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Room-temperature-grown rare-earth-doped GaN luminescent thin films

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Visible emission has been observed from rare-earth (RE)-doped GaN electroluminescent devices (ELDs) as-grown near room temperature on Si (50–100 °C): red from GaN:Eu, green from GaN:Er, and blue from GaN:Tm. Green emission at 537/558 nm from GaN:Er ELD had a measured brightness of ~230 cd/m^2 at 46 V bias. X-ray diffraction indicates that the low-temperature-grown GaN:Er structure was oriented with the *c* axis perpendicular to the substrate. Scanning electron and atomic force microscopy indicate that the films had a rough surface and a compact structure consisting of small grains. Electroluminescence intensity of GaN:RE was significantly improved with postgrowth annealing. For GaN:Er films, after 800 °C annealing, the green emission brightness efficiency increased by ~10×. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1406138]

Rare-earth (RE)-doped GaN has been shown[1] to be an extremely versatile optoelectronic material, with light emission throughout the visible spectrum and at important near-infrared wavelengths. GaN thin films are usually grown at relatively high temperature, such as 1000–1100 °C for chemical vapor deposition and 600–800 °C for molecular beam epitaxy (MBE). The standard MBE process for *in situ* RE doping of GaN has resulted[2] in the successful fabrication of electroluminescent devices (ELDs) with red, green, and blue color emissions using Eu, Er, and Tm, respectively. Growth of luminescent GaN:RE thin films at much reduced temperatures has many potential advantages, such as low cost, a simple process, many deposition options, and possible deposition on Si-based circuitry. Low-temperature growth of RE-doped GaN and resulting amorphous or polycrystalline films has certain advantages over high-temperature grown crystalline GaN: much lower stress due to lattice and thermal expansion mismatch, films which are relaxed after growth and can be easily scaled up for commercial production. Several groups have reported efforts to achieve this goal with GaN growth on oxide films or on glass substrates[3–6] being a main focus. For example, Yamada *et al.* recently reported[7] the successful growth of polycrystalline GaN on various substrates at high temperature. Gurumurugan *et al.* reported[8] visible cathodoluminescence and Dimitrova *et al.* reported[9] electroluminescent (EL) emission from amorphous AlN:Er films. In this letter, we report on visible EL emission—red, green, and blue—from GaN:RE thin films grown in the vicinity of room temperature: 50–100 °C.

GaN films were grown on *p*-type (111) Si substrate by MBE with a Ga elemental source and a nitrogen plasma source. Doping of individual RE elements (Er, Eu, and Tm) was performed *in situ* during growth from solid sources. GaN:RE layers were typically grown for 1 h, resulting in ~1 μm thickness. Reflection high energy electron diffraction during growth showed ring patterns which indicate amorphous or polycrystalline films. For EL measurements a simple ring-shaped Schottky electrode was fabricated on top of the GaN:Er film using indium–tin–oxide (ITO) sputtering and a lift-off process. The ITO film has more than 90% transmittance over the whole visible light range. The electrode has an area of 7.65×10^{-4} cm^2 and its detailed structure is reported elsewhere.[10]

We have obtained red, green, and blue emission from ELDs on GaN doped with Eu, Er, and Tm, respectively. Figure 1 shows the EL spectra from GaN:RE ELDs grown at 100 °C. Red light at 621 nm comes from the characteristic emission of Eu^{3+}, namely the $^5D_0\rightarrow{}^7F_2$ $4f-4f$ transition. Two green emissions, at 537 and 558 nm, are caused by two Er^{3+} transitions: $^2H_{11/2}\rightarrow{}^4I_{15/2}$ and $^4S_{3/2}\rightarrow{}^4I_{15/2}$, respectively. Blue emission at 477 nm comes from the Tm^{3+} transition: $^1G_4\rightarrow{}^3H_6$.

Figure 2 shows current–voltage ($I-V$) characteristics under dc bias and the brightness of a GaN:Er ELD. We have obtained a brightness of ~230 cd/m^2 at 46 V, which is higher than the requirement for cathode-ray tubes (~100 cd/m^2). The current flow, however, was significantly higher than from GaN:Er devices grown at high temperature. It was about 60 mA at 46 V, while in high-temperature-grown GaN:Er ELDs the same applied bias of the current flow is

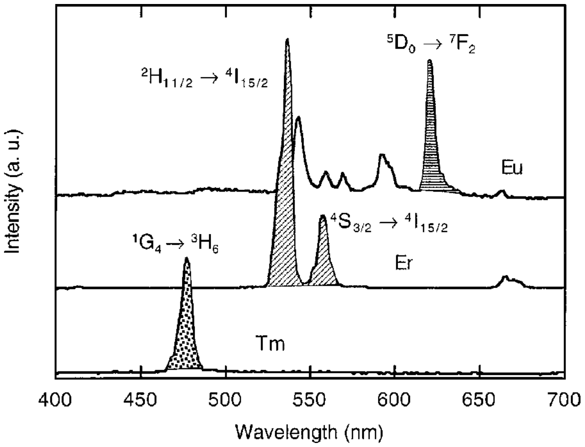

FIG. 1. EL emission spectra from ELDs on rare-earth-doped GaN as-grown at 100 °C. Red from GaN:Eu, green from GaN:Er, and blue from GaN:Tm. Characteristic $4f-4f$ innershell transitions are specified for each emission peak.

