Monitoring plasmon generated hot carriers one particle at a time

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The light emission from plasmonic nanostructures has attracted significant interest lately as several different mechanisms have been suggested. All of them agree that the surface plasmon enhances a weak emission as it acts like an efficient antenna. Here I will present results from our lab studying in particular single gold nanorods (AuNRs) [1-4]. Our most recent work shows that the emission from AuNRs can be viewed as a Purcell effect enhanced spontaneous emission of hot carriers. We developed a quantitative model and the simulated photoluminescence spectra capture both the main emission peak following the scattering lineshape and originating from the enhancement by the large photonic density of states (PDOS) of the longitudinal surface plasmon resonance as well as a weaker, short-wavelength emission close to the excitation energy. While the latter emission is assigned to radiative recombination of sp-band electrons and d-band holes, the main photoluminescence resonance originates from both radiative interband and intraband transitions. We were able to reach these conclusions by correlating the photoluminescence spectra and quantum yields of 80 single AuNRs for five excitation wavelengths covering both interband and pure intraband excitation. Importantly, although steady-state in nature, these measurements together with excitation power dependent studies indicate the involvement of hot carriers and indirectly illustrate their relaxation dynamics as the spectral position of the photoluminescence shifts due to changes in hot carrier distributions. While the different excitation wavelengths and powers prepared different initial distributions, the effect of the PDOS on the photoluminescence remained unchanged for the same AuNR, but could be tuned over a wide spectral range through its aspect ratio.

Reference