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

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We have realized full color integration on rare-earth-doped thin-film electroluminescent (EL) GaN using lateral integration. Tm, Er, and Eu dopants were *in situ* doped into GaN thin films during growth in order to obtain blue, green, and red emission, respectively. Three color pixel arrays have been fabricated using spin-on-glass films as the sacrificial layers for lift-off lithography. The pixel dimensions are $0.2\times0.7\ mm^2$, and the separation is 0.2 mm. dc EL devices were fabricated using indium tin oxide transparent electrodes. Typical applied voltage was 30–40 V. The blue emission from Tm-doped GaN has a peak at 477 nm, the green emission from Er-doped GaN has two peaks at 537 and 558 nm, while the red emission from Eu-doped GaN has a peak at 621 nm. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1539301]

High field thin-film electroluminescence (TFEL) devices[1,2] utilize hot electrons generated by the electric field to impact-excite light emitting centers in wide band gap semiconductors (WBGS). This phenomenon has been employed to fabricate flat panel display[3,4] emitting a variety of visible colors. Recently green,[5–7] blue,[8] and red[9] electroluminescence (EL) from rare earths (RE) *in situ* doped during growth into films of the III–V WBGS GaN have been reported. Other colors, such as turquoise[10] and yellow,[11] have also been obtained from GaN:RE through uniform codoping with Er and Tm and with Er and Eu, respectively. If the EL devices (ELDs) emitting these colors can be integrated on a single substrate, the integrated array could be regarded as the prototype model of future generations of large-scale flat panel displays (FPD). There are two color integration methods: vertical integration and lateral integration. In the former case, the phosphor films are placed layer-upon-layer, while in the later case the devices are arranged side-by-side. The advantage of vertical integration is that the structure is compact. Additional transparent electrodes need to be interleaved between the phosphor layers, complicating the overall fabrication process and limiting the high temperature exposure that can be utilized. A red and green integrated EL device with stacked layers of GaN:Eu and GaN:Er emitting green EL under positive bias and red EL under negative bias has been previously reported.[12] Lateral integration can be achieved with patterned phosphors for each color[13] or with a single unpatterned broadband emitting ("white") phosphor whose emission is separated into the primary colors by patterned color filters. The patterned filter approach is simple to fabricate, but does not produce pure primary colors, while the reverse is true for the patterned phosphor approach. Yamauchi *et al.*[14] reported a red and green TFEL device fabricated laterally by a combination of wet etching for the first phosphor layer and photoresist lift-off for the second phosphor layer. Other fabrication approaches for patterned phosphor multicolor TFEL devices include sputter etching.[15] Lee and Steckl[16] have reported two patterning fabrication techniques to obtain lateral two color integration in RE-doped GaN: use of shadow masks during 400 °C growth of GaN:RE films and photoresist lift-off in conjunction with <100 °C GaN:RE growth. The growth temperature of 400 °C is significantly lower than the optimized growth temperatures for GaN:RE films. RE-doped GaN has a very high potential in light-emitting devices and display application due to the advantage of GaN over other semiconductors.[17,18] In this letter, we report lateral three-color integration on RE-doped TFEL GaN.

Lateral integration of ELDs doped with different REs requires a repetitive sequence of thin-film growth and patterning. Because GaN thin films are chemically very robust, it is very difficult to use wet etching with chemical solutions for film patterning. Although it is possible to use plasma-based techniques to dry etch GaN:RE thin films to form pixels for different colors, the etched-region surfaces are quite rough making it very difficult to subsequently grow high quality RE-doped GaN thin films for the other two colors. Therefore, we have pursued the lift-off technique to integrate GaN:RE pixels for multiple colors. It is most important to select a suitable material as the sacrificial layer for the lift-off process used in three-color integration. The sacrificial layer needs to have the following properties: (a) ability to withstand high temperature annealing at more than 600 °C without emitting a significant quantity of (organic and/or inorganic) gases to avoid the contamination of the molecular beam epitaxy (MBE) growth chamber; (b) ability to be etched selectively with respect to the GaN layer and the substrate. We have investigated a spin-on-glass (SOG) liquid solution as the starting material for the sacrificial layer. SOG is an attractive choice: It is mostly silica and contains little or no organics, it can be annealed up to 900 °C, and it can be etched off in a HF solution in a few seconds.

