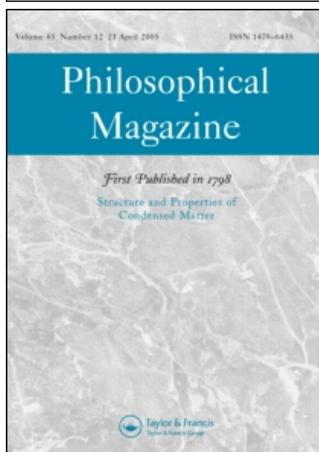


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High-brightness gallium nitride nanowire UV–blue light emitting diodes

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We report on high-brightness GaN nanowire UV–blue light emitting diodes (LEDs), which are fabricated by coupling of n-GaN nanowires and p-GaN substrates using two assembly methods, random dispersion (RD) and dielectrophoresis assisted assembly deposition (DAAD). These GaN nanowire LEDs have bright UV–blue emission (411–437 nm) from the n-GaN nanowire/p-GaN substrate junction and the light emission is strong enough to be observed with the naked eye even for a single GaN nanowire LED. The results reported here should have significant implications for the fabrication of highly efficient, low-cost UV–blue LEDs with low power consumption, as compared to conventional thin-film based GaN LEDs.

1. Introduction

Gallium nitride (GaN) nanowires are important building blocks for the fabrication of various optoelectronic as well as electronic devices of nanoscale dimensions [1–3]. They are highly suitable for light-emitting devices, e.g. light-emitting diodes (LED) and laser diodes (LD), due to their direct band gap structure and dislocation-free nature [2, 3]. A considerable bottleneck that previously prevented the widespread application of coloured nanowire LEDs has been the lack of high intensity LEDs and the lack of reproducible manipulation techniques to align and assemble the nanowires into well-defined arrays for highly integrated photonic devices [4, 5]. High-brightness blue LEDs are required to reproduce the full colour spectrum and achieve pure white light since blue is one of the three primary colours [6]. Recently, the hybrid heterojunction LEDs, which are based on the bottom-up assembly of nanowires (n-GaN nanowires) on top-down fabricated silicon structures (p-Si nanowires) from SOI (silicon–oxide–insulator) wafer, have been implemented as a new approach for introducing efficient photonic capabilities into integrated silicon electronics [5, 7]. However, these heterojunction LEDs have lower emission efficiency and brightness than homojunction devices (p-GaN/n-GaN) because an energy

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barrier formed at the heterojunction could decrease carrier injection efficiency [4, 5, 8, 9]. This problem can be potentially solved by fabricating homojunction devices and increasing the carrier injection efficiency with nanosized junctions. To meet this requirement, Kim *et al.* [10, 11] have shown that InGaN/GaN multi-quantum-well (MQW) nanorod LEDs exhibit enhanced light emission efficiency compared to thin-film based broad area LEDs and also suggested that the InGaN/GaN MQW nanorod LED is applicable to bright white LEDs. Despite their successful fabrication of a high-efficiency InGaN/GaN MQW nanorod LED, reproducible large-scale assembly techniques are still required for wafer-based large-scale integration.

To fabricate nanometre-scale devices with bottom-up techniques, a traditional approach is to start with random dispersion (RD) from a nanowire suspension, followed by electrode fabrication at known nanostructure location on the substrates [1–5]. In addition, several more advanced techniques have been reported for the manipulation of the nanostructures [12–14]. Among these techniques, the dielectrophoresis (DEP) process has become one of the most promising methods to align 1D nanostructures [12, 15–17]. Herein, we demonstrate high-brightness GaN nanowire UV–blue LEDs prepared using two assembly techniques, random dispersion (RD) and dielectrophoresis assisted assembly deposition (DAAD) [18]. Our approach to build up high-brightness GaN NW LEDs has two distinct features compared to the previous studies [5, 7, 10, 11]. First, our approach of combining dislocation-free GaN nanowires with the homojunction device structures produces high-brightness emission. Secondly, our approach for assembling the GaN nanowires using the DAAD technique is amenable for large-scale integration.

