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Photoluminescence study on Eu-implanted GaN

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The photoluminescence (PL) properties of Eu-implanted GaN thin films are studied. The experimental results show that the PL intensity is seriously affected by ion implantation conditions. The PL efficiency increases exponentially with annealing temperature increasing up to a maximum temperature of 1050°C. Moreover, the PL intensity for the sample implanted along the channelling direction is nearly twice more than that observed from the sample implanted along the random direction. The thermal quenching of PL intensity from 10K to 300K for sample annealed at 1050°C is only 42.7%.

Keywords: photoluminescence, metalorganic chemical vapour deposition, implanting, gallium nitride
PACC: 7855, 8115H, 6170T, 7280E, 7830L

1. Introduction

Rare earth ions doped materials have been widely studied for both optoelectronic and electronic applications.[1] Rare-earth ions doped in GaN can act as effective luminescence centres, which have a narrow linewidth and very small thermal quenching properties on account of intra-4f core-level transitions. Recently, Eu-doped GaN has been attracting much interest for its red-light emission , which has not been realized in commercial light-emitting diodes (LED) made from InGaN layers because indium enriched InGaN crystal is very difficult to prepare.[2−4] Eu-doped GaN can produce visible red emission at 621nm.[5,6] Although the Eu dopant introduced during molecular beam epitaxy of GaN has been studied, only a few papers have reported on PL properties of Eu implanted GaN.[7,8] The ion implantation doping method has more advantages. As we know, ion implantation can realize the selective doping into certain surface areas of a sample, the introduction of a large number of elements, and the precise control of dopant concentration and depth distribution. However, the implantation method needs annealing to achieve electrical or optical activation of rare earth ions and remove the damages induced by the ion implantation. Therefore, a lot of work needs to be done in order to study the optimal implantation and annealing conditions, and explore the fundamental features of Eu luminescence.

Nakanishi *et al*[9] have studied the influence of implantation conditions on luminescence features of Eu-doped GaN. However, further study needs to be done in order to explore the influence of some other implantation conditions. In this paper, we report that channelled ion implantation can enhance the PL intensity of Eu^{3+} greatly. Furthermore, we find that the implantation dose, annealing temperature and measuring temperature can affect the PL intensity and the structure of Eu-implantation GaN seriously.

2. Experiment

The GaN layer (∼1.0μm thick) used in this experiment were grown on a c-Al_2O_3 substrate by metal organic chemical vapour deposition (MOCVD) at 1000°C. Eu was implanted into GaN thin film at doses ranging from 5×10^{13} to 5×10^{15}cm^{-2} at the energy of 400keV. Random (at an angle of 10° to the c-axis, RI) and channelling implantations (along the c-axis, CI) were studied. After the implantation, the samples were cut into small pieces. The implanted GaN samples were annealed in a conventional resistance heating furnace for 30 min in nitrogen atmosphere at temperatures ranging from 800°C to 1150°C.

Photoluminescence (PL) was measured with the sample irradiated by a 325nm He-Cd laser at temperatures between 10K and 300K using a closed cycle helium cryostat. The incident power was about 10mW. The PL signal was collected and analysed with a JY-

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HRD1 double-grating spectrometer equipped with an RCA-C31034 photo-multiplier tube in a PAR-1140A photon counting mode.

3. Results and discussion

3.1. Dependence of PL intensity on ion implantation geometry

The so-called channelling implantation (CI) means that incident Eu^+ ions are implanted into a sapphire substrate along its $\langle 0001 \rangle$ axis, and the random implantation (RI) refers to the case where Eu^+ ions are incident at an angle of larger than 10° to the $\langle 0001 \rangle$ axis of sapphire substrate. PL spectra measured from channelling-implanted and random-implanted samples at room temperature are shown in Fig.1. The two samples both were implanted with $1\times10^{15}cm^{-2}$ Eu^+ ions and annealed at 1050°C for 30 min. At least four distinct transitions of the 4f shell can be seen in Fig.1. And their emission wavelengths are 543nm, 600nm, 621nm and 663nm respectively. Moreover, the strong red emission peak has two satellite peaks at 626nm and 630nm. A comparison of the peak positions with the standard energy diagram of Eu^{3+} indicates that the peak at 621nm should be attributed to the transition between the $^5D_0$ and $^7F_2$ states of Eu^{3+}. And the satellite peaks result from stark splitting of the levels corresponding to the main transition between the $^5D_0$ and $^7F_2$ states. The emission at 543nm originates from the transition between the $^5D_1$ and $^7F_1$ states of Eu^{3+}, the emission at 600nm comes from the transition between the $^5D_0$ and $^7F_1$ states and the emission at 663nm results from the transition between the $^5D_0$ and $^7F_3$ states.

