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Improved Light Output Power of GaN-Based Light Emitting Diodes by Enhancing Current Spreading Using Single-Wall Carbon Nanotubes

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Single-wall carbon nanotubes (SWCNTs) have been combined with indium tin oxide (ITO) to improve the output power of GaN-based light emitting diodes (LEDs). LEDs fabricated with the SWCNT/ITO contacts give a forward voltage of 3.61 V at 350 mA, which is slightly higher than that of LEDs with ITO-only contacts. The SWCNT/ITO and ITO-only contacts produce transmittance values of 91.5 and 94.4% at 460 nm, respectively. However, LEDs with SWCNTs show a higher output power by 60% at 20 mA compared to those without SWCNTs. Photoemission microscope analyses show that the well-dispersed SWCNT bundle efficiently serves as a current spreader.
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To realize high brightness GaN-based light emitting diodes (LEDs), the achievement of a high external quantum efficiency is indispensable. In conventional top-emission LEDs, the electrical property of p-type ohmic contacts affects the light output performance. Metal-based p-type ohmic contacts have been shown to produce low contact resistance. However, these metal-based schemes are semitransparent and, as a result, suffer from loss of light transmittance. To achieve a high optical transmittance, various transparent conducting oxides, such as indium tin oxide (ITO)- and ZnO-based oxides, have been investigated.[1,2] For example, ITO contacts on p-type $In_{0.1}Ga_{0.9}N$ (10 nm)/p-type GaN (p-GaN) and $In_{0.15}Ga_{0.85}N$ (10 nm)/p-GaN became ohmic with specific contact resistivities of 4.5×10^{-2} and 3.2×10^{-5} Ω cm^2 when annealed at 500 and 550°C, respectively.[3,4] ITO combined with thin interlayers of indium (10 nm), Cu-doped indium oxide (3 nm), Sn–Ag alloy (6 nm), Ag (1 nm), Ni (10 nm), and Au nanoparticles[1,5-9] resulted in specific contact resistances in the range of $\sim 1 \times 10^{-3}$ to $\sim 4 \times 10^{-4}$ Ω cm^2 when annealed at 500–630°C for 1–10 min in air. In addition, ZnO:Ga [10] contacts yielded ohmic behaviors with a typical specific contact resistance of 2.1×10^{-3} Ω cm^2 and gave a transmittance of $\sim$80% in the near UV and visible wavelength ranges even without annealing treatment. LEDs fabricated with the as-deposited ZnO:Ga p-contacts exhibited a light output nearly twice that of LEDs with oxidized Ni/Au contacts.[10] Indium oxide–doped ZnO (IZO) contacts exhibited transmittance values of 84–92% in the range of 400–600 nm and a specific contact resistance of 3.4×10^{-4} Ω cm^2 on p-GaN (3×10^{17} cm^{-3}) when annealed at 600°C for 5 min in a N_2 ambient.[11] The output power of LEDs with the IZO ohmic contacts was improved by 34% at an input power of 83 mW compared to that of LEDs with Ni/Au contacts. ZnO combined with a Ni interlayer (5 nm) gave a specific contact resistance of $\sim 1 \times 10^{-5}$ Ω cm^2 and a light transmittance of $\sim$76% in the range 400–550 nm when annealed at 450 and 550°C for 2 min in air.[2]

Single-wall carbon nanotubes (SWCNTs) have a superior electrical conductivity as well as good optical transparency characteristics.[12,13] Thus, SWCNTs have been used as a transparent conductor for various devices, e.g., field effect transistors[14,15] and organic LEDs.[16] In addition, homogeneous (mixed metallic and semiconducting) SWCNT films were applied as p-type ohmic contacts to GaN/InGaN quantum-well LEDs.[17] The 100 nm thick SWCNT film contacts produced a specific contact resistance of 1.1×10^{-2} Ω cm^2 upon annealing at 700°C for 60 s in N_2 gas ambient, which was somewhat better than that of N_2-annealed Ni/Au contacts. However, the SWCNT film gave a low transmittance of $\sim$60% at a wavelength of 434 nm. In this work, we also introduced SWCNTs at the interface between ITO p-contact and p-GaN to improve current spreading and, consequently, to enhance the output power performance of GaN-based LEDs. Among the various techniques for forming SWCNTs on substrates, such as drop-drying from solvent, airbrushing, Langmuir–Blodgett deposition, and filtering with a membrane, we used the most convenient, a simple dip coating technique, which enables SWCNTs to be also deposited on photoresist patterned LED structured samples. To effectively spread the current, individually dispersed SWCNTs (forming networks but not films) were employed, which could be controlled by dipping and rinsing processes.

