Spectral properties and dynamics of localized surface plasmon resonances in metal/insulator/metal nanostructures

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Metal/insulator/metal nanostructures indicate strong near-field enhancement based on the localization of electromagnetic field at nanogap and relatively longer dephasing time because light scattering is suppressed at an adequate wavelength region via the excitation of quadrupole plasmon mode [1]. The quadrupole plasmon mode is excited by a near-field interaction between gold nanostructures separated by the insulator. In this study, we have elucidated spectral properties and ultrafast dynamics of localized surface plasmon resonances in metal/insulator/metal nanostructures by time-resolved photoemission electron microscopy.

Ordered array of metal/insulator/metal nanostructures was fabricated by electron beam lithography and dry etching processes on a niobium doped titanium dioxide (0.05wt% Nb-TiO_2) single crystal substrate. Three layers composed of gold/alumina/gold (Au/Al_2O_3/Au) were deposited by sputtering for Au and atomic layer deposition for Al_2O_3 on the Nb-TiO_2 substrate. From the SEM image measured from the oblique angle, it was found that the metal/insulator/metal nanostructure shows a taper shape from bottom. The structures were considered to be a taper shape by the underetching phenomenon during the dry etching process. Therefore, a side length (top) of the upper structure is about 120 nm whereas that (bottom) of the lower structure is about 180 nm. The gap distance between upper and lower gold nanostructures is 15 nm.

An excitation of quadrupole plasmon mode was confirmed by far-field reflection spectroscopy as a Fano dip. On the basis of the measurements by multi-photon photoemission electron microscopy, stronger near-field enhancement was obtained at slightly shorter wavelength of Fano dip based on the excitation of quadrupole plasmon mode. Therefore, we have successfully demonstrated near field enhancement effects of dark state of quadrupole plasmon mode. The dephasing time of metal/insulator/metal nanostructures measured by time-resolved photoemission electron microscopy was estimated to be 7 fs which is slightly longer than ~3.5 fs which is dephasing time of gold nanoblock structures fabricated on the same substrate.

References:

[1] Q. Sun, H. Yu, K. Ueno, A. Kubo, Y. Matsuo, H. Misawa, *ACS Nano*, **2016**, *10*, 3835.