Time-of-Flight Measurement of a 355-nm Nd:YAG Laser-Produced Aluminum Plasma

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ABSTRACT

An aluminum target in air was irradiated by a 355-nm Nd:YAG laser with a pulse width of 10 ns and a repetition rate of 10 Hz. The emission spectra of the laser-produced aluminum plasma were investigated with varying distances from the target surface. The results show the presence of a strong continuum very close to the target surface, but as the plasma evolve in space, the continuum gradually disappears and the emitted spectra are dominated by stronger line emissions. The observed plasma species are the neutral and singly ionized aluminum and their speeds were investigated using an optical time-of-flight measurement technique. Results show that the speeds of the plasma species decreases gradually with distance from the target surface. Comparison of the computed speeds of the plasma species shows that the singly ionized species have relatively greater kinetic energy than the neutral species.

INTRODUCTION

Laser ablation of solids with short-pulsed, high-intensity light source has led to complicated light matter interactions. Some of the fundamental physical features of these interactions have not yet been fully understood, but nevertheless various practical applications have been developed in recent years, such as a cutting tool for metals, an elemental and chemical composition characterization technique (Corsi et al., 2001), and a promising chemical deposition technique (Richter, 1990).

Pulsed laser deposition of thin films has evolved into a well-recognized technique for a wide range of materials and in a variety of devices, especially in forming multicomponent films from stoichiometrically complex target source. Among other factors that affect the quality of the deposited film, a key parameter is the kinetic energy of the vaporized atoms and ionized species. The physical characteristics of the ejected species play a crucial role in the microscopic mechanism of film growth (Corsi et al., 2001; Willmott & Huber, 2000).

The study of laser-produced plasma (LPP) is essential to understanding the light-matter interactions and the numerous applications it entails. In this paper, we report a study that aims to investigate the parameters and characteristics of the plasma produced during this high-energy laser ablation of solid materials. The plasma parameters that were investigated were the spatial evolution of the plasma emission spectra and the velocities of the different species produced specifically by the neutral and singly ionized aluminum.

Optical emission time-of-flight measurement is the method used in the determination of the speeds of the plasma species. This emission spectroscopy method was chosen because it is non-intrusive, fast, and

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provides an effective and reliable determination between the produced neutral and ionized species (Willmott & Huber, 2000; Wang et al., 1996).

EXPERIMENTAL PROCEDURE

The experimental setup is shown schematically in Fig. 1. The 355-nm beam generated by the third harmonic frequency of a Q-switched Nd:YAG laser (Spectra Physics GCR-230-10) was focused with a 254-mm focal length quartz UV lens, L1, onto the surface of the rotating aluminum target. The laser was operated with a pulse width of 10 ns and a repetition rate of 10 Hz. The plasma plume was collected by a UV lens, L2, and imaged by another UV lens, L3, to a magnification of 5x. The emission spectra of the plasma was measured using an optical fiber mounted on a 1D micrometer translational stage to a monochromator (SPEX 1000M) with 1600 grooves/mm grating installed and equipped with a photomultiplier tube (PMT) detector (Hamamatsu R212). The slits of the monochromator were set at 200-mm widths for a 1.6 Å resolution. The signal was rid of noise and further amplified by a digital lock-in amplifier (SRS SR830), digitized, and finally analyzed by a personal computer. The optical time-of-flight measurements were done by connecting the PMT directly to a 500-MHz digitizing oscilloscope (Tektronix TDS 644B) triggered by the TTL signal synchronized with the Q-switching of the laser while the monochromator was centered to the observed peaks of aluminum. The optical fiber detector was placed on the image plane and aligned with the centerline of the plume to detect only the plasma species traveling axially above the target surface along the centerline of the plasma.

