Final Report on Laser Processing Analysis and Research Tracking Code 94-ERP-071

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Final Report on Laser Processing Analysis and Research

Tracking Code 94-ERP-071

Jim J. Chang, Charles D. Boley, and Earl R. Ault

Introduction

This Exploratory Research Project addressed the coupling between short pulse, short wave length, high-radiance laser beams and a solid substrate. The motivation for conducting this research stems from the ever increasing application of high-radiance lasers in processing materials for science and industry. As a result of extensive research and development activities at Lawrence Livermore National Laboratory (LLNL), there are now several lasers that can cut metals, ceramics, and composites rapidly with unmatched precision. Both gas and solid state lasers are now being employed in this area. The purpose of this project is to develop scientific understanding of the cutting process and to provide tools to optimize the laser system for various applications like precision hole drilling, cutting, welding, and ablative coating.

There is a significant amount of scientific data and model development that supports this area of study. Kar and Mazumder reported on studies of pulsed laser coupling for the creation of nano-particles.¹ This provided the initial motivation for our model. Here, we include a more detailed discussion of light absorption and propagation, and electron plasma effects and expansion into an ambient gas rather than only in a vacuum. Our first effort began in FY1994, which yielded the basic model. In FY1995, we refined the code and later identified a more appropriate equation of state (EOS). The experimental program and the modeling effort provided the essential parametric evaluations needed to benchmark the model. This program enabled us to discover several underlying plasma effects important to the cutting or ablation process. Extensive measurements of the plume of expanding vapor and optical emissions give plasma temperatures, densities, and hydrodynamic estimates of the vapor's neutral properties. In the first studies, both aluminum and iron are treated. Carbon studies were included in the final phases of this work.

Results

- Developed a comprehensive laser coupling and ablation model.
- Elucidated the parameter regime for ablative cutting using visible laser pulses of tens of nanoseconds in duration.
- Explained the reasons for poor coupling efficiencies when operating at high peak powers.
- Explained the reason for poor efficiency of cw or long pulse cutting.

- Showed the importance of beam quality and beam smoothing in the cutting process.
- Developed experimental techniques to diagnose the cutting process and optimize the laser system for various target materials.

Modeling Effort

DRILL is the code based on a hydrodynamic treatment of the solid surface, the melt layer, and vapor expansion. Plasma heating must also be included because of its major contribution to the energy balance of the problem. Appendix A (UCRL-JC-117078) of this report has the complete description of the code. During the first year of this project, DRILL was formulated and the numerics were worked out. Two variations are available for the one-dimensional treatment of the laser interaction with the surface: planar and spherical. By the end of that year, results were coming out of the code allowing comparison with experimental data being taken concurrently with stainless steel. The code accounts for all the expected physical effects including the time varying surface reflectivity obtained by measurement. As expected, we found that the use of visible laser light would be optimum in terms of coupling efficiency. At longer wavelengths, the plasma reflects more of the light thereby decreasing cutting efficiency. At shorter wavelengths, the photoionization of the metal vapor increases the plasma density, again reducing coupling efficiency. In the tens of nanosecond regime, the initial ablated solid is ionized and the further influx of heat is dominated by electron conduction from the critical surface of the plasma and the physical surface of the target.

The modeling activities in FY95 consisted of the following: investigating new effects with the code DRILL, extending the materials database of this code to include aluminum (in addition to the existing case of stainless steel), comparing results for aluminum with experiment, and identifying future physics additions to the code. These items are discussed in turn.

(1) New Effects

DRILL was used to study surface ablation where the vapor is modeled as expanding into a half sphere. (A previous applications dealt with deep hole drilling where planar expansion is most appropriate.) The description was still spatially one dimensional. The laser light travels to the target through a channel in the expanding gas, and the vapor heating is distributed uniformly in angle. Heat conduction into the target material was still considered planar. The results differed dramatically from those for deep drilling. Because of the increased volume available to the vapor, absorption in the vapor decreased and ablation per pulse increased. For illuminating stainless steel with green copper laser light of peak incident intensity 1 GW/cm^2 , the energy absorbed at the surface increased by a factor of about seven. With planar expansion, the

calculated surface temperature reached a maximum of 11,000°C and then fell within several nanoseconds to about 6000°C. Thereafter, it decayed over a period of several microseconds. With half-spherical expansion, on the other hand, the surface temperature rose quickly to about the same maximum, but fell much more slowly, reaching 6000°C only after the pulse length (about 70 ns). The amount of ablation increased by an order of magnitude, from 0.4 to 4 μm per pulse.

Another effect investigated was thermal expansion of the liquid. Liquid metals, such as iron and aluminum, expand by 25–30% when heated to boiling point; they are assumed to expand at about the same rate as they are heated further under pressure. With thermal expansion, DRILL predicted increased ablation (by about 25% at a power level of 1 GW/cm²) and a correspondingly thinner melt layer. Thus the ablation speed increases as the liquid edge density decreases, keeping the momentum into the material fixed.

