MODELING OF DYNAMICAL PROCESSES IN LASER ABLATION

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Code: E-MRS 1995 Spring Meeting, COLA '95, Symposium F, Paper F-I.2

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ABSTRACT

Various physics and computational approaches have been developed to globally characterize phenomena important for film growth by pulsed-laser deposition of materials. These include thermal models of laser-solid target interactions that initiate the vapor plume, plume ionization and heating through laser absorption beyond local thermodynamic equilibrium mechanisms, hydrodynamic and collisional descriptions of plume transport, and molecular dynamics models of the interaction of plume particles with the deposition substrate.

I. INTRODUCTION

The laser ablation technique for pulsed-laser deposition of thin films has proven extremely successful at growing high-quality films of many materials [1], including some as complex as high-temperature superconducting compounds. The physics ingredients that come into play in laser ablation are also quite complicated given that they involve laser-solid interactions at the target, plasma formation off the target, vapor/plasma plume transport toward the deposition substrate with its associated hydrodynamics and atomic physics, as well as plume-solid interactions at the deposition substrate.

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We have taken a global physics and computational approach to laser ablation that relies on thermal models to describe laser–solid interactions; on kinetic models of plasma formation in the plume; on an assorted variety of hydrodynamic, gas dynamic, and collisional models of plume transport; as well as on molecular dynamics methods to treat plume–substrate interactions. We have chosen to concentrate mostly on silicon to validate our models. The application of our physics results does however go beyond silicon, given the universality of many experimental observations, such as plume splitting for instance [2], for a wide variety of laser-ablated elemental materials and compounds.

For laser–target interactions, the vaporization threshold and the rate of evaporation have been addressed with our thermal models. For plasma formation, vapor breakdown during the laser pulse has been tackled with nonequilibrium, kinetic rate equations models. For plume transport, the differing character of plume dynamics in near-vacuum and in the presence of a higher pressure background gas [3] has been treated with our gas dynamic and collisional models. For plume–substrate interactions, film damage by highly energetic plume particles has been investigated with molecular dynamics calculations.

II. LASER-TARGET INTERACTIONS

Thermal models of laser–solid interactions have already been successfully applied to laser annealing of semiconductors. As implemented in the one- and two-dimensional (1- and 2-D) Laser8 computer programs [4], these models solve the enthalpy ($\rho h = \rho e + P$, with ρ the density, h the enthalpy, e the internal energy, and P the pressure) diffusion equation using finite differences

$$\frac{\partial}{\partial t}(\rho h) = \frac{\partial}{\partial x} \left(\kappa \frac{\partial T}{\partial x} \right) + S. \quad (1)$$

The laser energy input is specified at each instant of time and each point in space through the source term S related to the intensity of the laser pulse Φ :

$$S(x, t) = [1 - R(x, t)] \Phi(t) \alpha e^{-\alpha x}. \quad (2)$$

For silicon, the absorption coefficient α is taken to be $\alpha = 1 \times 10^6 \text{ cm}^{-1}$ in both the solid and liquid phases; the reflectivity R is set at 0.58 for the solid phase and 0.69 for the liquid phase; and the profile of the thermal conductivity κ is as in Ref. 4. Phase transitions are handled through the state array concept that determines the state of each cell in space and time according to the appropriate (enthalpy h , temperature T) state diagram. In addition to the solid-liquid transition suitable for laser annealing, we have extended the process to include the liquid-vapor transition necessary to model the initial stages of laser ablation. The extended thermal model also includes the effect of the pressure at the liquid surface P_s on the vaporization temperature T_v through the Clausius-Clapeyron equation:

$$T_v = \left[\frac{1}{T_0} - \frac{\ln(P_s/P_0)}{\Delta H} \right]^{-1}, \quad (3)$$

where T_0 is the vaporization temperature at atmospheric pressure P_0 and ΔH is the latent heat.

Results from calculations with the 1-D version of the Laser8 computer program are displayed in Fig. 1. The depth of vaporization and maximum recession speed at atmospheric pressure as a function of laser energy density are displayed on the bottom. Both are linear with energy density and indicate that the vaporization threshold is around 4 J/cm². The vaporization temperature and threshold laser energy density for onset of vaporization as a function of pressure at the liquid surface P_s are displayed on the top. Both have a $\log(P_s)$ pressure dependence and indicate that a lower vaporization temperature and a lower threshold laser energy density are obtained in near-vacuum than at atmospheric pressure in accordance with the Clausius-Clapeyron equation. In particular, the predicted threshold laser energy density decreases from ~4 J/cm² at atmospheric pressure to ~1.5 J/cm² at a pressure of 1 mTorr.