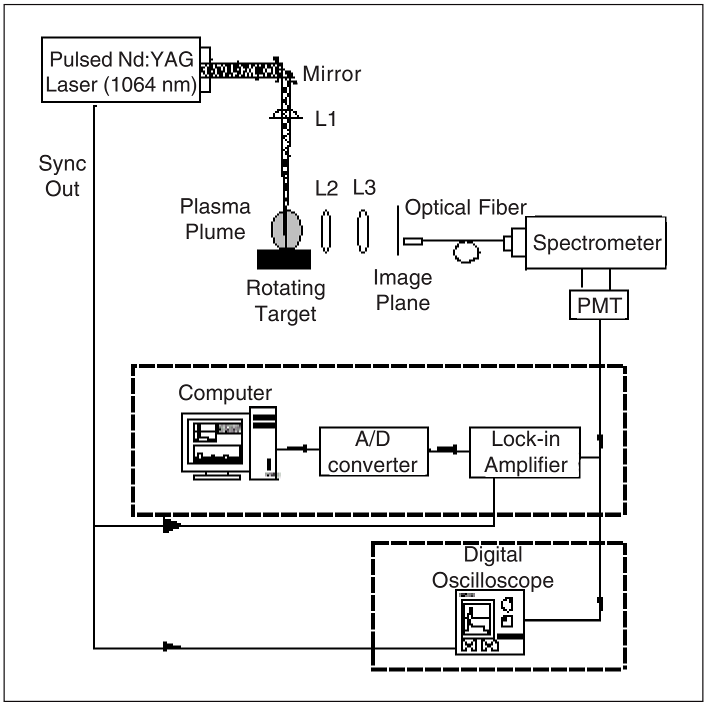

Fig. 1. Experimental setup.

RESULTS AND DISCUSSION

When a short-pulsed laser strikes a solid surface, the rapid rise in temperature leads to intense evaporation of atoms and molecules from the solid.

Even at relatively low intensities near the threshold for ablation, it is observed that the ablated material is significantly ionized, and the ions in the plasma plume can reach energies ranging up to several hundred eV. At the end of the laser pulse, the ablated material exists as a thin layer of plasma on the target surface. Initially, the expansion of the plasma plume is primarily driven by the plasma pressure gradients, but there may be additional contribution from Coulomb repulsion between the ions. When the plume has propagated more than a hundred micrometers from the target surface, the major part of the initial thermal energy in the plasma is converted to the directed kinetic energy of the ions (Willmott & Huber, 2000).

Plasma emission

The emission spectrum of the laser-produced aluminum plasma was recorded at different distances from the target surface. This was done by moving the optical fiber detector to increasing distances away from the target. Fig. 2 shows the spatially resolved and time-integrated spectra of selected neutral and singly ionized aluminum peaks. An intense continuum, very close to the target, was observed as the early development of the plasma. This continuum emission was attributed to both the elastic collisions of electrons with ions and atoms (free-free Brehmsstrahlung)

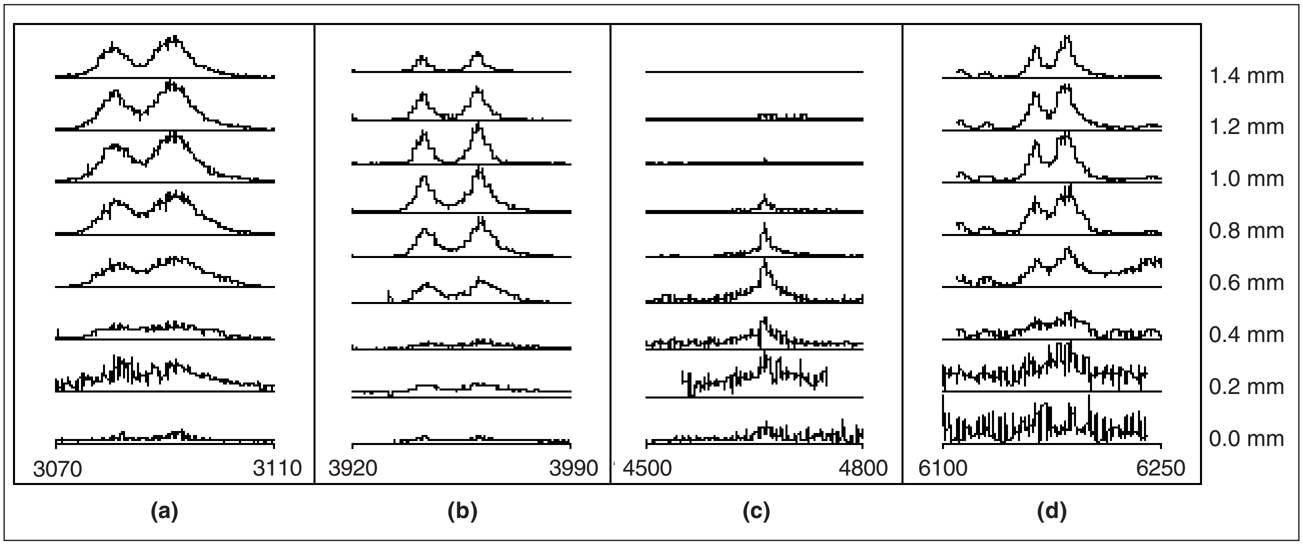

Fig. 2. Spatially resolved plasma emission lines of neutral (a & b) and singly-ionized (c & d) aluminum for distances of 0 mm to 1.4 mm from the target.

and the recombination radiation accompanying the electron-ion recombination (free-bound Brehmsstrahlung) (Willmott & Huber, 2000; Wolf, 1992). An example is the recombination process of the singly ionized species and an electron to form a neutral aluminum (Wolf, 1992).

