

Plasmonics for Photochemistry of Transient Molecules

Seminar

Monday,
October 28, 2024

3:00 – 4:00 p.m.

Beaupre Center,
Room 105

Plasmons hold great promise for photon conversion and coherent energy transport at the nanoscale. Following visible light excitation of collective conduction electrons near the surface of nanostructures, hot charge carriers are created via non-radiative relaxation pathways that are then harnessed to drive localized molecular transformations of individual molecules. Limited knowledge of the precise mechanisms underlying plasmon-mediated dynamics in nanogaps currently restricts widespread technological applications, which in turn has generated substantial fundamental research interest in surface molecule dynamics. This talk demonstrates the direct observation of the molecular anion radical probed using continuous wave single nanoparticle pump-probe surface-enhanced Raman spectroscopy (SERS) within optically confined near-field plasmonic substrates. Corroboration of the anion radical species formed within the solid-state is provided by open-shell density functional theory (DFT), resonance Raman, and electron paramagnetic resonance spectroscopy (EPR). Wavelength tunability of plasmon-driven electron transfer as well as the spectrally resolved polarization response of individual nanoparticles is correlated with far-field scattering and high-resolution transmission electron microscopy (TEM) with electronic properties inferred from 2D x-ray absorption near edge spectroscopy (2D-XANES). Implications for speeding up/slowing down excited-state dynamics in high-field confinement are discussed using spectroscopic visualization of kinetic lifetimes in transient absorption spectroscopy (fs-TA). Utilization of this new energetic pathway has profound implications for basic energy research with the potential for greatly enhancing next generation light harvesting nanoantennae in sustainable energy applications.



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