The thermal model allows us to specify the initial conditions for the plume formation and transport models to be described next.

III. PLUME FORMATION

Before considering gas dynamic and collisional models of plume transport from the target to the deposition substrate, let us briefly discuss the issue of plasma formation. From theoretical calculations based on local thermodynamic equilibrium, it has proven difficult to ionize the silicon vapor plume according to the Saha equation at the typical vaporization temperature of 0.3 eV and at neutral densities ranging from solid density to many orders of magnitude less than solid density. The lack of electrons makes absorption of laser light by the vapor unlikely, be it through electron-neutral inverse Bremsstrahlung [5], electron-ion inverse Bremsstrahlung [5], or plasma resonance absorption [6].

These considerations have prompted us to investigate nonequilibrium processes for plume ionization and absorption of laser light by the plume. Our breakdown model for 248-nm KrF laser light interacting with silicon within a pulse length of ~ 80 ns has been described in detail elsewhere [7] and will only be briefly reviewed here. It is based on a kinetic model of breakdown first applied by Rosen et al. [8] to coupling of 1- μ s pulses of 0.35- μ m XeF laser radiation to aluminum alloys. Rate equations for densities and temperatures of electrons, excited state neutrals, ground state neutrals, and singly charged silicon ions are evolved in time. These include dilution upon vapor expansion and resupply of particles from vaporization according to the Clausius-Clapeyron equation at the normal evaporation temperature and various density and temperature acceleration and deceleration mechanisms whose rates are taken from Zel'dovich and Raizer [9].

The key ingredient of the model is the two-stage process of electron impact excitation from the ground state to the excited state for neutrals at an energy of 5 eV and the subsequent electron impact ionization and photoionization of the excited state neutral to a singly charged silicon ion, which requires an additional 3.1 eV. This is to be contrasted with the jump from ground state neutral to singly charged ion at an energy of 8.1 eV, which is therefore more difficult to achieve with energies of 5 eV per photon in the KrF laser pulse. Mechanisms for absorption of laser light now include photoionization of the excited state [9], in addition to electron-neutral and electron-ion inverse Bremsstrahlung [5].

Results from the solution of these zero-dimensional rate equations are shown in Fig. 2 where the evolution in time of the densities of electrons (N_e), ground state neutrals (N_0), and excited state neutrals (N_*) is displayed for a laser energy density of 6 J/cm² on the top and 30 J/cm² on the

bottom. Acceleration of the electron density is clear in both cases from an initial state given by the Saha equation for the priming electrons and a Boltzmann distribution for the excited state silicon neutrals. Production of a significant number of electrons and, therefore, plume breakdown do occur within the laser pulse at both of these energy densities, within 50 nsec for 6 J/cm² and < 20 nsec for 30 J/cm². However, breakdown does not occur within the laser pulse for the lower energy density of 5 J/cm².

We will compare our transport modeling results with experimental observations at low fluences for which particulates or clusters are absent. Because ionization is difficult to achieve from a theoretical point of view at these low fluences, we have concentrated on neutral fluids for our models of plume transport.

IV. PLUME TRANSPORT

Experimental observations have shown marked differences between plume expansion in vacuum and in the presence of a higher pressure background gas. These observations are common to a wide range of ablated materials including silicon, carbon, yttrium, and high-temperature superconducting compounds such as YBCO. Ablation in high-pressure ambient gases results in shock waves and expansion fronts propagating through the background gases. Time-of-flight measurements also show two components in the ion probe signals, an energetic component that propagates at vacuum speed and another that is more or less significantly slowed down depending on the pressure of the background gas [2]. We have applied several hydrodynamic, gas dynamic, and collisional or scattering models to study plume expansion in vacuum and in a higher pressure background gas. Results from some of these models will be described starting from more qualitative but higher dimensional ones and proceeding to the more detailed and flexible 1-D ones.