InGaN/GaN multiple quantum-well LED structures ($\sim$460 nm in peak wavelength) were grown on *c*-face sapphire substrates by metallorganic chemical vapor deposition. The LED structure consisted of a 0.05 μm thick strained GaN layer, a 0.15 μm thick p-type GaN:Mg ($n_a = 5 \times 10^{18}$ cm^{-3}) layer, a 0.1 μm thick active layer, a 1.5 μm thick n-type GaN:Si ($n_d = 3 \times 10^{18}$ cm^{-3}) layer, and a 2.0 μm thick undoped GaN layer on the sapphire substrate. Before the fabrication of LEDs, the surfaces of the LED structure samples were ultrasonically degreased with acetone, methanol, deionized (DI) water, and a mixture of sulfuric acid (H_2SO_4) and hydrogen peroxide (H_2O_2) for 5 min in each step to remove organic contaminants. The LEDs (1050×1050 μm) were fabricated using photolithography patterning and inductive coupled plasma reactive ion etching. Before undergoing SWCNT deposition, the samples were treated with a diluted HCl solution, rinsed in DI water, and then blown dry with N_2. The SWCNTs grown via arc discharge were dispersed in dichlorobenzene ($C_6H_4Cl_2$, Sigma-Aldrich, 99%) through ultrasonication for 20 min at a dilute concentration of 0.5 mg/mL. The GaN samples defined by the standard photolithography technique were dipped in the SWCNT dispersed solution for 3 min and then rinsed in pure dichlorobenzene for 1 min. These processes were repeated three times to attach suitable amounts of SWCNTs on p-GaN, which were finally blown dry with N_2 gas (this process gave reproducible results at all times). ITO (220 nm thick) layers were sequentially deposited onto the samples with/without SWCNTs by radio-frequency magnetron sputtering. After the lift-off process, all the samples were rapid-thermal annealed at 600°C for 40 s in air. To characterize the SWCNT morphology on p-GaN, a field-emission-scanning electron microscope (Hitachi S-4300) was used. Current–

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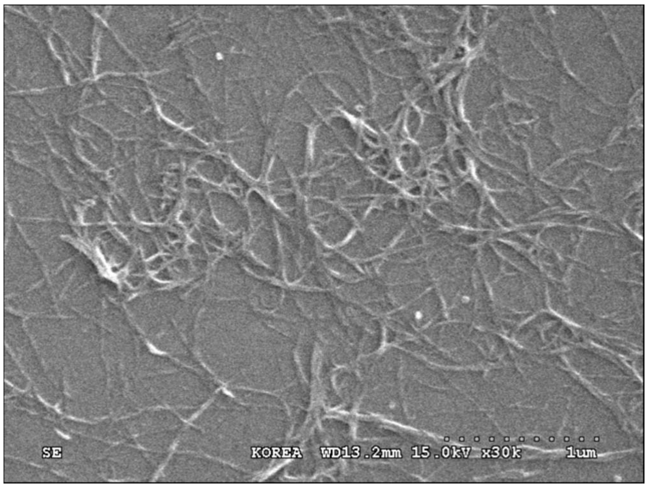

Figure 1. An SEM image of SWCNTs formed on p-GaN samples by a simple dipping in a SWCNT-suspended solution.