2. Experimental details

Gallium nitride nanowires were synthesized using a nickel (2 nm) catalyst deposited on c-plane sapphire wafer. Gallium (Ga) and nitrogen (N) components were supplied to the substrate by using metallic Ga and NH_3 gas. The system was heated to 900°C under a flow of NH_3 at a rate of $20\text{ cm}^3\text{ min}^{-1}$ for 6 h, and then cooled down to room temperature. The X-ray diffraction (XRD) pattern of the GaN nanowires was indexed to a wurtzite structure. Current–voltage (I – V) measurements were performed on different LED structures using a semiconductor parameter analyzer (HP 4156A) in the range of 20 fA–100 mA at room temperature. For LED structures, the p–n junction diodes were fabricated by coupling the n-GaN nanowires together with p-GaN substrate. First, the 200 nm thick oxide was deposited on the p-GaN substrate ($0.8 \times 0.8\text{ cm}^2$, 1 μm thick, see figure 1), which were prepared on the c-plane sapphire substrates using a metal–organic chemical vapour deposition (MOCVD). The resistivity, the mobility and the carrier concentration of the p-GaN substrate were determined from Hall measurements to be $3.28\ \Omega\text{ cm}$, $11.67\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ and $1.6 \times 10^{17}\text{ cm}^{-3}$, respectively. The circular-shaped anode contacts (Ni/Au = 30/150 nm, 100 μm diameter) were defined on p-GaN substrates after etching the oxide by dipping in diluted hydrofluoric (BHF) acid for 5 min. On the other hand, the doughnut-shaped cathode metal electrodes (Ni/Au = 30/150 nm, 500 μm

