Electronic Supplementary Information

Titanium Oxide Nanotube Arrays for High Light Extraction Efficiency of GaN-based Vertical Light-Emitting Diodes

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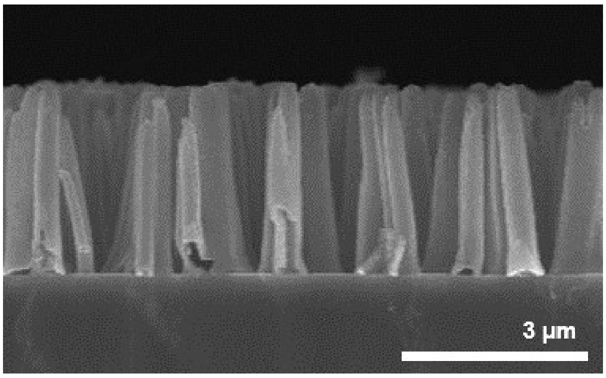

Fig. S1. Cross sectional view scanning electron microscopy (SEM) image of TiO_2 NTs on the n-GaN surface. Hollow regions are observed through the damaged bottom parts of TiO_2 NTs, which are formed when the sample was prepared for the SEM analysis.

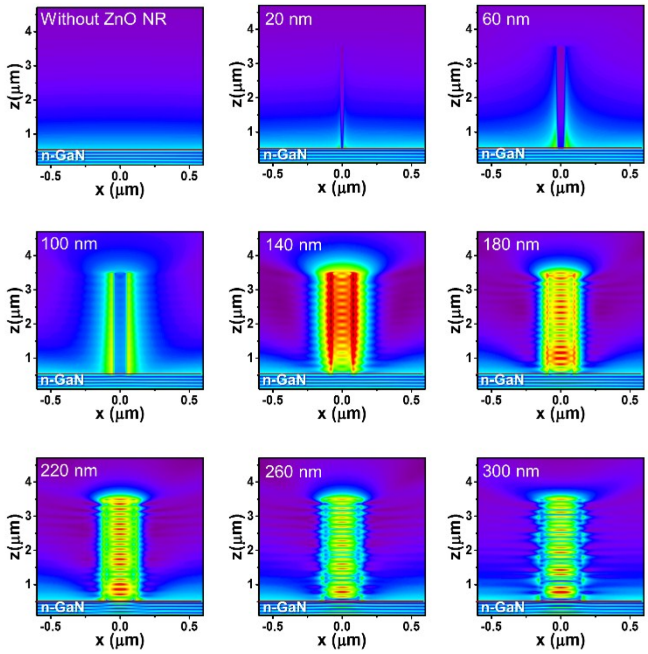

Fig. S2. Comparison of 3D FDTD simulation results for the electric field intensity of VLED for ZnO NRs with different diameters. Diameter of ZnO NR is increased from 20 nm to 300 nm.

As shown in Fig. S2, the electric field intensity at the flat end of the ZnO NR increases with increasing the diameter of NR up to 140 nm and the electric field intensity decreases thereafter until the diameter of 300 nm. The condition of light in the fundamental mode along the ZnO NR can be expressed in terms of the light wavelength λ and the NR diameter d as[1, 2]

$$\pi \frac{d}{\lambda}\left(n_{ZnO}^{2} - n_{air}^{2}\right)^{1/2} < 2.405 \qquad \text{(S1)}$$

where n_{ZnO} and n_{air} are the refractive indices of ZnO and air, respectively. It is known from eqn (S1) that the critical diameter of ZnO NR at the light wavelength of 440 nm is 194 nm. Because the amount of light in the fundamental mode guided outside as evanescent waves increases with decreasing the diameter of ZnO NR, the power fraction inside the ZnO NR sharply decreases below the critical diameter.[1] Thus, Fig. S2 shows that the intensity of electric field outside ZnO NRs is higher and broader as the diameter decreased below the diameter of 180 nm. Furthermore, the light at the interface between flat end of ZnO NR and air can be more efficiently extracted to air with decreasing the diameter of the NR because the transmittance at the flat end of ZnO NR abruptly increases as the diameter decreased below the critical diameter.[3] Although the light inside the ZnO NRs is more strongly extracted to air with decreasing the diameter, light injection from GaN layer to the ZnO NR is diminished due to the reduction of the effective contact area between the ZnO NR and GaN layers. As the ZnO NR diameter increased above critical diameter, light within the ZnO NR cannot be efficiently coupled to air due to high reflectance and low transmittance at the flat top surface of ZnO NR.[3] Therefore, these simulation results show that the diameter of the ZnO NRs should be maintained close to 140 nm to maximize the LEE of ZnO NR VLEDs.

