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J. Phys.: Condens. Matter **16** (2004) S5743–S5748 PII: S0953-8984(04)79473-4

Optical properties of GaN-based magnetic semiconductors

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Received 17 April 2004
Published 19 November 2004
Online at stacks.iop.org/JPhysCM/16/S5743
doi:10.1088/0953-8984/16/48/040

Abstract
Optical properties of GaCrN, GaDyN and GaGdN have been studied by means of photoluminescence spectroscopy having spectral and time resolution. Photoluminescence (PL) emission at around 3.29 eV was determined to be the band-to-band transition of GaCrN. The PL decay time of GaCrN depends on emission energy. For GaDyN, Dy^{3+} ions in host GaN exhibited several sharp peaks originating from the intra-4f orbital transitions of the series of ${}^4F_{9/2}$–${}^6H_J$ ($J = 15/2, 13/2, \ldots$), and the lifetime of the ${}^4F_{9/2}$–${}^6H_{13/2}$ transition was as long as 87 μs at 13 K. The broad defect level for GaDyN provides an efficient pathway for the carrier-mediated energy transfer between Dy^{3+} ions and the GaN host. However, no such evidence is found between Gd ions and GaN in the GaGdN layer.

1. Introduction

Semiconductor-based spintronics may present a greater wealth of possibilities, which is different from metallic spin devices that just provide new ways to store and read information in hard disks, tapes or MRAM. Optical properties of diluted magnetic semiconductors (DMSs) are of particular interest to offer a pathway between a magnetic information and an optical signal. However, the low Curie temperature of DMSs disturbs their application. We have attempted to grow transition-metal- (TM-) doped and rare-earth- (RE-) doped GaN in order to improve the Curie temperature. Ferromagnetic order was observed in GaCrN, GaGdN and GaDyN samples even at high temperatures [1, 2]. In this paper, we present the optical properties of GaCrN GaDyN, GaGdN.

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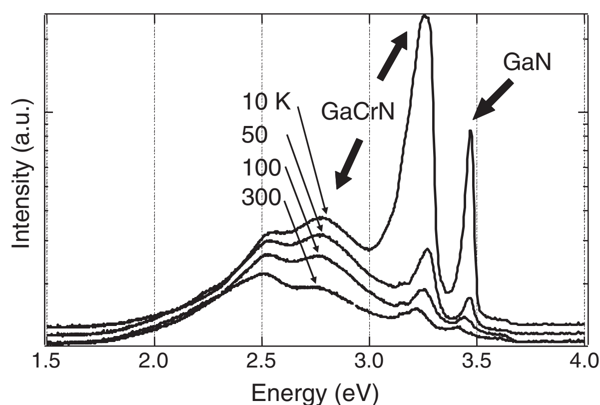

Figure 1. PL spectra for GaCrN sample with a Cr concentration of 0.5% from 10 to 300 K.

2. Experimental procedure

GaCrN epitaxial layers were grown on sapphire(0001) substrates by MBE with an ion-removal ECR radical cell [3]. Elemental Ga, Cr and ECR plasma-enhanced N_2 were used as sources. After thermally cleaning the sapphire substrate surface, a low-temperature GaN buffer layer was grown at substrate temperature (T_s) of 450 °C. On this low-temperature buffer layer, a GaN buffer layer was grown for 30 min at $T_s = 780$ °C. Finally, a GaCrN layer was grown at $T_s = 700$ °C for 1 h.

GaDyN and GaGdN epitaxial layers were grown on the (0001) Si face of a 6H-SiC substrate by radio-frequency plasma-assisted molecular-beam epitaxy (MBE). The layer structures were as follows: GaDyN (Dy $\sim$ 3%)(500 nm)/GaN(500 nm)/AlN(20 nm)/SiC substrate and GaGdN(Gd $\sim$ 6%)(250 nm)/GaN(500 nm)/AlN(20 nm)/SiC substrate. More detailed growth conditions are described elsewhere [4, 5]. On these samples, we have conducted the PL and time-resolved photoluminescence (TRPL) measurements. PL measurement was conducted with the 325 nm line of a 27 mW He–Cd laser as an excitation light source. For the time-resolved PL (TRPL), the wavelength, frequency, and full width at half maximum (FWHM) of the excitation light source were 266 nm, 1 kHz, and 150 fs, respectively.