GaN:RE films were grown in a Riber MBE-32 system on 2 in. *p*-Si (111) substrates. Solid sources were employed to supply the Ga (7 N purity) and rare earth (3 N) fluxes, while a SVTA radio frequency plasma source was used to generate atomic nitrogen. Lee and Steckl have extensively

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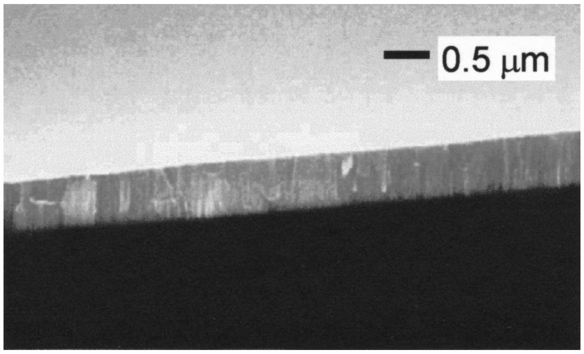

FIG. 1. Cross-sectional SEM micrograph of ∼0.6 μm SOG layer on Si wafer.

investigated the growth conditions for Er-doped GaN thin films.[19,21] To obtain strong EL green emission from GaN:RE ELD, three main parameters need to be selected appropriately: Ga flux,[19] substrate temperature,[20] and RE concentration.[21] For all GaN:RE growth runs, a GaN buffer layer was first deposited for 2 min, then GaN:RE layers were deposited for 30 min, and finally a GaN cap layer was deposited for 1 min. The total GaN film thickness was ∼0.5 μm. The parameters for nitrogen plasma were kept constant for all growth runs: the N_2 flow rate is 1.5 sccm and the plasma power is 400 W. The substrate temperature and the Ga cell temperature are selected based on optimized conditions for each RE dopant. For the GaN:Er growth, the substrate is at 600 °C, the Ga cell temperature is 930 °C, and the Er cell temperature is 860 °C. For the GaN:Eu growth, the substrate temperature is 500 °C, the Ga cell temperature is 920 °C, and the Eu cell temperature is 400 °C. For the GaN:Tm growth, the substrate is at 500 °C, the Ga cell temperature is 915 °C, and the Tm cell temperature is 600 °C.

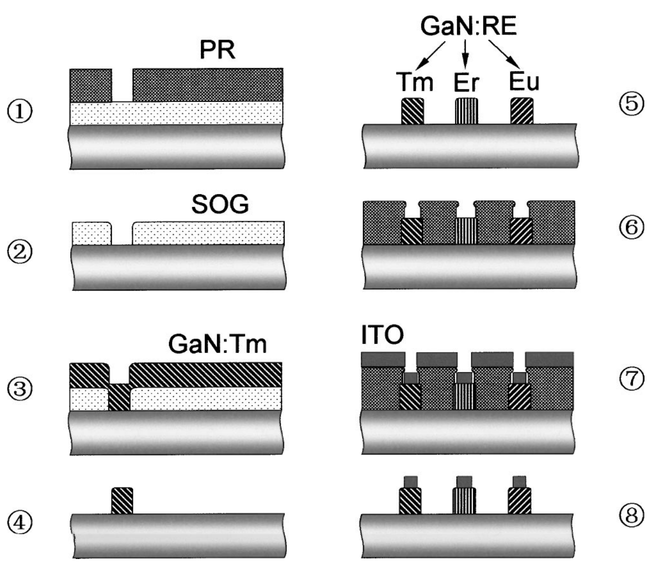

FIG. 2. Schematic diagrams indicating the steps for SOG lift-off process to obtain three-color integration: ① spin SOG on Si wafer twice, then coat with PR and expose the PR pattern; ② etch SOG with 0.1% diluted HF to form the SOG window for GaN:RE growth; ③ grow GaN:Tm in MBE system after the SOG was sufficiently outgassed; ④ SOG lift-off with HF revealing the GaN:Tm patterns; ⑤ repeat twice the procedures from ① to ④, using GaN:Eu and GaN:Er; ⑥ produce PR patterns for ITO electrodes on GaN:RE pixels; ⑦ sputter-deposit ITO thin films; ⑧ lift-off PR and anneal samples in N_2 ambient to form good contact.

