The band edge emission enhancement by the quadrupole surface plasmon-exciton coupling using direct-contact Ag/ZnO nanospheres

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S1. The absorption, scattering, and overall extinction cross sections of the Ag NB/ZnO HNS composite structure



Fig. S1. FDTD theoretical calculations of (a) absorption, scattering, and total extinction cross sections of Ag NB/ZnO HNS (the inset shows the structure diagram of the Ag NB/ZnO HNS); (b) the relationship between the SPR frequency and the etching durations.

With considering the influence of the dielectric environment, the absorption, scattering, and overall extinction cross sections of a Ag NB/ZnO HNS composite have also been stimulated as shown in the Fig. S1. A similar spectrum profile as that in a single Ag NB with the same size is obtained as shown in Fig. S1a, except for the little shifts in relevant peak positions due to the influence of the ZnO dielectric environment and the unresolved peak in UV region caused by the overlap of the ZnO BE absorption and the extinction of the Ag NB. Similar as that in an individual Ag NB, the dipole mode SPR frequencies in the Ag NB/ZnO HNS composite also undergo the red-shift with the NB's size increasing as shown in Fig. S1b. Compared with the dipole mode SPR, the quadrupole mode SPR displayed as an unresolved peak in UV region by overlapping with the ZnO BE absorption exhibits insensitivity with the Ag NB's size increasent.

S2. Near field distributions of a single Ag NB and the Ag NB/ZnO HNS composite



Fig. S2. FDTD simulation of the near-field distributions in (a) a single Ag NB normally irradiated with the light at 375 nm (3.31 eV) and the Ag NB/ZnO HNS composite (b) normally irradiated with the light at 375 nm (3.31 eV), (c) irradiated with the light by a 45° angle to the substrate at 375 nm (3.31 eV), and (d) normally irradiated with the incident light at 508 nm (2.44 eV). The sizes of the Ag NB and ZnO HNS are 146 nm and 258 nm in diameter as the same with the fabricated sizes in the experiments.

Near filed distributions of a single Ag NB and the Ag NB/ZnO HNS composite on silicon substrate were stimulated using the FDTD method under the light irradiation at different wavelengths and by different incident angles as shown in Fig. S2. In a single Ag NB, four-lobe field distribution pattern is distinguishable and can evidently prove the presence of the quadrupole mode SPR as shown in Fig. S2a. While in the Ag NB/ZnO HNS composite on the Si substrate, which is consistent with the practical structure in the experimental work, the four-lobe field distribution pattern shows a re-distribution with the intensive electric filed locating at the contact area between the Ag NB and ZnO HNS as shown in Fig. S2b. It is the strong evidence that the enhanced BE emission originates from the effective coupling between the quadrupole mode SPs of Ag NBs and excions in ZnO. In order to elucidate the influence of the incident light's angle and polarization on the near field distribution, a typical incident angle of 45° was applied in the simulation for a comparison as shown in Fig. S2c. It can be found that there is still a localized electric field at the contact area between the Ag NB and ZnO HNS, but with less intensity as that in the Ag NB/ZnO HNS composite irradiated with the normal incident light shown in Fig. S2b. Moreover, as shown in above Fig. S2 (c and d), the electric filed induced by the quadrupole mode SPR is much stronger at the contact area than that induced by the dipole mode SPR, which is well agreed with the high localization character of the quadrupole mode SPR.