3. Results and discussion

3.1. Optical properties of GaCrN

Figure 1 shows PL spectra for GaCrN sample from 10 to 300 K. The peak around 3.48 eV most likely corresponds to the exciton transition in the hexagonal GaN. A broad PL band with a maximum at about 2.9 eV (blue band) was observed in GaCrN, which originated from a donor–acceptor pair transition, because this peak exhibits a blueshift with increasing excitation power density [6]. However, the yellow band centred at about 2.3 eV showed no shift with increasing excitation power density under the same excitation conditions.

We found a PL peak having the strongest intensity appeared at about 3.29 eV. With increasing excitation power density, the energy of this peak also shows no shift and the integrated intensity also shows no saturation. The temperature dependence behaviour of this peak is similar to that of the GaN peak. Therefore, the 3.29 eV band is assigned to the emission due to the band-to-band transition in GaCrN.

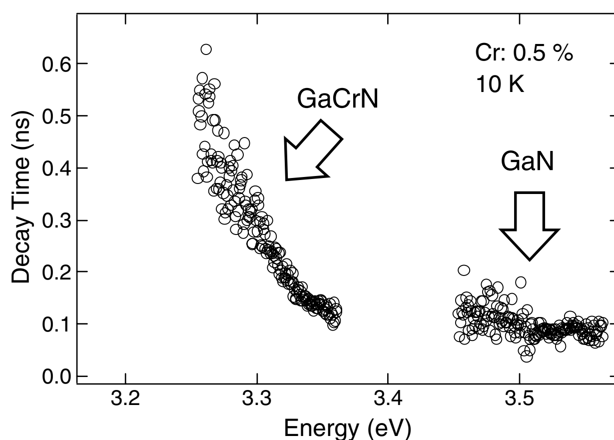

Figure 2. The PL decay times for GaCrN and GaN as a function of monitored emission energy at 10 K for the same sample in figure 1.

Figure 2 shows the PL decay times as a function of monitored emission energy for the GaCrN sample at 10 K. The monitored emission energy dependence of the PL decay times for the GaN buffer layer almost shows no dependence. However, for GaCrN, the decay time strongly depends on the monitored emission energy.

The PL peak from GaCrN was observed even at room temperature as shown in figure 1. On the other hand, from any of the established III–V-based DMSs (InMnAs, GaMnAs, etc), which were grown by low-temperature MBE to avoid phase separation, PL emission has previously never been observed. Therefore, the observation of PL emission from a GaN-based DMS is very important from the viewpoint of fabrication of new functional devices. This phenomenon may be explained in that there exists a piezoelectric field in the GaCrN layer. The piezoelectric field in GaCrN tilts the band edges, and electrons and holes are separately confined spatially. Therefore, a long lifetime was observed [7, 8].

3.2. Optical properties of GaDyN, DyN/GaN superlattice and GaGdN

Figure 3 shows PL spectra for GaDyN. They are classified into the broad peaks and the sharp peaks according to their forms. The former broad bands originate from crystal defects. The latter sharp lines can be assigned to those within a framework of intra-4f transitions, because these peaks are sharp, the peak energies are independent of temperature, and the lifetime of these emissions is 87 μs at 13 K. We assign these sharp peaks at 480 and at 580 nm to the following intra-4f orbital transitions: $^4F_{9/2}$–$^6H_{15/2}$ and $^4F_{9/2}$–$^6H_{13/2}$ transitions, respectively [9, 10].

Figure 4 shows the temperature variations of the integrated PL intensities of the 500 nm broad peak (open triangles) and the 580 nm sharp line (open circles). The PL intensity of the 580 nm sharp peak decreases with increasing temperature in proportion to that of the 500 nm broad peak. In general, the intra-4f orbital transition is independent of the thermal effects of the crystal. Therefore, this intra-4f orbital transition is related to the broad-peak luminescence. That is, the Dy ion absorbs luminescence photons from the defects. That excites the Dy ion to a higher level of 4f multiplets. Through the adequate paths, the excited electron relaxes to the initial state $^4F_{9/2}$ and produces the observed sharp luminescence.

Figure 5 shows PL spectra of GaGdN at various temperatures from 20 to 300 K. A broad peak having strong intensity around 500 nm also originates from the crystal defects. It is

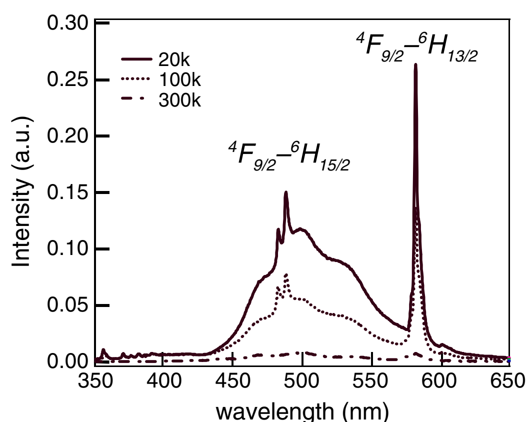

Figure 3. PL spectra of GaDyN with a Dy concentration of 3%.

