Effect of initial plasma geometry and temperature on dynamic plume expansion in dual-laser ablation

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Recent experiments have revealed the capability of large-area, uniform film growth using dual-laser ablation. The mechanism of this dynamic plume expansion is investigated in this letter. We report the critical role played by the initial geometry and temperature of the plasma in the subsequent expansion under dual-laser ablation. Initial plasma dimensions in the dual-laser ablation of ZnO are quantified by gated intensified charge-coupled detector-array imaging and combined with a hydrodynamic theoretical expansion model to yield radial thickness profiles for the deposited films. Comparisons with ellipsometric film thickness profiles indicate that the primary factors responsible for increased expansion of the dual-laser ablated plume are an extension of the initial plasma dimension in the axial direction as well as enhanced plasma temperature. © *1999 American Institute of Physics.* [S0003-6951(99)03011-9]

Ablation of targets by pulsed excimer lasers to grow thin films of a variety of materials, and the development of a basic understanding of the underlying mechanisms governing this process have been active research areas for over a decade.[1–4] Recent experiments on a dual-laser ablation technique for the growth of large-area, particulate-free thin films have indicated a dramatic change in the dynamic evolution of the excimer laser ablated plume upon incorporation of a second temporally synchronized CO_2 laser pulse on target. The observed increase in expansion of the laser ablated plume under dual-laser ablation was found to be critically dependent on a suitable adjustment of the interpulse delay on the tens of nanoseconds time scale.[5,6] We present, in this letter, the mechanism that is responsible for this enhanced expansion.

It is well known that the rapid deployment of laser energy into a thin layer of the target surface within the short excimer laser pulse duration in conventional single-laser ablation triggers an explosive evaporation. This leads to congruent evaporation that is considered to be one of the main advantages of pulsed laser ablation, especially for multicomponent film growth.[7] There are two distinct phases during the evolution of the laser ablated plume which include the generation of a highly collisional plasma followed by an essentially collisionless adiabatic plume expansion in vacuum. The first phase is initiated by the removal of a layer of material by the laser pulse which, depending upon the thermophysical properties of the target material, typically ranges from submicron to tens of microns in depth. As the laser pulse heats the evaporated material, a thin layer of plasma is formed on the surface of the target. The latter part of the laser pulse is absorbed into the plasma by the inverse bremsstrahlung process. While the lateral dimensions of the plasma (parallel to the target) resemble the laser spot size at the target, the axial dimension (perpendicular to the target) depends on the absorption by the plasma of the laser radiation. Typically, the hot initial plasma extends about 20–100 μm from the target surface.[4] The subsequent adiabatic expansion phase of the plasma, which follows, is governed by the pressure gradients across each dimension of the plasma. For a laser spot size with a large aspect ratio the transverse expansion profile tends to be elliptical with plasma along the shorter dimension expanding more than in the long dimension.[4] Since the plasma dimension in the axial direction is the smallest, the plume expansion is highly forward directed. Expansion profiles of the form $\cos^n \theta$ with n in the range of 6–70 have been reported for single laser ablated plumes.[8] As a result, the uniform area of film deposition is limited. In dual-laser ablation, the CO_2 laser is synchronized in time so that it is efficiently absorbed into the initial plasma layer. The consequence of this absorption is twofold. The inverse bremsstrahlung absorption that is enhanced at the larger CO_2 laser wavelength leads to an increase in plasma temperature as well as an increase in axial plasma dimension. We present an investigation of the role of these two effects on the mechanism of plume dynamics in dual-laser ablation.

The dual-laser ablation system used in our experiments has been described in detail in a previous publication.[5,6] The KrF excimer laser was focused to a $4.5\times 2\ \mathrm{mm}^2$ area on a hot-pressed high-density ZnO target in vacuum. The circular profile of the 2.5-mm-diam CO_2 laser pulse was overlapped with the rectangular excimer laser profile at the target. The laser energy fluence of the excimer and the CO_2 lasers at the target were 1 and 3 J/cm^2, respectively. The CO_2 laser by itself did not cause any noticeable ablation. A 50 ns interpulse delay between the onset of the CO_2 laser pulse and the excimer laser pulse, which was determined to be the optimum delay for oxide targets in a previous experiment,[6] was used in this study. Films were deposited on silicon wafers placed 4.5 cm from the target in a 10^{-5} Torr chamber pressure. The ZnO films deposited under these conditions were optically transparent at a substrate temperature of about 250 °C. The thickness profiles of the single laser and dual-laser ablated films along two orthogonal directions (y and z)

