Concentration quenching of Eu-related luminescence in Eu-doped GaN

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Introduction

GaN doped with rare-earth elements (RE) hold significant potential for applications in optical devices, since they show sharp and intense luminescence which is only minimally affected by temperature variations. Among the various RE dopants, Eu seems to be the most interesting, since it yields red luminescence (622nm) which has not been realized in commercially available light emitting devices (LED) that use InGaN active layers.

The red luminescence was analyzed and determined to be generated through a trap-level-mediated energy transfer from the host GaN, and the external emission efficiency was estimated to be about 0.18 at room temperature when the Eu concentration was about 2 at%. These results revealed that Eu-doped GaN could be a potential material for an active LED layer. However, the luminescence intensity abruptly decreased when the Eu concentration exceeded 2 at%. The cause of this luminescence quenching is not well understood. In this study, local structures around Eu atoms in Eu-doped GaN were investigated by extended X-ray absorption fine structure (EXAFS) measurements with a fluorescence mode, in order to study the cause of the concentration quenching of Eu-related luminescence in Eu-doped GaN.

Experimental and Result

The Eu-doped GaN thin films were grown on sapphire (0001) substrates by gas-source molecular beam epitaxy (GSMEB) using uncracked NH_3 gas with 6-nine purity as the nitrogen source. Metallic Ga with 6-nine purity and Eu with 3-nine purity were evaporated from conventional Knudsen effusion cells. The films were grown at 700°C and Eu doping concentration was controlled by varying the Eu cell temperature from 390 to 500°C. Eu concentrations in GaN were estimated, using Rutherford back scattering (RBS) spectrometry, to range from 0.1 at.%, to 16 at.%. The film thickness was about 1.0 µm for all samples.

Eu L_3-edge EXAFS measurements were carried out for the samples with Eu concentrations of about 0.1 at.%, 2 at.%, 16 at.% in fluorescence mode using beam line 12C at the High Energy Accelerator Research Organization (KEK) to study the local structures around Eu atoms. The EXAFS spectra were obtained in fluorescence detection mode at room temperature. A Fourier filtering technique was applied to the first nearest neighboring peak in the Fourier transform and the extracted $k^3\chi(k)$ on the shell was curve-fitted using the parameters calculated with FEFF8 program. The bond lengths, coordination numbers, and Debye-Waller factor were used as fitting parameters to yield optimum values.

Table I. Analytical results of EXAFS

Sample	Parameter	1^{st}neighbor atoms		2^{nd}neighbor atoms	
(A) Eu : 0.1%		N		Ga	
	C.N	1.4	1.8	11.2	
	R (Å)	2.3	2.5	3.4	
(B) Eu : 2%		N		Ga	Eu
	C.N	2.8	0.8	11.1	0.2
	C.N ratio			98%	2%
	R (Å)	2.3	2.5	3.3	3.3
(C) Eu : 16%		N		Ga	Eu
	C.N	0.2	3.0	5.4	2.9
	C.N ratio			65%	35%
	R (Å)	2.3	2.4	3.3	3.6

The analytical results show Fig1. They show most of the Eu atoms formed bonds with nitrogen with two different bond lengths of 2.3 and 2.5 Å, and the total coordination number for nitrogen was nearly four for all samples. These results reveal that most of the doped Eu atoms were incorporated into Ga lattice sites with distorted tetrahedral symmetry. For the second nearest neighbor atom, these results suggest the existence of Eu in addition to Ga. The atomic ratios for the second nearest neighbor atoms represent the concentration of the mixed crystal if the constituent atoms were randomly distributed. The Eu concentration value obtained by EXAFS analysis for sample (B) is 2 at.%. This value is same to that determined by RBS and indicates that Eu ions are almost uniformly distributed over the entire film. On the other hand, the analyzed Eu concentration for sample (C) is about 35 at.% and this value is clearly different from the RBS result (16 at.%). The larger value of Eu content obtained by EXAFS compared with that by RBS suggests phase separation of EuN from GaN.

Most of the red-emission is generated through excitation of GaN[1], therefore it may be difficult to generate red-emission in EuN since the energy transfer from GaN to EuN is not very efficient. Therefore one of the causes of the quenching of the Eu-related luminescence is the formation of EuN.

References

[1]S.Morishima et al., phys.status solidi (a)**176**,113(1999).

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