Lateral color integration on rare-earth-doped GaN electroluminescent thin films

D. S. Lee and A. J. Steckl[a)]
Nanoelectronics Laboratory, University of Cincinnati, Cincinnati, Ohio 45221-0030

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Lateral color integration has been obtained using GaN thin films doped with Er and Eu. These rare-earth doped GaN (GaN:RE) films were grown on Si (111) substrates by molecular beam epitaxy. Independent red and green emissions have been obtained from side-by-side Er and Eu electroluminescent devices. Photoluminescence and electroluminescence operation show green emissions at 537 and 558 nm from Er-doped GaN and red emission at 621 nm from Eu-doped GaN. Two patterning fabrication techniques have been investigated to obtain lateral integration: (a) use of shadow masks during 400 °C growth of GaN:RE films; (b) photoresist liftoff in conjunction with <100 °C GaN:RE growth. Devices fabricated by the shadow mask method were bright enough to be detected under the ambient light at a bias of 30 V. The GaN:RE films were clear and their surfaces were smooth with nanoscale GaN grains. The root mean square surface roughness was measured to be 5–10 nm. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1461884]

Starting with the first report[1] of visible emission from Er-doped GaN, the development of rare-earth (RE)-doped GaN has led to the successful fabrication of electroluminescent devices (ELD) with pure green,[2] red,[3] and blue[4] colors from Er-, Eu-, and Tm-doped GaN, respectively. Intermediate colors from GaN:RE ELDs have been reported through uniform codoping during growth, such as turquoise[5] from GaN codoped with Er and Tm, orange or yellow[6] from GaN codoped with Er and Eu. The first integration of primary colors in a GaN:RE ELD utilized[7] a device structure with two stacked layers of GaN:Eu and GaN:Er. Because of the current path through the device, the ELD emitted green light for positive bias and red light under negative bias. While vertical integration has the advantage of a compact structure it prevents optimum device performance because of the necessary bias conditions. In this letter, we discuss the lateral integration of individual color emitters in a GaN:RE device structure. Lateral integration in multicolor ELDs for flat panel displays enables[8–10] the deposition of films optimized for the emission of each color. The first structure investigated[11,12] for multicolor emission was a thin film electroluminescent (TFEL) device fabricated on the stacked phosphor films of ZnS:Tb,F and ZnS:Mn. Subsequently, Yamauchi *et al.*[13] reported a two-color (red/green) TFEL device fabricated laterally by a combination of wet etching and liftoff. Since then many groups have studied and improved multicolor TFEL integration mostly on II–VI hosts. Rare-earth-doped GaN has a very high potential in light-emitting devices and displays application resulting from a combination of the previously mentioned results with the advantages of GaN over other semiconductors: direct band-gap transition which is very important in optical applications, and a large energy band-gap which results in very low thermal quenching.[14,15] Moreover, GaN is a superior host for RE^{3+} ions than II–VI semiconductors, such as ZnS and SrS, in terms of charge neutrality, and chemical and thermal robustness.

GaN films were grown on *p*-type (111) Si substrate by molecular beam epitaxy (MBE) with a Ga elemental source and a nitrogen plasma source. Er and Eu doping was performed *in situ* during growth from solid sources. GaN:RE layers were typically grown for 1 h at either <100 or 400 °C substrate temperature. No buffer layers were grown for these particular films since the buffer layer is usually grown at 450–500 °C, which is higher than 400 °C. Room temperature growth of GaN:RE films was recently reported[16] to produce

