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Optical transmission measurements of explosive boiling and liftoff of a layer of micron-scale water droplets from a KrF laser-heated Si substrate

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Water plume velocities were measured in air by optical transmission as a function of laser fluence using a KrF laser for explosive boiling and liftoff of a layer of micron-scale water droplets from a laser-heated Si substrate of interest for laser particle removal. The thickness of the superheated water layer near the water/Si interface determines acceleration and removal of the water droplets from the Si substrate. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1555272]

INTRODUCTION

Deposition of a thin, submicron to several micron-thick liquid layer as an energy transfer medium (ETM)[1] on contaminated critical surfaces (e.g., Si masks,[1] telescope optics[2]) was found in the early 1990s to enhance laser cleaning of such critical surfaces.[1–3] Since then, explosive boiling of a liquid layer near a substrate surface and evaporation of the whole liquid layer together with contaminants in the form of a vapor-droplet plume was determined as the basic mechanism of this steam laser cleaning (SLC) technique. Threshold conditions (temperature, pressure and laser fluence) for the onset of boiling were experimentally measured for water, different alcohols and their mixtures using ''bulk'' (several mm thick) liquid layers.[4,5] In these experiments, the liquid layer is sufficiently thick that removal of the liquid does not occur. Many important experimental parameters of SLC, e.g., vaporization thresholds and plume velocities, have not yet been studied for different laser heating conditions and the thin liquid layers of variable thickness or water droplet layers actually used in laser particle removal,[5,6] where the liquid or droplet layer deposited is completely evaporated in the process. In this work velocities of a water plume produced in air by KrF laser heating of a Si substrate with a pre-deposited layer of micron-scale water droplets were measured at various laser fluences using an optical transmission technique.

EXPERIMENT

The beam from a 248 nm, 20 ns KrF excimer laser (Lambda Physik, LPX 210) with a 1 cm wide vertical slit aperture in its center was imaged by a cylindrical lens ($f = 10$ cm) at normal incidence onto a 0.25 mm thick Si(100) wafer with a pre-deposited thin layer of water (Fig. 1). The laser beam has nearly rectangular and Gaussian fluence, F, and distributions in the horizontal (X) and vertical (Y) directions (see the insets in Fig. 1), respectively, with characteristic dimensions of $x = 5$ and $\sigma_y^{1/e} = 1.5$ mm. Laser energy [0.2 J/pulse (±3%) after the aperture] was attenuated by color filters (Corning Glass Works) and was measured by splitting off part of the beam to a pyroelectric detector (Gentec ED-500). The water dosing system used[3] consisted of a source of pressurized nitrogen with a trigger valve, connected to a bubbler immersed in a glass flask filled with heated de-ionized water and directed onto the Si surface through a heated output nozzle. The dosing system (gas pressure of 0.7 bar, flask water and nozzle temperatures of 40 °C, dosing pulse of 0.5 s) was employed to deposit a layer of water droplets several microns in diameter on the Si wafer placed 5 cm from the nozzle. Micron-scale water droplets are formed because the native oxide film present on a Si substrate stored under ambient conditions does not wet.[1,3,7] The excimer laser was fired 0.05 s after the end of each liquid film deposition step. Deposition and removal of the water layer were measured in real time by observing the optical reflectance/scattering of a continuous wave (cw) HeNe laser focused on the center of the irradiated area. Since the micron-scale water droplets are relatively dense and the excimer laser beam is much larger than the droplet size, the plume that results from explosive boiling of the water droplets behaves as if it originated from a thin continuous layer of water. Propagation of the water plume along the normal to the Si surface was investiged using another cw HeNe laser beam tightly focused (waist diameter of 0.08 mm) with a 5 cm focal length lens at various distances Z above the center of the KrF laser spot on the wafer. The HeNe laser and focusing lens were mounted on a one-dimensional stage and shifted perpendicular to the Si wafer plane in 0.5 mm steps, along with the corresponding fast photodiode detector. A

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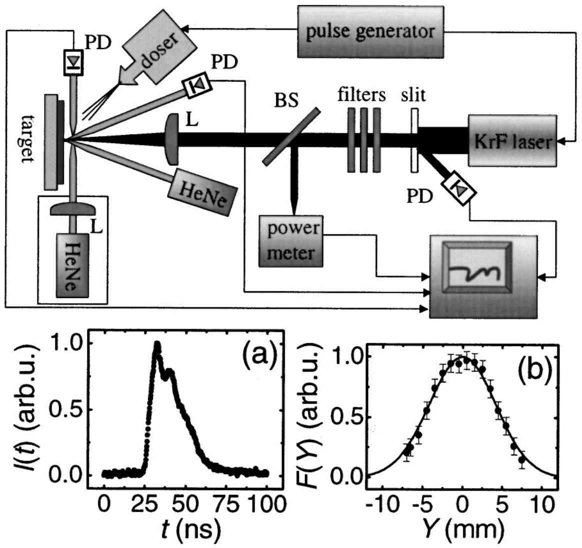

FIG. 1. Experimental setup for transient plume transmission studies: BS—beam splitter, L—focusing lenses, PD—fast photodiodes, target—Si wafer on a three-dimensional stage. Insets: (a) Laser power temporal dependence of the KrF laser and (b) Gaussian distribution of fluence F in the vertical (Y) direction.

