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Electroluminescence from GaN–polymer heterojunction

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ABSTRACT

Inorganic and organic semiconductor devices are generally viewed as distinct and separate technologies. Herein we report a hybrid inorganic–organic light-emitting device employing the use of an air stable polymer, Poly (9,9-dioctylfluorene-alt-benzothiadiazole) as a p-type layer to create a heterojunction, avoiding the use of p-type GaN, which is difficult to grow, being prone to the complex and expensive fabrication techniques that characterises it. *I–V* characteristics of the GaN–polymer heterojunction fabricated by us exhibits excellent rectification. The luminescence onset voltage is typically about 8–10 V. The device emits yellowish white electroluminescence with CIE coordinates (0.42, 0.44).

1. Introduction

The development and commercialisation of semiconductor light-emitting devices has made revolutionary progress due to the rapidly growing demand for displays, solid-state lighting and related applications. The progress in gallium nitride and organic semiconductor technologies are of particular significance [1–3]. GaN being a direct band gap semiconductor (3.4 eV) has attracted much attention since the development of bright blue light-emitting diodes (LEDs) by Nakamura and coworkers [4–9]. These light sources typically contain a diode that emits in the blue or ultraviolet region and phosphors to down-convert the energy giving rise to white light [10]. However, conversion with traditional phosphors results in poor colour rendering due to the relatively weak green and red emission from the respective components of the phosphor mixtures. In addition the phosphors are traditionally prepared in typical grain sizes of tens of micrometres inhibiting the development of advanced devices, where the emitter elements could be comparable to or smaller than the particle size of the phosphors. Organic semiconductor light-emitting diodes (LEDs) in turn can span the entire visible spectrum and have low-cost potential due to high-throughput manufacturing, offering new prospects for displays and lighting applications. These two semiconductor families have their own special advantages and disadvantages. Especially, inorganic materials exhibit excellent electrical properties but need complex and expensive fabrication techniques to characterise them. Their spectral coverage is also limited, although down-conversion with phosphors allows access to other colours and white-light emission. Comparatively organic semiconductors offer excellent luminescence properties, with a greater variety of emission wavelengths, but exhibit relatively poor electrical behaviour. Combining the desired features of both the families is clearly an attractive proposition.

Inorganic and organic semiconductor junction devices are generally viewed as distinct and separate technologies, but few examples of mixing the two such electronic materials exist. Hyunh et al. [11] have shown charge transfer between colloidal II–VI nanocrystals immersed within an organic conducting polymer matrix, while Coe et al. [12] have fabricated a LED where II–VI nanocrystals were imbedded within a planar organic junction. Heliotis et al. [13] have reported the use of fluorene-based polymer semiconductor films as efficient InGaN wavelength conversion media, using radiative coupling where emission from InGaN LED is absorbed by the polyfluorene film and subsequently re-emitted at longer wavelengths. These authors have reported optically pumped light emission but electrically pumped light emission would be more practical than optically pumped emission for lighting and laser applications. Konenkamp et al. [14] prepared ZnO nanowire/polymer heterojunctions and white-coloured electroluminescence from 350 to 850 nm were observed. In order to achieve electroluminescence from ZnO a few researchers have prepared LEDs based on nano-structured ZnO and p-type polymers, which are used to substitute for p-type ZnO [15]. Chang et al. [16] reported electroluminiscence from a ZnO nanowire/polymer p–n junction as well. In their work an insulator material is used to coat the ZnO nanowires, which is

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little complicated, and the electroluminescence produced is weak. To take advantage of GaN blue-light emission, development has begun of so-called hybrid p–n heterojunction LEDs, which are composed of heterojunctions between oxide-semiconductor nanowires and GaN p-type thin layers [14,17,18]. But in order to fabricate GaN Blue LED, one has to grow p-type GaN, which is difficult to grow. This inspired us to make white light from n-type GaN where we have used a p-type polymer as an alternate to p-type GaN to create heterojunction. The polymer used was F8BT Poly(9,9-dioctylfluorene-alt-benzothiadiazole), which is air stable and it can be spin-coated over GaN. In addition to act as a p-type layer; it also emits yellow light, which when combined in proper composition with GaN gives rise to white light.

2. Experiment

In the present study, we have used a commercial silicon-doped GaN (0 0 0 1)-oriented layer grown on sapphire with the *c*-axis perpendicular to the substrate. The GaN layer was 8 μm thick and the dopant concentration was 2×10^{18} cm^{-3} with emission wavelength ~360 nm. The n-GaN (0 0 0 1) substrate was degreased in trichloroethylene at 50 °C for 10 min, subsequently cleaned for 5 min in warm acetone at 60 °C in an ultrasonic bath, 10 min in methanol at room temperature in an ultrasonic bath and finally rinsed with high-purity millipore water (resistivity of 18.2 MΩ cm). Then the film was dried in brisk flow of nitrogen. F8BT polymer was spin-coated from a toluene solution with the concentration of 10 mg/mL (thickness ~80 nm) at 2000 rpm. Subsequently, a layer of PEDOT/PSS (poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate)) was deposited on top of the polystyrene layer serving as a hole-injecting contact. The layer was then heated at 110 °C for 10 min to remove the residual water solvent. Finally aluminium (100 nm) and silver (80 nm) were evaporated over GaN film and PEDOT:PSS, respectively, without breaking the vacuum. High resolution X-ray diffraction (D8 DISCOVER) was used to confirm the single crystal GaN film. The current–voltage characteristics were measured with a Keithley 6430 source metre. UV–vis measurement was performed using Perkin-Elmer Lambda 35 UV/vis spectrometer and photoluminescence measurements were carried out using JobinYvon Flurolog-3 spectrometer. The electroluminescence spectra recorded with an Avantes AVS USB2 2048 spectrometer in air.