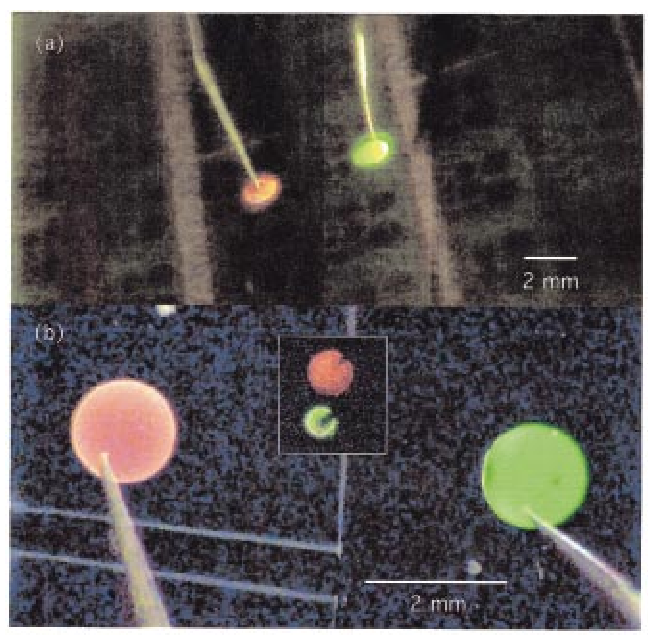

FIG. 1. (Color) Simultaneous red (Eu) and green (Er) emission from two-color integrated GaN:RE ELDs: (a) devices in operation under ambient lighting, showing parallel array of devices fabricated by the shadow mask technique; (b) closeup of shadow mask and liftoff (inset) fabricated devices in operation.

[a)]Electronic mail: a.steckl@uc.edu

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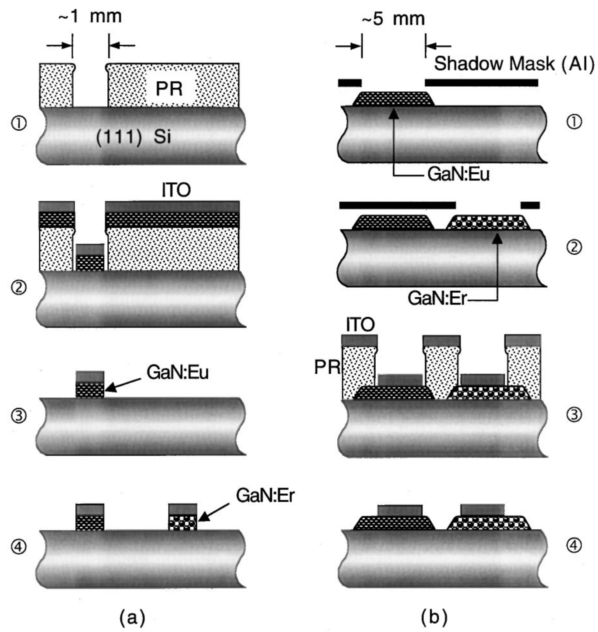

FIG. 2. Schematic diagrams indicating process steps for the two methods used to obtain two-color integration: (a) photoresist (PR) liftoff process with room-temperature growth of RE-doped GaN layers: ① PR patterning; ② GaN:Eu growth and ITO sputtering; ③ PR liftoff; ④ repeat ①–③ for GaN:Er; (b) 400 °C growth of GaN:RE and ITO liftoff: ① GaN:Eu growth with shadow mask; ② GaN:Er growth with second shadow mask; ③ PR patterning and ITO sputtering; ④ PR liftoff.

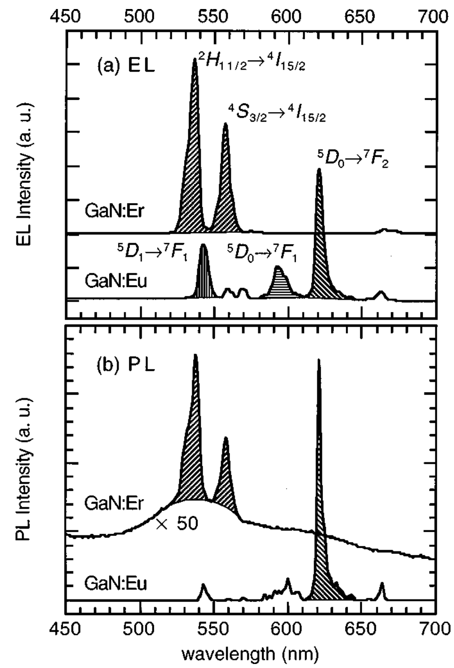

FIG. 3. EL and PL spectra from Er- and Eu-doped GaN films grown at 400 °C. Two green peaks are detected from the Er-doped GaN film and a dominant red peak from Eu-doped GaN.

