

# Nano-Optical Imaging and Application to Nanomaterials

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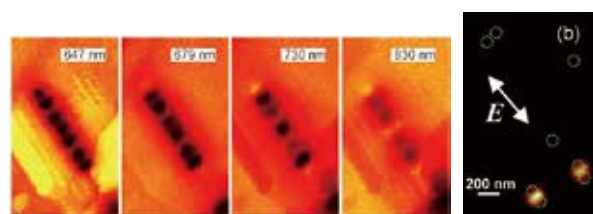
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Studies of local optical properties of molecular assemblies and materials are key to understanding nanoscale physical and chemical phenomena, and for construction of nanoscale functional devices. Nano-optical methods, such as scanning near-field optical microscopy (SNOM), enable optical imaging with spatial resolution beyond the diffraction limit of light. Combination of nano-optical techniques with various advanced spectroscopic methods may provide a methodology to analyze directly nanoscale functionalities and dynamics. It may yield essential and basic knowledge to understand origins of characteristic features of the nanomaterials systems. We have constructed nano-optical (near-field and far-field) spectroscopic and microscopic measuring systems, for the studies on excited-state properties of nanomaterials, with the feasibilities of nonlinear and time-resolved measurements. The developed apparatuses enable nano-optical measurements of two-photon induced emission, femtosecond time-resolved signals, and chiro-optical properties (as typified by circular dichroism), in addition to conventional transmission, emission, and Raman-scattering. Based on these methods, we are investigating the characteristic spatial and temporal behavior of various metal-nanostructure systems and molecular assemblies. Typical examples are shown in Figure 1. We succeeded in visualizing wave functions of resonant plasmon modes in single noble metal nanoparticles, confined optical fields in noble metal

nanoparticle assemblies. In recent few years, we have also succeeded in observing plasmon wave packet propagation dynamics with ultrafast time-resolved near-field imaging, local chiro-optical properties of chiral and achiral metal nanostructures, and so forth. The information on nano-optical properties of the materials are also relevant to exploration of novel optical manipulation principles, which is another research topic of the research group.



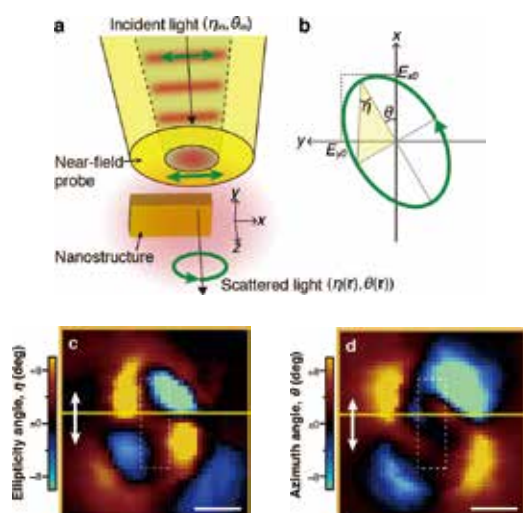
**Figure 1.** (Left four panels) Near-field transmission images of gold nanorod ( $20 \text{ nm}^D \times 510 \text{ nm}^L$ ). The wavelengths of observation were 647, 679, 730, and 830 nm from left to right. The spatial oscillating features were attributed to the square amplitudes of the resonant plasmonic wave functions. (Right) Near-field two-photon excitation image of dimers of spheric gold nanoparticles (diameter 100 nm) observed at 785 nm. The arrows indicates the incident light polarization. Dotted circles represent approximate positions of the particles.

### Selected Publications

- H. Okamoto, T. Narushima, Y. Nishiyama and K. Imura, "Local Optical Responses of Plasmon Resonance Visualized by Near-Field Optical Imaging," *Phys. Chem. Chem. Phys.* **17**, 6192–6206 (2015).
- H. Okamoto and K. Imura, "Visualizing the Optical Field Structures in Metal Nanostructures," *J. Phys. Chem. Lett.* **4**, 2230–2241 (2013).
- H. Okamoto and K. Imura, "Near-Field Optical Imaging of Enhanced Electric Fields and Plasmon Waves in Metal Nanostructures," *Prog. Surf. Sci.* **84**, 199–229 (2009).