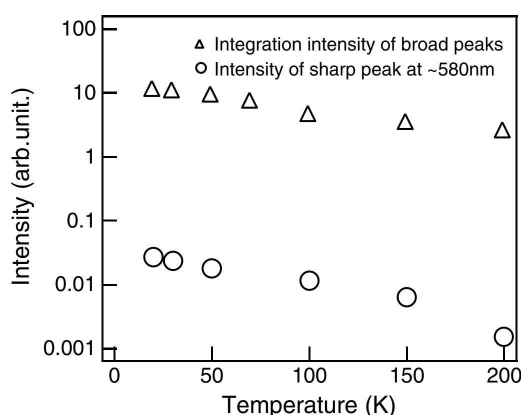

Figure 4. Temperature variations of PL intensities of two peaks from GaDyN sample.

interesting that a sharp line at about 650 nm, which also shows temperature independence of peak energy, was observed. The PL decay time of this peak is about 30 μs at both 13 K and RT. Although the 4f electron transition of Gd^{3+} dominated by the $^6P_{7/2}$–$^8S_{7/2}$ transition is at around 318 nm [10, 11], this emission is characterized as originating from the inter-4f transition of Gd ions judging from these properties. The emission mechanism is under study. Figure 6 shows the temperature variations of the integrated PL intensities of the 500 nm peak (open triangles) and the 650 nm line (open circles), which are different from those of GaDyN as shown in figure 4. This result provides evidence for the existence of the defect-related trap levels that are not involved in the energy transfer between the GaN host and the Gd ions.

4. Summary

We have studied optical properties of GaCrN, GaDyN and GaGdN. PL emission at around 3.29 eV was found from the GaCrN, in which PL decay time depends on emission energy. The intra-4f orbital transitions of Dy^{3+} ions were observed, and the lifetime of the $^4F_{9/2}$–$^6H_{13/2}$

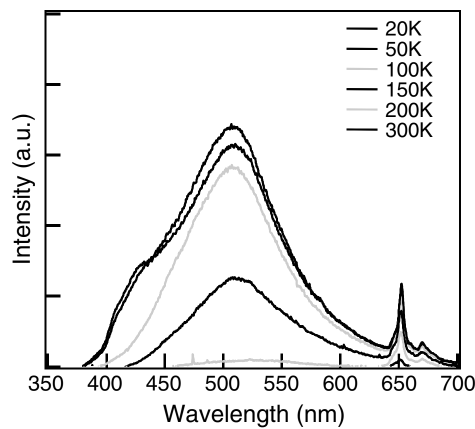

Figure 5. PL spectra of GaGdN with a Gd concentration of 6%.

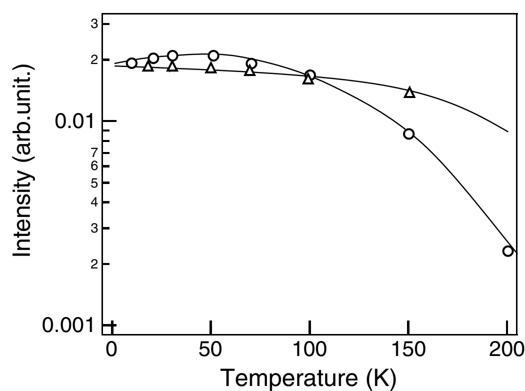

Figure 6. Temperature variations of PL intensities of two peaks from GaGdN sample.

transition was as long as 87 μs at 13 K. An unexpected sharp line was observed from the GaGdN sample; probably from the Gd ion.

Acknowledgments

The authors would like to express their thanks to Dr N Teraguchi of Sharp Corp. and Professor A Suzuki of Ritsumeikan University for the GaGdN and GaDyN samples. This work was supported by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan through MEXT Special Coordination Funds for Promoting Science and Technology (Nanospintronics design and realization, NDR) and in part by a Grant-in-Aid for Scientific Research on a Priority Area and the 21st COE Program from MEXT, Japan.

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