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Plasmonics at the Space-Time Limit

MARTIN AESCHLIMANN, Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, Germany

The optical response of metallic nanostructures exhibits fascinating properties: local field interference effects that lead to strong variations of the near field distribution on a subwavelength scale, local field enhancement, and long lasting electronic coherences. Coherent control in general exploits the phase properties of light fields to manipulate coherent processes. Originally developed for molecular systems these concepts have recently been adapted also to nano-optical phenomena. Consequently, the combination of ultrafast laser spectroscopy, i.e. illumination with broadband coherent light sources, and near-field optics, opens a new realm for nonlinear optics on the nanoscale. To circumvent the experimental limitation of optical diffraction we use a photoemission electron microscope (PEEM) that has been proved to be a versatile tool for the investigation of near field properties of nanostructures with a spatial resolution of only a few nanometers and that allows for new spectroscopy techniques with ultrafast time resolution [1,2]. We introduce a new spectroscopic method that determines nonlinear quantum-mechanical response functions beyond the optical diffraction limit. While in established coherent two-dimensional (2D) spectroscopy a four-wave-mixing response is measured using three ingoing and one outgoing wave, in 2D nanoscopy we employ four ingoing and no outgoing waves. This allows studying a broad range of phenomena not accessible otherwise such as space-time resolved coupling, transport, and Anderson localized photon modes [3, 4]. [1] M. Aeschlimann et al, Nature **446**, 301 (2007) [2] M. Aeschlimann et al, PNAS **107** (12), 5329 (2010) [3] M. Aeschlimann et al, Science **333**, 1723-1726 (2011) [4] M. Aeschlimann et al, Nature Photonics **9**, 663, (2015)