## 1. Imaging Chirality of Optical Fields near Achiral Metal Nanostructures Excited with Linearly Polarized Light<sup>1)</sup>

Chiral systems respond differently to left- and right-handed circularly polarized light macroscopically. As a consequence, only chiral materials show intrinsic macroscopic optical activity, and only chiral systems generate circularly polarized light from linearly polarized incident light. In the nanoscopic regime, in contrast to this general rule for macroscopic cases, it is theoretically expected that achiral (nonchiral) systems can locally generate circularly polarized fields. Here, we report experimental evidence for that situation in achiral systems consisting of gold nanostructures and linearly polarized incident light. The local circularly polarized fields were visualized by near-field polarimetry imaging. In this novel method, linearly polarized near-field radiation illuminates the sample, and the polarization state (ellipticity and rotational angle of polarization) of the far-field scattered light was measured at each position of excitation on the nanostructure, to construct the image. We found that highly circularly polarized fields were generated in the peripheries of the nanostructures, and the spatial features of the observed circularly polarized fields were qualitatively reproduced by a simple oscillating dipole model. The present results may provide a novel technique to produce controllable circularly polarized optical fields in nanospaces, the demonstration of which is now under way.

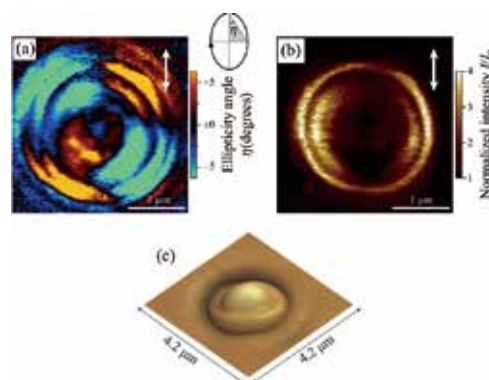


**Figure 2.** (a) Scheme of near-field polarimetry. (b) Definitions of ellipticity ( $\eta$ ) and rotation ( $\theta$ ) angles. (c) Ellipticity and (d) optical rotation images (at 800 nm) of nano-rectangle. Scale bar: 100 nm.<sup>1)</sup>

## 2. Nanoscale Chiral Surface Relief of Azopolymers Induced by Near-Field Optical Angular Momentum Light<sup>2)</sup>

An optical vortex with orbital angular momentum (OAM) can be used to induce micrometer-scale chiral structures in

various materials. Such chiral structures enable the generation of a near-field OAM light on a sub-wavelength scale, thereby leading to nanoscale mass-transport. We found formation of a nanoscale chiral surface relief (diameter  $\sim 400$  nm) in azopolymers due to near-field OAM light based on this principle. We analyzed near-field intensity and polarization state distributions in the periphery of the chiral structures using the near-field polarimetry imaging.



**Figure 3.** a) Near-field ellipticity, (b) near-field extinction, and (c) topographic images of left-handed spiral relief structure of azopolymer fabricated with OAM light (532 nm).<sup>2)</sup>

## 3. Super-Resolution Optical Trapping: Theoretical Prediction of Nanoparticle Manipulation Using Nonlinear Optical Response<sup>3)</sup>

Optical manipulation of nanoparticles (NPs) with nanoscale precision is one of goals of nanomaterials science. A way to realize this is the usage of localized surface plasmon resonances. The electric fields near metallic structures are highly localized, which generate sufficient force to trap NPs, and the optical nonlinearity of NPs appears at the same time. In this study, we propose a scheme of spatially highly confined (beyond the diffraction limit) optical trap of the NP into a particular hotspot of the metallic nanostructure array. The scheme is based on the optical nonlinearity of NPs, and utilizes two kinds of structured light: Gaussian and Laguerre-Gaussian beams. The principle resembles that of stimulated emission depletion microscope, one of the major techniques of super-resolution microscope. The simulation results show the significant role of the optical nonlinearity in plasmon trapings. They are expected to open up new degrees of freedom to manipulate NPs.

### References

- 1) S. Hashiyada, T. Narushima and H. Okamoto, *ACS Photonics* **5**, 1486–1492 (2018).
- 2) K. Masuda *et al.*, *Opt. Express* **26**, 22197–22207 (2018).
- 3) M. Hoshina, N. Yokoshi, H. Okamoto and H. Ishihara, *ACS Photonics* **5**, 318–323 (2018).

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