

Revealing higher order surface plasmon resonances and their dynamics by PEEM

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Localized surface plasmon resonances (LSPRs) are collective charge oscillations confined to the surface of metallic nanoparticles (NPs). The properties of LSPRs depend on the size, shapes, metal material of the NPs, and surrounding media. When the size of a metallic NP is much smaller than the wavelength of the incident light field, the free electrons in the NP can oscillate with the same phase forming a dipole mode ($l = 1$). In contrast, when the size is comparable to or larger than the light wavelength, the free electrons at different locations of the NP can oscillate with different phases resulting in the excitation of higher order quadrupole ($l = 2$) or multipolar modes ($l > 2$)^[1,2]. Recently, we demonstrated the near-field mapping of dipole LSPRs on various gold NPs by multiphoton photoemission electron microscopy (PEEM)^[3]. In this study, we investigate the near-field properties of higher order LSPRs using PEEM. For thin gold nanoblocks (side length: 200 nm, thickness: 24 nm), we explore that the dipole and quadrupole modes can be selectively excited under oblique incidence. With p-polarized light excitation the quadrupole mode cannot be excited and the dipole dominates the plasmonic field enhancement; in contrast, with s-polarized light excitation the quadrupole mode dominates as shown in Fig. 1(a). Furthermore, the dephasing of the quadrupole mode has been investigated by time-resolved PEEM and is compared with that of dipole mode. In addition, we also successfully observe the near-field images of higher order LSPRs on 600 nm-long gold nanorods as can be seen in Fig. 1(b). This study opens the door to investigate complex plasmonic nanostructures such as those with Fano resonances, where the interaction between dipole and higher order LSPRs occurs, using this unique PEEM technique.

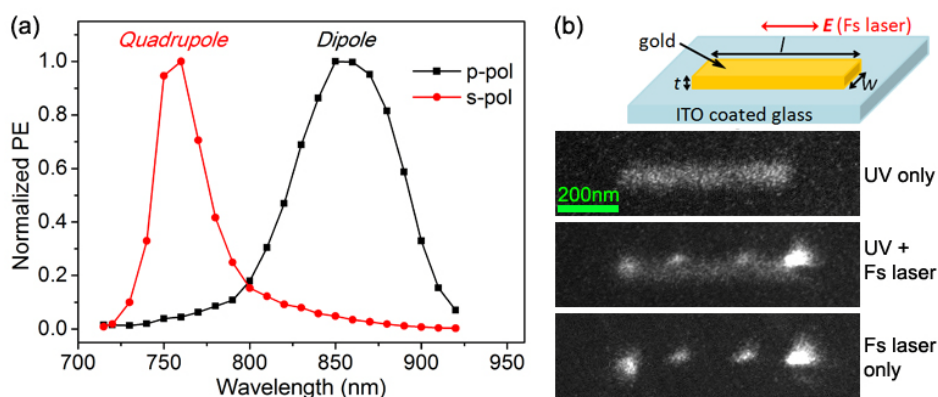


Figure 1. (a) Polarization dependent near field spectra of a gold nanoblock array, showing selective excitation of dipole and quadrupole LSPRs. (b) PEEM images of a long gold nanorod under different excitation conditions, exhibiting higher order ($l = 3$) LSPRs under femtosecond (Fs) laser excitation at 840 nm, the insert shows the schematic drawing of the nanorod ($l = 600$ nm, $w = 120$ nm, and $t = 24$ nm).

References

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