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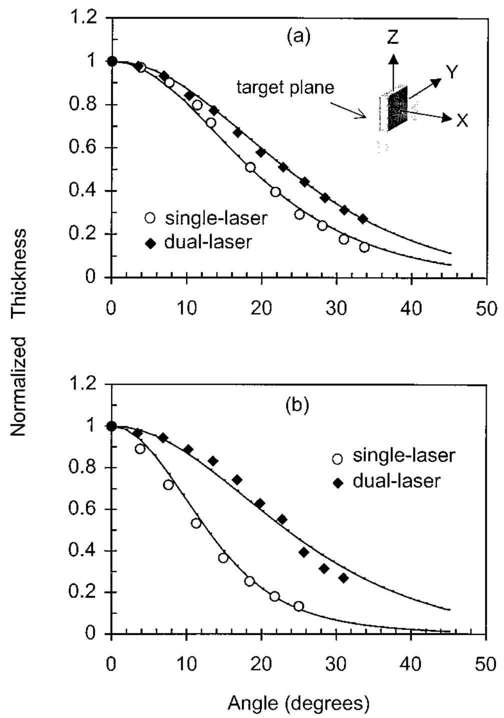

FIG. 1. Ellipsometric film thickness profiles along (a) the y axis and (b) the z axis for ZnO grown on Si. Solid lines indicate the theoretical fit.

were determined by using ellipsometric measurements. The angular variation of the normalized film thickness along the two transverse directions is presented in Fig. 1. The geometry of the plume propagation in the transverse y-z plane and the axial x direction is also shown as an inset in Fig. 1. The thickness variation of the single laser deposited film in the z direction is more pronounced than in the y direction as expected for an excimer laser spot of proportionately higher aspect along the y direction. The exponents obtained by fitting these profiles to a $\cos^n \theta$ functional form were $n=12$ and $n=28$ in the y and z directions, respectively. The high n values are characteristic of typical highly forward directed single laser ablated plumes. As illustrated in Figs. 1(a) and 1(b), the dual-laser process transformed the plume to a correspondingly less forward-directed expansion leading to a much more uniform film profile with exponents $n=8$ and 8.5 in the y and z directions. Furthermore, the close to unity aspect ratio of the dual-laser deposited film profile, in comparison to the aspect ratio of $\sim$2 for the single laser deposited film, indicates symmetric expansion of the plume corresponding to the symmetric area of overlap between the two lasers on target.

The mechanisms involved in the expansion process were investigated by using a theoretical model based on a system of gas dynamic equations applied to the single and dual-laser ablated plasmas. The model, previously used for single laser ablation,[4] incorporates an initially isothermal collisional plasma followed by an adiabatic expansion.

The isothermal plasma is initiated by the generation of a thin 1 μm layer of heated material on the target at the onset of the excimer laser pulse. During the 20 ns laser pulse τ, this plasma layer continues to absorb laser energy while receiving additional material from the surface of the target. The expansion of this self-regulated isothermal regime can be

