurements indicate metal ions are produced with charge states of Al^{2+} with electronic temperatures of 3.3 eV. LAAPD (of current density J) in a transverse magnetic field (B) also provide a potential means to accelerate a plasma plume in a preferred direction by the $J\times B$ force. This magnetized LAAPD technique might provide a means to separate ions from particulate or to vaporize particulate which is observed with conventional laser ablation.[14]

Figure 1(a) shows a simplified experimental configuration. The aluminum ablation plume is produced by focusing a KrF excimer laser (248 nm, 40 ns, $\leqslant$0.4 J) onto a 99.999% pure solid aluminum target. The local spot size is 0.06 cm^2 giving a typical fluence of 4 J/cm^2. The laser pulse energy is monitored by splitting off a fraction of the incident beam onto a calorimeter with a quartz flat. Figure 1(b) shows a magnified view of the target-electrode configuration. The target was made with a smooth, hemispherical shape to minimize electric field enhancement at the edges. The target functions as the grounded electrode while a copper (99.9% pure) annulus 0.5 cm from the target forms the high-voltage electrode. The discharge power is provided by a three-element L-C pulse-forming network (PFN) charged to 2.5–7.5 kV and switched by a triggered spark gap switch. A 10 Ω resistor placed in parallel with the target-electrode gap allows the discharge voltage to be monitored. PFN output current and plasma current are measured with (0.1 V/A) Pearson current transformers. The transverse dc magnetic field ($\leqslant$620 G) is generated by an electromagnet with pole pieces and is uniform to better than 2% in the plume expansion region. Time-resolved-gated-emission spectra were taken by focusing the plasma light from 1 mm in front of the ablation spot onto the slit of a Jarrell–Ash spectrograph coupled to a Tracor–Northern intensified optical multichannel analyzer. The spectrograph/detector spectral response combined with the use of glass optics only allowed spectra to be taken over the range of 380–600 nm. Evacuation of the glass four-way vacuum chamber was accomplished with a mechanically

[a]To whom correspondence should be addressed.

 0003-6951/94/65(5)/531/3/$6.00

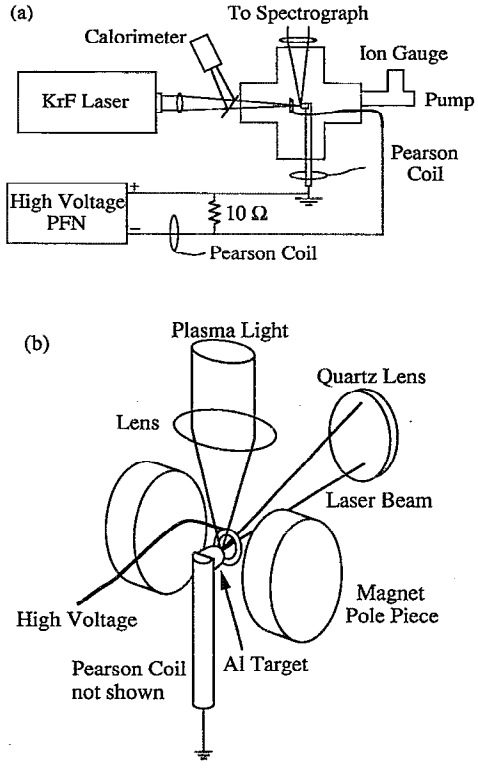

FIG. 1. (a) Simplified experimental configuration. Magnet pole pieces and return yoke not shown. (b) Close-up view of optics and target-electrode configuration.

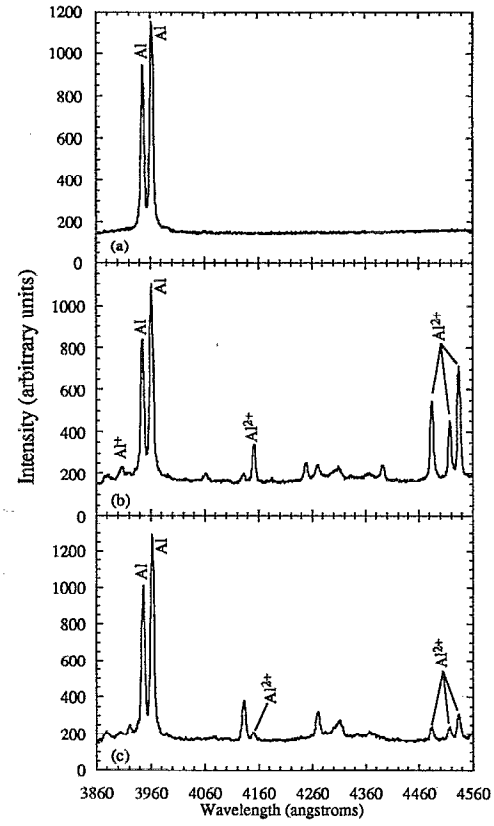

FIG. 2. Typical emission spectra: (a) laser ablation only; (b) laser ablation with 3 kV, 760 A discharge and no magnetic field; (c) laser ablation with 1.2 kV, 280 A discharge and 620 G field.

backed turbomolecular pump, while the pressure was periodically monitored with an ionization gauge. Ultimate pressures attained for this experiment were 1.3×10^{-4} Torr limited by the necessary use of a long, small-diameter pumping pipe.

