This talk describes recent projects where we have used a combination of experiments and theory to study the interaction of plasmons with excited states of organic molecules. Plasmon excitation can produce enhanced electric fields near the nanoparticle surface, and this can result in enhanced fluorescence intensities and shortened fluorescence lifetimes. A recent project with Chad Mirkin shows how this works for relatively large nanoparticle structures fabricated using on-wire lithography, and we show that classical electrodynamics provides an accurate description of the results.

When the nanoparticles are only a few nm in diameter, ligands surrounding the nanoparticle can influence time-resolved absorption spectrum, as demonstrated in a project done with Emily Weiss. Here we used electronic structure calculations to show that the ligands contribute significantly to the plasmonic density of states, and also influence the electron-phonon coupling strength. As a result it is possible to alter the rate of energy flow between electrons and phonons, and also the flow of energy to the surrounding solvent. In the third project, we have collaborated with Teri Odom’s group to study the time evolution of exciton populations in dye molecules that are near to plasmonic particle array structures. Here we modeled the dye excited states using a 4-level model that incorporates plasmon-enhanced fields that are obtained from FDTD calculations. We find that femtosecond pumping leads to inverted populations in the dye, and emission above an intensity threshold that is strongly coupled to lattice plasmon modes.