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## **Electronic Supplementary Information**

## Gap Surface Plasmon-Enhanced Photoluminescence from Upconversion Nanoparticle-Sensitized Perovskite Quantum Dots in Metal-Insulator-Metal Configuration under NIR Excitation

Minju Kim<sup>a</sup>, Youngji Kim<sup>a</sup>, Kiheung Kim<sup>a</sup>, Wen-Tse Huang<sup>b</sup>, Ru-Shi Liu<sup>b</sup>\*, Jerome K. Hyun<sup>a</sup>\* and Dong Ha Kim<sup>a,c,d</sup>\*

<sup>a</sup>Department of Chemistry and Nano Science, Ewha Womans University, 52, Ewhayeodae-gil, Seodaemun-gu, Seoul 03760, Republic of Korea <sup>b</sup>Department of Chemistry, National Taiwan University, Taipei 10617, Taiwan <sup>c</sup>Basic Sciences Research Institute, Ewha Womans University, 52, Ewhayeodae-gil, Seodaemun-gu, Seoul 03760, Republic of Korea <sup>d</sup>Nanobio·Energy Materials Center (National Research Facilities and Equipment Center), Ewha Womans University, 52, Ewhayeodae-gil, Seodaemun-gu, Seoul 03760, Republic of Korea

\*Corresponding author: Ru-Shi Liu, Jerome K. Hyun, Dong Ha Kim E-mail address: <u>rsliu@ntu.edu.tw, kadam.hyun@ewha.ac.kr</u>, <u>dhkim@ewha.ac.kr</u>



**Fig. S1** (a) TEM image of CsPbBr<sub>3</sub> PeQDs. (d) XRD pattern of the pre-synthesized PeQDs and corresponding standard card JCPDS: 54-0752. (c) PL spectrum of the pre-synthesized PeQDs (1mg mL<sup>-1</sup> in cyclohexane) and a digital photograph taken under 365 nm UV irradiation (inset). (d) Absorption spectrum of the pre-synthesized PeQDs (1mg mL<sup>-1</sup> in cyclohexane).



**Fig. S2** (a) TEM image of  $\beta$ -NaYF<sub>4</sub>:25% Yb<sup>3+</sup>, 0.3% Tm<sup>3+</sup> UCNPs. (b) XRD pattern of the pre-synthesized UCNPs and the corresponding standard card JCPDS: 16-0334. (c) UCL spectra of the pre-synthesized UCNPs (1mg mL<sup>-1</sup> in cyclohexane) and a digital photograph taken under 980 nm excitation (inset).



**Fig. S3** (a) PL spectrum of the UCNPs/PeQDs composite in cyclohexane (UCNPs: 1.6 mg  $mL^{-1}$  and PeQDs: 5 mg  $mL^{-1}$ ) under 365 nm UV irradiation. (b) Digital photograph taken under 980 nm excitation.



**Fig. S4** (a) Schematic illustration of  $PS_{60K}$ -*b*-P2VP<sub>60K</sub> capped AuNR. (b) TEM image of the pre-synthesized  $PS_{60K}$ -*b*-P2VP<sub>60K</sub> capped AuNRs. (c) Absorption spectra of the CTAB capped AuNRs (black) in water and  $PS_{60K}$ -*b*-P2VP<sub>60K</sub> capped AuNRs (red) in ethyl acetate. (d) FT-IR spectrum of the pre-synthesized  $PS_{60K}$ -*b*-P2VP<sub>60K</sub> capped AuNRs.



Fig. S5. High-magnification TEM image of AuNRs encapsulated by the  $PS_{60K}$ -*b*-P2VP<sub>60K</sub> ligand.



**Fig. S6** (a) AFM image and (b) depth profile of the UP layer on the Ag film ( $R_q = 3.59$  nm). (c) Top-down SEM image of the UP layer on the Ag film.



**Fig. S7** Enlarged top-down SEM images of the MUPM configuration (different regions), showing the presence of (a) 33 and (b) 29 AuNRs resting on the UP layer.



Fig. S8 Absorption spectra of the (a) MUPG and UPG and (b) MUPM and UPM.

	MUPM	MUPG	UPG	UPM
PLQY	45.7	34.8	33.9	4.5
[%] <sup>a)</sup>				

**Table S1.** PLQYs of the four configurations (MUPM, MUPG, UPG, and UPM) under 365 nmUV irradiation.

<sup>a)</sup> PLQY is the ratio between the emission intensity of the configuration and the difference between the intensity of excited light in blank and the intensity of excited light in the configuration.



**Fig. S9** (a) Schematic of different configurations: MUM (AuNRs-UCNPs-Ag film), MUG (AuNRs-UCNPs-glass), UG (UCNPs-glass), UM (UCNPs-Ag film). (b) UCL spectra of the different configurations under 980 nm excitation. (d) Emission enhancement factors from the MUM, MUG, and UM configurations with respect to the UG configuration.



**Fig. S10** (a) PL spectra under 365 nm UV irradiation and (b) UCL spectra under 980 nm excitation (45.54 W/cm<sup>2</sup>) of different plasmonic configurations: UP layer on AuNRs layer (red), single UCNPs/PeQDs/AuNRs layer (blue), and UPG (black).



Fig. S11 The simulated field distributions of the (a) electric field component,  $E_x$  and (b) magnetic field component,  $H_y$  for the MUPM configuration.



Fig. S12 The simulated field distributions of the (a) electric field component,  $E_x$  and (b) magnetic field component,  $H_y$  for the MUPG configuration.



Fig. S13 (a) AFM image and (b) depth profile of the PVP spacer on the Ag film ( $R_q = 0.46$  nm).



**Fig. S14** (a) PL spectra under 365 nm UV irradiation and (b) UCL spectra under 980 nm excitation of the different configurations: MUPM, MUPG, UPG, and UPM. (c) Emission enhancement factors from the MUPM, MUPG, and UPM configurations with respect to the UPG. The PVP spacer was inserted between the Ag film and the UP layer.



**Fig. S15** The simulated electric field distributions of the MUPM configuration with more AuNRs distributed in the top metallic layer.