Supplementary Information (SI) for Nanoscale.


Supplementary Information for

# Laser Patterning Captured in Real-Time: Surface Modifications of Multilayer Thin-Films Under Nanosecond Laser Heating

*Tugba Isik[1], Mason Freund[2], Will Linthicum[2], Bryan D. Huey[2], Volkan Ortalan[2]**

[1] School of Mechanical Engineering, Purdue University, West Lafayette, Indiana 47907, USA

[2] Department of Materials Science & Engineering, University of Connecticut, Storrs, Connecticut 06269, USA

E-mail: vortalan@uconn.edu

**Experimental Details**

The Ti/Ni multilayers were deposited on TEM grids with silicon nitride membranes (TedPella) using electron beam evaporation at the UConn Nanoelectronics Laboratory. Grids were fixed on a silicon substrate holder using Kapton tape, and the chamber was evacuated to a pressure of $5x10^{-6}$ Torr before the deposition. The multilayered structure was created by sequential deposition of high purity nickel and titanium pellets (99.995 %, Kurt Lesker) with an alternating Ti-Ni stacking order. Evaporation of Ti and Ni crucibles was performed with 0.8 and 1 A/s deposition rates, respectively, and the substrate rotation was set to 60 rpm. Two bilayers, four layers in total, were deposited on 15 nm and 50 nm thick SiN membranes. The thicknesses of individual metal layers were chosen to achieve 2:3 atomic ratio (Ti:Ni) and the total thickness of the metal film was kept around 50 nm. Electron diffraction analysis of the as-deposited bilayers demonstrated that the samples were polycrystalline and consisted of hexagonal Ti and face-centered cubic Ni.

Single-shot images were collected with high temporal resolution using photoemitted electron pulses. The sample excitation laser of the UTEM system had a 532 nm wavelength, Gaussian profile, and 1 ns pulse duration. The mixing of the multilayers was initiated with a single pulse of the sample excitation laser. The delay between these two lasers was set to different times with a digital delay generator to capture the transient dynamics. The timing jitter between the two lasers is ±3.56 ns. The delay times provided represent the mean time delays set by the digital delay generator. The laser spot was positioned either at the center of the observation point or ~20-25 μm away from it, so the edge of the laser-affected region could be imaged, as well as the central region. The evolution of the multilayer Ti/Ni film under laser heating was then examined by single-shot images collected at different time delays.

Further TEM characterization of the samples after single-shot experiments was performed using an FEI Talos TEM operated at 200 kV. Bright-field TEM images and high-angular annular dark field (HAADF) STEM images were collected to study the final morphology of the sample along with energy dispersive x-ray spectroscopy (EDX) maps. In addition, the final surface structure of the samples was investigated in an FEI Verios SEM operated at 30 kV, and AFM measurements were performed at the Huey Lab, providing information about the changes in the surface structure after laser heating. Thermal finite element simulations were conducted using COMSOL Multiphysics software to estimate the temperature resulting from laser heating. Mechanical buckling analyses were carried out using the Abaqus finite element analysis software. In buckling analysis, the 15 nm thick silicon nitride membrane was modeled with an elastic modulus of 183 GPa[1] and a Poisson's ratio of 0.27[2].

Lift-out TEM samples were prepared using an FEI Helios Dual Beam SEM/FIB system to investigate the cross-sectional profile of the thin films after laser heating. Since the total thickness of the sample was extremely small (around 65 nm) for the standard lift-out procedure, deposition of additional layers at the region of interest (ROI) before preparing a TEM lamella was required. Special care was taken to protect the surface structure of the sample by starting with 500 nm electron beam deposition of carbon (5 kV, 3.2 nA) on both sides of the membrane, followed by an additional 3 μm carbon deposition with the ion-beam (30 kV, 0.43 nA). After the total thickness of the ROI was increased to around 7.5 μm with these depositions, the standard lift-out procedure was followed for TEM lamella preparation.

