EXAMINING THE NANOWORLD USING A MOLECULAR SPECTROSCOPIST'S TOOLBOX

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I will describe recent advances in understanding the influence of nanoscale structure on plasmon-mediated electron dynamics. Steady-state extinction spectra of plasmonic nanoparticle networks are accurately described using hybridization models reminiscent of molecular orbitals. We have extended these molecular-based descriptions to account for nanoparticle electron dynamics by quantifying the coherence dephasing times of collective inter-particle plasmon modes of single nanostructures. In particular, we demonstrate that interference between plasmon modes of different angular momenta leads to increased coherence times. These observations are consistent with a model based on superpositions of molecular-like electronic states. These fundamental studies are important for understanding the structure-photonic-function relationship of plasmonic nanoparticles. This is because the spectroscopically determined coherence times reflect mode quality factors, which determine achievable amplification factors of optical signals. These new insights are made possible by recent advances in single-nanoparticle/molecule spectroscopy based on interferometric nonlinear optical detection. I will describe how the generation of sequences of phase-locked femtosecond laser pulses (33mrad phase stability) and their integration to an optical microscope were critical for this research.