Phase-propagation of localized surface plasmons probed by time-resolved photoemission electron microscopy

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PEEM in combination with nonlinear photoemission has just recently attracted considerable attention due to its high and local sensitivity to collective electron excitations in silver nanoparticles (localized surface plasmons - LSP) [1]. A highly promising aspect in this context is the potential of two-photon photoemission electron microscopy to be performed in a time-resolved stroboscopic mode enabling real-time experiments at a temporal resolution in the femtosecond-regime [2]. This allows one to directly monitor the spatio-temporal dynamics of the local field distribution associated with the decay of the LSP mode. Even more, in a phase-resolved 2PPE mode accurate information on the relative phase of the LSP-mode to an oscillating reference field such as the driving external light field can be achieved [3].

In this paper we demonstrate that the sampling of the LSP-dynamics by means of timeand phase-resolved PEEM enables detailed insight into the propagation processes associated with these excitations. In phase-integrated as well as phase-resolved measurements we observe spatiotemporal modulations in the photoemission yield from a single nanoparticle. These modulations are assigned to local variations in the electric near-field as a result of the phase propagation of a plasmonic excitation through the particle. Furthermore, the control of the phase between the femtosecond pump and probe laser pulses used for these experiments can be utilized for an external manipulation of the nanoscale electric near-field distribution at these particles.



Time-resolved PEEM of a silver nano-particle (particle diameter: 200 nm) highlighting particle-internal 2PPE-yield variations as function of the temporal delay between pump and probe laser pulse.

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