SUPPORTING INFORMATION

Enhanced waveguide-type ultraviolet electroluminescence from ZnO/MgZnO core/shell nanorod array light-emitting diodes via coupling with Ag nanoparticles localized surface plasmons

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Optimization of the deposition conditions of Ag-NPs

The Ag-NPs were deposited on the CS-NRs' surface by a simple sputtering method. The extinction spectra of Ag-NPs, as well as their size and density, can be modulated by the deposition condition, such as sputtering power (which is usually described by the sputtering current, I_s), deposition pressure of Ar gas (P_{Ar}), and deposition time (t_d). Three series of Ag-NPs samples were prepared at different I_s, P_{Ar}, and t_d, and their extinction spectra were measured (see Figure S1 below). In general, it is observed that with increasing I_s, t_d and decreasing P_{Ar}, the extinction peak of Ag-NPs gradually red-shifts from ~430 to ~620 nm and considerably broadens in the long wavelength region. These changes can be attributed to (1) the increase in the average size of Ag-

NPs, and (2) dipole-dipole interaction between the neighboring Ag-NPs due to the increased density. An "anomalous" phenomenon noted in Figure S1(c) is that the extinction peak tends to shift towards higher energy when t_d>60s. Possible reason is that a long enough deposition time makes most of Ag-NPs form a continuous film, and its LSP effect is weakened. The blue-shifted extinction peak may result from some discrete, small-sized Ag-NPs. For the fabrication of LSP-enhanced LED devices, the deposition conditions of Ag-NPs are set at I_s=6 mA, P_{Ar}=20 Pa, and t_d=30 s to make the LSP resonance wavelength be as short as possible.

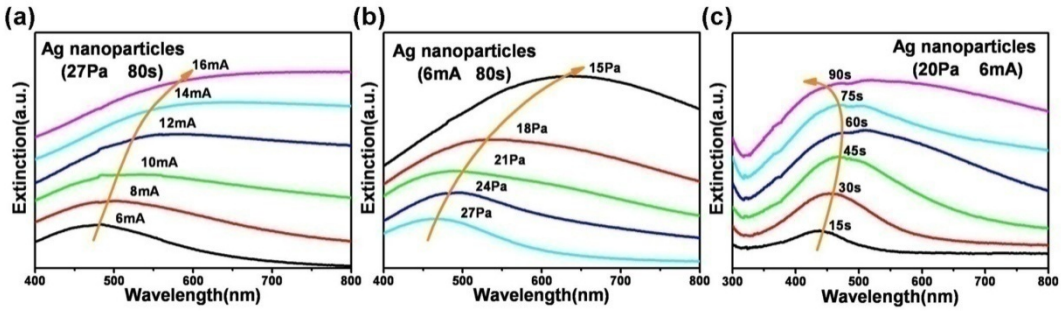

Figure S1 Extinction spectra of three series of Ag-NPs samples deposited at different conditions. Series I: I_s=6-16 mA, P_{Ar}=27 Pa, t_d=80 s; Series II: I_s=6 mA, P_{Ar}=15-27 Pa, t_d=80 s; Series III: I_s=6 mA, P_{Ar}=20 Pa, t_d=15-90s.

Calculation of the penetration depth of Ag-LSP fringing field into MgZnO

The penetration depth (Z) of Ag-LSP fringing field can be estimated from $Z = \frac{\lambda}{2\pi}\left|\frac{\varepsilon_{MgZnO}+\varepsilon_{Ag}}{-\varepsilon_{MgZnO}^{2}}\right|^{\frac{1}{2}}$, where ε_{MgZnO} and ε_{Ag} are the real part of the dielectric constants for MgZnO and Ag (-6.5), and λ is the wavelength of the LSP resonance. [1-3] Considering that no dielectric constant is available for MgZnO alloy, we assume that it follows a linear relationship $\varepsilon_{Mg_xZn_{1-x}O} = x\varepsilon_{MgO} + (1-x)\varepsilon_{ZnO}$, where ε_{MgO} and ε_{ZnO} are the dielectric constants of MgO (9.7) and ZnO (8.4), and x is the molar concentration of Mg element (in our case, $x = 0.2$). Based on the

above data, the penetration depth (Z) of Ag LSPs in $Mg_{0.2}Zn_{0.8}O$ is ~30-50 nm in the wavelength range of ~380-700 nm.

