

Ultrafast Electron Microscopy: Nanophotonics at the Limit of Space and Time

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Seeing is believing, and one of the best tools we have today for understanding nanoscale phenomena is the transmission electron microscope (TEM). The TEM has evolved beyond a tool for atomic-scale imaging and diffraction. Modern electron microscopes are now fully integrated instruments capable of measuring the dynamic material behavior at the nanoscale under various conditions and biases. One notable advance has been the combination of the TEM with ultrafast lasers and the development of ultrafast electron microscopy (UEM).[1] UEM allows one to investigate light-matter interactions with a time resolution of hundreds of femtoseconds and a spatial resolution at the nanometer scale. In this presentation, photon-induced near-field electron microscopy (PINEM) will be introduced as a novel spectroscopic approach that pushes the limits of space and time for understanding the dynamics of nanophotonics structures. PINEM is a technique that employs an optical pump pulse and a time-gated electron wave packet to probe the dynamics of photon-induced near-fields near resonant nanostructures.[2]

This presentation will detail recent experimental results and theoretical insights into understanding near-field excitation and decay from both plasmonic and dielectric resonators. Earth-abundant plasmonic aluminum nanocrystals were studied, both as pristine nanoparticles and when wrapped with SiO₂ shells. PINEM experiments showed plasmonic near-fields driven by a 515nm pump laser as well as polarization-dependent responses from the optical near-fields for all plasmonic nanoparticles. However, no variations in near-field behavior were observed across multiple samples due to the SiO₂ shell. For plasmonic nanoparticles, the UEM time resolution was limited to >500 fs, which did not facilitate the observation of plasmon excitation and decay dynamics as a function of their oxide interface.

In an attempt to explore nanophotonic systems with longer resonance lifetime and to observe decay properties, Mie resonant dielectric materials were studied with PINEM under similar conditions. Silicon nanoparticles were generated through the disproportionation of silicon monoxide between 1400-1600°C followed by hydrofluoric acid etching to liberate 50-200nm spheres with calculated Mie resonances in the visible. PINEM experiments once more confirmed polarization-dependent scattering, but to a lesser degree than their plasmonic counterparts. Due to the polydispersity of the sample, no extended Mie resonances have been observed to date. However, to overcome this limitation, we have developed a new method to produce monodisperse rutile TiO₂ with strong visible resonances. Upcoming experiments seek to test these well-defined nanophotonic resonators for their near-field dynamics with UEM.

This research is expected to produce insights into energy transfer dynamics of nanophotonic heterostructures and produce near-field decay dynamics of nanophotonic materials with applications in photocatalysis.[3]

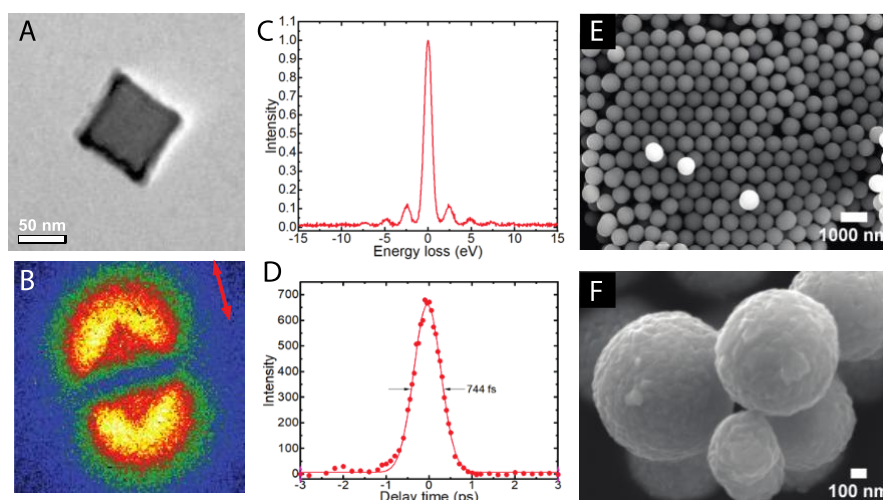


Fig. 1 Photon-Induced Near-field Microscopy for optical nanostructures. (A) Isolated plasmonic Al nanocube with 5nm thick silica shelling (B) Polarized PINEM field map of Al nanocube nearfields (C) Electron energy gain and loss spectra for PINEM measurements in (B) (D) Time dependence of PINEM fields (E) scanning electron micrograph (SEM) of monodisperse TiO₂ nanospheres which exhibit Mie resonances in the visible and (F) higher magnification of TiO₂ nanospheres. Part of this work was performed at the Center for Nanoscale Materials, a U.S. Department of Energy Office of Science User Facility, was supported by the U.S. DOE Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

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