To investigate the gross hydrodynamic features of plume transport in vacuum and in background gas, we have used a 2-D gas dynamic model that solves conservation equations for mass density (ρ), momentum ($\rho \bar{v}$), and energy ($\rho e + 1/2 \rho v^2$):

$$\frac{\partial}{\partial t} \begin{Bmatrix} \rho \\ \rho \bar{v} \\ \rho e + \frac{1}{2} \rho v^2 \end{Bmatrix} = -\bar{\nabla} \cdot \begin{Bmatrix} \rho \bar{v} \\ P \bar{I} + \rho \bar{v} \bar{v} \\ \bar{v} \left[\left(\rho e + \frac{1}{2} \rho v^2 \right) + P \right] \end{Bmatrix}. \quad (4)$$

They are augmented by the following equation of state for internal energy e and pressure P :

$$P = (\gamma - 1)\rho e = \frac{\rho}{M} k_B T, \quad (5)$$

with $\gamma = 5/3$.

This model is numerically implemented using finite differences in space and the Rusanov scheme [10] in time. Results from the 2-D model are displayed in Fig. 3 for plume expansion in vacuum (left) and in background gas (right). Contours of total density (plume plus background) are shown as time progresses from top to bottom. It is evident that strong shocks are generated as the plume expands in presence of the background gas. The plume does in fact snowplow the background gas, giving rise to the crescent feature at the leading edge, which is clearly seen in Fig. 3. We note that the gross characteristics of the expansion in vacuum and in background gas are remarkably similar to the light emission patterns experimentally detected using a gated charged coupled device (CCD) camera [2]. We have also produced similar features [11] using a novel 2-D particle-in-cell hydrodynamic model [12] of plume transport in which particles representing elements of the fluid are followed in space and time in the pressure gradient field.

Characteristics of plume expansion in vacuum and in background gas have been examined in more detail using a 1-D gas dynamic model that solves the same conservation equations as the 2-D model, but includes ionization through the Saha equation and energy input through laser light absorption. The set of equations used in the 1-D model follows:

$$\frac{\partial}{\partial t} \begin{Bmatrix} \rho \\ \rho v_x \\ \rho e + \frac{1}{2} \rho v_x^2 \end{Bmatrix} = - \frac{\partial}{\partial x} \begin{Bmatrix} \rho v_x \\ \rho v_x^2 + P - \mu \frac{\partial v_x}{\partial x} \\ v_x \left[\left(\rho e + \frac{1}{2} \rho v_x^2 \right) + P - \mu \frac{\partial v_x}{\partial x} \right] \end{Bmatrix} + \begin{Bmatrix} S_p \\ 0 \\ \alpha \Phi(t) e^{-\alpha x} + S_e \end{Bmatrix}, \quad (6)$$

with μ denoting the viscosity and $P = (1 + \eta) \rho \frac{k_B T}{M}$. The ionization fraction η is determined by a simultaneous solution of the Saha equation:

$$\frac{\eta^2}{1 - \eta} = \frac{M}{\rho} \left(\frac{2 \pi m k_B T}{h^2} \right)^{\frac{3}{2}} e^{-\frac{I_p}{k_B T}}, \quad (7)$$

and the equation of state:

$$\rho e = \frac{\rho}{M} \left[\frac{3}{2} (1 + \eta) k_B T + \eta I_p \right], \quad (8)$$

where I_p is the ionization potential. This model is similar to the 1-D models of Vertes and coworkers [13,14], except that the Rusanov scheme is again used to solve the equations [10]. Our model also contains source terms for mass density and energy input denoted by S_p and S_e , respectively, in the mass density and energy equation. These source terms allow us to start the calculations with a clean slate and input mass and energy into the transport model according to the results from calculations of laser-target interactions using the Laser8 computer program. The mass density source is then given as $S_p = n_{liq} M v_{rs}$ and the energy one as $S_e = n_{liq} M k_B T_v / (\gamma - 1)$, with v_{rs} the recession speed, T_v the vaporization temperature, and n_{liq} the liquid density. For the duration of the laser pulse, these terms provide a dynamic source of mass and energy into the system.