Figure 2(a) shows a typical spectrum of a unmagnetized laser-ablation aluminum plasma without the discharge. The light was collected for 5.2 μs starting from the KrF laser pulse. Although the plasma light is only produced during the first 400 ns after the ablation pulse,[15] the long gate-pulse length was chosen to be consistent with that used during the discharge shots since the evolution of the current and voltage was on a much longer time scale. It is seen that the only spectral lines present are the 394.40 and 396.15 nm Al neutral transitions. Additional spectra taken to cover the 380–600 nm range show no additional lines, confirming that the fractional ionization in the plume is very low, as observed by other researchers.[6–8,16]

Figure 2(b) shows a typical LAAPD spectra for 760 A peak plasma current and 3 kV peak discharge voltage without magnetic field. The maximum instantaneous power is 1.1 MW which results in 4.4 J deposited in the plume and electrodes during the 5.2 μs gate pulse. The PFN output current, the plasma current, the laser pulse as observed on a PIN photodiode, and the OMA gate pulse are shown in Fig. 3. The discharge spark-gap switch is triggered 100 ns after the KrF laser pulse. It should be noted that the discharge would not initiate without the laser ablation plume. The spectrum in Fig. 2(b) shows that, with the discharge, Al^{2+} charge states are created as evidenced by the Al^{2+} spectral lines at 415.02, 447.99, 451.26, and 452.92 nm. Additionally, there is a small peak at 390.07 nm from singly ionized aluminum (Al^{+}) atoms. Spectra taken in different wavelength ranges show strong Al^{2+} lines at 569.66 and 572.27 nm as well as a small

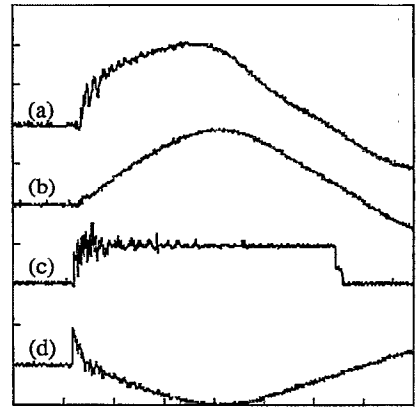

FIG. 3. Typical LAAPD waveform data: (a) PFN output current (400 A/div); (b) plasma current (400 A/div); (c) spectrometer gate pulse (4 V/div); (d) PIN diode signal of KrF laser pulse (2 V/div). Time scale is 1 μs/div.

TABLE I. Accepted Al^{2+} wavelengths, corrected measured line intensities, statistical weights, transition upper energy levels and Einstein coefficients (see Ref. 18).

λ_{ki} (nm)	I_{ki} (a.u.)	g_k	E_k (eV)	A_{ki} (10^8 s^{-1})
415.02	284	14	23.53	2.19
451.26	261	4	20.55	2.15
452.92	544	6	20.55	2.54
569.66	151	4	17.81	0.88
572.27	72	2	17.80	0.87

466.68 nm Al^+ intensity. Comparing Figs. 2(a) and 2(b) it is observed that the intensity of the Al neutral spectral lines are approximately the same. Statistical data from many shots confirms this, implying that no additional neutral atom excitation is caused by the discharge. Neutral atom emission occurs during the first 400 ns after laser ablation; during this time, the discharge voltage and current have not reached appreciable levels, so the neutral emission and the ion emission events can be separated in time. At the time of the experiment, delay between the gate pulse and laser/discharge was not possible; characterization of the emission as a function of time will be reported in a following article. The additional peaks in the spectrum of Fig. 2(b) are from impurities in the vacuum chamber, possible flashover of a polyethylene insulator, and copper atoms from the annular electrode.

After correcting the measured spectral-line intensities for the spectral response of the optical system, the relative intensities of the various Al^{2+} lines can be compared to produce an atomic-Boltzmann plot which gives an estimate of the Al^{2+} electronic temperature. To determine an estimate for a species electronic temperature the following relation is used:[17]

$$\ln\left(\frac{I_{ki}\lambda_{ki}}{g_k A_{ki}}\right) \propto \frac{E_k}{k_B T}, \tag{1}$$

and plotting the natural logarithm versus the right-hand side should give a straight line with the negative inverse of the slope being the electronic temperature for the particular species. Although this is only valid for equilibrium, it can provide a reasonable estimate for the Al^{2+} species in the discharge. Temperatures for other species were not possible to determine due to the lack of sufficient spectral lines with separated upper energy levels. Table I shows the Al^{2+} corrected measured line intensities, statistical weights, transition upper energy levels, and spontaneous emission Einstein coefficients.[18] The intensities shown are the average of 15 shots taken under identical conditions to correct for the shot-to-shot variation in spatial laser fluence leading to varying discharge quality. For the discharge (at 3 kV, 760 A) without the magnetic field, the atomic-Boltzmann plot gives an Al^{2+} electronic temperature of 3.3 eV. Due to insufficient statistical data for the 569.66 and 572.27 nm lines with the magnetic field present, an estimate for the temperature was not possible.