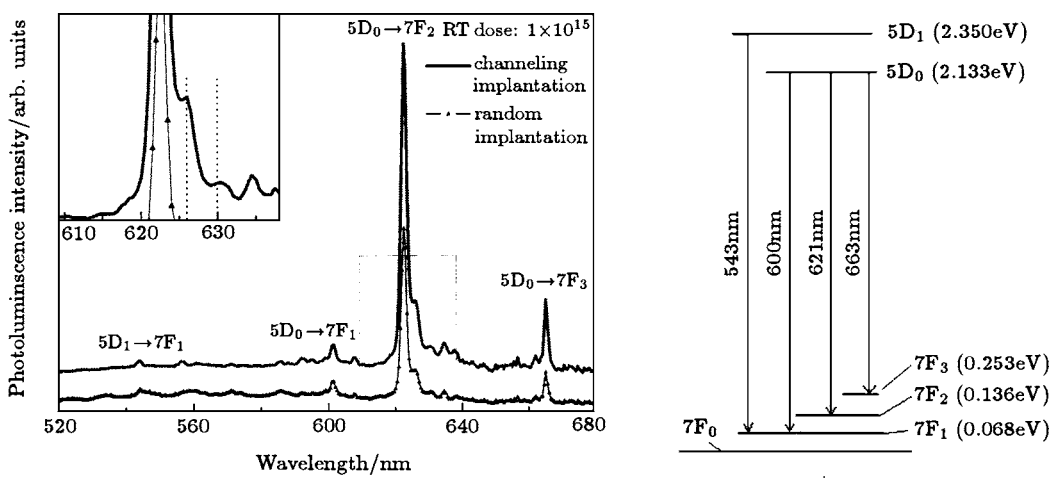


Fig.1. PL spectra of samples (annealed at 1050°C) implanted along channelling and random directions at room temperature.

It is also observed in Fig.1 that the PL intensity of the luminescence peaked at 621nm for the channelling-implanted sample is nearly twice higher than that for the random-implanted sample. In order to explore the dependence of PL on the implantation-induced damage, we continue to conduct Rutherford backscattering (RBS) measurement of the two samples. Three $\langle 0001 \rangle$ aligned RBS spectra and one random spectrum are illustrated in Fig.2. Of them, the backscattering yield of the aligned RBS spectrum of as-grown GaN is the lowest and has only one small Ga peak ($1^{\#}$ peak) coming from the surface layer, which indicates that crystalline lattice of the sample is quite perfect. There are two peaks ($1^{\#}$ peak and $2^{\#}$ peak) for random-implanted and channelling-implanted Eu–GaN samples. The $1^{\#}$ peak comes from the scattering from Ga atoms on the sample surface. The $2^{\#}$ peak corresponds to the direct scattering from displaced Ga atoms in the bulk. Both $1^{\#}$ and $2^{\#}$ peaks of the random-implanted sample are stronger than those of the channelling implantation, which means that the channelling implantation causes less damage to crystal than the random implantation. The main reason is that the channelling implantation can not only decrease the collision probability between implanted ions and host atoms, but also increase the implantation depth. Therefore, the channelling implantation can reduce the implantation damage and improve the Eu^{3+} photoluminescence effectively.