**Supplementary Figures**

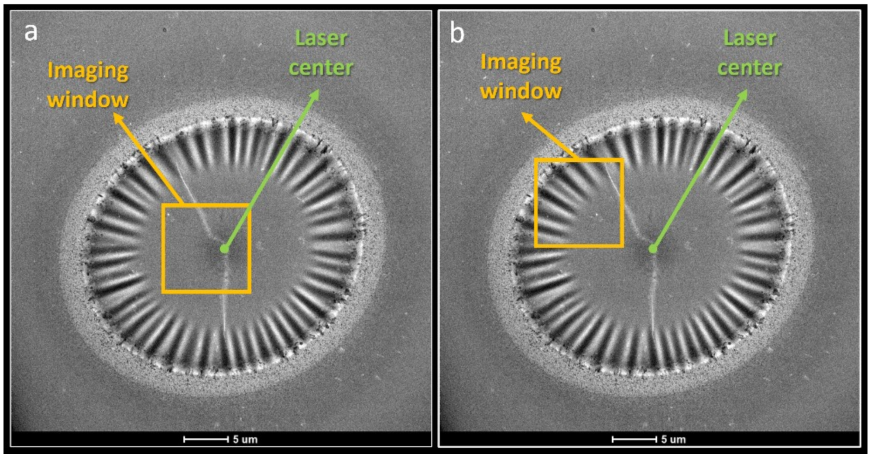


**Fig. S1** Schematic representations of two different imaging conditions used in the single-shot study, (a) imaging near the laser center, (b) imaging at the boundary of the laser-irradiated region.

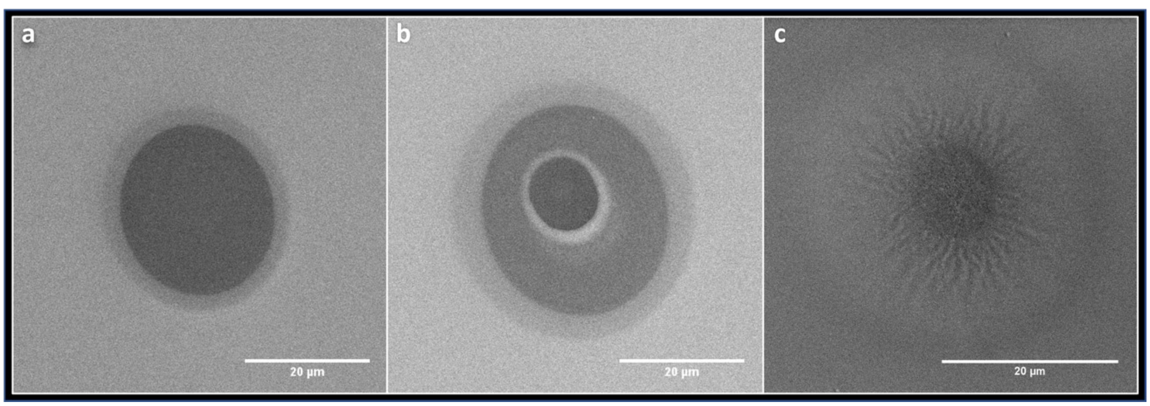


**Fig. S2** Comparison of surface deformation after laser heating for different membrane thicknesses and metal coating, (a) Ti/Ni bilayer coated 50 nm SiN membrane (lower laser fluence), (b) Ti/Ni bilayer coated 50 nm SiN membrane (higher fluence), (c) SEM image of only Ni coated 15 nm SiN membrane.

## References

**1)** T. S. Hickernell, F. M. Fliegel and F. S. Hickernell, in *IEEE Symposium on Ultrasonics*, IEEE, Honolulu, HI, USA, 1990, pp. 445–448.

**2)** Z. Gan, C. Wang and Z. Chen, *Surfaces*, 2018, **1**, 59–72.