(2) Database Additions

The quantities needed in extending the database to aluminum included the thermal parameters of the solid and liquid as well as the EOS of the vapor. The former quantities, insofar as they are known or can be approximated, were taken from the literature. For the EOS, we require the ionization fraction and the internal energy of the neutrals and ions as functions of the density and temperature for gaseous densities and temperatures up to several electron volts. This regime is sufficiently removed from that considered by theories, such as QEOS of More, et al. that a separate EOS code was written during LDRD activities in FY94. The code calculates Saha equilibrium using the empirical ionization energies and an assumed hydrogenic excitation spectrum. As expected, the pressure and energy for aluminum is somewhat less than for iron, since only three electrons contribute in aluminum.

A minor change in the materials database was the incorporation of a surface absorption model appropriate for metals.² While this model is only schematic, it includes the marked increase in absorption for temperatures above the melting point. The absorption thereafter saturates at a value which, in DRILL, had previously been used at all surface temperatures. The impact in calculations was small.

(3) Comparison with Experiment

Data taken by J. J. Chang include the positions of the shock surface and vapor-air interface as functions of time, and the electron density and temperature at selected points as functions of time. These were taken for surface ablation of aluminum. Comparison was made for measurements at an approximate peak power of 0.8 GW/cm^2 .

The earliest point for the measured shock surface at 100 ns corresponds to a speed of 4000 m/s. This is followed by a decay to ambient sound speed after about $8 \mu\text{s}$. The vapor-air interface initially expands at about half the shock speed and becomes stationary after about a microsecond. DRILL predicts a shock speed that agrees with experiment at the earliest and latest times but, the shock speed tends to decay somewhat more slowly at intermediate times. The interface shows a behavior similar to experiment except that it stabilizes in about $0.6 \mu\text{s}$ rather than a full microsecond.

In contrast with the relatively good agreement between shock and interface measurements, DRILL predictions of the electron temperature and density compare poorly with experiment. While the measured electron temperature near the surface is a few eV at early times, the code gives a temperature lower by an order of magnitude. Since this temperature lies far below the ionization threshold, the calculated electron density is insignificant compared with experiment. (Of course the calculated temperature and density increase with higher intensities.) The reason for this discrepancy seems to be the presence of nonthermal electrons, reminiscent of the situation in Inertial Confinement Fusion (ICF). The DRILL model assumes local thermodynamic equilibrium (LTE) and a single temperature for both electrons and atoms/ions.

(4) Future Directions

DRILL is complete and running. It is a valuable tool for studying laser ablation and cutting. In the future, the code could be further improved by incorporating the following suggestions. While the inclusion of nonthermal electrons should be considered at some point, our attention has also been focused on a second issue. This arises when laser intensities approach 1 GW/cm^2 and the liquid-vapor interface appears to be driven to the vicinity of the critical point. The following considerations are suggested.

First, imperfect gas corrections become important in the vapor near the ablation surface, since the critical density is typically a factor of two or so below that of the unheated liquid. At present, the vapor is treated as an ideal gas, in the sense that the pressure is the sum of the ideal pressures of the constituents. A natural EOS candidate is the Van der Waals theory, along with the Maxwell construction within the two-phase region. This EOS requires two parameters, which can be taken as the best established pair

among the critical temperature, pressure, and density. Setting this up for aluminum, one obtains seemingly realistic isotherms (e.g., the boiling point is reasonably accurate). The Van der Waals pressure could simply be added to the Saha pressure, with the ideal gas pressure subtracted so it is not counted twice. This combination of equations of state appears especially attractive in view of the minimal overlap between the regimes of importance of the two theories.

A second, more difficult, consideration is the problem that the surface temperature calculated in DRILL can formally exceed the critical temperature for sufficiently high intensities. Because the model assumes the existence of a distinct liquid-vapor interface given by the coexistence curve with only the vapor treated hydrodynamically, this situation calls for correction. One possible approach, which retains the simplicity of the vapor-pressure treatment and avoids the complexities of two-phase hydrodynamics, is to divide the medium into two regions: one below the critical temperature and one above it. Since the temperature gradient is large, the latter region is much thinner than the former, typically a fraction of a micron. It might prove possible to set up an approximate hydrodynamic treatment in the latter region, with the former region still subject to simple heat conduction.

Experimental Effort

In the experiment of laser processing analysis and research, we have investigated the hydrodynamics of vapor expansion, plasma density and temperature, laser-plasma interaction, and mechanisms of material removal with copper vapor lasers (CVLs). This report presents our findings on laser interaction with aluminum and carbon steel at laser intensity of 10^8 - 10^{10} W/cm², which is mostly commonly used in CVL material processing.