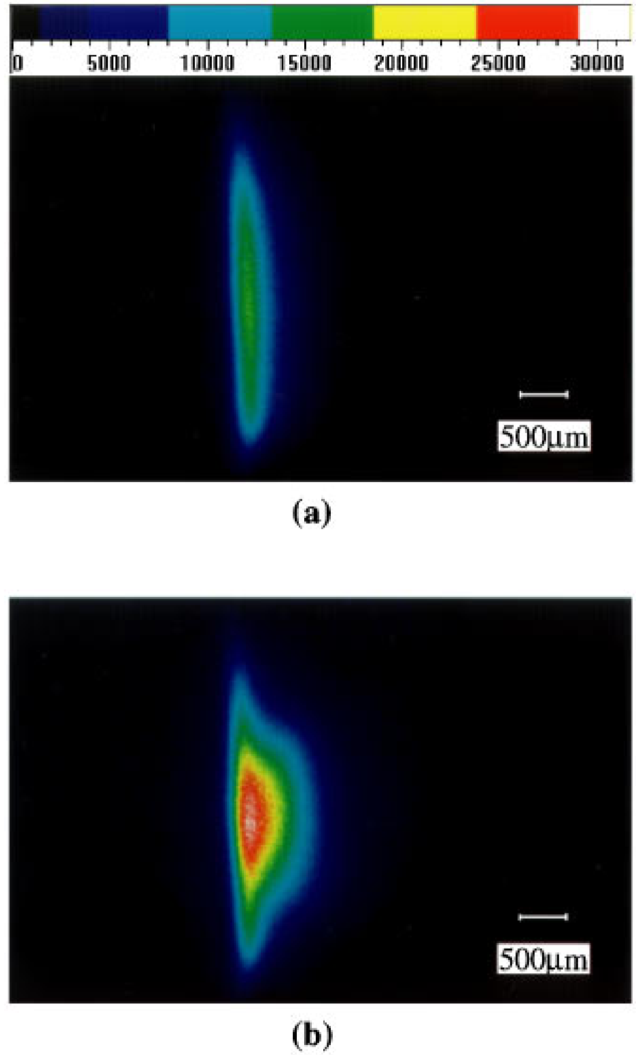

FIG. 2. Gated ICCD image of the first 150 ns of plasma emission in the x-z plane following: (a) single KrF laser and (b) dual-laser ablation.

described by the equation of continuity and the equation of motion. The dimensions of the plasma X, Y, and Z at time t are solutions to the following set of similarity equations:[4]

$$X(t)\left(\frac{1}{t}\frac{dX(t)}{dt}+\frac{d^2X(t)}{dt^2}\right)=Y(t)\left(\frac{1}{t}\frac{dY(t)}{dt}+\frac{d^2Y(t)}{dt^2}\right)=Z(t)\left(\frac{1}{t}\frac{dZ(t)}{dt}+\frac{d^2Z(t)}{dt^2}\right)=\frac{kT_0}{M} \quad t\leqslant\tau, \tag{1}$$

where T_0 is the initial plasma temperature, M is the atomic mass of the plume species, and k is the Boltzmann constant. The density and the velocity of the plasma are given by

$$n(x,y,z,t)=\frac{t}{\tau}\frac{n_0(t)}{X(t)Y(t)Z(t)}\times\exp\left(-\frac{x^2}{2X(t)^2}-\frac{y^2}{2Y(t)^2}-\frac{z^2}{2Z(t)^2}\right) \quad t\leqslant\tau, \tag{2}$$

and

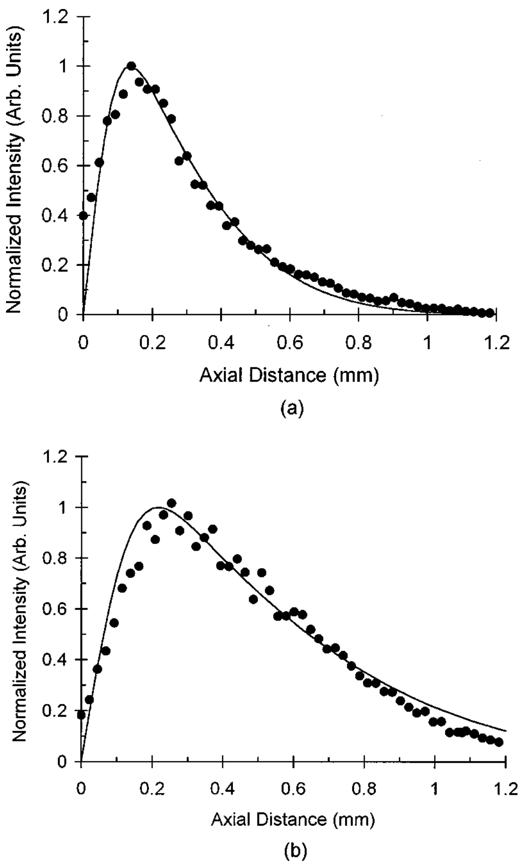

FIG. 3. Axial plume profile of the plasma emission under (a) single laser and (b) dual laser ablation. Dots indicate experimental data from ICCD imaging while the theoretical fits are shown as solid lines.

