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*Optical Society of America
CLEO/QELS '96
June 2-7, 1996
Anaheim, California*

Dynamics of Laser Ablation for Thin Film Growth by Pulsed Laser Deposition

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February 1996

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managed by
LOCKHEED MARTIN ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-96OR22464

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Fundamental gas dynamic and laser-material interactions during pulsed laser deposition are explored through sensitive imaging and plasma spectroscopic diagnostics. Two recent phenomena, plume-splitting in background gases and the unusual dynamics of graphite ablation for amorphous diamond film growth, will be presented.

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Fast imaging and sensitive spectroscopic investigations of laser ablation plume propagation reveal fundamental collisional phenomena relevant to film growth by pulsed laser deposition and optimized cluster growth via laser vaporization. Two phenomena will be detailed. The first involves the splitting of the ablation plume into distinct high and low energy components as a weak shock front forms during ablation into a low-density background gas. The second involves the dynamics of graphite ablation for vacuum deposition of tetrahedrally coordinated amorphous diamond films.

Spatially- and temporally-resolved (~ 0.1 mm, ~ 5 ns) plasma diagnostic techniques of optical emission spectroscopy, optical absorption spectroscopy, fast Langmuir probe analysis, and gated-ICCD fast photography are combined to provide a more complete picture of the laser ablation plume initiation and propagation. Spectroscopic imaging was performed using a tunable liquid crystal filter with 5 nm bandwidth across a 400–720 nm wavelength range. Gated photon counting spectroscopy was employed to explore extremely weak plasma luminescence following the propagation of the initial ablation plume in vacuum and during the 'rebound' of the plume with a substrate during pulsed laser deposition of amorphous diamond.

Ex situ film analyses have shown that ArF-laser (193-nm) irradiation produces higher quality amorphous diamond films than does KrF (248-nm) laser irradiation of pyrolytic graphite in vacuum. Three principal regions of plume emission have been characterized: (1) a bright luminescent ball ($v \sim 3\text{-}5 \text{ cm}/\mu\text{s}$) displaying nearly entirely C^+ emission which appears to result from laser interaction with the initial ejecta, (2) a spherical ball of emission ($v \sim 1 \text{ cm}/\mu\text{s}$) displaying neutral carbon atomic emission lines and, at early times, jets of excited C_2 , and (3) a well-defined region of broadband emission ($v \sim 0.3 \text{ cm}/\mu\text{s}$) near the target surface first containing emission bands from C_2 , then weak continuum emission from C_3 and possibly higher clusters and/or blackbody emission from hot clusters or nanoparticles. The evolution of these three regions is shown in Figure 1. Diamond-like film quality correlates directly with the presence of the high-velocity ion ball and minimization of the cluster emission at the target surface.

Addition of background gases strongly enhances the third (cluster) component, in accordance with plume-splitting phenomena, a general effect which will be described for several materials. As shown in Fig. 2, during expansion into low-pressure background gases the ion flux in laser ablation plasma plumes is generally observed to split into distinct "fast" and "slow" components. The fast component is target material which penetrates the background gas in accordance with a scattering model, while the slow component is material which has undergone momentum-changing collisions with the background gas, or with other plume atoms. When the multicomponent graphite plume of Fig. 1 encounters low-pressure argon several collisional overlap regions between plume atoms are created, as shown in Fig. 2(b), which leads to additional clustering near the target surface.

The combination of sensitive imaging and photon-counting diagnostic techniques permit an understanding of the importance of gas dynamic effects on the

time-of-flight distributions of species arriving during the deposition of thin films in both vacuum and background gases.

This research was sponsored by the Oak Ridge National Laboratory, managed by Lockheed Martin Energy Research Corp. for the U.S. Department of Energy under contract number DE-AC05-96OR22464.

Figure Captions

Fig.1. Gated-ICCD photographs of the total visible luminescence at the indicated times following ablation of pyrolytic graphite in vacuum using (a), (b), (c), (e) ArF-laser (6.7 J/cm^2) and (f) KrF-laser (17.7 J/cm^2) irradiation (incident angle 30° from the left). The 5-grayscale palette (bottom) is normalized to the maximum numbers of counts for each image. (d) Schematic indicating the different luminescent plume components discovered by ICCD-imaging (see text).