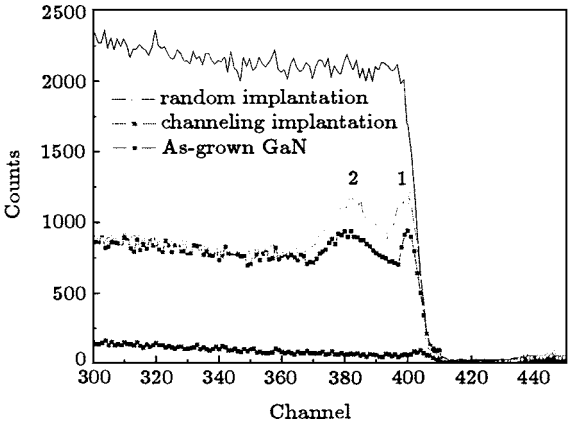


Fig.2. Random and ⟨0001⟩ aligned RBS spectra of as-grown sample, random-implanted sample and channelling-implanted sample after annealing at 1050°C.

3.2. Dependence of PL intensity on Eu^{3+} ion dose

The Eu^{3+} dose affects the Eu^{3+} PL intensity seriously. Eu^{3+} ions at doses ranging from 3×10^{14} to $5\times10^{15}cm^{-2}$ were separately implanted into different samples. It is found that the PL intensity from the sample increases with Eu^{3+} dose increasing. When the Eu^{3+} dose rises to $1\times10^{15}cm^{-2}$, the PL intensity increases up to a maximal value. However after this point, the PL intensity descends with Eu^{+} dose increasing. At a higher implantation dose, redundant Eu^{+} creates polycrystalline EuN compound, which not only enhances the Eu^{+} ions inter-collision process without light emission, but also is inactive to emit light by itself.[10] Generally, most of the red emission is evoked by the energy transferred from GaN, but the energy from GaN is not enough to excite Eu^{3+} in EuN to emit light. Furthermore, high dose implantation introduces such serious damages to sample that residual defects are still there even after annealing at high temperatures. The residual defects capture photo-carriers, causing the decrease in PL intensity.

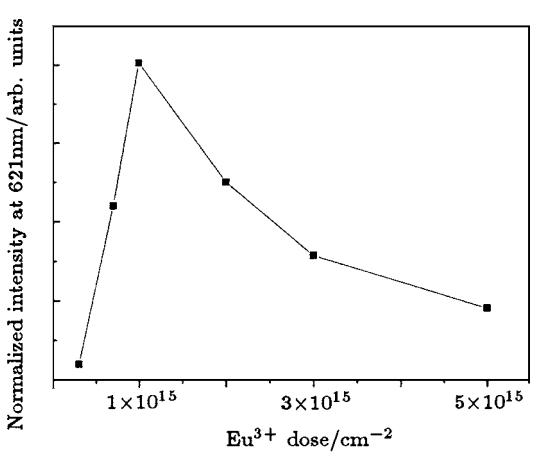


Fig.3. Photoluminescence (PL) spectra of Eu-implanted GaN with Eu dose.

3.3. Dependence of PL intensity on annealing temperature

Annealing temperature is a main factor to affect the PL emission intensity from Eu-implanted GaN samples. Figure 4 shows PL spectra measured from GaN samples implanted at a dose of 3×10^{15} cm^{-2} Eu^{3+} ions and annealed at temperatures ranging from 800°C to 1050°C in N_2 atmosphere. As can be seen from the figure, the peaks at 543nm and 600nm disappear because a luminescence quenching phenomenon takes place at the dose. And the peaks at 621nm and 663nm increase with annealing temperature increasing. The inset shows the integrated PL intensities as a function of annealing temperature for three transitions from 621 to 630nm. We find that the PL intensity increases exponentially with annealing temperature increasing up to 1050°C, and the further increase in annealing temperature causes the PL intensity to degrade. In the annealing temperatures ranging from 800°C to 1050°C, the implantation-induced damage can be gradually removed and Eu^{3+} ions are optically activated, thus the PL intensity increases with annealing temperature rising. However, the GaN thin film begins to dissociate at temperatures higher than 1050°C, causing the decrease in PL intensity.