Results from 1-D gas dynamic calculations of silicon plume transport in vacuum with the dynamic source effect and of calculations where the density and temperature profiles at the end of the laser pulse are taken as initial conditions and allowed to freely expand are displayed in Fig. 4. These calculations were performed without ionization of the vapor and without absorption of laser

light by the vapor plume. The time evolution of the pressure at the solid surface with the source effect and for free expansion are shown on the top, while the plume front position as a function of time in both of these cases is shown on the bottom. It is clear from Fig. 4 that high pressure at the surface is maintained for a longer time due to continuous ablation for the duration of the laser pulse compared to the free expansion case where the pressure at the surface rapidly drops as $1/t$. As a result, the plume expands with a higher maximum velocity than in the free expansion case as also shown in Fig. 4 where the plume front speed is $\sim 1 \times 10^6$ cm/sec with the dynamic source effect compared to $\sim 5 \times 10^5$ cm/s in the case of free expansion. More details on the dynamic source effect for plume transport in vacuum, including analytical expressions for the steady-state density profile and maximum front velocity, can be found in a companion paper [15].

Plume free expansion in vacuum and in a higher pressure background gas has also been studied with our 1-D gas dynamic model, albeit for silicon expanding into a silicon background. For the calculations with background gas presented in Fig. 5, the background density n_b was set at $n_b/n_p = 5 \times 10^{-3}$ compared to the plume density n_p . Comparison of the density profiles in vacuum and in background gas displayed in Fig. 5(a) and (c) shows that the plume snowplows the background gas, which is pushed ahead of the plume, while the expansion of the plume is slowed down in presence of the background gas. The particle fluxes displayed in Fig. 5(b) and (d) show that the snowplowed background gas reaches the probe at 5 cm from the target at plume vacuum speed, with the plume arriving later at the detector because of slowing down from interaction with the background gas.

The results of pursuing the time evolution further are shown in Fig. 6 for a calculation with dynamic source effect and a silicon background gas pressure of 200 mTorr. The density and velocity profiles are displayed as a function of distance from the target at four different times in the calculations up to 500 μ s. Snowplowing of the background gas at the leading edge (a); rarefaction of the plume (b); slowdown and turnaround of the plume peak, the peak between target and front, by the snowplowed and piled-up background gas at the leading edge (c); and the subsequent reflection of the plume peak from the target (d) lead to multiple shocks between target and front, as observed in experiments performed in high background gas pressure [3].

Other approaches that show promise in elucidating the phenomena that take place when the plume interacts with a background gas include a hydrodynamic model with two distinct species and collisional drag between them [16], another model first proposed by Koopman and Goforth [17], and also a scattering model we have recently developed. Fluxes obtained from the two-species hydrodynamic model with collisional drag show that the plume component has penetrated through the background gas and reaches the detector placed at 5 cm at essentially vacuum speed, with the dragged background gas reaching the detector some time later. The model of Koopman and Goforth relies on a fully ionized layer of background gas to scatter the plume ions through ion-ion collisions. It is only by including this interaction that two velocity components are observed in the plume fluxes detected 5 cm from the target in the numerical calculations.

Our scattering model includes plume-plume, plume-background, and background-background collisional interactions with cross sections that depend on the difference of the velocities to various inverse powers. The calculated fluxes at a distance of 2 cm from the target are displayed in Fig. 7. They indicate that either the background gas or plume gas can get to the probe first depending on whether the background gas is lighter, such as helium, or heavier, such as argon, than the silicon plume, with velocities exceeding plume vacuum speed in the helium case under conditions appropriate for elastic collisions. Splitting of the plume into two energetic components is also perceptible in Fig. 7 in the case of an argon background. This effect would have been stronger if the scattered material had been tracked as yet another species throughout the calculation. Extensions of this promising scattering model along these lines are currently under way.

V. PLUME-SUBSTRATE INTERACTIONS

Time-of-flight measurements [3], as well as the model calculations presented here, clearly show that material reaches the deposition substrate at speeds equal to and even exceeding plume speed in vacuum even in the presence of a background gas, followed by a slower moving component whose velocity depends on background pressure. It is expected that the fast-traveling component may cause film damage due to the high kinetic energy of these particles. In the case of deposition of copper films, time-of-flight measurements indicate that the fast-moving particles can have kinetic

energies in excess of 200 eV, while the energies of the slow-moving particles are < 100 eV under normal ablation conditions.