$$\mathbf{V}(x,y,z,t)=\frac{x}{X(t)}\frac{dX(t)}{dt}\mathbf{i}+\frac{x}{Y(t)}\frac{dY(t)}{dt}\mathbf{j}+\frac{x}{Z(t)}\frac{dZ(t)}{dt}\mathbf{k}. \qquad (3)$$

The subsequent adiabatic expansion of the plasma, initially confined to a small volume with dimensions X_0, Y_0, and Z_0, is obtained by simultaneously solving the equations of motion, continuity, adiabatic state, and temperature, which would lead to the following set of similarity equations:[2,4]

$$X\frac{d^2X}{dt^2}=Y\frac{d^2Y}{dt^2}=Z\frac{d^2Z}{dt^2}=\frac{kT_0}{M}\left(\frac{X_0Y_0Z_0}{XYZ}\right)^{\gamma-1}, \qquad (4)$$

where γ is the ratio of specific heat ($\sim$1.6 for a monatomic gas). The thickness profiles for the deposited films were computed by time integrating the flux, $n(x,y,z,t)\cdot\mathbf{V}(x,y,z,t)$ at points (y,z) on the substrate. An initial plasma temperature of 7000 K was used in the simulation. The result of the calculated thickness profiles are also shown in Fig. 1. As shown in Fig. 1, close agreement with the ellipsometric profiles is obtained for the experimental excimer laser spot size on targets of $Y_0=1.0$ mm and $Z_0=2.25$ mm in the case of single excimer laser ablation. The axial dimension of the plasma at the conclusion of the isothermal phase (X_0) is calculated to be 28 μm. This is consistent with the expected range for the initial plasma dimension in a typical single laser-target interaction.[4]

Modeling the dual-laser ablated plume is complicated by the presence of the second CO_2 laser pulse. There are two distinct stages to the initial isothermal expansion in the dual-laser case. The first stage, during the excimer laser pulse, may be modeled similarly to the single laser expansion with a higher temperature corresponding to increased absorption of CO_2 laser energy. After the conclusion of the excimer laser pulse, the continuing absorption of the CO_2 laser by the plasma for approximately 150 ns is modeled by Eqs. (1)–(3) without including additional incorporation of material from the target into the plume by setting $t=\tau$ in Eq. (2). The final adiabatic expansion is modeled by Eq. (4). The corresponding best theoretical fits shown in Fig. 1 for the dual-laser deposited film profile were obtained for an initial plasma temperature of 25 000 K and a transverse laser spot size of $Y_0=1.0$ mm and $Z_0=1.0$ mm corresponding to the experimental area of overlap of the CO_2 and excimer lasers on the target. Our calculations indicate a dual-laser axial plume dimension of 54 μm at the conclusion of the excimer laser pulse. The transverse dimension is consistent with the symmetric overlap of the two lasers on target and accounts for the overall symmetry of the films deposited using dual laser ablation. Significantly, the calculations indicate a factor of 2 increase in the initial axial extent of the plasma, from 28 μm for single laser ablation to 54 μm for its dual laser counterpart.

We have used time gated intensified charge-coupled detector-array (ICCD) imaging to obtain the initial dimensions of the single and dual-laser ablated plasmas directly. Images of the plasma captured with a 150 ns gate and zero delay in the x-z plane at the target are shown in Fig. 2. Excellent agreement between the observed axial emission profile and the calculated profile obtained from the previous model by integrating the emission over the 150 ns observation window is shown in Fig. 3. Therefore, the model incorporating a higher temperature and increased extent of the initial plasma under dual-laser ablation consistently explains both the emission and thickness profiles.

In summary, CO_2 laser absorption into the initial excimer laser ablated plasma causes both increased temperature and axial extension. Control over this phase of the ablation process significantly affects the subsequent plume expansion and the uniformity of deposited films. Experiments are currently in progress to tailor the temporal profile of the CO_2 laser pulse to study the resultant effect on plume dynamics.

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