(1) Hydrodynamics of Vapor Expansion

We applied shadow and schlieren techniques with a 20-ns frequency-doubled YAG laser to visualize plume formation and to measure its expansion speed. The vapor plume was produced by focusing the laser light onto a rotating target (rod) under ambient air. A pulsed flashlamp (~ 1 μ s) has also been utilized in this experiment because of its superior density resolution during the later phase of vapor expansion. Temporal resolution of the flashlamp-produced plume image is achieved with a 20-ns gate camera.

The measurement of vapor expansion on aluminum reveals that strong plasma absorption-induced ignition occurs at laser intensity higher than ~ 2 GW/cm². This plasma ignition induces detonation wave with a speed of 10–25 km/s, which is 2–4 times faster than the primary vapor expansion speed. Assuming the hydrodynamic approximation allows the vapor density at the end of laser pulse is estimated to be $\sim 3 \times 10^{20}$ at the vapor front and

$\sim 10^{21}/\text{cm}^3$ near the surface. A surface pressure of a few thousand atmospheres is indicated. The average vapor temperature is estimated to be around 10,000 K when there is no laser heating. The observed plasma heating above the laser ignition threshold significantly raises the vapor front temperature to 30,000–40,000 K (i.e., detonation front) and lowers the vapor density to $\sim 2 \times 10^{19}/\text{cm}^3$.

The initial expansion of evaporated target material generates a spherical wavefront leading to a shock wave that compresses the air between the vapor and shock fronts. During the expansion, the vapor front slows and the shock gradually detaches from the vapor. During this time, the shock moves away from the target at almost the speed of sound in air leaving the air behind the shock is close to the ambient condition. The separation between vapor and shock starts at about 1 μs after the laser pulse. Increasing the laser radiance from 0.83 GW/cm² to 10 GW/cm² size of the vapor plume from ~ 1.2 to ~ 3.3 mm. The ablated vapor stays attached on the target surface and slowly mixes with the ambient air through Rayleigh-Taylor instability and binary gas diffusion. The entire process of vapor depletion takes a few milliseconds to complete.

(2) Plasma Characteristics of the Vapor Plume

We used emission spectroscopy to determine the plasma temperature and density. A gated (20 ns) OMA spectrometer with a 1200-groove/mm grating was employed. For the case of aluminum, emission lines of Al(I), Al(II) and Al(III) have all been observed. Substantial self-absorption occurs at the resonance lines of Al(I) due to the existence of a hot central core in the plume due to laser heating. The existence of a large temperature difference between the central core and the outer plasma leads to a plasma emission dominated by its core region when it is optically thin. In this experiment, we used three Al(II) emission lines (281.6, 358.6, and 466.3 nm) to construct a Boltzmann plot. The peak electron temperature at 10 GW/cm² is indicated to be ~ 3.5 eV (i.e., at 150 ns), which is very close to the estimated temperature of the detonation wave based on hydrodynamics. The electron temperature drops to ~ 1.8 eV at a laser radiance of 0.5 GW/cm². This temperature is substantially higher than the temperature derived from hydrodynamic data. Although the reason for this discrepancy is still unknown, it may be caused by unaccounted residual laser heating when the gas temperature is derived for the laser conditions below the threshold of a detonation wave. One of the distinct features we find in this measurement is that the electron temperature at the region half way from the surface is noticeably higher than at the surface. This is most likely a result of laser heating that occurs even at laser intensities below the plasma ignition threshold.

In this experiment, measurement of Stark broadening of the 358.6 nm emission line enabled us to determine the electron density of the blow off

plasma. The electron density during the initial 100 ns can not be estimated with this technique because of poor line resolution caused by broad band radiation. One hundred and fifty nanoseconds after the laser pulse, the electron density is $\sim 4.2 \times 10^{18}/\text{cm}^3$ at a laser intensity of $10 \text{ GW}/\text{cm}^2$. At $0.5 \text{ GW}/\text{cm}^2$ electron density drops to $2.3 \times 10^{18}/\text{cm}^3$. The spatial variation of electron density between the material surface and expansion front is relatively small, even though the neutral density drops rapidly. This would indicate that the degree of ionization at the expansion front is about 5–10 times higher than that near the surface. This is apparently caused by laser heating.