Optimization of MgZnO spacer-layer thickness

The coupling with LSPs is a near-field interaction, which strongly depends on the distance. In the present experiment, MgZnO spacer-layer thickness is optimized by the variation of ZnO PL intensity with MgZnO thickness. The PL spectra of the Ag-NPs decorated ZnO NRAs without MgZnO coating layer and with different thicknesses of MgZnO layer were measured and shown in Figure S2 below. Even if there is no MgZnO layer and the Ag-NPs directly contact with the ZnO NR's surface, the UV emission can also be enhanced by ~2 times. It is known that the introduction of a dielectric spacer layer can suppress the undesired charge transfer and nonradiative Förster resonant energy transfer between Ag and ZnO, and thus results in further luminescence enhancement. As the MgZnO spacer-layer thickness increases, ZnO UV emission intensity gradually increases, and a maximum enhancement is obtained at the MgZnO thickness of ~15 nm. Further increasing its thickness leads to the reduction of luminescence enhancement due to the evanescent wave nature of LSPs. Such a variation trend is also illustrated in the inset of Figure S2. The above results suggest that there is an optimized MgZnO thickness of ~15 nm, which is used in our LED device design.

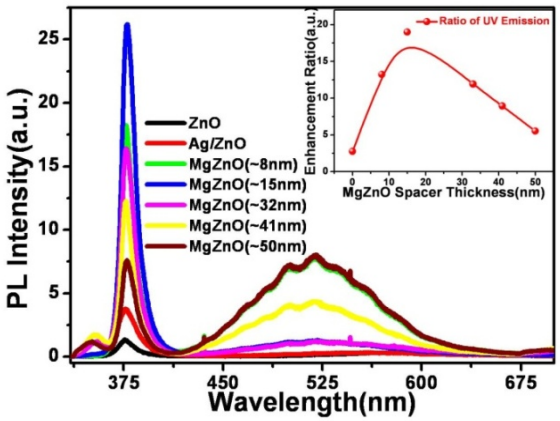

Figure S2 PL spectra of Ag-NPs decorated ZnO NRAs with different thicknesses of MgZnO spacer layer. The inset shows the variations of UV emission enhancement ratio with the MgZnO thickness.

Influence of the leakage current on EL intensity comparison

A non-negligible factor is that there is undesirable leakage current through the LED devices. Thus, the forward injection current I can be divided into two parts, one is the effective current contributing to the EL process (I_{eff}), and the other is the leakage current (I_{leak}). For both devices with and without Ag-NPs, their forward injection currents can be expressed as:

$$I^* = I^*_{eff} + I^*_{leak} \qquad \text{(with Ag-NPs)}$$

$$I = I_{eff} + I_{leak} \qquad \text{(without Ag-NPs)}$$

The *I-V* curves in Figure 3(c) show that the LED device with Ag-NPs has larger leakage current than the one without Ag-NPs (that is, $I^*_{leak} > I_{leak}$). In our experiments, the comparison of EL intensity between the two LEDs was performed at the same injection current. When the injection current is the same for both devices ($I^* = I$), the effective current of the Ag-decorated device will be smaller than that of the undecorated one ($I^*_{eff} < I_{eff}$) because of $I^*_{leak} > I_{leak}$. It means that, even at relatively low effective current I^*_{eff}, the device with Ag-NPs decoration shows obvious EL enhancement compared to the one without Ag-NPs. Thus, the comparison of EL

intensity is meaningful, though the devices have non-negligible leakage current.

Optical absorption and PL spectra of the ZnO NRA

Figure S3 below shows the optical absorption and PL spectra of the ZnO NRA, and their spectral overlap suggests a possibility of self-absorption during the EL propagation along the NR waveguide.

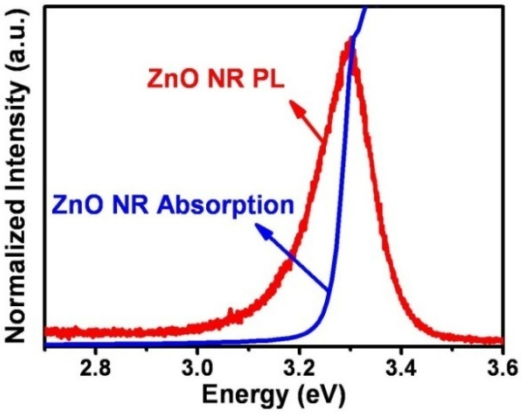

Figure S3 Optical absorption (blue line) and PL spectra (red line) of ZnO NRAs.

References

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