visible emission ELDs with the same colors as higher temperature growth. The Ga flux was 3.9×10^{-7} Torr for <100 °C growth and 5.5×10^{-7} Torr for 400 °C growth. Eu-doped GaN films were grown with Eu cell temperatures of 410 °C resulting in a concentration of 0.1 to 1 at. %. Er-doped GaN films were grown with an Er cell temperature of 860 °C, which resulted[17] in an Er concentration of 0.5 to 1 at. %. The nitrogen flow parameters were fixed for all growths: 1.5 sccm flow rate and 400 W plasma power. Simple circular Schottky electrodes were fabricated for EL measurements on top of the GaN:RE films using indium–tin oxide (ITO) sputtering. The ITO film has more than 90% transmittance over the whole visible light range. Diameters of the electrodes range from ~100 μm to ~1.5 mm.

Figure 1 shows two-color emission from laterally integrated ELDs fabricated on 400 °C-grown GaN films doped with Eu and Er. The photograph in Fig. 1(a) was taken under ambient light to show the devices under test as well as their photoemission. Figure 1(b) is a closeup micrograph showing bright emission from two ELDs with a diameter of ~1.5 mm. The green emission from the GaN:Er ELD has CIE coordinates of (0.27, 0.71). The emission from the GaN:Eu ELD is reddish-orange because of a green component (as discussed below). The corresponding CIE coordinates are (0.61, 0.36). The inset in Fig. 1(b) shows emission from ELDs fabricated on <100 °C-grown GaN films. The ELDs were biased negative on the ITO electrode, and positive on the Si substrate. The EL emission mechanism is impact excitation of Er^{3+} and Eu^{3+} by injected electrons through the GaN host. Therefore, it is important to grow sufficiently crystalline GaN for a good host material.

Two fabrication methods have been investigated for two-color lateral integration: photoresist (PR) liftoff and shadow mask deposition. The main fabrication steps for each technique are shown schematically in Fig. 2. The liftoff process was applied to both GaN:RE films and to the ITO electrode [Fig. 2(a)]. This integration approach is relatively easy to implement but does require that the GaN deposition occur at a temperature below 100 °C. Otherwise, considerable outgassing of solvent from PR and a resulting high base pressure in the growth chamber produce unreliable GaN films. The liftoff method consists of the following fabrication steps: ① After PR patterning, the sample is inserted into the MBE system; ② GaN:Eu film is grown for 1 h at ~20 °C followed by the sputter deposition of the ITO film; ③ PR liftoff with PR stripper and postcleaning produce a first ELD for red color from the GaN:Eu film; ④ Repeat steps ①–③ for the GaN:Er film, resulting in side-by-side ELDs for red and green emission on GaN:Eu and GaN:Er films, respectively. Since only simple alignment was used during the fabrication process, edge lines and even some gaps are seen between the regions of the grown films as shown in Fig. 1. The light-colored stripes in (a) are gaps between the grown films and lines in (b) are due to the edge of the shadow mask.

Figure 2(b) shows the steps contained in the shadow mask approach. In this method the growth of GaN:RE takes place at 400 °C. The temperature was chosen for the following reasons: (a) the GaN:RE ELD brightness increases with growth temperature; (b) the GaN crystallinity also improves with growth temperature, but relatively slowly above 400 °C; (c) the Al shadow masks begin to deform significantly below the melting point of Al (660 °C). The shadow mask ELD fabrication procedure consists of the following major steps: ① place sample with the first Al shadow mask in the MBE system and grow GaN:Eu at 400 °C; ② grow GaN:Er at 400 °C with the second shadow mask blocking the already grown GaN:Eu film; ③ produce PR patterns for ITO electrodes on both GaN:Eu and GaN:Er regions and sputter de-