(3) Plasma Absorption

The measurements of electron temperature, electron density, and neutral density enable us to examine the plasma absorption mechanisms. The two dominant mechanisms responsible for plasma absorption at laser intensity used in this work are inverse bremsstrahlung and photoionization. Inverse bremsstrahlung absorption, which is mainly determined by electron density, has an absorption depth of $\sim 10 \text{ mm}$ for an aluminum plume even at a laser intensity of $10 \text{ GW}/\text{cm}^2$. This absorption is negligible for a $\sim 1\text{-mm}$ -size plasma. On the other hand, the degree of photoionization, which depends on the neutral density and temperature, is found to have prominent effects on plasma absorption, especially at higher laser intensities. Its absorption depth decreases significantly from 1 m to merely $20 \mu\text{m}$ as the laser intensity increases from $\sim 0.15 \text{ GW}/\text{cm}^2$ to $10 \text{ GW}/\text{cm}^2$. Evidently, photoionization absorption is the dominant effect at higher laser intensity because of the high density of excited neutral species that can be photoionized by copper laser photons. The marked increases of plasma absorption as laser intensity increases agrees with our observation of the plasma ignition threshold ($\sim 2 \text{ GW}/\text{cm}^2$). The increased plasma shielding at laser intensities above plasma ignition threshold prevents further laser-material interaction and limits the coupling of laser energy to the surface. Therefore there is an optimum operating point for a given material and laser spot size.

(4) Mechanisms of Material Removal

We found that mechanisms of material removal using copper lasers were sensitive to the material thermal properties. For materials with high thermal diffusivity and low melting point, such as aluminum, we find that expulsion of molten material by the recoil developed by the expanding vapor is more important than evaporation. Formation of liquid droplets during liquid ejection occurs ($1\text{--}2 \mu\text{s}$ delay) after the laser pulse on aluminum. This is likely to be caused by the formation of a relatively thick melt layer ($20\text{--}50 \mu\text{m}$). At higher laser repetition rates, the effect is even more pronounced because of larger thermal energy stored on the material surface

layer between pulses. Liquid ejection from aluminum leads to an approximately three times higher laser drilling speed than in the case of carbon steel. Metals with relatively low thermal diffusivity and higher melting point, such as carbon steel, have less tendency to form a thick melt layer. Consequently, expulsion liquid droplets are less pronounced, and material removal by evaporation dominates the process.

Since the removed material is mostly in the form of a dense vapor, this strongly suggests that the surface temperature during laser pulse is well above the material critical temperature such that surface tension is negligible for a prolonged period of time. This observation agrees quite well with our estimation of surface temperature based on the hydrodynamics of vapor expansion. In Appendix B (UCRL-JC-123313), we provide a preprint of a paper describing this work on metals submitted to the *Journal of Applied Physics Letters*.

(5) Carbon Ablation

With the installation of a small vacuum chamber, experimental studies of laser ablation from a graphite surface were undertaken. The purpose of the studies is to understand the ablation process and to study the nature of the resulting carbon vapor and plasma plume. This material has good commercial potential for producing diamond-like films or field emitters. By measurement we were able to determine the temperatures and densities leading to an optimization of the charge states of the carbon plasma. It has been determined that C^+ concentration is important to the formation of sp^3 diamond-like bonds at the surface of the substrate to be coated.³

We found that the quality of diamond-like coating (DLC) produced by laser ablation of graphite is sensitive to the laser intensity on the target surface. Best coating morphology has been produced at a laser power density of approximately 0.5 GW/cm^2 when using a copper vapor laser. The coating quality degrades substantially as laser intensity increases to 5 GW/cm^2 . Our measurements based on schlieren and optical spectroscopy relate DLC quality to the dynamics and composition of the ablated carbon plume, as described in the following paragraphs:

1. It has been found that surface micro explosions occur at or on graphite laser intensities greater than 1 GW/cm^2 . This micro explosion takes place long after the laser ablation process, which lasts for only 100–200 ns. At $\sim 500 \text{ ns}$ after the 60-ns laser pulse, particulates are ejected from the carbon surface for 5–10 μs . It's believed that subsurface superheating caused by absorbed laser energy followed by rapid surface cooling by evaporation leads to an internal superheated region that subsequently explodes late in time. The explosion produces

large amount of micro particulates at a temperature of 2500–3000 K and a speed of a few tens of meters per second. This observation agrees well with the increased micro particles on the DLC at higher laser intensity.

2. The optical emission from the carbon plume indicates that C and C₂ are the dominant species when the laser intensity is ~0.5 GW/cm². Above 1 GW/cm², C and C⁺ dominate. The larger amount of C₂ at lower laser power has been found to be the result of lower electron temperature and density of the carbon plume. This means the dissociation of C₂ is less likely (i.e., dissociation energy of C₂ is 5–6 eV). However, time of flight measurements indicate that the kinetic energy of the dimmers is 3–4 times higher at high laser intensities (5 GW/cm²) allowing the dimmers and electrons to flow together for a longer period of time causing additional dissociation. We believe this explains why we see lower dimmer densities by optical spectroscopy than expected at higher laser intensities even though greater dimmer production would be expected at the higher levels. The high-quality film produced under the condition of large amount of energetic C₂ signifies the importance of plasma characteristics in laser ablative coating.

Further data analysis of the carbon data is continuing with the results to be submitted for publication in the Spring of 1996.

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