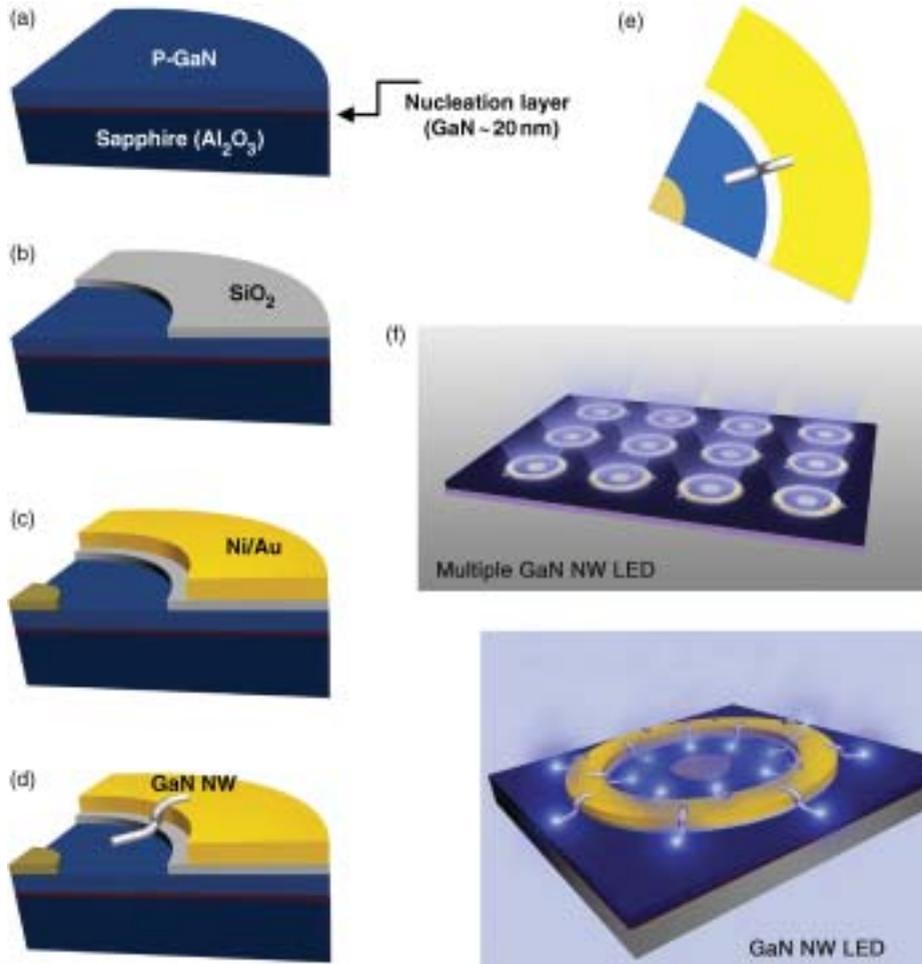


Figure 1. Schematic diagram of the process (steps a–d) for the fabrication of the homojunction GaN nanowire LED (n-GaN nanowire/p-GaN substrate) structure formed by dielectrophoresis assisted assembly deposition (DAAD) and random dispersion (RD) on the p-GaN/sapphire substrate. (a) p-GaN/GaN nucleation layer structures on sapphire substrate. (b) Plasma-enhanced chemical vapour deposition (PECVD) of the oxide (SiO_2) on p-GaN/n-GaN nucleation layer/Sapphire substrate. (c) Patterning of the anode and cathode metal (Ni/Au = 30/150 nm), and annealing of the anode metal at 650–750°C for 30 s. (d) Dispersion of n-GaN nanowires by two deposition methods, RD and DAAD. (e) Top view of the DAAD prepared GaN nanowire LED. (f) 3-dimensional (3-D) illustration of the homojunction GaN nanowire LED structures on large-scale wafer using DAAD technique and single homojunction GaN nanowire LED structure. For DAAD experiment, we used an electric field of $15 \text{ V}_{\text{p-p}}$ at a frequency of 10 kHz.

diameter) for contact to n-type GaN nanowires were patterned on the oxide by the standard photolithography process. All of the samples were annealed at temperatures of 650–750°C for 30 s to achieve ohmic contacts to GaN. One sample ($0.8 \times 0.8 \text{ cm}^2$) consists of 25 doughnut-shaped p–n junction LED structures, whose

geometry includes a 100 μm gap between two electrodes (anode and cathode). The p–n junction (n-GaN nanowire/p-GaN substrate) was obtained by two different ways, random dispersion (RD) and dielectrophoresis assisted assembly deposition (DAAD). A continuous-wave He–Cd laser (Kimmon) at 325 nm was used for photoluminescence (PL) studies at room temperature. Electroluminescence (EL) was investigated with fibre optic spectrometer (SD 2000, Ocean optics, Inc.). EL images were taken with a charged coupled device (CCD) camera built in microscopes (I-pinnacle Co) as well as digital camera (Canon Power Shot G2).

3. Results and discussion

Figures 1a–e show the fabrication process for our homojunction GaN nanowire LED structures which are formed by assembling n-GaN nanowires on a patterned p-GaN substrate by means of random dispersion (RD) and dielectrophoresis assisted assembly deposition (DAAD). Three-dimensional illustrations of the homojunction GaN nanowire LED structures prepared by the DAAD are shown in figure 1f.

Photoluminescence (PL) spectra of the homojunction GaN nanowire LED structures (n-GaN nanowire/p-GaN substrate) together with PL of the as-made GaN nanowires on sapphire substrate are shown in figure 2a. The measurements were performed with a He–Cd laser (325 nm) as an excitation source at room temperature. The band edge emission at 3.38 eV (367 nm) with a narrow full width half maximum (FWHM) of 18 nm was observed in both as-made n-GaN nanowires and GaN nanowire LED structures while Mg-doped p-GaN blue emission at 2.83 eV (438 nm) was also observed in GaN nanowire LED structures. The 438 nm emission is attributed to a radiative recombination related to Mg acceptors in p-GaN [6].

Current–voltage (I – V) measurements (figure 2b) show well-defined current rectifying behaviour, as expected from homojunction p–n diodes with a turn-on voltage of ~ 3.4 V. Little leakage current and no breakdown were observed for reverse bias up to -25 V. The observed reverse leakage current was $\sim 5 \times 10^{-4}$ A at 25 V of reverse bias voltage at room temperature. To illustrate the reproducibility of the n-GaN nanowire/p-GaN substrate junction LED structures, we have fabricated 25 samples under the same conditions of the fabrication processes and found that all of the junctions exhibit similar rectifying behaviour and clearly function with low leakage current, as shown in figure 2b. The on-series resistances (R_S) for these random dispersion (RD) prepared p–n junction LEDs were determined to range from 20 k Ω to 25 k Ω from the inverse of the slope in the I – V curve in figure 2b. This value is relatively high compared to the thin-film based GaN LED [6, 19]. This high on-series resistance can be attributed to large resistivity of metal contacts with the n-GaN nanowire and p-GaN substrate.

