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Eu³⁺ luminescence properties of Eu- and Mg-codoped AlGaN



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ABSTRACT

We investigated the effect of Mg codoping on luminescence properties of AlGaN:Eu to improve emission through synergy effect between an increase in bandgap by AlGaN and the Mg codoping technique. The luminescence properties of AlGaN:(Eu, Mg) are strongly influenced by the Mg concentration and Al composition. Mg codoping in AlGaN was observed to contribute to increasing photoluminescence (PL) integrated intensity and to improve thermal quenching from 7.3% to 60% while the dominant optical site remained site B (622.3-nm peak) with low excitation cross section. The total concentration of optically activated Eu at 25 K was a constant at for either optical site, indicating that Mg codoping did not affect the formation of optical sites. The PL decay times at room temperature (RT) increased with Mg concentration because of suppression of the back-transfer process. For optimized Mg concentration, an increase in the Al composition contributed to the total activated Eu concentration and changed the dominant optical site from A (620.3-nm peak) to B. The activation energy E_{a} , which is the difference in energy between the ⁵D₀ energy level and the trap level in the host material, was estimated from temperature dependence of PL decay time. The E_a for site A was larger than that for site B, suggesting that the back-transfer rate for site A was less than that for site B.

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1. Introduction

Electronic materials embedded with rare-earth (RE) ions are widely applied in optoelectronics devices, such as YAG:Nd solidstate laser, display phosphors and Er-doped optical-fiber amplifiers. The RE ion luminescence is based on the 4f-4f transition of innershell electrons shielded by outer shell electrons, showing a sharp line emission and thermal stability of the emission wavelength. However, miniaturization and integration of optical devices based on conventional RE-ion material are difficult because of the inability to excite RE ions through current injection. RE-doped semiconductors are potential candidates for realizing next-generation devices, such as optical devices with thermally stable emission wavelength and opto-spintronic devices. When a narrow-bandgap semiconductor (Si, GaAs, etc.) is selected as the host material, large thermal quenching and low photoluminescence (PL) intensity were observed at room temperature (RT) were observed [1]. Therefore, RE-doped nitride semiconductors with wide bandgaps have attracted attention for improving luminescence properties. Samples

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http://dx.doi.org/10.1016/j.jlumin.2015.04.036 0022-2313/© 2015 Elsevier B.V. All rights reserved. have been prepared through ion implantation and annealing [2,3], in addition to crystal growth through molecular beam epitaxy (MBE) [4–8] and organometallic vapor phase epitaxy (OMVPE) [9]. RGB-color-integrated RE-doped GaN thin film electroluminescence (EL) devices, and light-emitting high electron-mobility transistors (HEMTs) with spatially selective Eu doping region have been fabricated [10,11]. Light-emitting diodes (LEDs) with Eu doped GaN (GaN:Eu) and Er doped InGaN active layers showing red and infrared emission have been demonstrated [12–14]. For further development of device performance, a fundamental technology for optically activating RE ions in a crystal is required.

To develop such technology, selection of the host material and control of optical sites are key factors. It is well known that an increase in the bandgap of the host material contributes to suppressing the thermal quenching of PL intensity [1]. In fact, when the optical properties of Eu-, Tb-, Er-, and Tm-implanted AlGaN were evaluated, an increase in the Al composition resulted in an increase in the PL intensity from RE ions at RT owing to the suppression of the back-transfer process [2,15–17]. Therefore, the use of AlGaN with larger bandgap is promising to improve the luminescence efficiency. On the other hand, several emission peaks were observed in the Eu luminescence in GaN corresponding to ${}^5D_0-{}^7F_2$ electron transition, indicating the formation of several

optical sites [18–22]. From combined excitation–emission spectroscopy (CEES), it was clarified that each optical site in GaN has a different excitation and emission efficiency [19]. Therefore, the control of optical sites can contribute to improve the optical properties. For Er-doped GaAs, oxygen codoping facilitated the formation of a specific local structure and resulted in the unification of the emission peak [23]. We developed an Mg-codoping technique for GaN:Eu grown by NH₃-source MBE, which achieved the selective activation of one site and enhancement of PL intensity from Eu ions at RT [24,25]. The improvement of optical properties by Mg codoping has also been reported for OMVPEgrown Eu- and Mg-codoped GaN (GaN:(Eu, Mg)) and Eu-implanted Mg-doped GaN [26,27].

Based on these reports, in this study, we investigated the effect of Mg codoping on optical properties of AlGaN:Eu to improve emission through synergy effect between an increase in bandgap by AlGaN and the Mg codoping technique.

