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Inside Out: Visualizing intercalation-driven phase transformations within individual nanoparticles

Abstract: A number of energy and information-relevant processes, ranging from battery charging to memory storage and retrieval, rely on nanomaterial phase transitions induced by solute intercalation. However, many of these phase transitions are poorly understood, since observing them in nanomaterials – and in particular in individual nanoparticles – is extremely challenging. This presentation will describe a novel technique to visualize intercalation-driven phase transitions within individual nanoparticles, based on *in-situ* environmental transmission electron microscopy (TEM) and plasmon electron energy loss-spectroscopy (EELS). As a model system, we focus on the hydrogenation of palladium nanoparticles. We use the plasmon-EEL signal at varying hydrogen pressures as a proxy for hydrogen concentration in the particle. First, we investigate the hydriding properties of single-crystalline particles, free from defects and grain boundaries, and free from elastic interactions with the substrate. We obtain single particle loading and unloading isotherms for particles ranging from approximately 10 nm to 50 nm, allowing us to address outstanding questions about the nature of phase transitions and surface energy effects in zero-dimensional nanomaterials. We find that hydrogen loading and unloading isotherms of single crystals are characterized by abrupt phase transitions and macroscopic hysteresis gaps. These results suggest that thermodynamic phases do not coexist in single-crystalline nanoparticles, in striking contrast with ensemble measurements of Pd nanoparticles. Then, we extend our single-particle techniques to explore the hydriding properties of polycrystalline and multiply-twinned nanoparticles, including Pd nanorods and icosahedra. In contrast to single crystalline nanoparticles, these particles exhibit sloped isotherms and narrowed hysteretic gaps. Based on these results, we develop a model to deconvolve the effects of disorder and strain on the phase transitions in nanoscale systems. Lastly, we describe techniques to generate high-resolution plasmon-EELS (and hence phase) maps of nanoparticles. These mapping studies promise unprecedented insight into the internal phase of nanomaterials, and can be complemented with diffraction and dark-field imaging studies. We will discuss how these results could be used to interpret the thermodynamics of Li-ion insertion in battery electrodes, hydrogen absorption in state-of-the-art metal hydride catalysts, or ion exchange reactions in quantum dot syntheses.

Bio: Jennifer Dionne is an assistant professor in the department of Materials Science and Engineering at Stanford University. Jen received B.S. degrees in Physics and Electrical & Systems Engineering from Washington University in St. Louis in 2003, and a Ph.D. degree in Applied Physics from the California Institute of Technology in 2009, advised by Prof. Harry Atwater. She joined Stanford in 2010 following a postdoctoral research fellowship at the University of CA, Berkeley and Lawrence Berkeley National Laboratory, working with Prof. Paul Alivisatos. Jen's research develops new nano and optical materials for applications ranging from high-efficiency solar energy conversion to bioimaging and manipulation. This research has led to demonstration of negative refraction at visible wavelengths, development of a subwavelength silicon electro-optic modulator, development of quantum plasmonic materials, design of new optical tweezers for nano-specimen trapping, and demonstration of a metamaterial fluid. She was recently awarded the Sloan Foundation Fellowship (2015), the Presidential Early Career Award for Scientists and Engineers

(2014), and the inaugural Kavli Nanoscience Early Career Lectureship (2013). She was also named one of Technology Review's TR35 - 35 international innovators under 35 tackling important problems in transformative ways (2011).