Fig.2. (a) Ion probe current (mV into 50Ω) induced by the ArF-laser (6.7 J/cm^2) generated plasma at $d = 5 \text{ cm}$ along the normal to the graphite target into 60, 70, 80 and 90 mTorr of Ar. In addition to the dominant fast component of the ion flux, a second component is observable at delays of $\sim 10 \mu\text{s}$. The insert shows the disappearance of the fast component and the evolution of the second component for vacuum ($7 \times 10^{-7} \text{ Torr}$) and argon pressures of 100, 160, 200, 260, and 300 mTorr. (b) ICCD image of luminescence (unfiltered) from the ArF-laser (6.0 J/cm^2) generated carbon plasma propagating into 70 mTorr of argon ($2.1 \mu\text{s}$ delay, $0.2 \mu\text{s}$ exposure). The grayscale palette used is shown. (c) Ion current versus time after laser pulse in 350 mTorr of Ar.

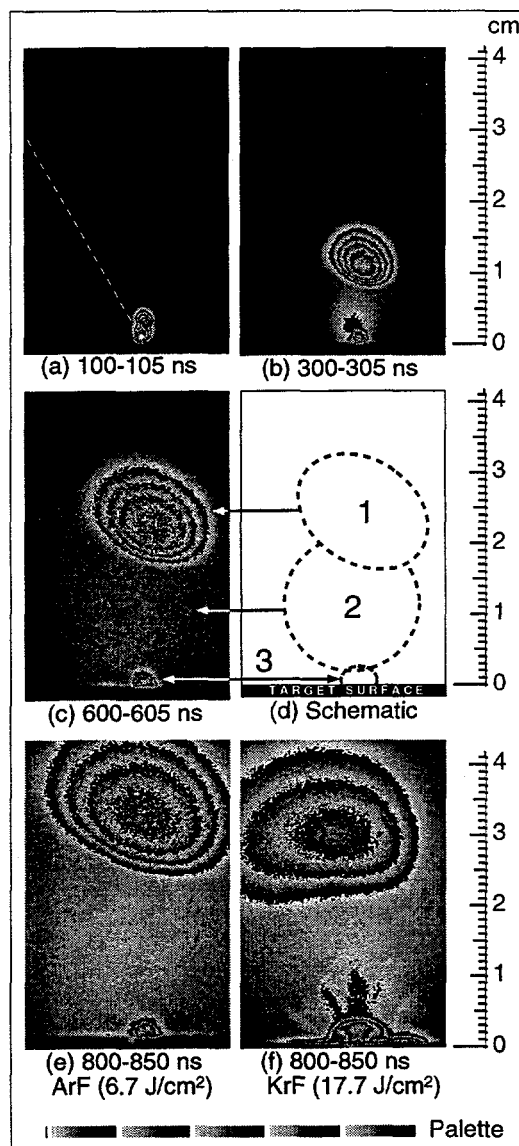


Figure 1

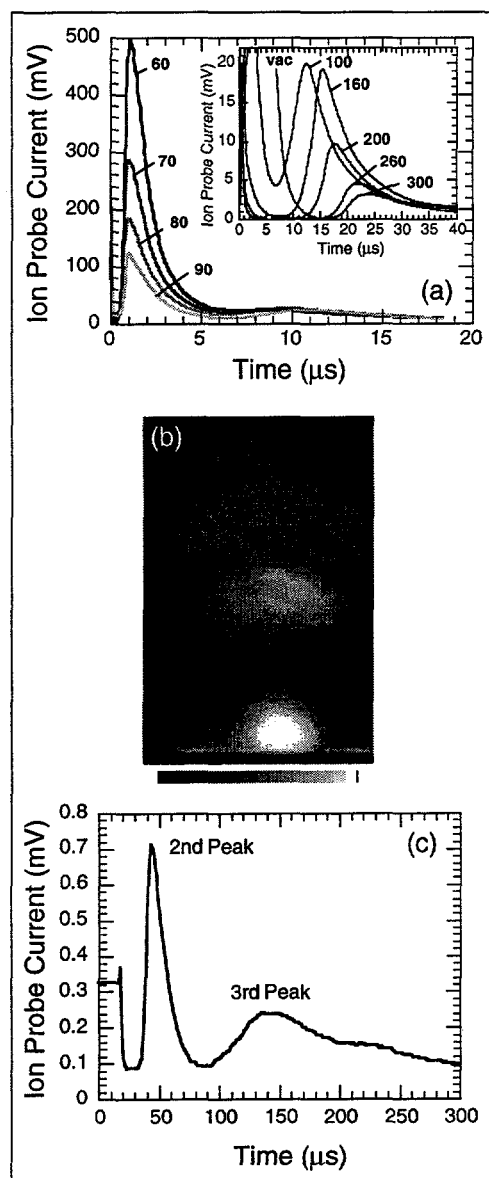


Figure 2