Laser-induced self-assembly of noble metal nanoparticles and EELS characterization

Yueying Wu[1], Guoliang Li[2], Charles Cherqui[3], Jon P. Camden[2], David J. Masiello[3], Jason D. Fowlkes[4], Philip D. Rack[1, 4]

1 Department of Materials Science and Engineering, University of Tennessee, Knoxville, TN37996, USA
2 Department of Chemistry and Biochemistry, University of Notre Dame, Notre Dame, IN46556, USA
3 Department of Chemistry, University of Washington, Seattle, WA98195, USA
4 Oak Ridge National Laboratory, Center for Nanophase Materials Science, Oak Ridge, TN37831, USA
Email: prack@utk.edu

Metallic nanostructures have played a central role in many important applications such as optical and magnetic information processing, catalysis, sensing, and photovoltaics. Recently, the self and directed assembly of nanoparticles via metallic thin film dewetting has proven to be an effective and low-cost approach to realizing multi-functional nanostructures. While the dewetting behavior of polymer films has been experimentally and theoretically studied in the past decades, the physical mechanisms underlying metallic thin film dewetting are not well understood due to the drastic difference in the physical properties of metal films. In our recent studies, the self and directed pulsed laser induced dewetting of unpatterned, as well as nanolithographically pre-patterned thin films of various shapes and sizes was investigated for the purpose of understanding how initial boundary conditions facilitate precise assembly. Specifically, we present a study on the self-assembly of noble metal thin films and also provide a comprehensive characterization of the resultant nanoparticles (See figure 1 for an over view of synthesized Ag nanoparticles and measurement/ simulation set up) through experiment using electron energy loss spectroscopy (EELS) and through simulation using the full-wave electron-driven discrete-dipole approximation (e-DDA) software package. We use truncated spherical nanoparticles as a model system to test the predictions made by Mie theory. From a synthesis point of view, the nanospheres (~20nm to 1μm) are in a deep metastable state and hence stable at high temperatures. Moreover, the liquid dewetting process yields nanoparticles with an ultra-smooth surface when compared to the traditionally rough surfaces of lithographically prepared nanoparticles. Both of our EELS experiment and theoretical simulations probes all electron density oscillations supported by the system, including the Mie, bulk, and surface modulated bulk modes. In this talk we give an overview of the proposed synthesis method and illustrate how correlated and highly precise patterns can be synthesized in both core-shell structure and 2-dimensions (figure 2 shows a series of 2D patterns as an example). We also present the theoretical and experimental EELS characterization of the nanoparticles. Finally we will present some concepts for advanced optical materials and devices based on directed assembly of complex nanoparticle arrays.

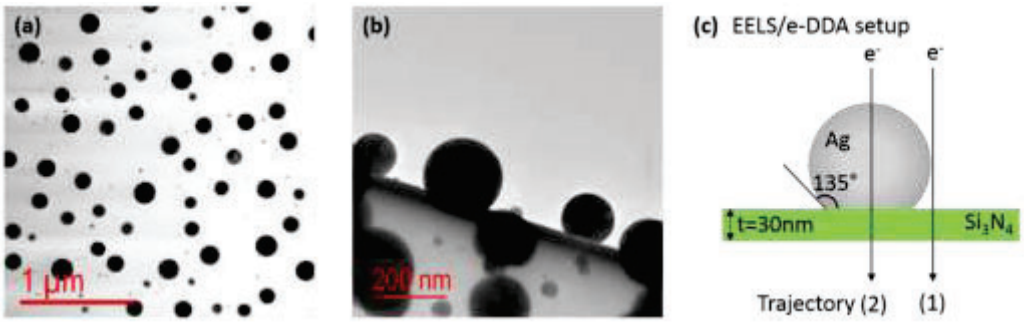

Figure 1. (a-b) Plane view and cross-section TEM images of Ag nanoparticles sitting on a Si3N4 substrate. **(c)** Schematic of the EELS experimental and e-DDA simulation set up. The electron beam was ~1 nm away from the particle surface, as is shown in the schematic.

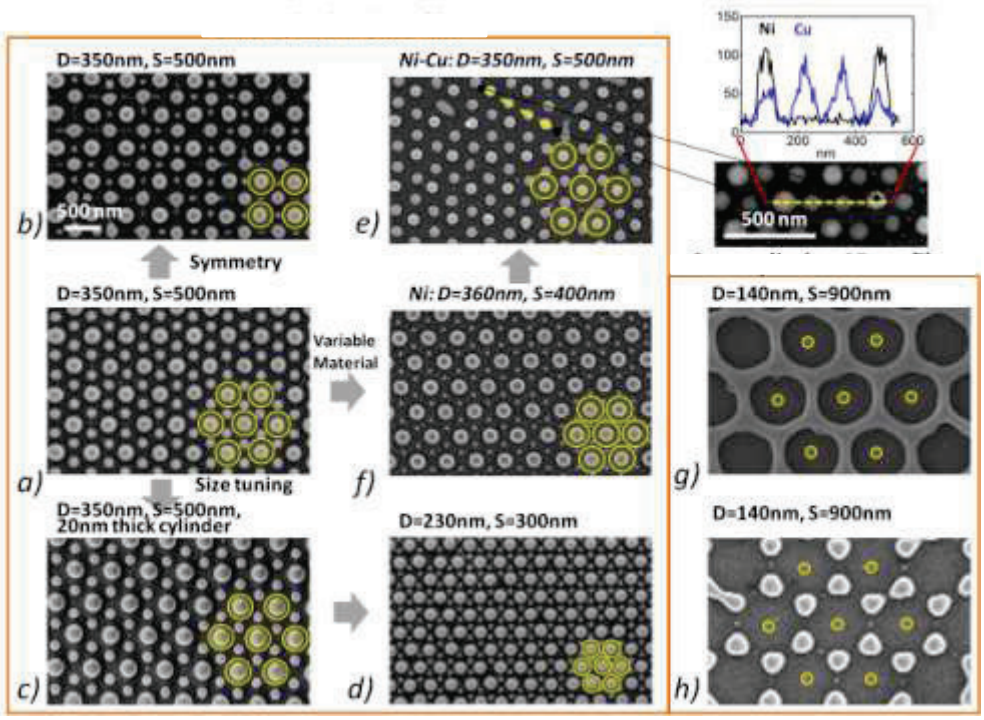

Figure 2. Collage of Cu-Ni nanoparticle arrays that have been directly assembled on Silicon dioxide substrates.