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1. 概要（Summary）

Near-field spectral properties of stacked nanogap gold (Au) nanostructures have been studied by multi-photon photoemission electron microscopy (PEEM) combined with a wavelength tunable femtosecond laser as an excitation source. The dark plasmon mode was excited by the near-field coupling between the upper and lower Au nanostructures separated by an alumina layer thickness of only 15 nm. A strong near-field enhancement effect was induced by the localization of the electromagnetic field between the upper and lower Au nanostructures and the longer plasmon dephasing time based on the excitation of the dark plasmon mode.

2. 実験（Experimental）

【利用した主な装置】

PEEM (Elmitec); electron beam lithography (EBL, ELS-F125-U, Elionix); Helicon sputtering (MPS 4000, ULVAC); Atomic Layer Deposition (ALD, Sunnale-R150, Picosun); ICP plasma etching system (RIE-101iPH, Samco); FE-SEM (JSM-6700FT, JEOL).

【実験方法】

The stacked nanogap Au nanostructures fabricated by sputtering, ALD, EBL, and drying etching techniques. The substrate is niobium-doped titanium dioxide crystal. Three layers of film were laminated by Au/Al_2O_3/Au with the thickness of 40 nm/15 nm/40 nm. The size of the top Au nanoblock was tuned between 100 nm and 120 nm to control the coupling strength between the upper and lower Au blocks. The near-field spectra and the dephasing time were measured by PEEM

3. 結果と考察（Results and Discussion）

SEM images clearly show the stacked nanogap metallic structures with three layers (Au/Al_2O_3/Au) as seen in Fig. 1 (a). Figure 1(b) shows both far-field reflection spectrum of an array of the stacked structures with size of the upper block of 120 nm. It can be found that the near-field enhancement peak locates around the reflection dip, indicating the formation of Fano resonance resulted from the near-field coupling between the upper and lowers blocks. The Fano dip wavelength redshift with increasing structural size. Time-resolved PEEM was used to measure the plasmon dephasing time. For the above-mentioned structures, a dephasing time of 10.5 fs could be obtained (Fig. 1(c)). Importantly, it was experimentally verified that the dephasing time of the dark plasmon mode is extended with decreasing the detuning degree. It was also confirmed that the dephasing time of all the stacked nanogap Au structures from is longer than 3.5 fs for the single layer Au nanoblocks. Namely, the dephasing time of the dark plasmon mode is extended 3-fold as compared with that of the plasmon mode of the Au nanoblocks at maximum in the measured structures.

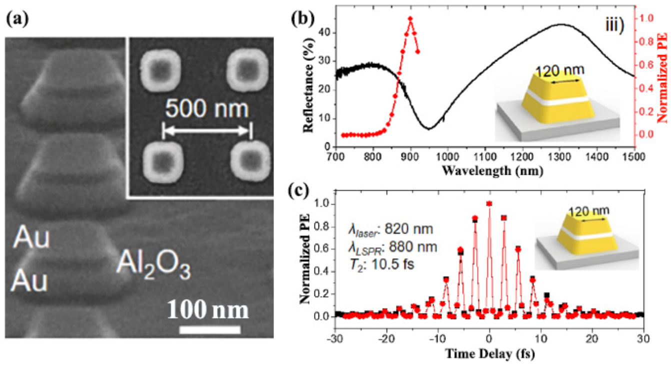

Fig. 1 (a) SEM images of the stacked Au nanostructures. (b) Reflection spectrum and near-field spectrum of the stacked structures. (c) Time-resolved autocorrelation photoemission curve to obtain the dephasing time.

4. その他・特記事項（Others）

N/A

5. 論文・学会発表（Publication/Presentation）

1) K. Ueno, J. Yang, et al., Appl. Mater. Today, 14, 159-165 (2010).
2) K. Ueno *et al.*, OSA Advanced Photonics Congress 2018, July 4, 2018, Zurich, Switzerland,
3) K. Ueno, J. Yang, R. Tatsumi, *et al.,* 第 66 回応用物理学会春季学術演講会, March 9, 2019, Tokyo, Japan.

6. 関連特許（Patent）

N/A