Femtosecond imaging of surface plasmon dynamics in nano-optics

Atsushi Kubo
Institute of Physics, University of Tsukuba
PRESTO, Japan Science and Technology Agency

Surface plasmon polariton (SPP), a coherent density fluctuation of electrons photoexcited and propagates along surfaces of metals, has a potential to carry digital information in sub-wavelength scaled devices. Microscopic studies with sub-wavelength spatial resolutions, such as near-field scanning optical microscopy (NSOM), have been providing crucial knowledge for understanding physical properties and behaviors of SPPs. However, these methods usually have very slow time-resolutions. To investigate transient dynamics of SPP wave packets in circuits and devices, ultrafast time-resolutions are required.

By combining time-resolved two-photon photoemission (TR-2PP) and photoemission electron microscopy (PEEM), we take movies of SPPs with 0.3-fs frame interval, 10-fs time resolution, and 50-nm spatial resolution. Samples are deposited silver films with optical patterns of SPPs (e.g. lenses) incused using a focused ion beam technique. 10-fs, 400-nm, and phase-correlated optical pulse pairs irradiate the sample with the incident angle of 65^{o} from the surface normal and launch SPPs in optical patterns. SPP wave packets propagate along the silver surface, while the optical pulses induce local polarizations within the light penetration length. As a consequence of the interference of the SPP and the light-induced filed, a grating of surface polarization is formed. The polarization gratings are microscopically imaged by mapping the polarization-induced two-photon photoemission with using the PEEM. The spatial distribution and the intensity of gratings evolves as a function of the pump-probe delay time, therefore, we can extract the dynamics of SPPs form a sequences of PEEM micrographs taken with using a pump-probe manner.

We take movies of some typical behaviors of SPPs such as propagation, interference, focusing and damping. At silver-vacuum interfaces, SPPs excited with 400-nm light have the propagation length of ~3μm and the group velocity of ~0.6*c* (*c*: speed of light in vacuum). These properties are consistent with the values derived from the complex wave vectors of SPP. More detail dynamics of SPPs including the decoherence processes can be investigated by analyzing the spectra of photoelectrons, and such measurements are achieved by combining PEEM and time-of-flight electron spectroscopy (TOF-PEEM).

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Atsushi Kubo
Assistant Professor,
Institute of Physics, Graduate School of Pure and Applied Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki, JAPAN.
PRESTO, JST, 4-1-8 Honcho, Kawaguchi, Saitama, JAPAN.
Telephone & FAX: +81-29-853-2918
E-mail: kuboa@sakura.cc.tsukuba.ac.jp

Personal History

2003	Ph. D. in Physics, University of Tsukuba.
2003-2005	Postdoctoral fellow, University of Pittsburgh.
2005-2007	Researcher of PRESTO, JST and Visiting researcher, University of Pittsburgh.
2008-	Assistant Professor, University of Tsukuba.