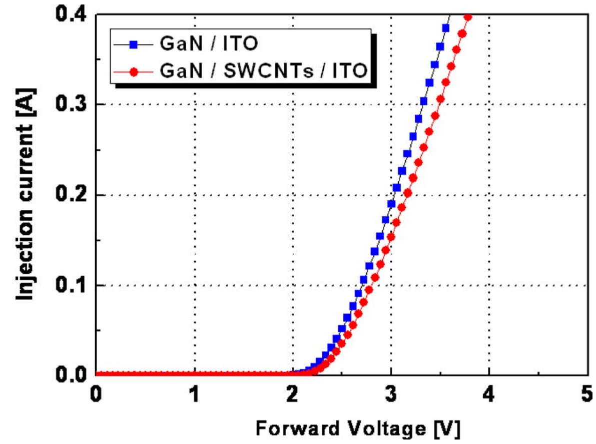

Figure 3. (Color online) The forward *I-V* characteristics of blue (460 nm) LEDs ($1050 \times 1050\ \mu m$) fabricated with ITO-only and SWCNT/ITO contacts, which were heat-treated at 600°C for 40 s in air.

voltage (*I-V*) data were measured at room temperature using a high current source-measuring unit (Keithley 238). The light output power was measured by a Newport dual channel power meter. Transmittance measurements were carried out using a UV/visible spectrophotometer (Shimadzu, UV-1800). The current-spreading features of LEDs with/without SWCNTs were characterized through a photoemission microscope (MoDooTek, PHEMOS-1000).

Figure 1 shows a scanning electron microscopy (SEM) image of SWCNTs formed on p-GaN samples by a simple dipping in a SWCNT-suspended solution. There are randomly distributed SWCNTs forming networks. The density of the SWCNTs was measured to be of the order of $10^9\ cm^{-2}$. Considering their thickness, they consist of SWCNTs and a SWCNT bundle (30–80 nm diameter), indicating that they have a mixture of semiconducting and metallic characteristics. Furthermore, the existence of excessively bundled SWCNTs (or films) between ITO and p-GaN caused the ITO electrode to peel off during heat-treatment (not shown). This implies that the optimization of the dipping process for forming SWCNT networks (having a density of $\sim 10^9\ cm^{-2}$) is essential for use as an electrode in GaN-based LEDs. The density could be controlled by monitoring the dipping and rinsing process.

Figure 2 exhibits the light transmittance of ITO-only and SWCNT/ITO layers on p-GaN samples, which were annealed at 600°C for 40 s in air. The SWCNT/ITO contacts give a little bit lower transmittance compared to the ITO-only contact across the whole wavelength region of 360–500 nm. For example, the transmittance at 460 nm was measured to be 91.4 and 94.4% for the SWCNT/ITO and ITO only contacts, respectively. Individually dispersed SWCNTs without bundled SWCNTs gave almost the same transmittance as that of ITO-only contacts (not shown). This indicates that the presence of the bundled SWCNTs plays a major role in absorbing visible light. So, the optimization of the dipping and repetition process for obtaining proper networks is important for the improvement of the output performance of LEDs.

Figure 3 shows the forward *I-V* characteristics of blue (460 nm) LEDs ($1050 \times 1050\ \mu m$) fabricated with ITO-only and SWCNT/ITO contacts, which were heat-treated at 600°C for 40 s in air. The LEDs fabricated with the SWCNT/ITO contacts give a forward-bias voltage of 3.61 V at an injection current of 350 mA, which is a bit higher than that (3.44 V) of the LEDs made with ITO-only contacts. The increase in the forward-bias voltage of the LEDs with SWCNTs may be explained in terms of the presence of SWCNTs in contact with p-GaN. The SWCNT/ITO contacts produce inhomogeneous interfaces having two different contacts on p-GaN, i.e., SWCNT/p-GaN and ITO/p-GaN. Since CNTs have a smaller work function (4.7 eV) than that of ITO (4.8–5.0 eV),[18-20] the SWCNTs could contribute to an increase in the surface barrier height, leading to an increase in the contact resistivity.

Figure 4 shows the current–light-output-power behaviors of the

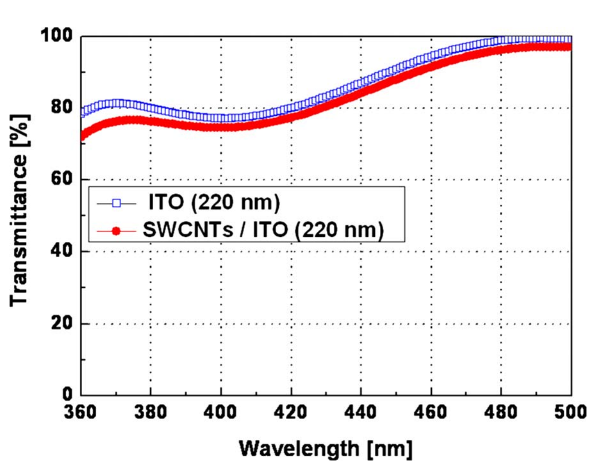

Figure 2. (Color online) The light transmittance of ITO-only and SWCNT/ITO layers on p-GaN samples, which were annealed at 600°C for 40 s in air.

