Near-field Distribution under Modal Strong Coupling with Coherent Interaction

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The modal strong coupling system between localized surface plasmon and Fabry–Pérot (FP) nanocavity modes was demonstrated to enhance the photocurrent generation efficiency and the quantum efficiency of the water splitting photoreaction.[1,2] Moreover, we have discovered the enhancement of the hot carrier generation efficiencies in the Au nanodisks (Au NDs) on thin-film TiO_2/Au-film (ATA) FP nanocavity modal strong coupling structure. We hypothesized that the coherent interaction between Au NDs through the FP nanocavity enhances hot carrier transfer efficiencies. In the present study, we further applied photoemission electronic microscopy (PEEM) to investigate the coherent interaction and the coherence area by discussing the near-field properties and the photoemission distributions.

In order to quantitively observe the coherence area, we fabricated the ATA structures with different particle numbers (PNs) of the 80-nm Au NDs hexagonal clusters. First, the near-field intensity spectra were observed by the PEEM measurement, and we found the near-field intensity peaks of the ATAs were at 710-730 nm. Next, we took the photoemission distribution images of the Au NDs clusters with different PNs at the peak wavelength excitation. We found that the Au NDs located in the rim part of the cluster showed obviously stronger photoemission intensities than the central part when PNs were larger than 7. This result indicates that Au NDs clusters exhibit the different modes from the individual plasmonic mode of single Au ND due to the coherent interaction through the FP nanocavity. Similar phenomena were also observed in the numerical simulation using the finite element method.

1) X. Shi, K. Ueno, T. Oshikiri, Q. Sun, K. Sasaki and H. Misawa, *Nat. Nanotechnol.* **2018**, 13, 953.
2) Y. Suganami, T. Oshikiri, X. Shi, H. Misawa, *Angew. Chem. Int. Ed.* **2021,** 60, 18438.