Additional spectra taken for 1.2 kV and 280 A unmagnetized discharges for the wavelength range in Fig. 2 show little evidence of the Al^{2+} emission as seen in Fig. 2(b) although the Al^+ emission at 390.07 nm is greater than observed with the 3 kV, 760 A unmagnetized discharge. The lack of visible Al^{2+} emission implies the fraction of Al^{2+} ions is small because the plasma temperature is much lower. Figure 2(c) shows a plasma spectrum with the application of a 620 G transverse magnetic field (at 1.2 kV, 280 A); the visible wavelength Al^{2+} emission resembles Fig. 2(b) (at 3 kV, 760 A) and the Al^+ emission at 390.07 nm intensity is reduced over that seen without the magnetic field at the lower voltage. This suggests that the magnetic field enhances the excitation and ionization of the plume through improved electron confinement. Enhancement of laser ablation plume (without discharge) excitation and ionization due to the presence of a transverse magnetic field has also been observed with ablation of MgO by Dyer *et al.*[10] Experiments studying the characterization of the emission as the discharge evolves and the dependence on discharge parameters are currently underway and will be reported in a future article.

This research is supported by National Science Foundation Grant CTS-9108971 funded jointly by DDM. J. S. L. has a National Science Foundation Graduate Fellowship. The authors acknowledge the technical assistance of A. Edson, C. Pae, R. Spears, and R. Kishek and the spectroscopy advice of Professor M. L. Brake.

[1] G. L. Doll and J. A. Sell, Phys. Rev. B **43**, 6816 (1991).
[2] R. K. Singh, J. Narayan, A. K. Singh, and J. Krishnaswamy, Appl. Phys. Lett. **54**, 2271 (1989).
[3] L. Ganapathi, S. Giles, R. Rao, J. P. Zheng, and H. S. Kwok, Appl. Phys. Lett. **63**, 2552 (1993).
[4] A. N. Zhirikin, V. N. Bagratashivili, O. V. Boyarkin, and V. N. Burimov, *Laser Radiation Photo-Physics,* edited by B. Braren and M. N. Libenson (SPIE, Bellingham, WA), Vol. 1856, p. 92 (1993).
[5] Q. Y. Ying, D. T. Shaw, and H. S. Kwok, Appl. Phys. Lett. **53**, 1762 (1988).
[6] R. A. Lindley, R. M. Gilgenbach, and C. H. Ching, Appl. Phys. Lett. **63**, 888 (1993).
[7] R. M. Gilgenbach, C. H. Ching, J. S. Lash, and R. A. Lindley, Phys. Plasmas **1**, 1619 (1994).
[8] R. W. Dreyfus, J. Appl. Phys. **69**, 1721 (1991).
[9] J. T. Dickinson (private communication).
[10] P. E. Dyer, L. Dirnberger, S. R. Farrar, and P. H. Key, *Proceedings of the Second International Conference on Laser Ablation* (AIP, New York, 1993).
[11] C. B. Collins, F. Davanloo, E. M. Jeungeman, W. S. Osborn, and D. R. Jander, Appl. Phys. Lett. **54**, 216 (1989).
[12] J. Krishnaswamy, A. Rengan, J. Narayan, K. Vedam, and C. J. McHargue, Appl. Phys. Lett. **54**, 2455 (1989).
[13] R. M. Gilgenbach, O. E. Ulrich, and L. D. Horton, Rev. Sci. Instrum. **54**, 109 (1983); M. L. Brake, R. M. Gilgenbach, L. D. Horton, and J. E. Tucker, Plasma Chemistry and Plasma Proc. **3**, 367 (1983).
[14] D. B. Geohegan, Appl. Phys. Lett. **62**, 1463 (1993).
[15] M. L. Brake, J. Meachum, R. M. Gilgenbach, and W. Thornhill, IEEE Trans. Plasma Sci. **PS-15**, 73 (1987).
[16] R. Poprawe, M. Wehner, G. Brown, and G. Herziger, in *High Power Lasers* edited by E. W. Kreutz, A. Quenzer, and D. Schuocker (SPIE, Bellingham, WA, 1987), Vol. 801, p. 191.
[17] H. R. Griem, *Plasma Spectroscopy* (McGraw–Hill, New York, 1964).
[18] W. L. Wiese, M. W. Smith, and B. M. Miles, *Atomic Transition Probabilities, Vol. II: Sodium Through Calcium* (National Bureau of Standards, Washington, 1969).