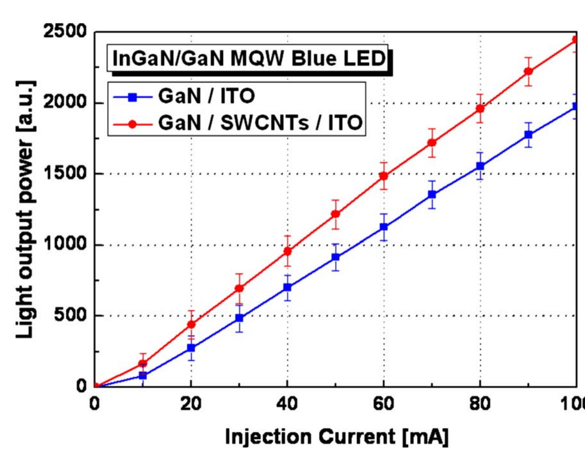

Figure 4. (Color online) The current–light-output-power behaviors of the LEDs fabricated with the ITO-only and SWCNT/ITO contact layers, which were annealed at 600°C for 40 s in air.

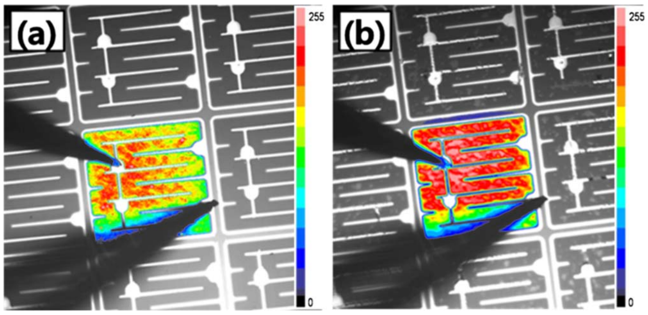

Figure 5. (Color online) The light-emitting patterns (under a forward bias of 4 μA) of the LEDs fabricated with the (a) ITO-only and (b) SWCNT/ITO contact layers, which were annealed at 600°C for 40 s in air.

LEDs fabricated with the ITO-only and SWCNT/ITO contact layers, which were annealed at 600°C for 40 s in air. It is evident that the LEDs with the SWCNT/ITO contacts produce a much higher output power compared to the LEDs with the ITO-only contacts. For example, the use of the SWCNT/ITO contacts results in the improvement of the light output power at 20 mA by 60% compared to that of the LEDs without the SWCNTs. This is inconsistent with the combined results of the light transmittance and the forward-bias voltage (Fig. 2 and 3, respectively). The discrepancy can be attributed to the fact that the SWCNT bundle having metallic electrical characteristics serves as an effective current spreader, conveying an electric charge from the ITO electrode to p-GaN,[13] as described below.

To compare the current-spreading phenomena in the LEDs (1050×1050 μm) fabricated with/without the SWCNTs, the emitted patterns of chips under a forward bias of 4 μA were characterized by a photoemission microscope. Figure 5 shows the light-emitting patterns of the LEDs fabricated with the ITO-only and SWCNT/ITO contact layers, which were annealed at 600°C for 40 s in air. For the LEDs with the ITO only contacts (Fig. 5a), the photoemission is nonuniform and localized. In other words, the light emission is crowded near the p-probe. As shown in Fig. 5b, however, the LEDs with the SWCNT/ITO contacts exhibit a uniform light emission across the whole chip area. This indicates that the SWCNTs incorporated into the ITO contacts play a critical role in spreading the injected current and serve as an efficient current spreader, leading to a much higher light output power.