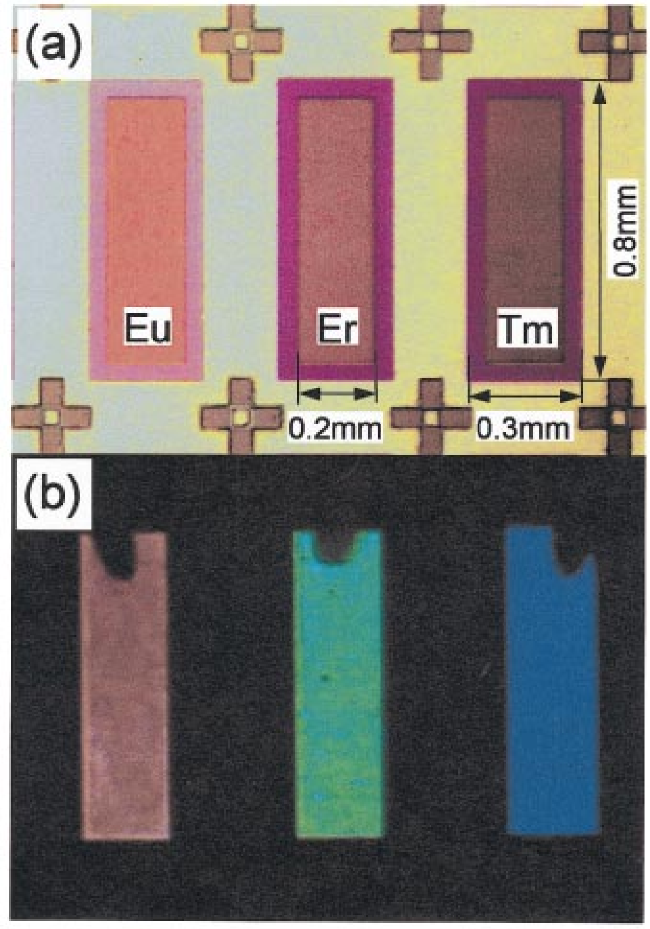

FIG. 3. (Color) Laterally integrated GaN:RE TFEL containing the three primary colors fabricated with the SOG lift-off technique: (a) optical microscopy photograph of the GaN TFEL showing the three-color integration; (b) blue, green, and red emission under dc bias from TFEL GaN devices doped with Tm, Er, and Eu, respectively.

Simple rectangular Schottky electrodes were deposited for EL measurements on top of the GaN:RE pixels using indium tin oxide (ITO) sputtering. The ITO film has more than 85% transmittance over the whole visible light range.

A scanning electron microscope (SEM) photograph of the cross section of the SOG film on a Si wafer is shown in Fig. 1. The thickness of the SOG film is about 0.6 μm after two applications and is quite uniform in thickness over the entire Si wafer. The main fabrication steps for three-color integrated devices are shown schematically in Fig. 2.

Figure 3(a) contains an optical photograph of the laterally integrated three-color TFEL GaN:RE devices. Each GaN:RE device is 0.3 mm wide and 0.8 mm high. The size of the ITO electrodes is 0.2 mm wide and 0.7 mm high. The EL devices are separated from each other by 0.2 mm. The edge of the GaN:RE pattern is very sharp. Figure 3(b) shows the GaN:RE ELDs in operation under dc bias. Blue, green, and red emission is observed from the laterally integrated TFEL GaN devices.

EL spectra of blue, green, and red emission from GaN:Tm, GaN:Er, and GaN:Eu pixels, respectively, are shown in Fig. 4(a). The spectrum of visible emission from GaN:Tm consists of a dominant blue peak at 477 nm, caused by the electronic transition from the $^1G_4$ level to the $^3H_6$ ground state of Tm.[8] The emission spectrum of GaN:Er con-

