

# Nano-Optical Imaging and Chiral Light-Matter Interaction in Nanomaterials

## Center for Mesoscopic Sciences Division of Supersensitive Measurements



**OKAMOTO, Hiromi**  
Professor  
[aho@ims.ac.jp]

### Education

1983 B.S. The University of Tokyo  
1991 Ph.D. The University of Tokyo

### Professional Employment

1985 Research Associate, Institute for Molecular Science  
1990 Research Associate, The University of Tokyo  
1993 Associate Professor, The University of Tokyo  
2000 Professor, Institute for Molecular Science  
Professor, The Graduate University for Advanced Studies

### Award

2012 The Chemical Society of Japan (CSJ) Award for Creative Work  
2020 The Commendation for Science and Technology by the MEXT Awards for Science and Technology Research Category

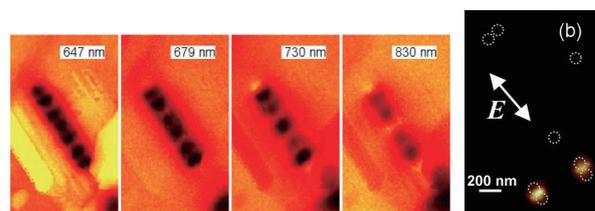
### Member

Research Assistant Professor  
YAMANISHI, Junsuke  
CHENG, An-Chieh  
Project Assistant Professor (NINS)  
AHN, Hyo-Yong  
Technical Support Staff  
ISHIKAWA, Akiko  
Secretary  
ITO, Atsuko

**Keywords** Nano Optics, Plasmons, Chirality

Studies of local optical properties of molecular assemblies and materials are the keys to understanding nanoscale physical and chemical phenomena, and for construction of nanoscale functional devices. Optical microscopic methods, in particular nano-optical methods, such as scanning near-field optical microscopy (SNOM) which enables resolution beyond the diffraction limit of light, reveals essential characteristics of the materials and develop novel properties of them. Combination of microscopic techniques with various advanced spectroscopic methods may provide a methodology to analyze nanoscale functionalities and dynamics directly. 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 polarization dependence and nonlinear/time-resolved measurements. The developed apparatuses achieved 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, plasmon wave packet propagation dynamics, local chiro-optical properties of chiral and achiral metal nanostructures, and so forth. We also developed far-field high-precision circular dichroism microscope that facilitate chirality analysis of materials in a wide range of research areas. The information on nano-optical properties of the materials is 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 spherical 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, "Local Optical Activity of Nano- to Microscale Materials and Plasmons," *J. Mater. Chem. C* **7**, 14771–14787 (2019).
- 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).

## 1. Local Chiro-Optical Effects in Gold Nanostructures Investigated by Chiral Photoinduced Force Microscopy

Photoinduced force microscopy (PiFM) is an optical near-field imaging technique based on the technique of atomic force microscopy (AFM). By irradiating light on a system consisting of a sample and a probe tip, and detecting the force arising from interactions between the light-induced polarizations of the sample and the probe, PiFM enables the visualization of local optical near-fields with nanometer-scale resolution. By employing left-handed and right-handed circularly polarized light as the excitation source, the differential force signal measured between the two polarization states (referred to as chiral PiFM) is expected to reflect chiro-optical effects in the near field. We previously demonstrated the feasibility of this approach through experimental measurements using pseudo two-dimensional chiral metallic nanostructures.<sup>1)</sup>

In the present study, we extend this methodology to investigate the general applicability of chiral PiFM and to gain deeper insights into its measurement principles. To this end, we perform chiral PiFM measurements and analyses on a variety of gold nanostructures with different geometries. In the case of three-dimensional chiral gold nanoparticles, near-field signals that correlate with the handedness of the particles were observed. For achiral assembled particle systems, local chiro-optical responses were observed in the peripheral areas, exhibiting spatially oscillatory patterns with alternating positive and negative signals. In spherical nanoparticles, where no chiro-optical effect is theoretically expected under circularly polarized illumination, weak signals were still observed in the chiral PiFM measurements. The result suggests that such signals may arise from symmetry-breaking effects involving the probe tip itself, and further analysis is ongoing to elucidate the underlying mechanisms.

## 2. Development of Far-Field Circular Dichroism Microscopy

Circular dichroism (CD) spectroscopy is a powerful technique widely employed for the detection and characterization of materials chirality. However, in anisotropic samples, signals arising from linear dichroism and other polarization-dependent effects often interfere with CD measurements. Consequently, the application of conventional CD measurement techniques to microscopic imaging has faced significant challenges to ensure signal accuracy and precision. Only very few reports on CD-based microscopic imaging have been published. In a previous study, we developed a high-precision far-field CD microscope based on a novel circular polarization modulation method, which enabled CD microscopic imaging with sufficiently suppressed interference from linear polarization effects.<sup>2)</sup>

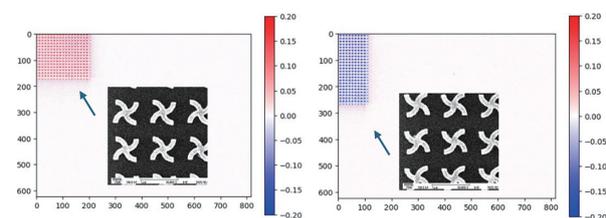
### Awards

YAMANISHI, Junsuke; 56<sup>th</sup> Excellent Presentation Award from Japan Society of Applied Physics (2024).

YAMANISHI, Junsuke; Nagoya University Ishida Prize (2025).

AHN, Hyo-Yong; Award for Promotion of the Most Excellent Young Researcher from Plasmonic Chemistry Research Group (2025).

To apply CD imaging to a wider range of research, diagnostics, and so forth, it is desirable to reduce measurement time and to simplify the optical alignment procedures. The previously developed system required a certain level of expertise for optical alignment and typical measurement times on the order of several minutes per image, indicating a need for further development. In the present work, we propose a new approach to CD microscopy aimed at substantially reducing imaging time and improving ease of operation. As a result of one such attempt, we have succeeded in acquiring CD images at several frames per second, with a slight compromise in signal accuracy compared to the earlier system configuration. This improvement represents a significant step toward the realization of real-time or live CD imaging, which is expected to broaden the applicability of CD microscopy in both fundamental and applied research.



**Figure 2.** Examples of CD images of pinwheel-shaped gold nanostructure arrays. Red and blue parts indicate positive and negative CD signals.

## 3. Toward the Observation of Chirality-Induced Dynamics

In recent years, it has been reported that the irradiation of achiral plasmonic materials with circularly polarized light can induce the formation of chiral nanostructures through photochemical reactions. This process is believed to involve several key steps: The excitation of chiral plasmonic resonances on achiral structures under irradiation of circularly polarized light; generation of chiral local electromagnetic fields; the spatial arrangement and orientation of reactant molecules in response to these fields; and, finally, the photoexcitation (or thermal excitation) of the reactants followed by chemical reactions to form chiral nanostructures. To understand the mechanisms of the processes, we have constructed an experimental apparatus for time-resolved measurements of chiral optical responses. Using this setup, we are currently investigating the dynamics of chirality-induced optical responses in plasmonic materials.

### References

- 1) J. Yamanishi, H.-Y. Ahn and H. Okamoto, *Nano Lett.* **23**, 9347 (2023).
- 2) T. Narushima and H. Okamoto, *Sci. Rep.* **6**, 35731 (2016).