Time-resolved analysis of thickness-dependent dewetting and ablation of silver films upon nanosecond laser irradiation
Dongfeng Qi, Dongwoo Paeng, Junyeob Yeo, Eunpa Kim, Letian Wang, Songyan Chen, and Costas P. Grigoropoulos

View online: http://dx.doi.org/10.1063/1.4952597
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/108/21?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in
Nanoparticle generation and transport resulting from femtosecond laser ablation of ultrathin metal films: Time-resolved measurements and molecular dynamics simulations

Solvent annealing induced phase separation and dewetting in PMMA/SAN blend film: Film thickness and solvent dependence

Structuring by field enhancement of glass, Ag, Au, and Co thin films using short pulse laser ablation

Pulsed laser dewetting of patterned thin metal films: A means of directed assembly

Time-resolved electron shadowgraphy for 300 ps laser ablation of a copper film
Arrays of micron- or nano-sized holes in metallic films exhibit local field enhancement, strong plasmonic response, and high electron emission.\(^1,2\) During the last decade, such structures have attracted growing interest, owing to promising applications in optoelectronics\(^3,4\) as well as other fields.\(^5\)

Direct laser ablation and dewetting is an interesting method for machining holes in metallic specimens. Compared to conventional photolithography that is combined with reactive ion etching or chemical etching, nanosecond laser irradiation\(^6\) has shown advantages in terms of its non-contact characteristics, low cost, and flexibility with respect to the target material. By tuning the incident laser power, it is possible to machine holes of different dimensions and shapes on metallic surfaces.\(^7\)

Fundamental understanding of laser-induced dewetting and ablation processes is important in order to enable improved prediction and optimization of laser processes.\(^8,9\) Several optical probing techniques have been developed for studying laser-material interactions.\(^10,11\) For example, time-resolved shadowgraphy has been utilized to directly observe the ablation process in the nanosecond time regime.\(^12–14\) Capturing the ejected plume and shockwave emission at certain delay times.\(^15\) Nevertheless, even in the simplest case of metal films, the induced phase change and the ensuing dewetting and ablation processes have not yet been fully investigated.

In this letter, time-resolved images revealed the evolution of different morphologies during the ablation and dewetting processes. We selected silver as the sample of interest because of its well-known properties and use as electrode material in electronics. Silver thin films of different thicknesses (50, 80, and 350 nm) were prepared by electron beam evaporation on quartz substrate. The time-resolved imaging system was set up as shown in Figure 1(a). Nd:YAG laser pulses of 532 nm wavelength and 5 ns temporal width impinged on silver film targets. The laser beam was focused by a 2× infinity corrected, non-achromatic long working distance objective lens at normal incidence. Nanosecond flash lamp (NANOLITE KL-K flash lamp, flash duration = 7 ns) was employed as an illumination source to provide temporally resolved images. These images were captured by a charge coupled device (CCD) camera via a 20× infinity objective lens. A digital delay generator was used to control the delay time between the processing laser and the image acquisition. The oscilloscope was used to record the actual delay time of the processing laser signal and the flash lamp signal. To ensure true representation, at least six images were examined at each delay setting.

We estimated the ablation threshold fluences of each film by linear curve fitting as shown in Figure 1(b). Above the ablation threshold fluence, \(F_{th}\), the relationship between the ablated spot radius, \(r_a\), and the laser fluence, \(F\), is well characterized by a Gaussian beam profile, \(r_a^2 = 2r_f^2 \ln(F/F_{th})\), where the 1/e beam radius, \(r_f\), is about 31 μm. If \(r_a^2\) is plotted vs \(\ln(F)\), extrapolation to \(r_a^2 = 0\) yields the ablation threshold. For different thickness silver films (50, 80, and 350 nm), the ablation thresholds \(F_{th}\) were estimated as 0.92 J/cm\(^2\), 2.59 J/cm\(^2\), and 8.45 J/cm\(^2\), respectively.

Next, we focus on investigating the surface morphologies produced by the exposure of the Ag films to near ablation threshold fluences. Figure 2(a) presents optical dark-field images, SEM images, and time-resolved images of the 50 nm Ag film surface. For low laser fluence below 0.89 J/cm\(^2\), droplet-like structures are formed in the central area, implying melting and dewetting. As the fluence increases, the central droplet-like structures disappear, and melted material is dewetted radially outward. For the thicker samples (80 nm and 350 nm), as shown in Figures 2(b) and 2(c), droplet-like structures can also be formed.
upon irradiation at fluences of 2.31 J/cm² and 7.71 J/cm², respectively. At higher laser fluences, peripheral rims and droplet fingers appear in the outskirts of the holes.