Significantly, electroluminescence (EL) studies on n-GaN nanowires/p-GaN substrate junctions demonstrate that the homojunction GaN nanowire LED structures exhibit UV–blue light emission in forward bias voltage (up to 40 V). Figure 3a shows that the EL spectrum comprised a much broader distribution (centred at 420 nm with a FWHM of 58 nm at the drive voltage of 20 V) than the emission from thin film based GaN p–n diodes [6, 19]. The EL data for the single

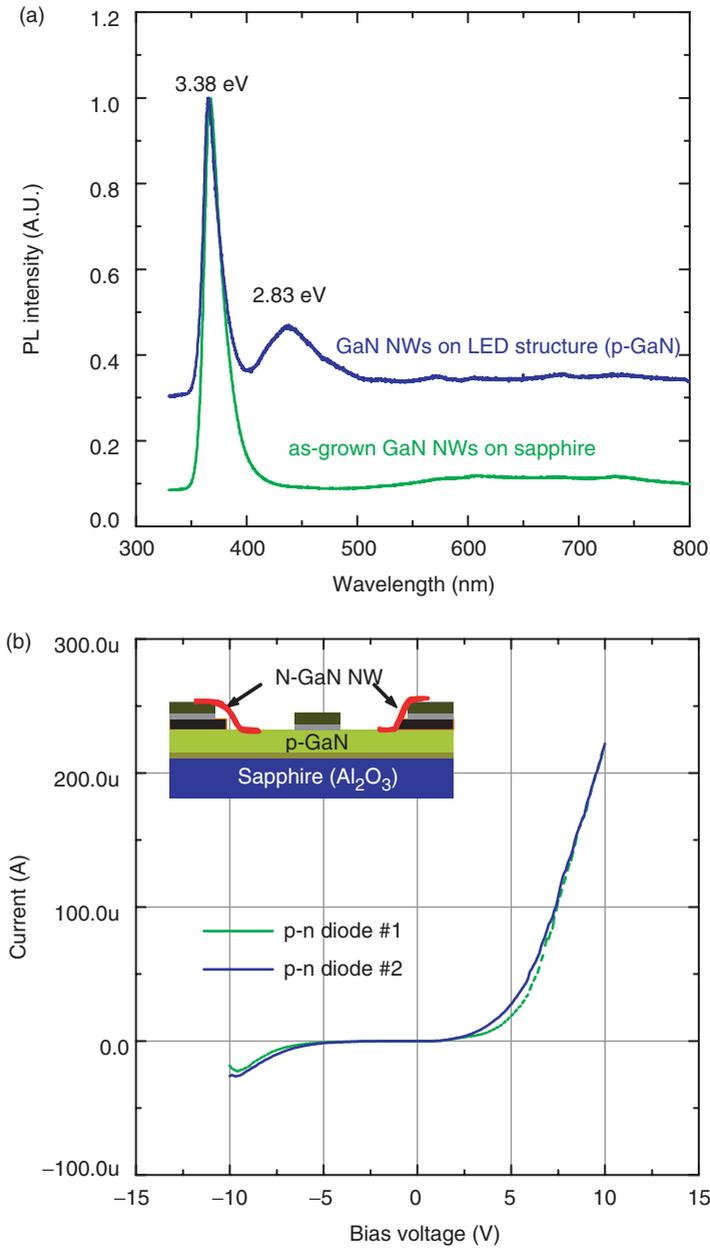


Figure 2. Optical and electrical characterization of homojunction GaN nanowire LED structures. (a) Photoluminescence (PL) spectra of as-grown n-GaN nanowires on sapphire and n-GaN nanowires on LED structures. The spectrum peaks are at 365–367 nm (band edge emission of n-GaN nanowires) and 438 nm (Mg-doped GaN substrate). (b) Current–voltage (I – V) characteristics of random dispersion (RD) prepared GaN nanowire LEDs. The measurements were performed with two GaN nanowire LEDs located in different regions on the same chip ($0.8 \times 0.8 \text{ cm}^2$), consisting of 25 LEDs. All of the LEDs on the chip show clear rectifying behaviour as shown in (b).

