The University of Texas at San Antonio

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Understanding the role of the hot-electrons in the photophysical properties of nanomaterials

The surface plasmon resonance of the photoexcited plasmonic nanoparticles dephases in a few femtoseconds, emitting photons that are observable as scattering, and generating electron-hole pairs that cause light absorption. The electrons are excited to energy states higher than the Fermi level, suggesting a nonthermal distribution according to Fermi-Dirac statistics. The nonthermal hot electrons can excite electronic or vibrational transitions in molecules adsorbed on the surface of the nanoparticles, thereby inducing photochemical reactions. Within a few hundred femtoseconds, due to electron-electron scattering and electron scattering with the surface, the remaining hot electrons thermalize to temperatures that are hundreds of degrees above the surrounding lattice. One picosecond after the initial excitation of the plasmonic nanoparticle, the thermally hot electrons relax and transfer their energy to the lattice through electron–phonon interactions. Finally, the thermal energy dissipates to the surrounding medium by thermal conduction through lattice vibrations. The hot electrons of the plasmonic nanoparticles can induce photochemical reactions. It is not completely clear whether plasmonic photothermal heating has an impact on the photochemical reaction induced by nonthermal hot electrons. The lattice vibrational modes of nanoparticles are related to the intrinsic mechanical properties of the material and the shape of the nanoparticle. Ultrafast pump-probe spectroscopy is used to study these features. Vibrations cause a periodic change in the nanoparticle size and thus a corresponding shift in the localized surface plasmon resonance (LSPR) spectrum. This time-dependent shift is followed by measuring the changes in the intensity of the spectrum at a fixed wavelength at different delay times after the excitation. The mechanical properties of different hollow (single and double shells) and solid nanoparticles of different shapes and sizes will be discussed.

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