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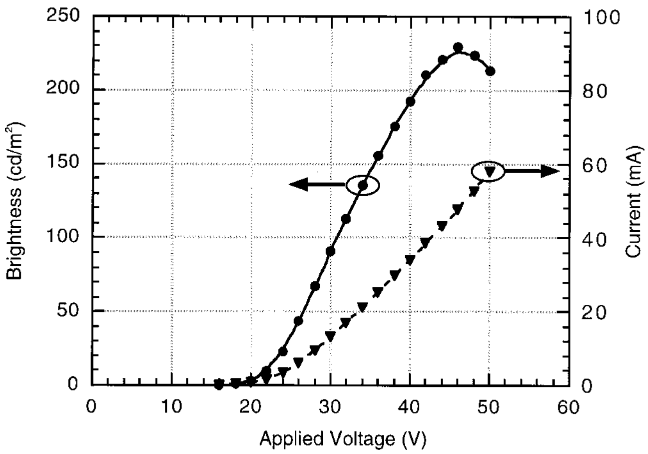

FIG. 2. $I-V$ characteristics of a GaN:Er ELD and its corresponding brightness. Measured brightness was ~230 cd/m^2 under 46 V of negative bias.

usually 1 order of magnitude smaller. One possible explanation for this may come from the high density of defects and dislocations present in low-temperature-grown GaN:Er, resulting in electrically leaky characteristics.

We have performed x-ray diffraction (XRD) measurements to investigate the GaN crystalline quality. Figure 3(a) shows the preferential (0002) orientation of hexagonal GaN at $2\theta = 34.5°$, indicating that *c*-axis preferred growth occurs even at this low temperature. However, the intensity is very weak compared to the Si(111) peak, which indicates that the film is weakly polycrystalline. Scanning electron microscope (SEM) and atomic force microscope (AFM) images of a GaN:Er film are contained in Fig. 3(b). Both images show that the film has a fairly rough surface with small grains. The SEM cross section exhibits a tightly packed film with columnar grains. This is consistent with the XRD of preferential growth orientation perpendicular to the substrate surface. The small grains are probably due to the low Ga surface mobility at this very low growth temperature. The resulting grain size seems to be in the range of 10–100 nm. The rms roughness of this film measured by AFM was 9.4 nm, which is ~4 times higher than that of high-temperature-grown GaN:Er. To investigate the material characteristics of the film in more detail, we have carried out Fourier transform infrared (FTIR) measurement. The FTIR spectra of GaN:Er films grown both at low (100 °C) and high (700 °C) temperature are shown in Fig. 4. Each has a peak at 573 cm^{-1}, which is attributed to Ga–N stretching vibration in the hexagonal GaN crystals and is similar to results obtained for GaN on Si (111) using a BN buffer layer.[11,12]

Postgrowth annealing was investigated at temperatures from 600 to 1000 °C, in 100 °C steps. The films were annealed for 2 min using rapid thermal annealing under N$_2$