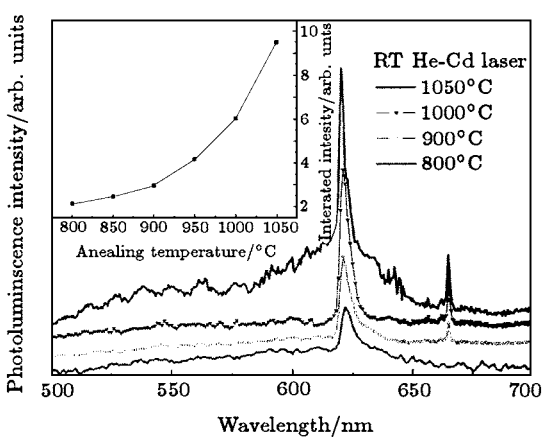


Fig.4. PL spectra measured from Eu-implanted GaN samples annealed at temperatures from 800°C to 1050°C. The inset shows the dependence of the integrated PL emission between 621 and 630nm at room temperature on the sample annealing temperature.

3.4. Dependence of PL intensity on sample temperature

Favennec *et al*[11] demonstrated a reduction in thermal quenching with the increase of band-gap energy of the semiconductor. As we know, GaN is a wide-bandgap semiconductor, we expect GaN:Eu to have a lower thermal quenching. Thermal quenching measurements of the GaN sample random-implanted with Eu and annealed at 1050°C at a dose of 1×10^{15} cm^{-2} was performed with a liquid He cooled coldfinger. Figure 5 shows the dependence of the Eu^{3+} emission on measuring temperature of the sample. Five distinct transitions at 543nm, 600nm, 621nm, 663nm and 703nm can be seen in the 10k spectrum, but only four distinct transitions at 543, 600, 621 and 663nm can be found in 300k spectrum. All transition line widths do exhibit thermal broadening. Figure 6 shows the dependence of each PL peak on measured temperature in detail. The integrated PL intensity at 621nm, 600nm and 663nm reduce to 57.3%, 84.7% and 92.6% when the temperature increases from 10K to 300K, respectively. The peak at 543nm is almost unchanged. This good thermal stability will be of benefit to the manufacture of high temperature optoeletronic devices.

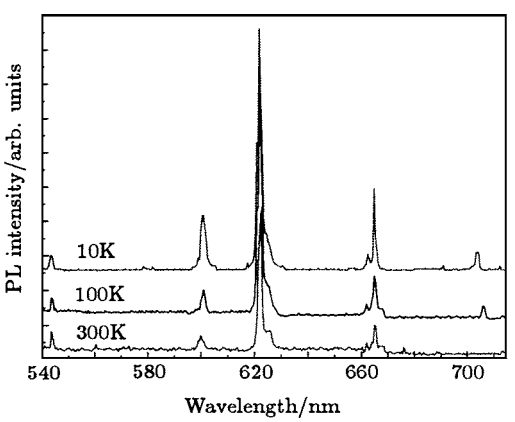


Fig.5. PL spectra measured at 300K, 100k and 10K from the Eu-implanted GaN sample (RI) annealed at 1050°C.

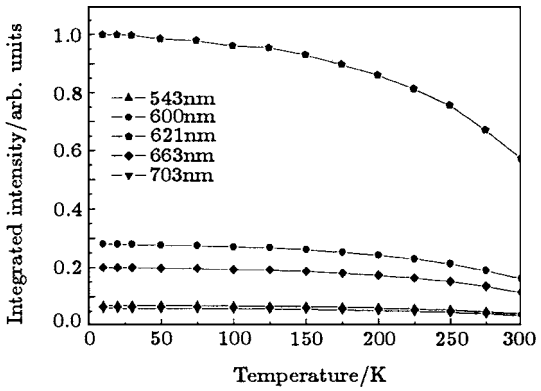


Fig.6. Temperature dependence of the integrated PL intensity at 543nm, 600nm, 621nm, 663nm and 703nm.

4. Conclusion

In the current work, we have studied photoluminescence properties of Eu-implanted GaN epilayers. The PL intensity is seriously affected by ion implantation geometry. The PL intensity for the sample implanted along the channelling direction is nearly twice more than that for the sample implanted along the random direction. The dependence of the PL intensity on Eu^{3+} dose is also studied. The PL intensity increases with the increase of dose when the dose is less than 1×10^{15} cm^{-2}, and decreases when the dose is more than 1×10^{15} cm^{-2}. The PL intensity increases exponentially with annealing temperature increasing up to 1050°C, and the further increase in annealing temperature causes the PL intensity to degrade. The thermal quenching of PL intensity for the sample annealed at 1050°C from 10K to 300K is 42.7.%.

Acknowledgement

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