Growth temperature dependence of optical modal gain and loss in GaN:Eu active medium on Si

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Abstract: The dependence of optical modal gain and loss on GaN:Eu growth temperature is reported. GaN:Eu thin films were grown on Si substrates with AlGaN transition and cladding layers at temperatures ranging from 600°C to 850°C. The modal gain and loss in the GaN:Eu layer were a strong function of the optically active Eu atomic concentration and of the interface quality between the active layer and the top cladding layer, which in turn depended on the growth temperature. Optimum optical properties of maximum modal gain of ~ 100 cm^{-1} and minimum loss of ~ 46 cm^{-1} were obtained for growth at 800°C.

OCIS codes: (140.3380) Laser materials; (160.5690) Rare earth doped materials

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1. Introduction

Rare earths (RE) ions display very narrow light emission lines due to inner shell transitions [1]. Light emitting applications [2] of RE-based lumophores range from phosphors in flat panel displays to amplifiers in telecommunications to solid-state lasers. Solid state laser systems based on RE ions are a versatile light source, primarily in the infrared.

The doping of GaN with trivalent rare earth ions (RE^{3+}) has been shown [3] to produce excellent emission at visible and infrared wavelengths. We have previously reported visible lasing action (λ = 620 nm) with in-situ doped GaN:Eu thin films grown by molecular beam epitaxy (MBE) on sapphire [4] and Si [5] substrates. The use of a Si substrate for GaN:RE laser operation will accelerate the development of Si-based "light chips" which integrate light sources, modulators, detectors, etc. In integrating GaN:RE and Si technology, one of the main concerns is the effect of GaN related thermal processing on the underlying Si devices. Therefore, establishing the growth temperature necessary for optimum lasing properties is very important.

In this letter, we report on how the GaN growth temperature affects the optical gain and loss of the GaN:Eu active layer through its materials characteristics . The GaN:RE growth temperature on conventional Si substrates is known to strongly effect photoluminescence (PL) properties for films deposited by either MBE [6] or sputtering [7]. Reflection high-energy electron diffraction (RHEED), X-ray diffraction (XRD), and scanning electron microscopy (SEM) were used to investigate the effect of growth temperature on material characteristics of GaN:Eu active layer.

2. Material preparation

The planar waveguide laser structures demonstrated here were grown and characterized at University of Cincinnati on AlGaN templates supplied by Nitronex that have (111) Si as substrate. The structure consists of a ~1μm thick $Al_xGa_{1-x}N$ multilayer stack with different Al compositions serving as bottom cladding and transition layers, a Eu-doped GaN film as active medium, and an $Al_xGa_{1-x}N$ top cladding layer. The bottom cladding and transition layer stack was grown on 100 mm float-zone Si (111) substrates in a vertical, cold-wall, rotating disc metal organic chemical vapor deposition (MOCVD) reactor. [8] The reactor design enables control of temperature and flow across multiple zones allowing for development of uniform processes. For the Group III elements, trimethylgallium (TMG) and trimethylaluminum (TMA) precursors were carried by Pd-diffused hydrogen. Ammonia (NH_3) was used as the N precursor. The resistivity of the substrate was >10 kΩ-cm. Growth was nucleated with AlN at 1020ºC. A strain-compensating (Al,Ga)N transition layer was employed to accommodate mismatch between the substrate and the epilayers. For the structure used in this study, the bottom cladding layer was unintentionally doped ~30% AlGaN with a thickness of ~ 250 nm. The alloy content of the bottom cladding layer was measured using room temperature photoluminescence.

The Eu-doped GaN active layer was grown at several growth temperatures from 600 to 850 °C for 1 hr in a solid source molecular-beam epitaxy (MBE) system, resulting in GaN:Eu layers of ~550nm. Other growth parameters were kept constant at 3.6×10^{-7} Torr for the Ga flux (880°C Ga cell temperature), 8.9×10^{-7} Torr for the Al flux (1025°C Al cell temperature), 470°C for the Eu cell temperature and 1.8 sccm N2 flow for nitrogen plasma using 400W RF power. The Ga cell temperature and resulting flux is known [8] to have an important role in the structural quality of GaN: RE film. In the results presented here for growth on modified Si substrate, the optimum Ga flux was in a relatively narrow range $(3.5 – 3.7) \times 10^{-7}$. The growth process was monitored by a 30keV reflection high-energy electron diffraction (RHEED) system. The top cladding layer was typically grown for 30min at the same temperature as that used for the growth of the GaN:Eu active layer. XRD measurements indicated that the Al composition in the top cladding layer of all sample had an approximately constant value (~30%). The final process step was annealing at 675°C for 100min.