LeCroy 9360 storage oscilloscope triggered by an electrical pulse from a fast photodiode, which detected part of the excimer beam scattered directly from the slit, was used to record water plume transmission transients delayed by the time of flight, t_D, of the plume to the HeNe probing distance and to measure the laser pulse energy in each pulse. The gas valve and excimer laser were triggered manually in single-shot mode with corresponding delays using a pulse generator (Stanford Research Systems DG 535).

RESULTS AND DISCUSSION

The water plume transmission transients for constant water dosing conditions, different probing distances and peak laser fluences of $F_{\mathrm{max}}=0.32-0.76\ \mathrm{J/cm}^2$, i.e., above the explosive boiling threshold of water on a Si surface (0.2 $\mathrm{J/cm}^2$),[8] but below the ablation threshold of Si (about 1.4 $\mathrm{J/cm}^2$),[8,9] are presented in Fig. 2. The maximum probe distance, Z_{max}, is approximately 3 mm, where detection of the plume transmission transients becomes very unstable because of three-dimensional rarefaction, dispersion, vaporization, turbulence and other instabilities in the plume[5] as well as the decelerating effect of viscous air drag. As shown in Fig. 2, the plume transmission varies much less at larger distances ($Z=2.5$ mm) than it does near the Si surface ($Z=0.5$ mm).

A characteristic parameter of the transmission transients is the plume arrival time at the HeNe probe position, t_D, depending on Z and defined in Fig. 2 as the onset of the drop in HeNe transmission. The experimental data as $Z-t_D$ trajectories for several F_{max} values are given in Fig. 3(a). These $Z-t_D$ curves were fit with an exponential function $f(t)=V_0T_{\mathrm{damp}}[1-\exp^{-t/T\mathrm{damp}}]$. The initial plume velocity at $Z=0$, V_0, and the viscous drag velocity damping time, T_{damp}, which takes into account the decelerating viscous drag force in air for the water vapor plumes were used as variable parameters. In order to have a simple analytical description of the vapor plumes we employed a one-dimensional model of continuous viscous flow assuming thin, homogeneous initial liquid layers.

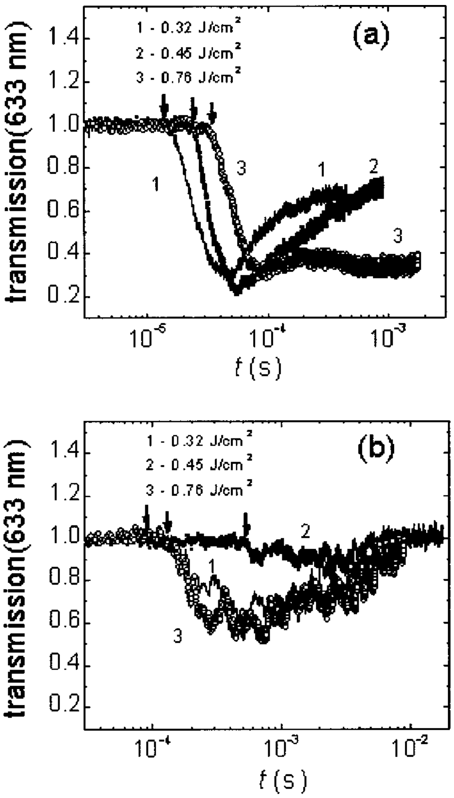

FIG. 2. Characteristic water plume transmission transients at different fluences F_{max}: $Z=$(a) 0.5 and (b) 2.5 mm. The delay time values, t_D, are shown for different fluences by several arrows.