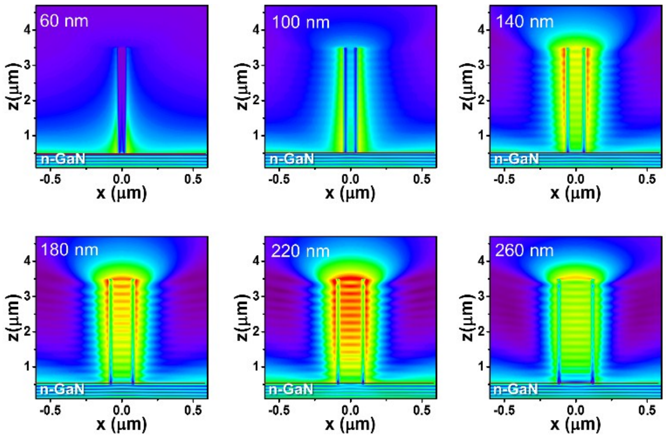

Fig. S3. Comparison of 3D FDTD simulation results for the electric field intensity of VLED for TiO_2 NTs with different diameters. Diameter of TiO_2 NT is increased from 60 nm to 260 nm and wall thickness is 20 nm. The intensity of the electric field at the end of the TiO_2 NT increases with increasing the NT diameter up to 220 nm and the electric field intensity is decreased at the diameter of 260 nm. Compared to ZnO NR with a diameter of 140 nm in Fig. S2, TiO_2 NT with a diameter of 220 nm depicted in Fig. S3 clearly illustrates that a relatively higher light extraction is observed. These results indicate that TiO_2 NTs grown on GaN-based VLEDs are more effective light-extracting nanostructures than ZnO NRs for high-efficiency VLEDs.

