Ultrafast photoemission electron microscopy: Imaging nonlinear plasmonic phenomena on the femto/nano scale

Light interacting with a metal surface can excite both single-particle (e-h pair) and collective (plasmon) excitations. By angle-resolved photoemission spectroscopy and photoemission electron microscopy, we investigate the coherent ultrafast dynamical processes in interaction of light with silver metal surfaces. To establish the physics of nonlinear multiphoton photoemission, we model the fundamental response of silver to intense optical excitation through its known dielectric properties and momentum resolved band structure. Furthermore, we employ the two photon photoemission process to image plasmonic phenomena in Ag metal films. By means of interferometric time-resolved photoemission electron microscopy (ITR-PEEM), we can create spatial maps of two-photon photoemission excited in nanostructured Ag films. We fabricate specific nanoscale structures for the coupling of surface plasmon polaritons (SPP), the electromagnetic modes of a metal/dielectric interface, and we image their effect on the coupling, propagation, interference, and focusing of SPP waves. By advancing the delay between identical and collinear pump and probe pulses with interferometric precision, we are able to record movies of SPP wave propagation, and nonlinear interactions with ~50 nm spatial resolution and 330 attosecond/frame temporal precision. Based on simple theoretical models, we discuss the imaging process, the optics of SPP wave packets, and the prospects of ultrafast microscopy of plasmonic phenomena.