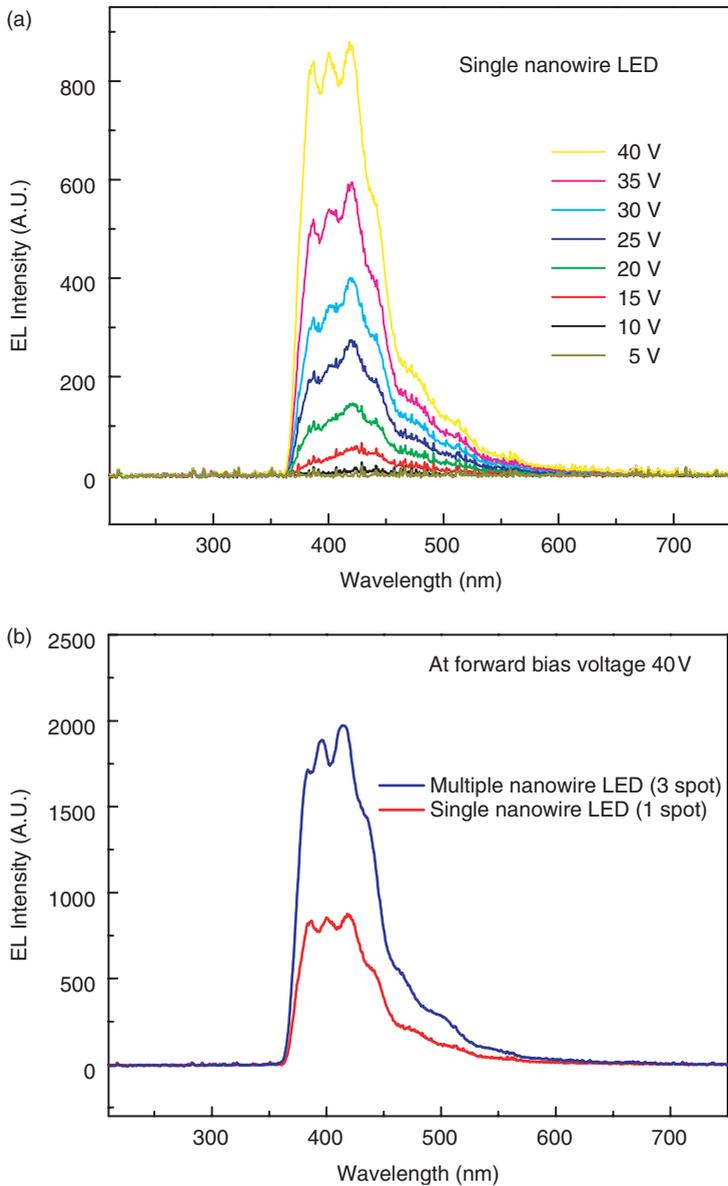


Figure 3. (a) Electroluminescence (EL) spectra of the homojunction single GaN nanowire LED structures recorded from forward bias voltage in the range of 5 V to 40 V. The samples were prepared by random dispersion (RD) technique. (b) The EL spectra for the single and multiple (3 nanowires) LED structures at the forward bias voltage of 40 V. For colour, see online.

GaN nanowire LED (see figure 3a) strongly suggested that the UV and blue emission in EL spectrum originate from electron–hole recombination at the n-GaN nanowire/p-GaN substrate interface [6, 20, 21]. A relatively strong UV peak (387 nm) in the n-GaN nanowire/p-GaN thin film junction may be due to a poor crystal quality and

low hole concentration of the p-GaN substrate (in our case $1.6 \times 10^{17} \text{ cm}^{-3}$) that gives rise to the apparent broad EL spectrum in n-GaN nanowires/p-GaN thin film. Similar results have been observed in thin film based GaN p-n diodes [6, 21]. In our GaN nanowire LED structures, light emission was observed around the n-GaN nanowires where they made contact to p-GaN substrate.

Figures 4a–e show that as we increased the forward voltage to the single GaN nanowire LED, we observed strong UV-blue light emission, which can be readily observed with the naked eye. The first image of single GaN nanowire LED we observed is shown in figure 4e, although previous studies on single crossed GaN nanowire junction LEDs often need to be imaged with charged-coupled device (CCD) camera [15]. We suggest that the high EL intensity could be explained in terms of the enhancement of the carrier injection for our homojunction nanowire devices. This carrier enhancement in GaN nanowire LEDs could be due to the size effect of the n-GaN nanowires on p-GaN substrates. High electric fields could be induced on p-GaN beneath n-GaN nanowires due to the small size of n-GaN nanowire and small junction area, which could reduce the depletion width of the p-GaN and increase the tunnelling probability [22]. Consequently, the EL intensity could be enhanced due to the increased contribution of the tunnelling carrier (current) to the total current. The effect of the multiple nanowires in LED structures was also studied. We have collected the relative EL spectra from the single and multiple (3 nanowires) LED structures. Figure 3b shows the EL spectrum from the multiple nanowire LED structures, which further illustrates the importance of the nanowire/substrate junctions for the emission we observed. Our results point towards the exciting possibility that single GaN nanowire LED can be used for nanoscale light sources in nanoscale integrated photonic circuits.

