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In-situ, nanometer-scale visualization of nanoparticle phase transitions and light-matter interactions in 2- and 3-D

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We present new spectroscopic techniques that enable visualization of nanoparticle phase transitions in reactive environments and light-matter interactions with nanometer-scale resolution. First, we directly monitor hydrogen absorption and desorption in individual palladium nanocrystals. Our approach is based on *in-situ* electron energy-loss spectroscopy (EELS) in an environmental transmission electron microscope. By probing hydrogen-induced shifts of the palladium plasmon resonance, we find that hydrogen loading and unloading isotherms are characterized by abrupt phase transitions and macroscopic hysteresis gaps. These results suggest that alpha and beta phases do not coexist in single-crystalline nanoparticles, in striking contrast with conventional phase transitions and ensemble measurements of Pd nanoparticles. Then, we then extend these techniques to monitor nanoparticle reactions in a liquid environment. By constructing a flow chamber, we directly monitor growth and assembly of colloidal plasmonic metamaterial constituents induced by chemical catalysts. Lastly, we introduce a novel tomographic technique, cathodoluminescence spectroscopic tomography, to probe optical properties in three dimensions with nanometer-scale spatial and spectral resolution. Particular attention is given to reconstructing a 3D metamaterial resonator supporting broadband electric and magnetic resonances at optical frequencies. Our tomograms allow us to locate regions of efficient cathodoluminescence across visible and near-infrared wavelengths, with contributions from material luminescence and radiative decay of electromagnetic eigenmodes. The experimental signal can further be correlated with the radiative local density of optical states in particular regions of the reconstruction. Our results provide a general framework for visualizing chemical reactions and light-matter interactions in plasmonic materials and metamaterials, with sub-nanometer-scale resolution, and in three-dimensions.