NONLINEAR FOURIER-TRANSFORM SPECTROSCOPY OF SINGLE GOLD NANOPARTICLES

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Plasmonic metal nanoparticles exhibit strong and tunable light-matter interactions which have enabled a variety of applications. Alongside their strong linear extinction, plasmonic nanoparticles have also been noted for their nonlinear optical properties. Here we present a study of multiphoton absorption in plasmonic gold nanorods using Fourier-transform spectroscopy with nonlinear optical detection. The excitation spectra of single gold nanorods is retrieved from an interferometric multiphoton photoluminescence (MPPL) signal which is generated by a pair of phase-locked pulses with attosecond interpulse delay precision. The resulting Fourier-transform spectra reveal the two- and three-photon excitation pathways that lead to emission from gold nanorods. Investigation of the spectra demonstrates the connection between resonance frequency and nonlinear absorption cross-section in gold nanorods, which is attributed to the influence of gold's electronic structure.[a]

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