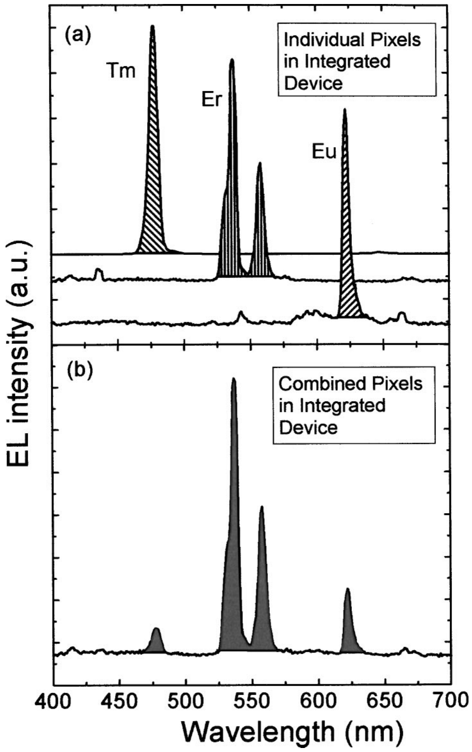

FIG. 4. EL emission spectra from integrated blue, green, and red pixels of GaN:RE: (a) individual spectra from Tm, Er, and Eu pixels at a dc bias of 30 V with current of 33, 3, and 5 mA, respectively; (b) spectrum from all three pixels with simultaneous emission, at a dc bias of 30 V and current of 45 mA.

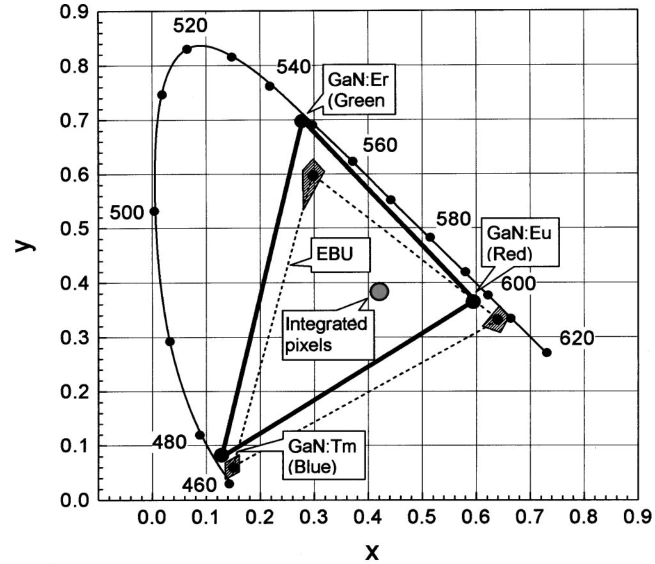

FIG. 5. CIE $x-y$ chromaticity diagram showing the locations of the blue, green, and red emission from the individually biased pixels in an integrated device and from simultaneously biasing all three pixels in the device. Also shown are the coordinates of the EBU-recommended phosphors.

sists of two strong lines at 537 and 558 nm, which provide the green emission color. The two green lines have been identified as Er transitions from the $^2H_{11/2}$ and $^4S_{3/2}$ levels to the $^4I_{15/2}$ ground state.[6,7] The GaN:Eu emission contains a major peak at 621 nm from the $^5D_0$ to $^7F_2$ transitions of the $4f$ shell of Eu^{3+} ions.[9] Minor peaks at ~540 nm and around 600 nm add a green-yellow component to the main Eu red peak at 621 nm. The combined simultaneous emission spectrum from all three pixels is shown in Fig. 4(b). The EL brightness of the red, green, and blue pixels from the three-color integrated device is approximately 14, 45, and 3 cd/m^2, respectively.

Figure 5 illustrates the full color capability of RE-doped GaN TFEL integrated devices using the Commission International d'Eclairage (CIE) chromaticity diagram. The blue emission has CIE coordinates of (0.13, 0.09). The CIE coordinates of the green and red emission are (0.28, 0.70) and (0.60, 0.37), respectively. The combined emission from all three integrated pixels results in CIE coordinates of (0.42, 0.38). These coordinates correspond to Planck emission at a temperature of ~3000 K and a color rendering index of ~85. The solid triangle in the diagram connects and defines the full color capability of emission from GaN doped with Tm (blue), Er (green), and Eu (red). The CIE coordinates recommended by the European Broadcasting Union (EBU) are given by the dashed line triangle.

In summary, a lateral integration method was investigated to realize blue, green, and red color integration on RE-doped GaN electroluminescent thin films. Spin-on-glass films were used as sacrificial layers for a lift-off process. The CIE coordinates of the GaN:RE phosphors demonstrate their capability for full color displays.

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