

Characterization of Laser Ablation Dynamics for Nickel Thin Films on Silicon Using Movie Mode Dynamic TEM

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Laser ablation processes, which involve the removal of a material from a substrate have important industrial applications in laser machining of structural components and micro-electronic devices. [1] Limited data is available in the literature about direct imaging of the laser ablation process that occurs for metal films with thicknesses below 100 nm since time resolutions in the nanosecond range or faster are required [1]. Dynamic TEM is an ideal tool to directly image laser ablation processes on the nanometer length scale, and provides a fundamental insight to the mechanisms that govern material removal from solid substrates during laser irradiation.

Nickel thin films with a thickness of 64 nm were sputter deposited at room temperature on the (100) surface of silicon substrates that were covered with a native oxide layer. Plan-view TEM samples were prepared by conventional grinding, polishing, dimpling, and subsequent Ar⁺ ion-milling for 3 hours at 3.5 kV with a final ion beam cleaning for 30 minutes at 0.5 kV. [2] Real time observation of laser ablation was performed with a Dynamic Transmission Electron Microscope (DTEM) equipped with a pump-probe laser system. The DTEM is installed at Lawrence Livermore National Laboratory (LLNL) [3]. The experiments were carried out with a 532 nm wavelength pump laser and a 12 ns FWHM pulse duration. The laser beam had a Gaussian beam profile with a diameter of $135 \pm 5 \mu\text{m}$, and an energy of 12 μJ per pulse. Nine laser pulses were used to acquire a total of 9 consecutive micrographs of the thinnest, i.e., most electron transparent, area of the sample to capture the evolution of the thin nickel film. Prior to the DTEM experiments the geometry of each TEM sample was characterized by valence electron energy-loss spectroscopy using a JEOL-JEM 2100F/Cs operated at 200 keV. The temperature distributions, heat transfer and resulting mechanical stress due to laser irradiation was modeled for the sample geometry using spatio-temporal simulations with the COMSOL Multiphysics software.

Figure 1 shows a series of bright field TEM images recorded by DTEM, and reveal the transient process of laser ablation caused by a laser power density below 10^8 W/cm^2 [4]. Figure 1A shows that the as-deposited nickel thin film was continuous prior to laser irradiation. Figure 1B displays the surface reorganization that was observed 20 ns after the laser pulse. The image intensities representing a cellular structure of the nickel film indicate the onset of liquid dewetting of the nickel film in this area [5]. Figure 1C demonstrates the formation of round nanoparticles and substrate fracture 115 ns after laser irradiation. Figure 1D, taken minutes after laser irradiation, shows the substrate fractured in the thin region where nickel film originally dewetted (Figure 1B). It has been shown that laser ablation processes with low power density are accompanied by particle formation where round particles are liquid droplets ejected through hydrodynamic processes and irregularly-shaped particles with smaller sizes are formed via thermal-induced stresses and fracture [6]. Since direct temperature measurements are not possible during DTEM experiments, the temperature distribution in the sample was modeled as a function of time (see Figure 2). Figure 2 shows a series of time-temperature profiles where the Gaussian laser pulse is centered at the edge of the sample; the temperature profiles are given for different distances from the

edge of the sample. The simulated temperature profiles indicate that, for the laser energy used in these experiments, the temperature exceeds the melting temperature of nickel thin film, which is ~ 1680 K. The dynamics of the low power density laser ablation was observed with high temporal and spatial resolution through capturing the early stages of liquid dewetting of the nickel film followed by nano-sized particle formation and substrate fracture, which will be interpreted with thermal-induced stress simulation via COMSOL.

References

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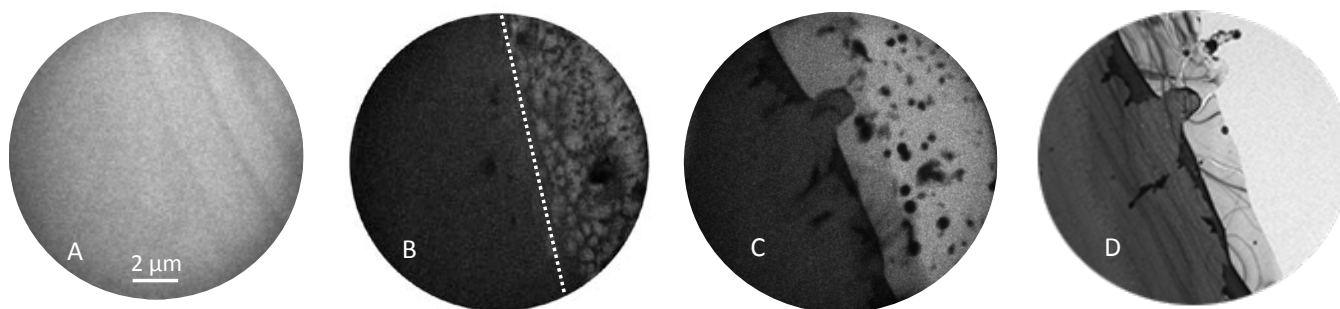


Figure 1. Bright field images from 12 ns electron pulses demonstrating the dynamics of laser ablation. A) as-deposited Ni film on a silicon substrate [4]. B) morphological instabilities developed 20 ns after laser heating showing the nickel film dewetting to the right of the dotted line [4]. C) nanoscale particle formation and substrate fracture captured at 115 ns [4]. D) the same region minutes afterwards [4].

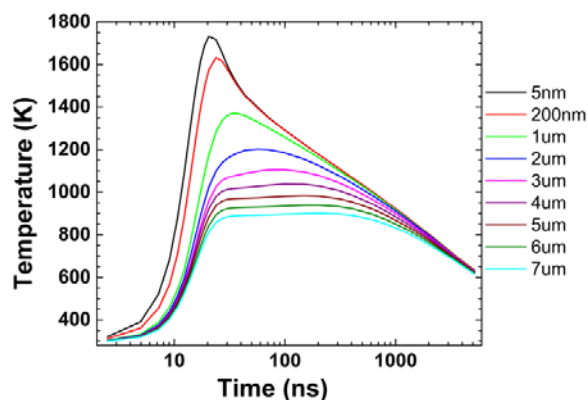


Figure 2. Spatio-temporal COMSOL simulation of the temperature evolution as a function of time for a laser pulse centered at the edge of the wedge-shaped sample [4]. The distances are relative to sample edge.