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3. Results and discussion

We conducted *in-situ* monitoring of RHEED patterns to analyze the thin film structure and the interface roughness between the top cladding and the GaN:Eu active layer. Figure 1 displays RHEED patterns observed after GaN:Eu growth at different temperatures (but prior to the growth of the top cladding layer). The RHEED pattern can easily distinguish the crystalline structure of the thin film, such as cubic and hexagonal phases [10]. The RHEED patterns in Fig. 1 indicate that GaN:Eu films have hexagonal structure. The RHEED pattern of the starting AlGaN template (Fig. 1(a)) shows the streaky pattern characteristics of a well-ordered smooth surface. At low growth temperature (600 and 650°C), the RHEED patterns [Fig. 1(b) and (c)] show spotty features indicating a three-dimensional growth that represents surface roughness on greater than atomic scale. GaN:Eu layer having this spotty pattern will likely exhibit crystal defects observable with XRD. As the growth temperature increases, the RHEED pattern begins to change from spotty to streaky. The sample grown at 800°C has such a streaky pattern [Fig. 1(e)], indicating two-dimensional growth and a return to a smooth surface [10]. Above this temperature, the surface becomes somewhat irregular, resulting again in a spotty pattern. The occurrence of the spotty pattern at 850°C [Fig. 1(f)], could be explained by the migration [10] of excess nitrogen species (N-rich growth condition) due to Ga desorption on the surface at this temperature.

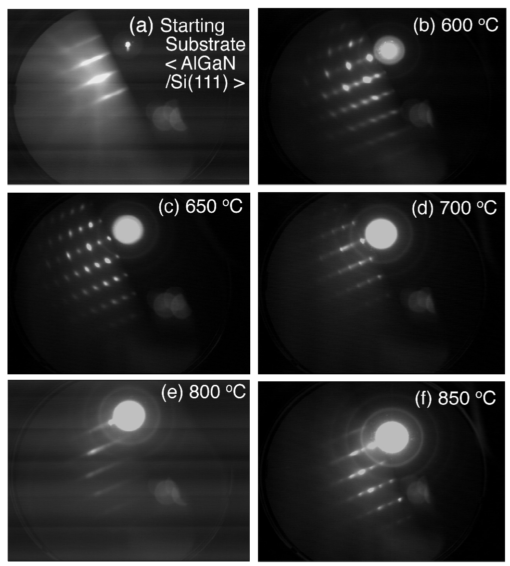

Fig. 1. RHEED patterns of Eu doped GaN on Si substrate at different temperatures: (a) starting substrate < AlGaN/Si(111) >; (b) 600°C; (c) 650°C; (d) 700°C; (e) 800°C; (f) 850°C;

XRD measurement was also used to analyze the structural quality of the GaN:Eu films. The XRD spectra of the films showed the same peak position at $2\theta = 34.5°$ for all growth temperatures, indicating that the films have the preferential (0002) orientation of hexagonal GaN and is in agreement with the RHEED data. The variation of peak intensity and the full width at half maximum (FWHM) of the peak are plotted in Fig. 2. For growth at low temperature (600 - 650°C), the XRD data shows a relatively weak and broad peak that indicates an amorphous to polycrystalline structure [6]. As the growth temperature increases to 700°C, the structural quality of the GaN:Eu film is greatly improved with much narrower FWHM and increased XRD peak intensity. Figure 2 points to a minimum growth temperature for high quality GaN:Eu film on AlGaN/Si templates. By comparison, GaN:RE film grown directly on Si experience a gradual structural improvement with growth temperature [6].

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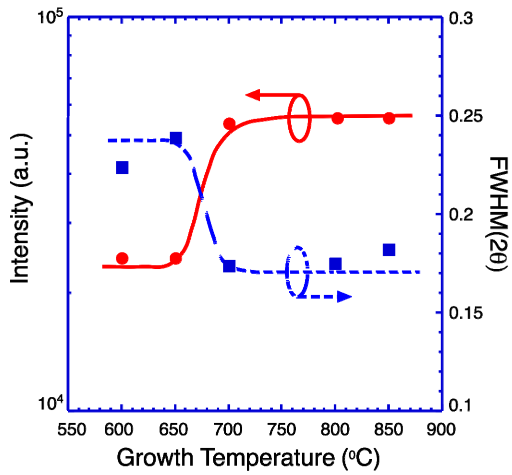

Fig. 2. XRD peak intensity and FWHM variation as a function of growth temperature.

Figure 3 contains SEM microphotographs of the cross-section of GaN:Eu thin films grown at different temperatures. The films grown at low temperature (600 - 650°C) have highly columnar structure and rough interfaces [Fig.3 (a), (b)]. This result is consistent with the corresponding RHEED patterns [Fig.1 (a), (b)] and is supported by a weak and broad XRD peak at the same growth temperatures (Fig. 2). Above 650°C, the GaN:Eu film acquires a more dense structure and has a well-defined interface with the AlGaN top cladding layer. As expected, the the MBE grown GaN:Eu film displays a more compact crystal structure than that deposited by the sputtering method. [7] The cross-section of the structures as revealed in the SEM photographs of Fig. 3 support the existence of abrupt transition of structural quality of film indicated in the XRD data of Fig.2.