We have performed calculations using the embedded atom method of molecular dynamics to assess the effect of particle kinetic energies on the film quality by pulsed-laser deposition from a copper target. A copper particle above the Cu(100) surface is allowed to hit the surface at velocities corresponding to kinetic energies ranging from 100 eV to 200 eV. The most probable point on the surface for the incoming particle to hit was found to be the open crystal channel along the $\langle 100 \rangle$ direction on Cu(100). Trajectories of the particles and substrate atoms are recorded as a function of time. The critical energy for penetration of a copper particle into the Cu(100) surface is found to be ~ 150 eV, as shown in Fig. 8. At this energy, the particle touches the surface atoms but does not penetrate. An energetic copper particle with a kinetic energy of 200 eV can however penetrate the first layer of the Cu(100) surface, as seen in Fig. 8. The hopping distances for this 200 eV particle are large initially and gradually decrease as the particle loses its kinetic energy. It finally comes to rest after hopping about 5 Å along the $\langle 110 \rangle$ surface channel. It is therefore possible for 200 eV particles to cause permanent mechanical damage according to these molecular dynamics calculations.

VI. SUMMARY

We have described a global approach to modeling of the laser ablation process with several physics and computational models applied to laser-target interactions, plume transport in vacuum and in background gas, and plume-substrate interactions. The key issues of evaporation rate and threshold, plasma formation, plume transport in near vacuum and in presence of higher pressure background gases, and mechanical damage to films by energetic particles have been addressed with a measure of success. Quantitative comparisons with experiments will continue, as well as extensions of the current models to treat not only elemental materials but also compounds.

VII. ACKNOWLEDGMENTS

This research is sponsored at the Oak Ridge National Laboratory (ORNL) by Laboratory Directed Research and Development Funds and by the Division of Materials Sciences, U. S.

Department of Energy, under contract DE-AC05-84OR21400 with Lockheed Martin Energy Systems, Inc. K. R. Chen and Chun-Li Liu are supported by an appointment to the ORNL Research Associate Program, administered jointly by the Oak Ridge Institute for Science and Education and ORNL. Alex Puzetky is on assignment to ORNL from the Institute of Spectroscopy, Russian Academy of Sciences, Troitsk, Russia.

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Figure Captions

Fig. 1 Results from 1-D Laser8 for silicon: vaporization temperature and laser energy density threshold for vaporization as a function of surface pressure (top) and vaporization depth and maximum recession speed as a function of laser fluence at a liquid surface pressure of 1 atm (bottom).

Fig. 2 Results from kinetic model of breakdown for silicon: ground state neutral (N_0), excited state neutral (N^*), and electron (N_e) densities in the plume as a function of time for laser energy densities of 6 J/cm^2 (top) and 30 J/cm^2 (bottom).

Fig. 3 Results from 2D gas dynamic model: contours of density for plume expansion in vacuum (left) and in background gas (right) as a function of time (top to bottom).

Fig. 4 Results from 1-D gas dynamic model with dynamic source effect from continuous ablation of material within the laser pulse: pressure at the solid surface with dynamic source effect and for free expansion (top) and position of plume front in these same two cases (bottom) as a function of time.

Fig. 5 Results from 1-D gas dynamic calculations of plume free expansion in vacuum and in background gas: a) density profile of plume in vacuum at $10 \mu\text{s}$; b) plume flux in vacuum as a function of time at 5 cm from the target; c) total, plume, and background density profiles at $10 \mu\text{s}$; d) total, plume, and background fluxes as a function of time at 5 cm from the target.

Fig. 6 Results from 1-D gas dynamic calculations of plume expansion in background gas at a pressure of 200 mTorr: density (full curve) and velocity (dashed curve) profiles as a function of distance from the target at various times during the evolution.

Fig. 7 Results from collisional scattering model for a silicon plume into argon (top) and helium (bottom) background gases: fluxes at 2 cm from the target for the plume in vacuum and for the plume in 200 mTorr background gases, and background fluxes.

Fig. 8 Results from molecular dynamics calculations using the embedded atom method of film damage by energetic particles: vertical trajectories of a copper plume particle with kinetic energies of 150 eV (top) and 200 eV (bottom).

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Figure 1 [Leboeuf et al.]

Figure 2 [Leboeuf et al.]

Vacuum

Background Gas

Figure 3 [Leboeuf et al.]

Figure 4 [Leboeuf et al.]

Figure 5 [Leboeuf et al.]

Figure 6 [Leboeuf et al.]

Scattering model, $\sigma = 5 \times 10^{-17} \text{ cm}^2$

Figure 7 [Leboeuf et al.]

**Critical Kinetic Energy for Penetration: 150 eV
for Neutrals Bombardment of Cu(100)**

**200 eV Neutrals Bombardment of Cu(100)
Trajectory of the Energetic Particle**

Figure 8 [Leboeuf et al.]