

The Light Years: Combined optical and environmental electron microscopy to visualize dynamic photochemical processes with atomic-scale resolution

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Pearl Jam's hit, "The Light Years," declares "We were but stones, light made us stars." Bringing light to the transmission electron microscope (TEM) promises to transform our understanding of materials, enabling direct observation of dynamic photochemical processes with near-atomic scale resolution. To help achieve this goal, my lab is developing capabilities for concurrent optical and electron microscopy within an environmental TEM. This presentation will describe our efforts to visualize a variety of dynamic photochemical processes *in situ* with nanometer-to-atomic scale resolution. We focus in particular on i) observing plasmon-driven chemical transformations in nanoparticles; ii) detecting quantum light emission from two-dimensional materials; and iii) developing TEM-Raman with nanometer-scale resolution. First, we study the photocatalytic dehydrogenation of Au-Pd systems, in which the Au acts as a plasmonic light absorber and Pd serves as the catalyst. We find that plasmons modify the rate of distinct reaction steps differently, increasing the overall rate more than ten-fold. Plasmons also open a new reaction pathway that is not observed without illumination, laying a foundation for site-selective and product-specific photocatalysts. Secondly, we use scanning transmission electron microscopy and cathodoluminescence spectroscopy to investigate color centers in two-dimensional hexagonal boron nitride, a wide bandgap material capable of room-temperature, high-brightness visible quantum emission. We find that sharp, single-photon emission peaks are usually associated with regions of multiple fork dislocations, facilitating design of next-generation two-dimensional materials and devices. Finally, we describe a new technique for vibrational spectroscopy in the TEM: electron- and light-induced stimulated Raman scattering (ELISR). In particular, we leverage the electron beam as a highly-localized Angstrom-scale source to locally excite the plasmonic resonances of individual nanoparticles, which stimulate local Raman scattering. We show how ELISR can be used to map molecular signatures with nanometer-scale resolution.