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Non-equilibrium hot carrier dynamics in plasmonic nanostructures PRINEHA NARANG, RAVISHANKAR SUNDARARAMAN, ADAM JERMYN, Caltech, EMILIANO CORTES, STEFAN A. MAIER, Imperial College, London, WILLIAM A. GODDARD, III, Caltech — Decay of surface plasmons to hot carriers is a new direction that has attracted considerable fundamental and application interest, yet a fundamental understanding of ultrafast plasmon decay processes and the underlying microscopic mechanisms remain incomplete. Ultrafast experiments provide insights into the relaxation of non-equilibrium carriers at the tens and hundreds of femtoseconds time scales, but do not yet directly probe shorter times with nanometer spatial resolution. Here we report the first ab initio calculations of non equilibrium transport of plasmonic hot carriers in metals and experimental observation of the injection of these carriers into molecules tethered to the metal surface. Specifically, metallic nanoantennas functionalized with a molecular monolayer allow for the direct probing of electron injection via surface enhanced Raman spectroscopy of the original and reduced molecular species. We combine first principles calculations of electron-electron and electron-phonon scattering rates with Boltzmann transport simulations to predict the ultrafast dynamics and transport of carriers in real materials. We also predict and compare the evolution of electron distributions in ultrafast experiments on noble metal nanoparticles.

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