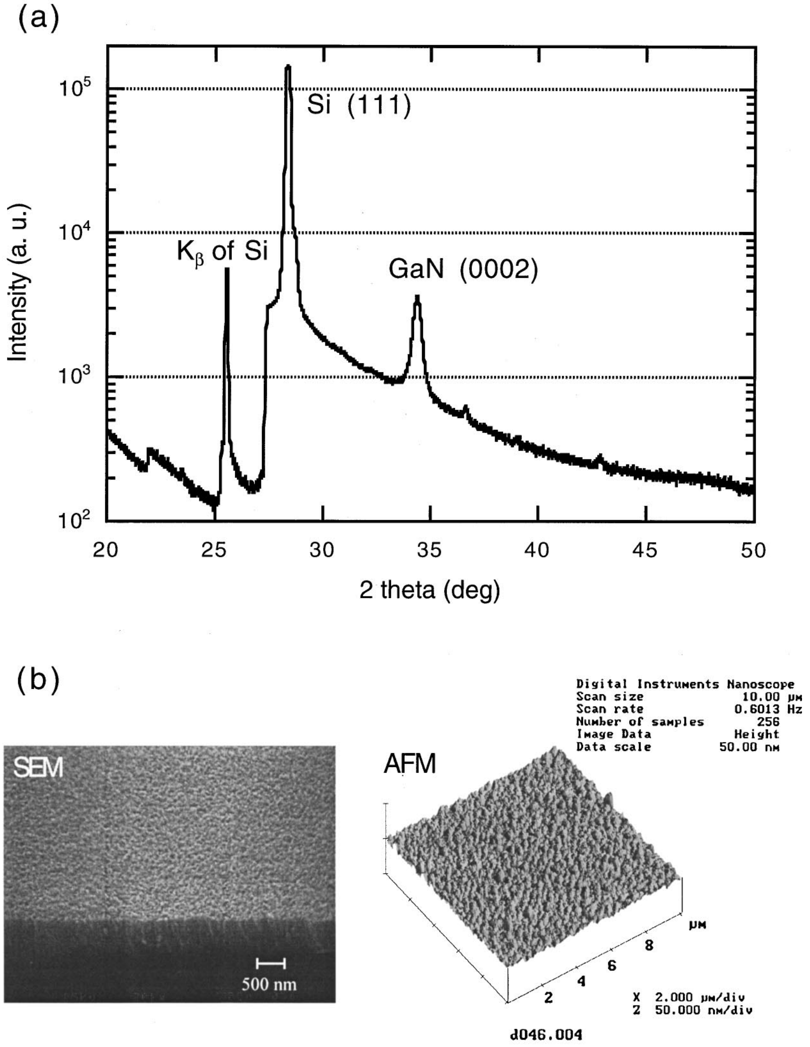

FIG. 3. Crystallinity and morphology of low-temperature-grown GaN: (a) XRD spectrum, showing preferential (0002) orientation of hexagonal GaN; (b) SEM and AFM images, showing rough surface with small columnar grains.

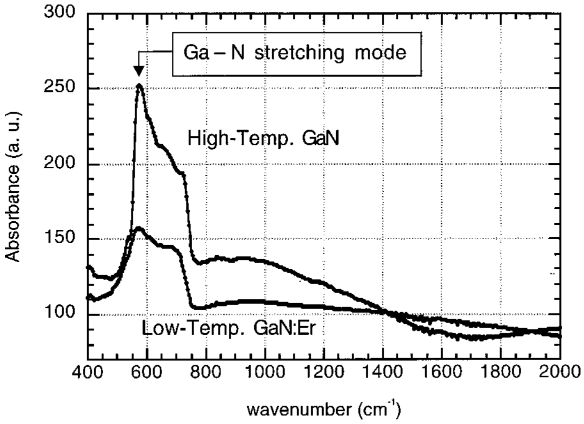

FIG. 4. FTIR spectra of a low-temperature-grown GaN:Er film and a high-temperature-grown GaN film as a reference. Peak at 573 cm^{-1} is attributed to Ga–N stretching vibration in the hexagonal GaN crystal.

environment. The resulting normalized brightness versus applied voltage is plotted in Fig. 5 for different anneal temperatures. BIV is defined as "brightness normalized by current flow versus voltage." Since the EL brightness was influenced by many factors including those associated with EL device fabrication, the current-normalized brightness is a more appropriate parameter to evaluate rather than the raw brightness, since it provides an indication of light emission independent of current flow. The brightest EL emission was obtained from 800 °C-annealed films and was about ~7 times higher than that from the as-grown film. With increasing temperature above 800 °C, the brightness decreased and finally no emission was detected at 1000 °C. XRD measurements performed before and after annealing found no change in the XRD intensity. The brightness improvement after annealing could come from increased Er activation in GaN, as reported[8,9] in the case of AlN:Er deposited by sputtering followed by high temperature annealing. Another possibility is that annealing may improve the crystalline quality only near the surface region. More work is needed to explain this improvement.

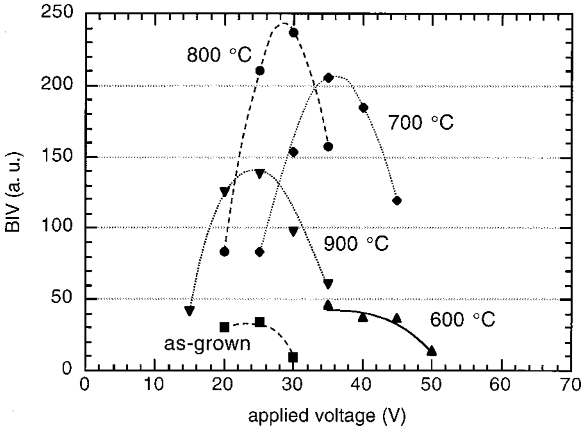

FIG. 5. BIV vs applied bias for GaN:Er ELDs annealed at various temperatures. Maximum brightness was obtained after annealing at 800 °C and was about 1 order higher than that of as-grown material.

In summary, we have shown that optically active GaN:RE films can be grown at surprisingly low temperatures (≤100 °C). EL emission was observed from as-grown films. It appears that the N_2 rf plasma and Ga evaporation cell provide enough energy to form nanoscale grains of GaN. However, due to the low substrate temperature, migration of Ga adatoms on the surface is very limited. As a result, three-dimensional growth with very small grains is most probably achieved in this growth regime. Each GaN grain is still assumed to be subject to the conditions for good GaN "films." In other words, the factors affecting the quality of GaN films are also applicable to these small grains: stoichiometric growth (i.e., V/III ratio), rf-plasma power, and so on. More importantly, stoichiometric growth is assumed to be the key factor in obtaining strong EL-emitting GaN:RE even for low temperature growth.

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