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Resonant secondary light emission from plasmonic Au nanostructures at high electron temperatures created by pulsed laser excitation DAVID CAHILL, JINGYU HUANG, WEI WANG, CATHERINE MURPHY, University of Illinois — We study continuous-wave (cw) and pulsed laser excitation of aqueous suspensions of Au nanorods (AuNRs) as a model system for secondary light emission from plasmonic nanostructures. Resonant secondary emission contributes significantly to the background commonly observed in surface-enhanced Raman scattering (SERS) and to the light emission generated by pulsed laser excitation of metallic nanostructures that is often attributed to two-photon luminescence (TPL). The intensity of anti-Stokes emission collected using cw laser excitation at 785 nm is described by a 300 K thermal distribution of excitations. Excitation by sub-picosecond laser pulses at 785 nm broadens and increases the intensity of the anti-Stokes emission in a manner that is consistent with electronic Raman scattering by a high temperature distribution of electronic excitations predicted by a two-temperature model. Broadening of the pulse duration using an etalon reduces the intensity of anti-Stokes emission in quantitative agreement with the model. Experiments using a pair of sub-picosecond optical pulses separated by a variable delay show that the time-scale of resonant secondary emission is comparable to the time-scale for equilibration of electrons and phonons.

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