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Visualizing plasmons by near-field spectroscopy

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1. Introduction

Plasmons have attracted extensive interest due to their potential applications in various fields. The potential arises from ability of optical field confinement and hence enhancement of the optical field in the vicinity of the plasmonic material. Enhanced fields are utilized for surface enhanced Raman scattering (SERS), for example. To design SERS substrate for sensing applications, deep understanding of plasmon is essential. To reveal spectroscopic, spatial, and dynamical properties of plasmons, observation of plasmons with high spatio- and temporal resolution is crucial. We developed near-field spectroscopic methods, which enable to visualize plasmons spatio-temporally [1]. By using the microscope, we studied spatial and dynamical features of plasmons excited in various nanostructures. In this study, visualization of plasmons with various imaging techniques will be described.

2. Experimental

We used aperture near-field optical microscope, which combines various light sources depending on spectroscopic purposes. For Raman and transmission measurements, a cw laser and a Xe lamp, and for non-linear and time-resolved measurements, a mode-locked Ti:s laser is used as a light source. Spatial resolution of the microscope is < 100 nm, and time resolution is < 20 fs.

3. Results and discussion

Figure 1(a) show a near-field transmission spectrum observed on a gold nanorod (dia. 20 nm × length 510 nm). The spectrum exhibits multiple plasmon resonances. Figures 1(b,c) show near-field transmission images observed at the resonance wavelength of 656 nm and 676 nm, respectively. The image shows periodic spatial oscillation along the long axis of the nanorod, and the period gets longer for the longer wavelength. These spatial features are assigned to plasmon modes resonantly excited. We revealed that the near-field optical imaging is useful to visualize plasmon modes excited in other nanostructures such as gold plates [2], gold nanovoid [3], and so on.

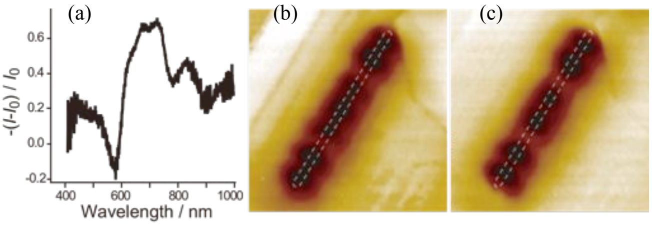

Figure 1. (a) Near-field transmission spectrum of a gold nanorod. (b,c) Near-field transmission images of the rod observed at 656 nm and 676 nm, respectively.

We found that optical field distribution can be visualized by detecting non-linear optical signal such as two-photon luminescence. Figure 2(a) show a two-photon excitation image of rough gold film (thickness 20 nm). Bright part indicates that optical field is enhanced by local excitation of plasmons. Figures 2(b,c) show near-field Raman excitation images of the same area in Fig. 2(a) observed at 1363 cm^{-1} and 1515 cm^{-1}, respectively. We found from these images that Raman activity is, in general, high at enhanced optical fields. We also performed fringe-resolved autocorrelation measurements on the same sample to obtain dephasing time of plasmons [4], and found that the Raman activity has a correlation with the dephasing time of the plasmons.

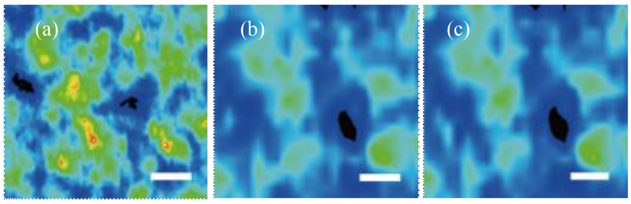

Figure 2. (a) Near-field two-photon excitation image of a rough gold film. (b,c) Near-field Raman excitation images of the same sample observed at 1363 cm^{-1} and 1515 cm^{-1}, respectively.

4. Conclusions

We have utilized near-field optical spectroscopy to visualize plasmon modes and optical fields excited in various plasmonic materials. Visualized spatial features varied depending on the resonance wavelength and hence plasmonic waves excited in the structure. To characterize plasmonic functions in detail, dynamical aspect of plasmon should be also characterized. Time-resolved measurements revealed that dephasing time of plasmons is from several fs to a few tens fs. By visualizing plasmon dephasing time and Raman active sites, we found that the Raman enhancement is higher at the plasmon exhibiting longer dephasing time.

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