To summarize, we investigated the effect of SWCNTs on the electrical and optical characteristics of high power GaN-based LEDs fabricated with ITO ($\sim$220 nm) contact layers. The presence of the SWCNTs at the ITO/p-GaN interface insignificantly degraded the light transmittance, whereas the LEDs made using the SWCNTs resulted in a bit higher forward-bias voltage compared to the LEDs without the SWCNTs. Despite the somewhat inferior properties, however, the LEDs with the SWCNT/ITO contacts produced a much higher output power by $\sim$60% at 20 mA compared to those with the ITO-only contacts. This indicates that the use of the SWCNTs could represent an effective process for fabricating high performance GaN-based LEDs.

Acknowledgments

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References

1. J.-O. Song, H.-G. Hong, J.-W. Jeon, J.-I. Sohn, J.-S. Jang, and T.-Y. Seong, *Electrochem. Solid-State Lett.*, **11**, H36 (2008).
2. J.-O. Song, K.-K. Kim, S.-J. Park, and T.-Y. Seong, *Appl. Phys. Lett.*, **83**, 479 (2003).
3. K.-M. Chang, J.-Y. Chu, and C.-C. Cheng, *Solid-State Electron.*, **49**, 1381 (2005).
4. J.-S. Jang and T.-Y. Seong, *J. Appl. Phys.*, **101**, 013711 (2007).
5. J.-O. Song, K.-K. Kim, H. Kim, H.-G. Hong, H. Na, and T.-Y. Seong, *Electrochem. Solid-State Lett.*, **10**, H270 (2007).
6. J.-O. Song, J. S. Kwak, Y. Park, and T.-Y. Seong, *Appl. Phys. Lett.*, **86**, 213505 (2005).
7. J.-O. Song, K.-K. Kim, H. Kim, Y.-H. Kim, H.-G. Hong, H. Na, and T.-Y. Seong, *Mater. Sci. Semicond. Process.*, **10**, 211 (2007).
8. R.-H. Horng, D.-S. Wuu, Y.-C. Lien, and W.-H. Lan, *Appl. Phys. Lett.*, **79**, 2925 (2001).
9. L.-C. Chen and S.-F. Lu, *Phys. Status Solidi A*, **203**, 2451 (2006).
10. K. Nakahara, K. Tamura, M. Sakai, D. Nakagawa, N. Ito, M. Sonobe, H. Takasu, H. Tampo, P. Fons, K. Matsubasa, et al., *Jpn. J. Appl. Phys., Part 2*, **43**, L180 (2004).
11. J.-H. Lim, D.-K. Hwang, H.-S. Kim, J.-Y. Oh, J.-H. Yang, R. Navamathavan, and S.-J. Park, *Appl. Phys. Lett.*, **85**, 6191 (2004).
12. Q. Cao, H.-S. Kim, N. Pimparkar, J. P. Kulkarni, C. Wang, M. Shim, K. Roy, M. A. Alam, and J. A. Rogers, *Nature (London)*, **454**, 495 (2008).
13. D. Hecht, L. Hu, and G. Grüner, *Appl. Phys. Lett.*, **89**, 133112 (2006).
14. R. Martel, T. Schmidt, H. R. Shea, T. Hertel, and Ph. Avouris, *Appl. Phys. Lett.*, **73**, 2447 (1998).
15. Z. Wu, Z. Chen, X. Du, J. M. Logan, J. Sippel, M. Nikolou, K. Kamaras, J. R. Reynolds, D. B. Tanner, A. F. Hebard, et al., *Science*, **305**, 1273 (2004).
16. D. Zhang, K. Ryu, X. Liu, E. Polikarpov, J. Ly, M. E. Tompson, and C. Zhou, *Nano Lett.*, **6**, 1880 (2006).
17. K. Lee, Z. Wu, Z. Chen, F. Ren, S. J. Pearton, and A. G. Rinzler, *Nano Lett.*, **4**, 911 (2004).
18. X. Cui, M. Freitag, R. Martel, L. Brus, and P. Avouris, *Nano Lett.*, **3**, 783 (2003).
19. W. Kim, A. Javey, R. Tu, J. Cao, Q. Wang, and H. Dai, *Appl. Phys. Lett.*, **87**, 173101 (2005).
20. T. Minami, *Semicond. Sci. Technol.*, **20**, S35 (2005).