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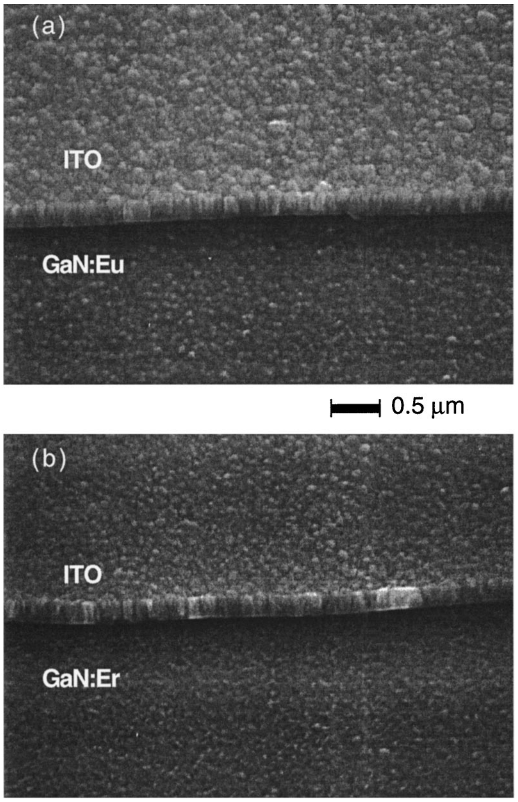

FIG. 4. SEM micrographs showing the morphology of the GaN:RE films grown at 400 °C and the sputtered ITO electrodes. Note the comparatively flat surfaces and compact structures with small grains.

posit ITO thin films; ④ PR liftoff producing side-by-side ELDs for two-color integration.

Figure 3(a) shows EL spectra taken for both red and green emissions from GaN:Eu and GaN:Er films grown at 400 °C, respectively. The visible emission of GaN:Er has two characteristic green peaks at 537 and 558 nm, which are attributed to two $4f-4f$ Er^{3+} inner shell transitions: $^2H_{11/2} \rightarrow ^4I_{15/2}$ and $^4S_{3/2} \rightarrow ^4I_{15/2}$, respectively. The characteristic red emission of Eu^{3+} ions comes from the dominant $^5D_0 \rightarrow ^7F_2$ transition. As reported[3] previously, Eu^{3+} ions in GaN ELDs also have significant emission in the green at ~543 nm ($^5D_1 \rightarrow ^7F_1$) and orange at 590–600 nm ($^5D_0 \rightarrow ^7F_1$). The combined effect is to produce an EL emission color which is orange-red. The bias voltage of 30 V resulted in current flow of 43 mA for the GaN:Er ELD and 167 mA for the GaN:Eu ELD. The ELDs were very stable under these bias conditions and the EL emission was constant for many hours of operation. The somewhat conductive nature of these films is due to the relatively low growth temperature and it can be improved by optimization of growth parameters and conditions leading to increased brightness. The ELDs fabricated on <100 °C-grown GaN:RE films also showed red and green EL. However, the device lifetime was relatively short and emission not as bright as in the ELDs on 400 °C-grown films. The electrical properties ($I-V$) of GaN:Er ELDs and their temperature dependence was recently published,[18] along with a model for their operation.

Photoluminescence (PL) was carried out at room temperature by above-band-gap excitation using a HeCd (325 nm) laser. As shown in Fig. 3(b), the same characteristic transitions are also observed here as in EL for the GaN:RE samples grown at 400 °C. No PL was detected for the samples grown at 100 °C because of the poor crystalline quality. Note that the red emission PL intensity from a GaN:Eu film is ~50 times stronger than the green emission intensity from a GaN:Er film. Interestingly in PL unlike EL, GaN:Eu emits pure red because of the dominant 621 nm peak.

The surface morphology was investigated by scanning electron (SEM) and atomic force microscopy (AFM). In Fig. 4, SEM photographs show the surface morphology of the GaN:RE films grown at 400 °C. The films appear to have comparatively flat surfaces and compact structure with nanoscale grains. Root mean square (rms) roughness of these films was ~5 nm in AFM. Room-temperature-grown films also had a compact structure with tiny grains but with somewhat rougher surfaces, ~10 nm rms roughness.

In summary, we have fabricated integrated EL devices emitting two colors: red and green, on GaN:Er and GaN:Eu films grown separately on the same substrate by two different fabrication techniques: liftoff and shadow masking. ELDs on 400 °C-grown films had an emission sufficiently bright to be useful under ambient lighting conditions. The ELDs on the <100 °C-grown films are much less bright. However, the EL emission intensity can be improved by postannealing at ~800 °C previously reported[16] for room-temperature growth of GaN:RE. Considering these results, we can reach the conclusion that three-color integration of GaN:RE ELDs is now possible for flat panel display (FPD) applications.

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