The $f(t)$ functions employed fit the $Z-t_D$ curves quite well [Fig. 3(a)]. At fluences of 0.32, 0.45 and 0.76 $\mathrm{J/cm}^2$ good agreement is observed between the calculated maximum plume propagation distance V_0T_{damp} (2.88 ± 0.14, 2.87 ± 0.14 and 2.89 ± 0.03 mm, respectively) and the experimentally observed value $Z_{\mathrm{max}}\approx3$ mm. The corresponding T_{damp} values are different (0.14 ± 0.02, 0.21 ± 0.03 and 0.25 ± 0.01 ms, respectively) and the calculated V_0 values decrease with an increase in F_{max} [Fig. 3(b)]. The different T_{damp} values that characterize viscous drag damping of water plume motion may mean that the simple one-dimensional viscous continuous flow plume propagation model used is not really applicable in this case. For example, we have assumed that the droplet structure on the surface does not affect vaporization and that the plume always consists of the same size and density droplet and vapor mixture. A more complex model would include these and other effects such as three-dimensional rarefaction, dispersion, turbulence and other instabilities in liquid plumes. The V_0 values obtained, however, seem to be a good estimate of plume initial velocities, at least for small propagation distances where $Z-t_D$ curves can be linearized and viscous drag and plume instabilities neglected. The resulting initial velocities, $V_0\approx10-20$ m/s, are very low relative to the supersonic velocities expected for condensation products formed during

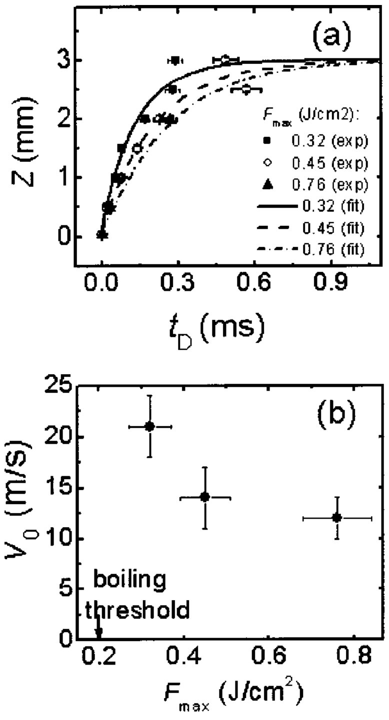

FIG. 3. (a) $Z-t_D$ trajectories and their exponential curve fits at different fluences $F_{\max}$; (b) dependence of the initial water plume velocity V_0 on $F_{\max}$.

adiabatic expansion of a molecular vapor, implying that liftoff of the entire water layer occurs as a result of explosive boiling of the near-surface water layer. Qualitatively similar results were obtained using the same experimental setup on several micron-thick, homogeneous layers of 2-propanol, which does wet the native oxide on Si.[10]

The observed trend of the $V_0(F_{\max})$ curve might be explained by the simultaneous explosive boiling and expansion of a superheated water layer near the water/Si interface in each droplet under spinodal conditions,[11] that acts as a vapor "piston" and imparts mechanical momentum to the top "cold" liquid layer of the droplet. The onset of explosive water boiling, t_{boil}, due to the instantaneous spinodal decomposition shifts gradually with an increase in $F_{\max}$ to the beginning of the heating laser pulse. This shortening of the explosive boiling delay time reduces the heat-affected zone, $L_{\text{dep}} \sim (\chi t_{\text{boil}})^{1/2} \sim 0.01\ \mu\text{m}$, in the water droplets (height $L \approx 1.5\ \mu\text{m}$), while the water thermal diffusivity, χ, slowly decreases as a function of the increase in water temperature near the water/Si interface. The velocity V_0 of the entire cold liquid layer that gives rise to the plume can be calculated using momentum conservation in the form of $L_{\text{dep}} \times V_{\text{exp}} \approx L \times V_0$, where $V_{\text{exp}} \sim C_l \times (\Delta V/V_0)$ is the thermal expansion velocity of the superheated water layer under spinodal conditions, $C_l \approx 1.5$ km/s is the longitudinal sound velocity in water at ambient conditions,[12] and the increase in water molar volume $(\Delta V/V_0) \approx 0.5-1.5$ at positive pressures along the spinodal curve up to the critical point.[11] Thus, V_0 should scale as $V_{\text{exp}} \times L_{\text{dep}}/L$, in qualitative agreement with theoretical predictions[13] and the experimental results of this work. The decrease of $V_0(F_{\max})$ with an increase in $F_{\max}$ is shown in Fig. 3(b) with lift-off (plume) velocities of 10–20 m/s for our experimental conditions.

CONCLUSIONS

Water plume velocities were measured in air by an optical transmission technique as a function of laser fluence for explosive boiling and liftoff of a thin layer of water droplets on a Si substrate heated by a KrF laser. The results are counterintuitive in that the initial velocity of the vapor/droplet plume decreases with an increase in laser fluence. The thickness of the near-surface superheated water layer in the droplets determines the acceleration and removal of the entire water droplet layer from the Si substrate. At higher fluences, the water in contact with the hotter laser-heated Si substrate reaches the critical temperature faster, and liftoff limits additional heating by the substrate. For lower fluences, the longer heating time results in a thicker heated layer and more total energy in the water droplets. These results help in understanding and optimization of laser particle removal via explosive boiling of a thin deposited liquid or droplet film.

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