As the plasma expands away from the target, it can be observed that the continuum gradually decreased and line emissions started to appear and become stronger than the continuum. At these distances, line emissions dominated the radiation process. This is manifested in Fig. 2, starting at distances 0.4 mm to 0.6 mm away from the target surface. However, as the plasma expands further starting from 1 mm, the intensity of the line emissions gradually decreased. It is shown that among the observed lines, Al (I) (4663 Å) decreased rapidly at 0.8 mm and diminished totally at 1.0 mm. This can again be attributed to the singly ionized ion-electron recombination, thereby producing a neutral aluminum (Griem, 1964).

Table 1 enumerates the observed line emissions of aluminum in the produced plasma. The theoretical values are the air wavelengths of aluminum (CRC Handbook of Physics and Chemistry, 2000) and the observed values are the average of the spatially resolved spectra. The deviation of the observed values from the theoretical ones can be attributed to the different line broadening and shifting mechanisms occurring in the plasma, such as the Doppler, pressure, and Stark broadening and shifting mechanisms (Bekefi, 1976; Griem, 1964). The ejection/expansion velocities of the radiating plasma species cause the Doppler shift while collisions in the plasma causes the pressure broadening. Stark broadening, which is the more prominent, is attributed to the interaction between the radiating species and the charged particles in the plasma.

Table 1. Observed Al (I) and Al (II) peaks.

Aluminum and their emission lines (Å)		Observed plasma emission lines (Å)
Al (I)	3082.15	3081.4 ± 4.2
Al (I)	3092.71	3091.6 ± 5.4
Al (I)	3944.00	3943.1 ± 3.3
Al (I)	3961.52	3960.8 ± 4.1
Al (II)	4663.05	4664.3 ± 8.1
Al (II)	6183.45	6185.0 ± 5.2

Expansion velocities of the ejected species

Fig. 3 shows the graph of the delay time of the Al (I) species relative to the laser pulse versus its probable location in the plasma. From this data, the speeds of the species were deduced. Estimates of their ejection speeds were obtained by determining their delay time as they propagate a certain distance along the plasma.

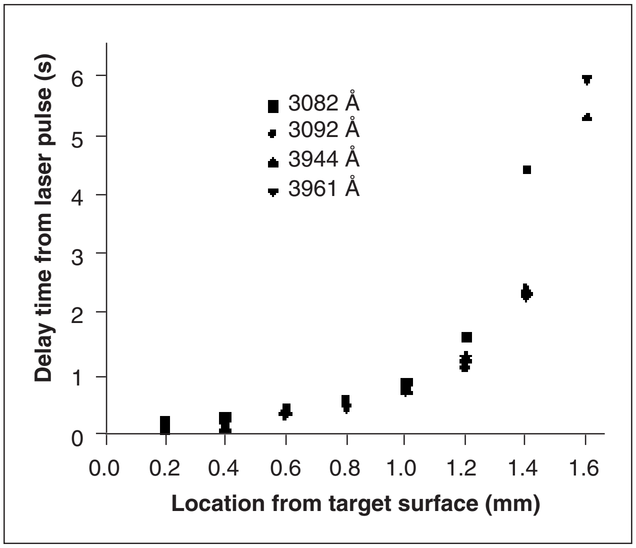

Fig. 3. Location of the Al (I) species and its delay time with respect to the laser pulse.

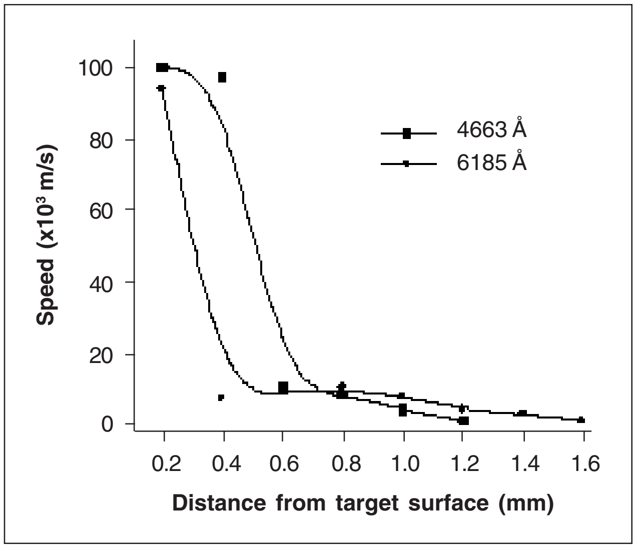

Fig. 5. Computed speeds for the singly ionized aluminum plasma species.