To maximize the light emission from the single GaN nanowire LED and to meet the requirement for a wafer-based large-scale integration, we also assembled the GaN nanowires across the electrodes (anode and cathode) by dielectrophoresis assisted assembly deposition (DAAD) [18]. A similar assemble approach has been published elsewhere [4, 5, 12, 16, 17]. Duan *et al.* [4] first demonstrated the possibility of using an electric field-assisted assembly technique (dc electrophoresis) to align and control InP nanowires in p-n junction structures. Recently, Lao *et al.* [17] reported on ZnO nanobelt Schottky diodes which are formed by ac dielectrophoresis. Our DAAD (ac dielectrophoresis method) can be readily used to deposit semiconductor nanowires in selected areas and positions by controlling the applied frequency and the electric field to the electrodes [23]. For the DAAD experiment, a drop of the GaN nanowire suspension ($\sim 3 \mu\text{l}$) was placed on the selected gap using a micropipette while the electric field was being applied across the electrodes. The anode (inside electrode, see figure 5c) was grounded while the cathode (outer doughnut-shaped electrode) was applied with a sinusoidal alternating current (ac) voltage. The electric field was continuously applied until the suspension completely dried out. Our previous studies showed that the alignment yield increased with increasing ac peak-to-peak voltage (ac electric field) [23]. To understand such behaviour observed in the GaN nanowires with the applied ac electric field, we can consider the well-known standard model of dielectrophoresis (DEP) exerted on nanowires. For a homogeneous cylindrical shape and a long wire with its major axis parallel