2. Effect of Mg codoping on optical properties of Eu-doped AlGaN

The samples were grown on a GaN template by using NH₃-source MBE. The group-III metals Ga (6N), Al (5N), and Eu (3N) as well as the Mg (6N) dopant were supplied from effusion cells. The beam equivalent pressure (BEP) of Ga, Al, and Eu were 6×10^{-7} , 1.5×10^{-7} , and 6×10^{-8} Torr, respectively. After cleaning with an organic solvent, the wafers were loaded into a chamber, and thermal cleaning was performed at 825 °C for 10 min. Subsequently, a GaN buffer layer was grown for 10 min, followed by the growth of an AlGaN:(Eu, Mg) layer with a NH₃ flow of 3.0 sccm for 1 h. Al composition of AlGaN:(Eu, Mg) was determined to be 11% from the XRD pattern. The Eu concentration was estimated to be 2×10^{20} cm⁻³ by referring to the Eu concentration of Eu-doped GaN under the same growth condition of the quaternary alloys. Here, to investigate the dependence of optical properties on the Mg concentration (N_{Mg}) , the Mg concentration was varied from 1.2×10^{17} to 8×10^{18} cm⁻³, which was estimated by using the secondary ion mass spectrometry (SIMS). In the PL measurement, a He-Cd laser (325 nm) and Xe lamp with band-pass filter (400 nm) were used as excitation sources.

First, PL measurements of GaN:Eu and $Al_{0.11}Ga_{0.89}N$:Eu were performed to clarify the effect of the bandgap of the host material on the PL properties. Fig. 1 shows PL spectra at RT for GaN:Eu and

AlGaN:Eu. Sharp emission corresponding to ${}^{5}D_{0}-{}^{7}F_{2}$ electron transition was observed around 620 nm for both samples. For GaN:Eu, three dominant peaks at 620.3 nm (shoulder peak), 622.3 nm and 633.8 nm were observed which are labeled peaks or sites A, B, and C for convenience, respectively. An additional other peak was observed at 625.5 nm. The difference in photon energy between peak B and this additional peak was approximately 12 meV, which corresponding to the value of energy localized to the Eu incorporation sites [19]. Therefore, the peak at 625.5 nm can be concluded to be a phonon replica of peak B, and it is labeled peak B.

On the other hand, for AlGaN:Eu, a peak at 617.6 nm was observed in addition to peaks A, B, and C, additional peak is labeled peak or site D. To classify the optical sites according to the excitation process, the PL spectra of AlGaN:Eu sample under above-bandgap excitation (λ_{ex} =325 nm) and below-bandgap excitation (λ_{ex} = 400 nm) were evaluated at RT. Peaks A, B, C, and D were observed under above-bandgap excitation, while only peak B was observed under below-bandgap excitation. This result suggests that the excitation process for sites A, C, and D is different from that for site B. The behaviors of optical properties corresponding to sites A and C were similar in this study. For example, the ratio of the PL intensity of peak A to that of peak C was almost constant among all samples despite of Mg concentration and Al composition. The excitation cross sections of sites A and C were almost the same. The origin of site D was not clear at this stage. However, the emission intensity corresponding to site D was sufficiently small compared with that of the dominant peak for either sample. Therefore, the optical property was discussed by focusing on the difference in behavior between sites A and B.

The PL integrated intensity for AlGaN:Eu was 3.5 times as strong as that for GaN:Eu. To evaluate the effect of thermal quenching, the temperature dependence of PL was measured from 25 K to 300 K. The ratio of PL integrated intensity at 25 K to that at 300 K for AlGaN:Eu and GaN:Eu was 7.3% and 3.6%, respectively. Additionally, the ${}^{5}D_{0}$ -level lifetime at RT estimated from the time-resolved PL measurement for AlGaN:Eu and GaN:Eu were 30 and 17 μ s, respectively. Therefore, AlGaN proved effective to improve the luminous efficiency.

Next, the optical properties of AlGaN:(Eu, Mg) with different Mg concentrations were evaluated to investigate the synergy effect between Mg codoping and a wider bandgap. Fig. 2 shows the PL spectra at RT for AlGaN:(Eu, Mg) with different $N_{\rm Mg}$ and GaN:(Eu, Mg) with an optimized $N_{\rm Mg}$ of 3×10^{18} cm⁻³ as a



Fig. 1. PL spectra at RT for GaN:Eu and Al_{0.11}Ga_{0.89}N:Eu.

Fig. 2. PL spectra at RT for Al_{0.11}Ga_{0.89}N:(Eu, Mg) with different Mg concentrations and GaN:(Eu, Mg) with optimized an N_{Mg} of 3×10^{18} cm⁻³.

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