Pulsed laser dewetting of thin metal annuli for experimental investigation of nanoscale hydrodynamics
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The dewetting of thin metal films is of interest for potential applications utilizing self- and directed-assembly at the nanoscale. Depending on the geometry of nanoscale structures, pulsed laser induced dewetting (PLiD) can induce the formation of a variety of arrangements and sizes of nanoparticles, with potential applications including plasmonics, catalysis of carbon nanotube growth, and others.

Experimental investigation of the behavior of nanoscale annulus structures after PLiD is performed, with a primary focus on annuli with internal radii less than 300nm, thickness less than 20nm, and width less than 450nm. Prior work [1] on larger scales has investigated the formation of arrays of multiple droplets, where particles are typically distributed within the original nanostructure footprint. At these larger dimensions, several factors compete to direct the formation of the particles, including the Plateau-Raleigh instability, thin film instabilities (spinodal or nucleation), and the retraction dynamics [2]. Recent work investigates the dimensional parameter space where dewetting of an annulus results in the competition of the collapse to a single particle versus circumferential instabilities which are operative at larger scales. Ultimately, the physical system tends to reach a local potential energy minimum, where the surface energy is minimized. This phenomenon is complicated by substrate interactions with the droplet. Theoretical models involving liquid-substrate and Van der Waals interactions have been developed [3] regarding the dewetting of thin metal nanostructures on substrates. PLiD has the advantage of causing very short liquid lifetimes, which allows the liquid and visco-inertial behavior to dominate. This is in contrast to surface diffusion driven processes that may dominate other dewetting methods, such as annealing by substrate heating.

Scanning electron microscope (SEM) imaging is performed to characterize initial morphology of the as-synthesized nanostructures and their post-laser treated pseudo-equilibrium state (Figure 1). Image processing techniques extract quantitative dimensional properties for both initial state and final state morphologies. As nanoscale dewetting is a stochastic process, analysis of many samples allows aggregation of statistical information for parameter space analysis (Figure 2). Describing the scale range most likely to result in annuli dewetting to unitary particles allows comparison with predictions of computational modeling, and a better understanding of hydrodynamic forces at the nanoscale.

Experimental methods
Fabrication is performed in a class 1000 cleanroom. PMMA is spin-coated onto 100 nm thick SiO$_2$ on Si wafers for 90 seconds at 4000 rpm. E-beam lithography exposes CAD patterns of a variety of ring dimensions. Exposed wafers are developed in 1:3 MIBK:IPA solution for 100 seconds, followed by an IPA rinse and N$_2$ dry. A 60 second plasma descum precedes sputtering. Copper films of 5 to 15 nm are uniformly sputtered at 8.8±1.0 nm/min in a 3.0± 0.1 mT argon atmosphere. Following deposition, liftoff is performed with a 7 min acetone bath and 15 to 30 min acetone sonication. Rinsed and dried samples are loaded for SEM imaging. After imaging, a 248 nm KrF excimer laser irradiates samples at 5 Hz with 18 ns (FWHM Gaussian) pulses and a fluence of 160 mJ/cm$^2$. Solvent rinse and N$_2$ dry precedes SEM post-pulse imaging.
References


[Figure 1] Two 15 nm thick annuli as-deposited (top) and after pulsing to equilibrium (bottom), with dimensions of initial internal and external radii. Similarly sized rings formed both unitary and binary particles, indicating some stochasticity in the dewetting process.

[Figure 2] Fabrication and analysis of 15 nm thick annuli over a variety of dimensions allows quantization of the average number of droplets formed from structures with specific combinations of internal radius and width. Collected from ~600 nanostructures.