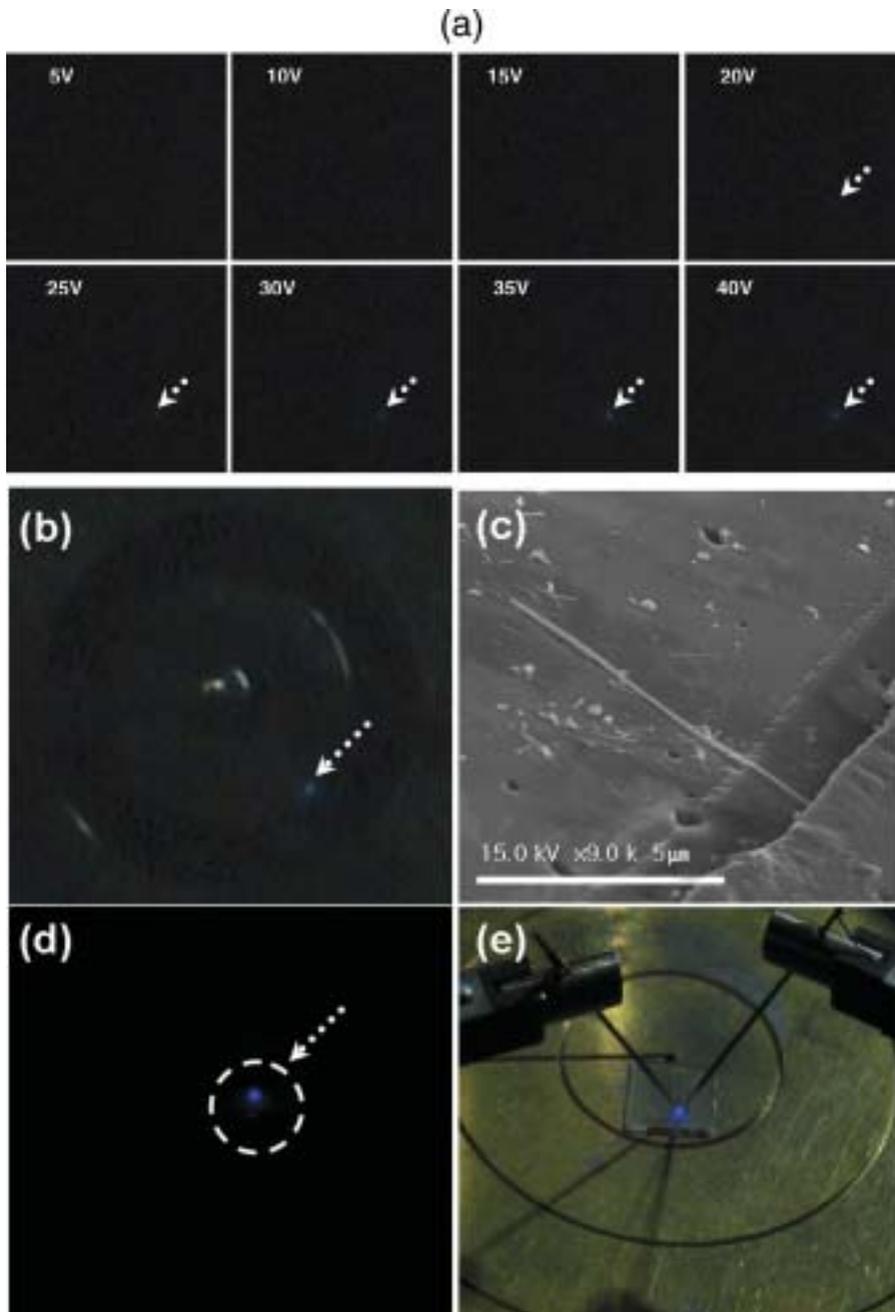


Figure 4. Electroluminescence (EL) properties for single GaN nanowire LED prepared by random dispersion technique. (a) EL images of single GaN nanowire LED with different forward voltage in the range of 5 V to 40 V. EL (b) and SEM image (c) from single GaN nanowire. This GaN nanowire in SEM image (c) is exactly what we used for EL study. EL images were taken by charged coupled device (CCD) camera. Digital EL images, without and with background light, from a single GaN nanowire are shown in (d) and (e).