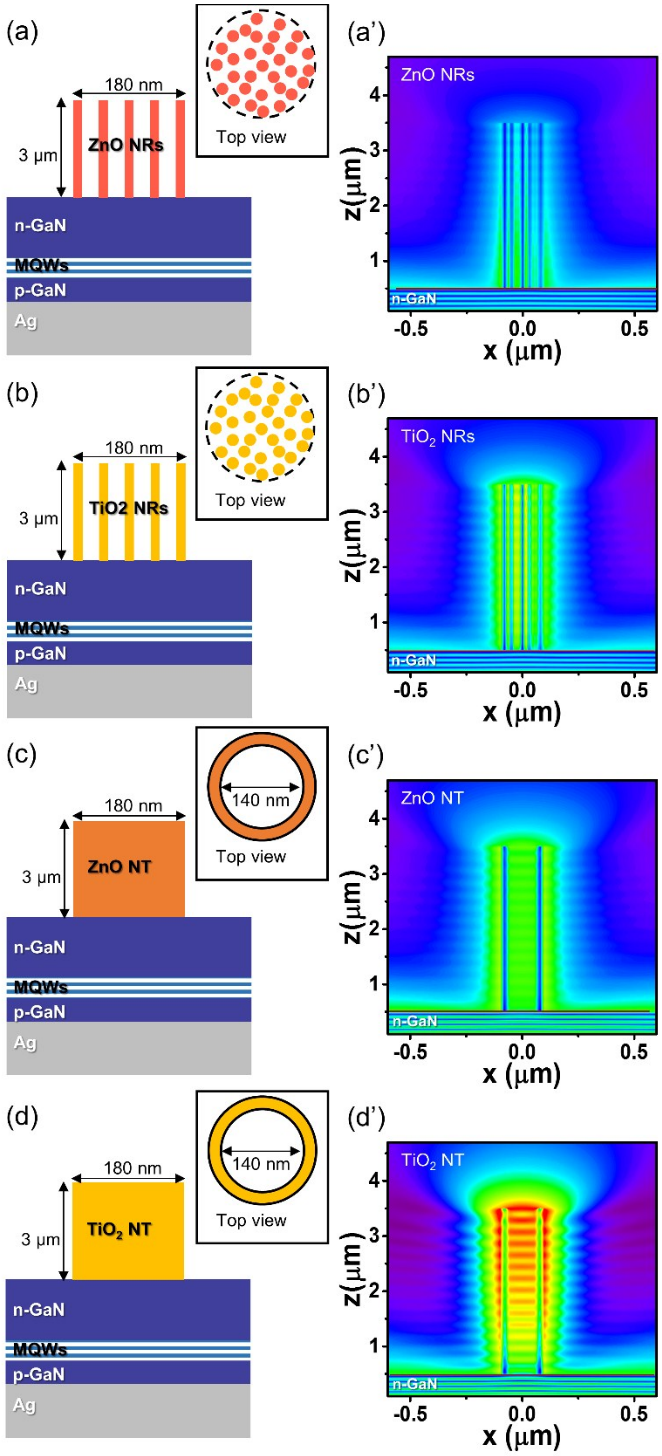

Fig. S4. Schematic diagrams of (a) ZnO NRs, (b) TiO_2 NRs, (c) ZnO NT, and (d) TiO_2 NT, respectively. The diameter of NR and wall thickness of NTs are 20 nm, respectively. The 3D FDTD simulation results for the electric field intensity of VLED are compared for (a') ZnO NRs, (b') TiO_2 NRs, (c') ZnO NT, and (d') TiO_2 NT, respectively. The total volume of the high density NRs is assumed to be same as that of a NT for a fair comparison.

Figs. S4a-d show geometries of simplified VLEDs with ZnO NRs, TiO_2 NRs, ZnO NT, and TiO_2 NT. As shown in Figs. S4a'-d', the intensity distribution of the electric field within each

VLED was simulated at a wavelength of 440 nm which corresponds to the EL emission peak at an injection current of 350 mA. For the VLEDs with TiO_2 NRs presented in the Figs. S4b', the intensity of the electric field above the apex of the TiO_2 NRs is higher than that above the ZnO NRs because the light generated from the active layers efficiently injects to TiO_2, compared to ZnO due to the absence of refractive index difference at the TiO_2/GaN interface. Compared to Fig. S4b', Fig. S4d' distinctly presents that a relatively higher intensity of the electric field is observed at the end the TiO_2 NT. This result implies that the light injection from the GaN epilayer to the TiO_2 NT is more efficient than the high density NRs, although the total area of the TiO_2/GaN interface of the NT configuration is equal to that of the high density NRs. Consequently, these results indicate that TiO_2 NT can more efficiently extract light from the GaN epilayer than three other surface nanostructures due to the refractive index matching and outstanding geometrical configuration for light extraction.

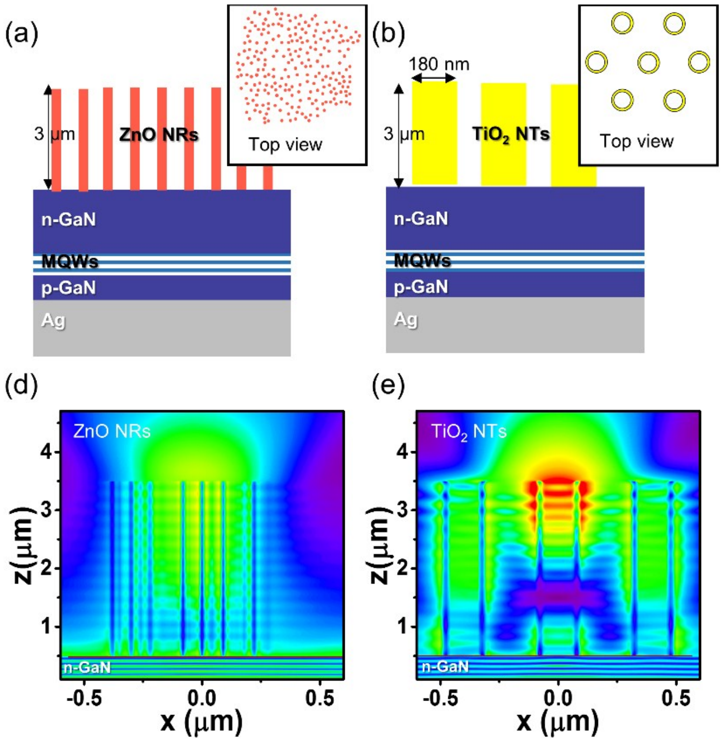

Fig. S5. Schematic diagrams of (a) ZnO NRs and (b) five TiO_2 NTs with wall thickness of 20 nm. A total of 160 ZnO NRs with a diameter of 20 nm are distributed on n-GaN layer, as shown in Fig. S5. Comparison of 3D FDTD simulation results for the electric field intensity of VLED for (c) ZnO NRs and (d) TiO_2 NT. The total volume of the high density ZnO NRs is assumed to be same as that of five TiO_2 NTs for a fair comparison. Interestingly, although a weak light intensity is detected within ZnO NRs with a diameter of 20 nm, which is too small to transfer electric field within ZnO NR, the light propagation is observed along the surfaces of ZnO NRs, resulting in an enhancement of light extraction above the ZnO NRs. However, these results indicate that TiO_2 NTs are more effective light-extracting configurations than high density ZnO NRs.

References

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