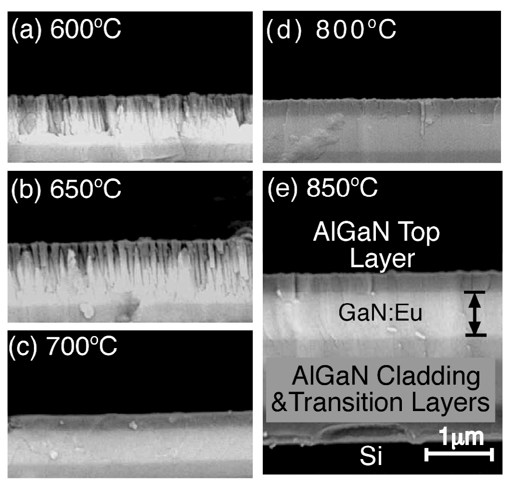

Fig. 3. SEM photographs of the cross-section of the planar waveguide structure for different GaN:Eu growth temperatures: (a) 600°C; (b) 650°C; (c) 700°C; (d) 800°C; (e) 850°C

The optical modal gain and loss of GaN:Eu films having different growth temperatures was measured at the peak Eu wavelength (~620nm) by the variable stripe length method [11] and the shifting excitation spot (SES) method [12], respectively. As shown in Fig. 4, the modal gain increases rapidly (~10×) for growth temperatures from 600°C to 800°C and then starts to decrease for growth at 850°C. The maximum modal gain obtained is ~ 100 cm^{-1}. The total Eu atomic concentration was roughly the same for all films at ~1 at.%. However, the reason for the modal gain vs. growth temperature trend can be partially explained by the variation of the *optically active* Eu concentration. The insert to Fig. 4 shows the Eu PL intensity over the same growth temperature range. The PL intensity, which is proportional to the active Eu concentration, shows a similar trend with growth temperature, as does the optical modal gain. This indicates that the number of incorporated active Eu ions in GaN:Eu is a strong function of temperature based on the proportional relation between PL intensity and the number of active ions.

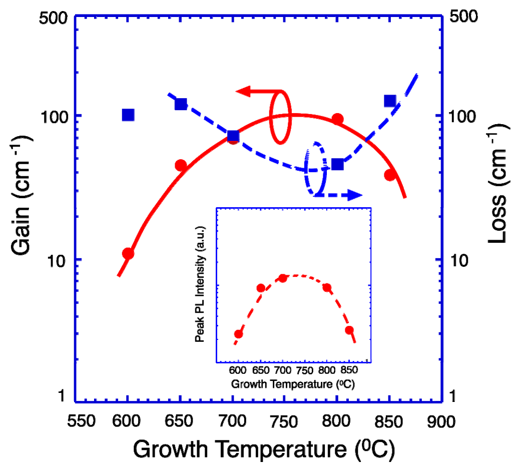

Fig. 4. Optical modal gain and loss in GaN:Eu planar waveguides grown at various temperatures. Inset shows the peak PL intensity as a function of growth temperature.

The trend in optical loss versus growth temperature, also shown in Fig. 4, is nearly opposite to that of the modal gain. The loss decreases with growth temperature up to 800°C and then starts to increase. The high loss at low temperature could be due to a combination of the rough interface properties caused by 3D growth [Fig.1 (a), (b)] and optical scattering at the boundary of columnar grains [Fig. 3 (a), (b)]. At a growth temperature of 700°C, the GaN:Eu film has good crystalline quality [Fig. 2] and a compact structure [Fig. 3 (c)]. The RHEED pattern, however, shows that the interface is still rough due to 3D growth, which therefore results in a relatively high loss. The sample grown at 800 °C has the minimum loss value, followed by an increase in loss at 850 °C. The growth temperature dependence of the optical loss can be explained by the interface quality. The scattering induced by surface roughness is the main loss component compared to other factors [13], such as substrate leakage loss, modal mismatch loss, etc. The structure that has high interface quality also has minimum loss. This also explains the difference in the growth temperature that produces the highest modal gain and the highest PL intensity. Although the active Eu concentration has a (slight) maximum for growth at 700°C, the highest modal gain occurs for growth at 800 °C because of the much improved interface quality.

4. Conclusion

In summary, we have determined the growth temperature dependence of optical modal gain and loss in the GaN:Eu active medium grown on Si with AlGaN transition/cladding layers. The quality of the GaN:Eu layer in terms of interface roughness and crystallinity strongly depends on the growth temperature. The concentration of optically active Eu ions appears to have a maximum for growth temperature from 650 to 800°C. The modal gain was strongly affected by the active Eu concentration. Interface and structural quality are the main factors controlling loss and modal gain in the GaN:Eu layer. The growth temperature of 800°C seems to be optimal condition to obtain the highest modal gain and lowest loss value, as well as a compact structure and smooth interfaces.

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