The transient evolution of the film surface morphologies at selected laser fluences are recorded using the time-resolved imaging setup and presented in Figure 3. As previously mentioned, micron-sized droplet-like particles are formed at lower laser fluences, i.e., around 0.89 J/cm² for the 50 nm film and 2.31 J/cm² for the 80 nm film. The time resolved images at the corresponding laser fluences in Figures 3(a) and 3(b) indicate that shiny small particles appeared in the central area of the irradiated zone at times of 200 ns and 500 ns after the laser irradiation, presumably formed by Ostwald ripening. Furthermore, since micron-sized metallic particles of size parameter \( \phi = \pi D / \lambda \), where \( D \) is the particle diameter and \( \lambda \) the probing wavelength, scatter radiation strongly in the forward direction and yield much weaker back-scattering, the central area appears dark in reflection images beyond an elapsed time of 630 ns, signifying aggregation to bigger micron-sized particles. At slightly increased laser fluence, holes with droplet-like or cylindrical rim structures around these edges are formed on the silver film surface, respectively. At higher fluences, i.e., at and above 1.38 J/cm² for the 50 nm thickness specimen, the dewetted material is pulled toward the edges of the holes forming peripheral droplet-like structures after an elapsed time of 700 ns. For the 80 nm thick sample, rim structures can also be formed if the laser fluence is at or above 2.31 J/cm². For the 350 nm thick sample, several distinct stages in Figure 3(c) are observed at different laser fluences. Evidence of ablation in the center and dewetting in the edge area is observed at the laser fluence of 8.82 J/cm² that is slightly higher than the ablation threshold of 8.45 J/cm². At the higher laser fluences of 9.92 J/cm² and 10.52 J/cm², radially outward droplet fingers are observed.

Figures 4(a)–4(c) depict the SEM images of Ag thin films for different laser fluences, where the dewetting and ablation edges are distinct. Figure 4(d) gives the schematic
diagram of ablation and dewetting distribution for 50 nm thickness sample under different laser fluences. If the laser fluence (red curve) exceeds the ablation threshold, the central ablation area (blue line) surrounded by the dewetting zone (green line) appears. The dewetting regime (green line) solely appears in the spot center if the laser fluence (black curve) is below the ablation threshold.

Figure 5 depicts distinct stages of the thin film modification process and the corresponding ablation and dewetting threshold, respectively. If the laser intensity in the spot center is below the ablation threshold, in the dashed line region, the nanosecond laser pulse causes local dewetting of the Ag layer in the spot center. In this case, nanoparticles grow larger through Ostwald ripening, wherein the larger particles scavenge smaller ones, broadening the size distribution in the dewetting area after an elapsed time of 650 ns for all the samples (Figure 5(a)). If the laser fluence exceeds the respective ablation threshold of each sample, the SEM and time-resolved images of 50 nm sample show ablative material removal in the spot center and the formation of droplet-like particles in the peripheral dewetting zone after an elapsed time of 700 ns (Figure 5(b)). Rim structures are then formed in the 80 nm and 350 nm thick samples after laser irradiation (Figure 5(c)). It is recalled that the surface temperature is higher at the center of the melt pool, and the absorbed energy induces surface tension driven flow due to the high temperature gradient. Consequently, due to the diminishing surface tension with increasing temperature for liquid metals, material should be transported radially outward by the positive surface tension gradient. However, the 50 nm film cannot supply significant mass transfer and only droplet-like particles are formed. Order of magnitude estimates for the 50 nm film and melt pool diameter of 60 μm yield O(0.1 m/s) thermocapillary driven melt velocity that in turn corresponds to O(10⁻⁵ N) inertia force on the liquid pool. On the other hand, estimates of the recoil force exerted on the melt pool for laser fluence in the range of 1 J/cm² yield O(10⁻¹ N) values. Although the inertia force scales approximately with the square of the film thickness, it is still much smaller than the recoil force even for the 350 nm film. One may therefore expect that the thermal gradient driven surface flow is of minor consequence. For the 350 nm thickness sample, as shown in Figure 5(d), the ablated splashed droplets shoot off the surface at higher laser fluence
The problem becomes more strongly coupled due to the fluid where the ablation is accompanied by the plasma effects and length. This is likely to be important at the higher fluences. However, the laser interaction present experiments, as it affects merely the magnitude of effect of the wavelength is not particularly significant for the different inorganic materials and comparable thickness. The mass transport, and the effect of the ablative material removal and their dependence in thin silver films. The time-resolved imaging setup helped elucidate the transient breakup of the dewetted melt pool, the surface tension induced mass transport, and the effect of the ablative material removal and their dependence on the imparted laser fluence.

(above 9.92 J/cm²), exerting strong recoil force and producing droplet fingers (Figure 5(d)). For higher laser fluence, the liquid Ag materials are squeezed out of the pool very quickly by the diminishing surface tension and pushed around the peripheral rim structure. Droplet fingers occur due to the recoil force induced acceleration around the rim.\(^{23,26,27}\) The same experimental trends would be observed in thin films of different inorganic materials and comparable thickness. The effect of the wavelength is not particularly significant for the present experiments, as it affects merely the magnitude of the energy input into the film. However, the laser interaction with the ejected matter would depend on the laser wavelength. This is likely to be important at the higher fluences where the ablation is accompanied by the plasma effects and the problem becomes more strongly coupled due to the fluid dynamics and radiative transport in the ejected plume.

In conclusion, we have carried out in-situ visualization of nanosecond laser-induced dewetting and ablation processes in thin silver films. The time-resolved imaging setup helped elucidate the transient breakup of the dewetted melt pool, the surface tension induced mass transport, and the effect of the ablative material removal and their dependence on the imparted laser fluence.

D. F. Qi acknowledges the support of the Chinese Scholars Council and the National Science Foundation of China (Grant Nos. 61474081 and 61534005).