Emission enhancement in MoS_2 flakes through coupling with plasmonic resonance visualized by cathodoluminescence

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I. Introduction

Transition metal dichalcogenides (TMDs) such as molybdenum disulfide (MoS_2) have unique optical properties such as the strong luminescence and the tunable bandgap. However, the small interaction distance is a limitation for their applications[1]. In this study, we report a significant enhancement of A- and B-exciton emissions in MoS_2 flakes by coupling with plasmonic Au nano-pyramid arrays fabricated by the colloidal lithography technique. We experimentally map local emission enhancement from the structure with a nanometer spatial resolution by cathodoluminescence (CL) technique. A strongly enhanced emission intensity within the MoS_2-Au pyramids was achieved due to the localized surface plasmon resonance confined in nano-pyramids and its interaction with the periodicity-based resonance of array structure, promising for applications in optoelectronic and nano-photonic devices.

II. Fabrication and Measurement

We use a JEM-2100F scanning transmission electron microscope (STEM) combined with a light detection system. The specimen is placed inside a parabolic mirror that collimates the light emitted from the specimen (Fig.1a). While the electron beam scans over the specimen, the CL signals generated through optical excitation by fast electrons are collected at each electron beam position to obtain a CL photon map[2].

We fabricate Au hexagonal nano-pyramid arrays by a combination of self-ensembly and colloidal lithography technique using 500 nm polystyrene colloids as nano-masks. To produce a combined structure with MoS_2 flakes, MoS_2 solution was drop-casted on top of the Au pyramid arrays. The final structure is illustrated in the inset of Fig.1d.

III. Results and discussion

The back-scattered electron image (BEI) and bright-field STEM image shown in Fig.1b and c confirm the combined structure of the MoS_2 nanoflake and the Au pyramid array. As shown in Fig.1d, a pristine Au pyramid array has a broad peak around 2 eV and the peak of a bare MoS_2 lies at 1.92 eV between A- and B- exciton emissions. The CL peak intensity of MoS_2-Au pyramid is significantly higher than that from the bare MoS_2, the pristine Au pyramid, or their sum intensity, which can be attributed to the optical coupling of the MoS_2 flake with Au pyramids. Importantly, the CL map in Fig.1e at the photon energy of 1.92 eV captured the field enhancement in the MoS_2 flake, which is distinct at the plasmonic Au pyramids. Thus, we have successfully shown the enhanced exciton emissions in MoS_2 flakes by coupling with plasmonic resonance, promoting applications in future nano-photonic devices.

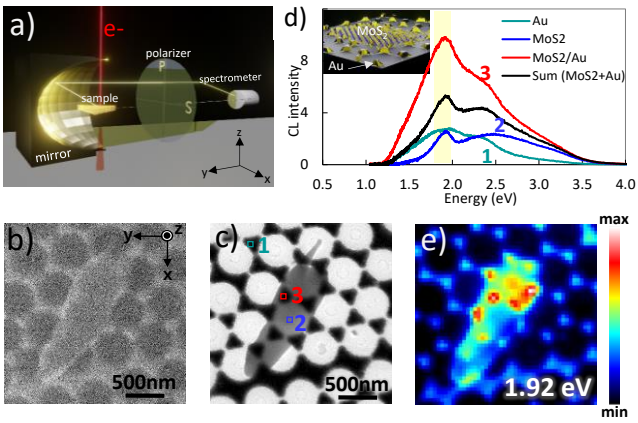

Fig.1: a) Schematic representation of cathodoluminescence setup. b) and c) BE image and bright-filed STEM image of MoS_2-Au pyramid structure. d) CL spectra of Au pyramid, bare MoS_2 and MoS_2-Au pyramid marked at positions 1, 2 and 3 shown in (c), respectively and sum curve of individual Au pyramid and bare MoS_2. e) CL photon maps corresponding to BEI and STEM images in (b) and (c) obtained at 1.92eV which is at the enhanced emission peak highlighted in yellow range in (d).

References:

1. Y. Dai et al., "Broadband Plasmon-Enhanced Four-Wave Mixing in Monolayer MoS2," Nano Lett. **21**(14), 6321–6327 (2021).
2. T. Sannomiya et al., "Coupling of plasmonic nanopore pairs: Facing dipoles attract each other," Light Sci. Appl. **5**(9), 1–7, Nature Publishing Group (2016).