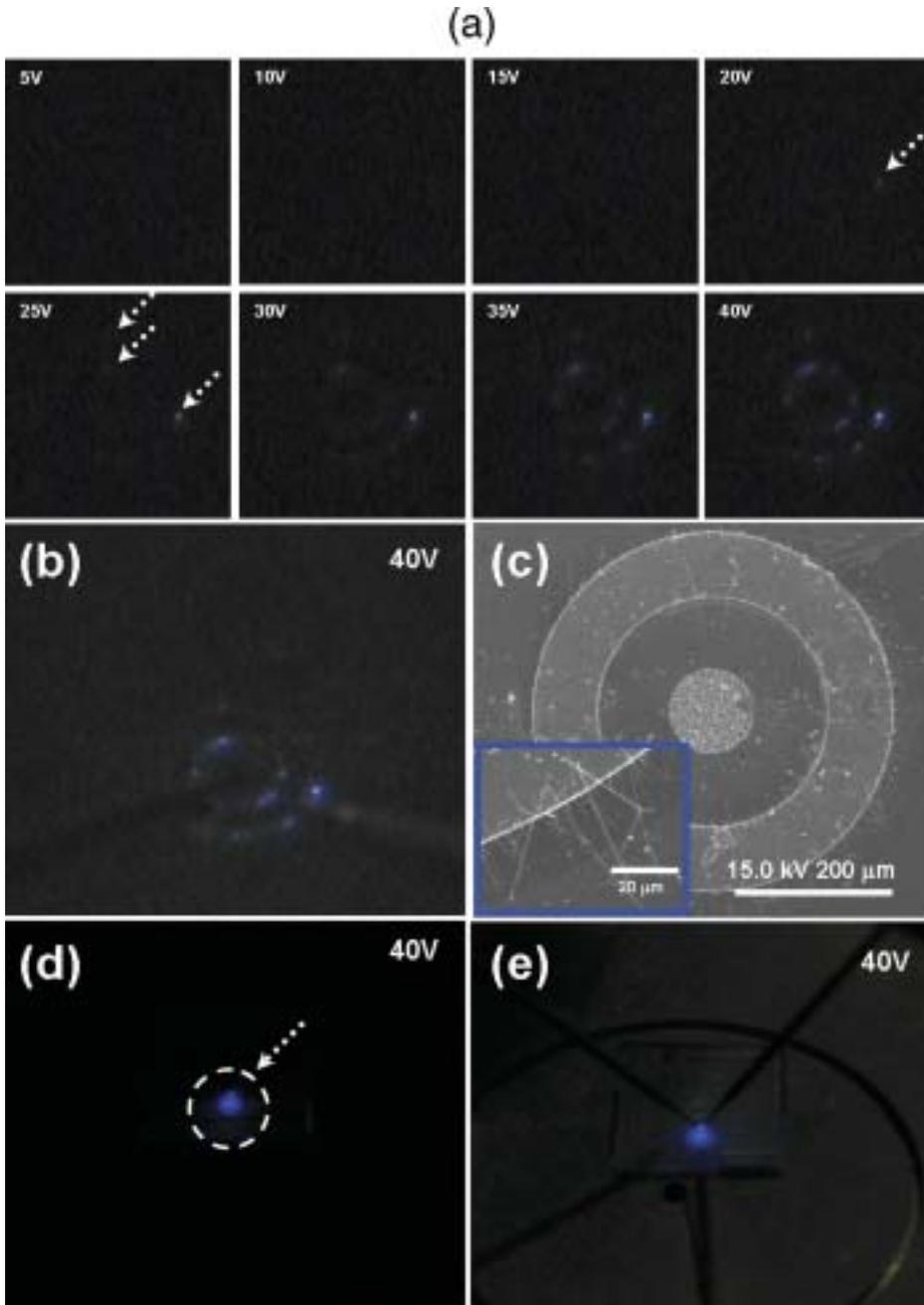


Figure 5. Electroluminescence properties for aligned GaN nanowire LED prepared by DAAD techniques. (a) EL images of aligned GaN nanowire LED with different forward voltage in the range of 5 V to 40 V. EL (b) and SEM image (c) from aligned GaN nanowire. Inset in (c) shows the aligned GaN nanowires around the cathode metal. EL images taken by digital camera without and with background light from the GaN nanowires are shown in (d, e).

to an inhomogeneous alternating electric field, the DEP force is given by [18, 23, 24]

$$\vec{F}_{\text{DEP}} = \frac{v}{2} \varepsilon_m K(\omega) \vec{\nabla} (\vec{E}_{\text{rms}}^2) = \frac{\pi r^2 l}{2} \varepsilon_m K(\omega) \vec{\nabla} (\vec{E}_{\text{rms}}^2), \quad (1)$$

where the v is the volume of the nanowires, $K(\omega)$ is the real part of the Clausius–Mosotti factor, r is the radius of the nanowires and l is the length of the nanowires. Equation (1) clearly indicates that the DEP force is highly dependent on the volume of the nanowires, the Clausius–Mosotti factor and the gradient of the electric field. For the GaN nanowires within the isopropyl alcohol (IPA) liquid medium, the theoretical calculation of $K(\omega)$ at the different angular frequency is bonded by the limits $1.7 \times 10^7 < K(\omega/2\pi) < 1.0$ in the frequency range of 1 kHz ~ 80 MHz. The sign of the real part of $K(\omega)$ denotes the direction of the electric field [24]. The theoretical calculation indicated that the value of $K(\omega)$ was positive (known a positive DEP) in the frequency range of 1 kHz ~ 80 MHz. When $\text{Re}[K(\omega)]$ is positive, the induced force points toward the high electric field at the electrode surface and thus the GaN nanowires are gathered around the electrode edges (in our case, cathode electrodes). The DAAD for the homojunction GaN nanowire LED structures was performed at a frequency of 10 kHz with 15 V_{p-p}, which is optimum condition for GaN nanowire suspensions with IPA [23]. As shown in figure 5c, the GaN nanowires are well-aligned around the cathode electrodes. Figure 5a–e show the EL properties for these aligned GaN nanowire LEDs. Similar strong UV–blue light emissions were observed from the DAAD prepared GaN nanowire LED structures as seen in single GaN nanowire LED structure above. For the forward voltage of 40 V in figure 5a, the clear multiple spots are observed in the DAAD prepared GaN nanowire LED structures. The results indicated that the light emissions could be intensified with the formation of multiple GaN nanowires junctions as we observed in figure 3b. This is also a solid evidence for the possibility for wafer-based large scale integration approaches and that these DAAD technique represent one of the most powerful assembly technique to align and manipulate the semiconductor nanostructures.

4. Conclusion

In summary, we have demonstrated the large-scale fabrication of high-brightness GaN nanowire UV–blue LEDs, assembled with n-GaN nanowires on p-GaN substrate using random dispersion or DAAD technique. The successful fabrication of homojunction p-GaN material/n-GaN nanowire UV–blue LED using a DAAD technique suggests that it is a noteworthy advance towards the realization of the large-scale addressable nanowire-based UV–blue light emitting source. This new approach also offers great opportunities for further fundamental research, as well as applications in solid-state lighting and biomedical areas.

Acknowledgments

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