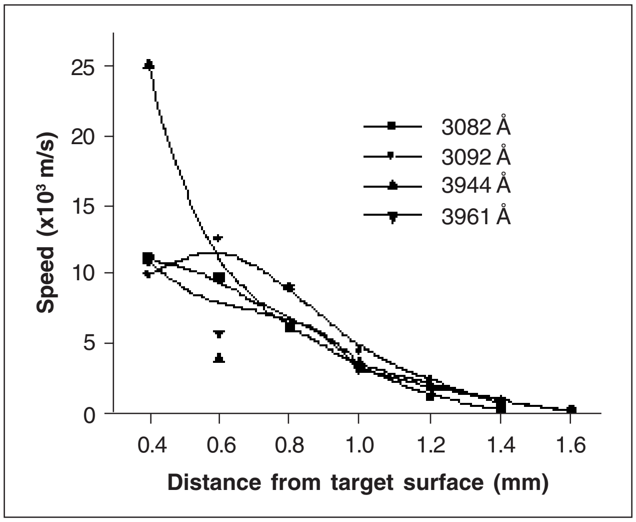

Fig. 4. Computed speeds of the neutral aluminum plasma species.

Thus, for the 3082 Å line with a distance between 1.2 to 1.4 mm from the target surface, we get an estimate of its ejection speed of 4.0 x 10^4 cm/s.

Figs. 4 and 5 show the computed speeds of the neutral and singly ionized aluminum species. It can be observed that the species acquired a very high ejection speed as it was exfoliated from the target surface and gradually decreased as it propagates along the plasma. This gradual slowing down of the plasma species is attributed to its collisions with the other plasma species, such as the electrons and the ions, and the presence of the surrounding air, which can cause an impediment to its expansion in space. It can also be observed that in comparison to the computed speeds of the selected plasma species, the singly ionized aluminum species have relatively higher speeds than the neutral aluminum species. The difference in their speeds can be attributed to the lesser mass of the singly ionized aluminum species due to the absence of an electron. Thus, it can be inferred that the singly ionized aluminum species have greater kinetic energy than the neutral species.

CONCLUSION

The spatially resolved spectra of the laser-produced plasma show that a strong continuum was generated close to the target surface, but as the plasma expands in space, stronger line emissions gradually appear. The "masking" of the line emissions near the target surface can be attributed to the many-body collisions, which occur as the plasma constituent species are ejected from the material surface. As the different species expand in space, collisions with other plasma species are reduced, and its further evolution in space is determined by its interaction with the surrounding air by elastic and inelastic collisions and recombination with air molecules and particles. The emission time-of-flight measurements were used to deduce the speeds of the individual species produced in the plasma. It was observed that the singly ionized species have greater kinetic energies than the neutral species. Continuing research is done on the investigations of the ejection speeds of the plasma species by observing as many species as possible to further verify our present results.

The current research is a part of a continuing project that aims to investigate the pulsed laser deposition parameters and eventually, the deposition of high quality thin films.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the assistance provided by Mr. Jonathan A. Palero and Ms. Marilyn A. Hui in the experiment. This research is funded by the Philippine Department of Science and Technology through the Engineering and Science Education Project and the Philippine Council for Advanced Science and Technology Research and Development (PCASTRD).

REFERENCES

Bekefi, G., 1976. Principles of laser plasmas. New York, Wiley.

Corsi, M., et al., 2001. A fast and accurate method for the determination of precious alloy caratage by laser-induced plasma spectroscopy. *Eur. Phys. J. D*. 13: 373-377.

CRC Handbook of Physics and Chemistry, 2000.

Griem, H.R., 1964. Plasma Spectroscopy. New York, McGraw-Hill.

Knudtson, J.T., et al., 1987. The UV-visible spectroscopy of laser-produced aluminum plasmas. *J. Appl. Phys*. 61(10): 4771-4780.

Richter, A., 1990. *Thin Solid Films*. 188: 275.

Wang, X.T. et al., 1996. Optical spectroscopy of plasma produced by laser ablation of Ti alloy in air. *J. Appl. Phys*. 80(3): 1783-1786.

Willmott, P.R. & J.R. Huber, 2000. Pulsed laser vaporization and deposition. *Rev. Mod. Phys*. 71(1).

Wolf, P., 1992. The plasma properties of laser-ablated SiO2. *Appl. Phys*. 72(4): 1280-1289.