

## Extrinsic Chirality by Interference between Two Plasmonic Modes on an Achiral Rectangular Nanostructure Using Multi-photon Photoemission Electron Microscopy

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Localized surface plasmon resonances (LSPRs), which are collective oscillations of conductive electrons at the surfaces of metallic nanostructures, harvest light incident on the nanostructures and enhance the local electric field, which is called the near field (NF). The NF induced by circularly polarized light (CPL) irradiation on the plasmonic structure is a hot scientific topic. Recently, Okamoto *et al.* observed a nanoscale two-dimensional chiral NF distribution on achiral rectangular gold nanostructures.<sup>1</sup>

In this study, we observed a chiral NF intensity distribution on a series of achiral gold nanorectangular structures (Au-NRs) under CPL irradiation by using multiphoton photoemission electron microscopy (MP-PEEM).<sup>2</sup> Additionally, the differential NF spectra under left and right CPL irradiation, which represent the asymmetry of the NF intensity distribution, were investigated. We propose an interpretation that the chiral NF intensity distribution on an achiral metallic nanostructure is extrinsically generated by the interference between two LSPR modes by combining state-of-the-art MP-PEEM techniques and the classical oscillator model. Not only the NF intensity distribution but also the observed peak wavelength of the DNF spectra in both the experiment and numerical simulation are well explained by our interpretation. Furthermore, it was proven that not only the dipole mode but also the hexapole mode could interfere with another mode and generate a chiral NF intensity distribution. A series of experiments revealed that the intensity of the NF and its phase angle of each mode are critical factors for the generation of the chiral NF intensity distribution. Importantly, both control factors can be estimated from the NF spectra under linearly polarized light irradiation without CPL irradiation.

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2) T. Oshikiri, Q. Sun, H. Yamada, S. Zu, K. Sasakai, H. Misawa, *ACS Nano* 2021, 15, 16802.