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Non-equilibrium and ultrafast processes in surface plasmon decays

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Despite more than a decade of intensive scientific exploration, new plasmonic phenomena continue to be discovered, including quantum interference of plasmons, observation of quantum coupling of plasmons to single particle excitations, and quantum confinement of plasmons in single-nm scale plasmonic particles. Simultaneously, plasmonic structures and widening applications in integrated nanophotonics, biosensing, photovoltaic devices, single photon transistors and single molecule spectroscopy. Decay of surface plasmons to hot carriers is a new direction that has attracted considerable fundamental and application interest, yet a theoretical understanding of ultrafast plasmon decay processes and the underlying microscopic mechanisms remain incomplete. Recently we analyzed the quantum decay of surface plasmon polaritons and found that the prompt distribution of generated carriers is extremely sensitive to the energy band structure of the plasmonic material. A theoretical understanding of plasmon-driven hot carrier generation and relaxation dynamics at femtosecond timescales is presented here. We report the first ab initio calculations of phonon-assisted optical excitations in metals, which are critical to bridging the frequency range between resistive losses at low frequencies and direct interband transitions at high frequencies. Employing a Feynman diagram approach here has been critical to determine the relevant processes. Previously a challenge in such calculations of metals has been to treat the intermediate virtual state as well as the energy conserving `on-shell' intermediate states that correspond to sequential processes. Here we compare the plasmon line width and decay rates estimated directly from the experimentally-measured complex dielectric functions with theoretical predictions for cumulative contributions from direct, surface-assisted, phonon-assisted transitions and resistive losses. Extending the theory methods developed for phonon-assisted optical transitions, we calculate multiplasmon and nonlinear processes in the ultrafast regime from the mid-IR to visible and in different geometries. Next we report ab initio calculations of non equilibrium transport of plasmonic hot carriers in metals. The full description of non-equilibrium

carrier transport involves several variables: the spatial and temporal evolution of energy and momentum distributions of the hot carriers. We examine plasmonic hot carrier dynamics from two orthogonal simpler perspectives: ultrafast dynamics in pump-probe measurements, where spatial dependence can typically be ignored, and the steady-state transport in nanostructures at continuous wave illumination, where time dependence is not important. 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 from single femtosecond to picosecond time scales.

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