3. Results and discussion

Fig. 1 shows the schematic diagram of the device. Fig. 2 shows the XRD pattern of the GaN film on c-plane sapphire. Fig. 3 shows the photoluminescence spectra of GaN film and F8BT with excitation wavelengths 325 and 450 nm, respectively. There is also contribution from the defect band related yellow emission from GaN [19]. UV–vis absorption spectrum of F8BT is shown as inset in Fig. 3. The current–voltage characteristics of the

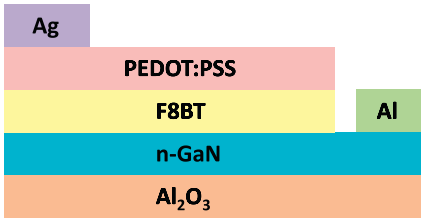

Fig. 1. Schematic diagram of the device.

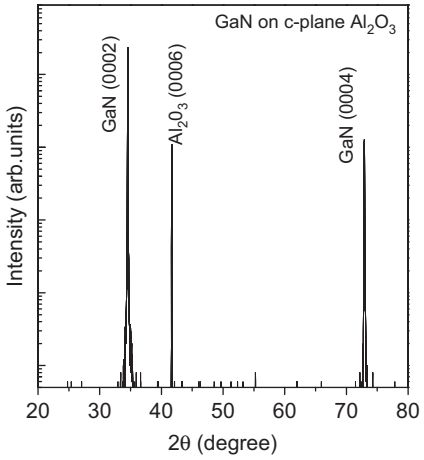

Fig. 2. Shows the HRXRD of the GaN film.

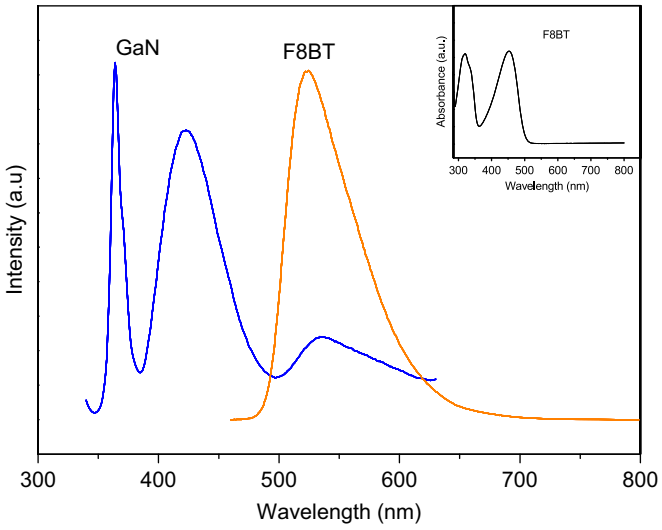

Fig. 3. PL spectra of GaN film and F8BT at 325 and 450 nm as excitation wavelengths, respectively. UV–vis absorbtion spectrum of F8BT is shown as inset.

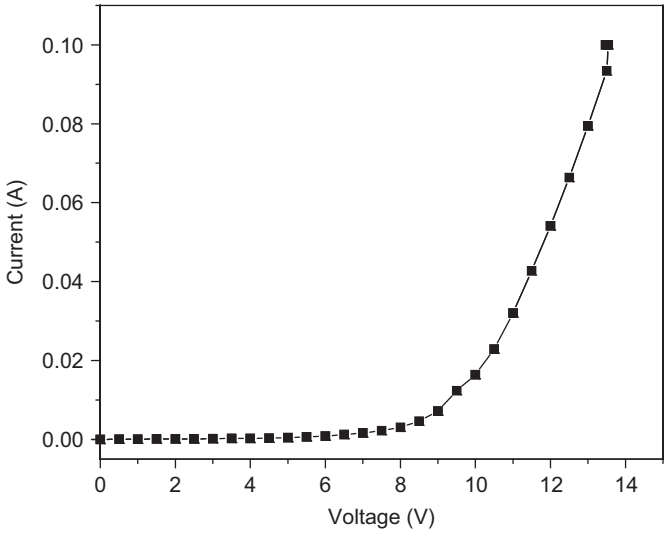

Fig. 4. Current–voltage characteristics of the device.

Al/GaN/F8BT/PEDOT:PSS/Ag heterostructure is shown in Fig. 4. *I–V* characteristics show excellent rectification; the luminescence onset voltage is typically about 8–10 V. The energy band diagram