Photoluminescence and excitation spectroscopy of the 1.5 μm Er-related band in MBE-grown GaN layers

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Abstract

The infrared photoluminescence at 1.5 m due to the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of Er^{3+} ions has been investigated for GaN:Er^{3+} layers grown by MBE. Low temperature high resolution measurements performed under continuous illumination at the wavelength $\lambda_{exc} = 532$ nm, resonant to one of the intra-4f-shell transitions, revealed that the 1.5 μm band consists of up to eight individual spectral components. In excitation spectroscopy, a temperature dependence splitting of resonant bands was observed. On the basis of these experimental results, a possible multiplicity of optically active centers formed by Er doping in GaN layers is discussed.
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1. Introduction

For over a decade, rare earth (RE) doped III–V semiconductors have been widely investigated in view of their photonic properties and applications in optoelectronics [1,2]. GaN as a host for RE ions is of interest due to its wide band gap, that guarantees luminescence at room temperature [3]. Er doping of GaN has been successfully used for

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fabrication of green light emitting devices [4]. Such a material is also interesting as regards luminescence at 1.5 μm, coincident with the minimum absorption losses in optical fibers. The 1.5 μm emission from GaN:Er^{3+} is the focus of this paper. In particular, we consider whether Er doping results in the formation of a multiplicity of optical centers which contribute to the 1.5 μm emission band. The GaN:Er^{3+} samples used for these experiments were kindly provided by Dr. A.J. Steckl, from the University of Cincinnati; these were grown at the University of Cincinnati on a p-type (111) Si substrate, using the solid source molecular beam epitaxy (SSMBE) method, after deposition of an AlN buffer layer. Details of preparation procedure can be found in Ref. [5]. The experiments described in this paper were carried out on a particular sample grown at an Er cell temperature of 860 °C, a Ga cell temperature of 935 °C, and a substrate temperature of 650 °C. The concentration of Er was [Er] ≈ 1 at.%.

2. Experimental results

2.1. Photoluminescence spectroscopy

Fig. 1 shows the integrated intensity of the 1.5 μm photoluminescence (PL) band originating from the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ Er^{3+} intra-4f-shell transition. The sample was excited resonantly with a Nd:YVO_4–based laser operating in a continuous mode at $\lambda_{exc} = 532$ nm, on–off modulated with a mechanical chopper. The sample was placed in a He gas flow cryostat and the measurements were taken in the range from 4.2 K to room temperature. The PL signal was dispersed with a high resolution 1 m spectrometer and detected with a Ge detector. A photomultiplier tube (PMT) was used to record the decay kinetics of the PL signal.

The black line in Fig. 1 presents the PL spectrum of the sample in the 1.5 μm range taken at 4.2 K. A phonon replica of the band can be seen at an energy lower by 14 meV. Such a separation does not coincide with the optical phonon in GaN, so a local phonon is probably involved. The grey line in the figure shows the PL spectrum at room temperature. A similar band is repeated at 14 meV above the main spectrum, representing anti-Stokes luminescence. The inset to the figure depicts a high resolution scan of the main PL band at 4.2 K: up to eight well resolved peaks can be distinguished.

2.2. Photoluminescence excitation spectroscopy

Fig. 2 shows the integrated room temperature PL intensity of the 1.5 μm band as a function of the excitation energy (PLE). The tunable sub-band excitation was provided by a Nd:YAG–pumped optical parametric oscillator (OPO). The 350.4 nm line of an Ar^+ ion laser was used for over-band-gap excitation. As can be seen, under below-band-gap excitation only energies resonant with one of the 4f shell transitions led to the 1.5 μm emission. The individual PLE bands can be identified with transitions from the ground state ($^4I_{15/2}$) to the $^4I_{11/2}$, $^4I_{9/2}$, $^2H_{11/2}$, and $^4F_{7/2}$ excited states, respectively. In addition, we note that three of the PLE bands split into two components; their separations differ with the band.
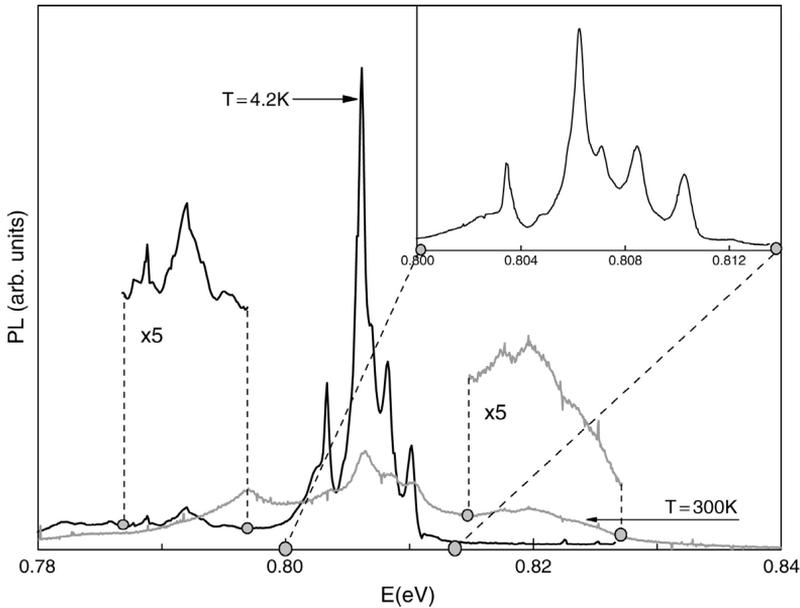
Fig. 1. The $^4I_{13/2} \rightarrow {}^4I_{15/2}$ emission spectrum from GaN:Er^{3+} under excitation to the $^2H_{11/2}$ multiplet (λ_{exc} = 532 nm), recorded at 4.2 K (black line) and room temperature (grey line); the phonon replica and anti-Stokes luminescence are shown five times amplified. Inset: the high resolution spectrum of the main band at 4.2 K (range: 0.800–0.814 eV).

3. Discussion

The observed splitting of the PLE bands—Fig. 2—together with the multiple structure reported in the high resolution spectrum—Fig. 1—could indicate the existence of more than one type of Er-related optically active center in the sample studied. We investigated this possibility further by monitoring the 1.5 μm band under selective excitation tuned to individual components of the PLE bands. Identical spectra have been obtained with E_{exc} = 1.23, 1.25, 1.48, 1.51, 2.31, 2.35 and 2.50 eV. Also for over-band-gap excitation (3.54 eV) no difference has been seen in the structure of the 1.5 μm PL band. We also investigated the decay characteristics of individual components of the 1.5 μm band under selective excitation and found no differences, as nearly identical decay times were measured independently of the excitation energy. Finally, the PLE spectra taken over a range of temperatures from 10 K to room temperature are depicted in Fig. 3. We observe that the relative intensity of the two components for each PLE band changes with temperature. While the higher energy component of the band enhances at lower temperatures, the lower energy component quenches and disappears.

Taking into account the Er^{3+} level splitting due to the crystal field, we will try to interpret the double structure of the PLE bands and its temperature dependence considering thermalization within the ground state. At low temperatures, only the lowest level of the ground energy state is populated. At a higher temperature, Er^{3+} ions thermalize and
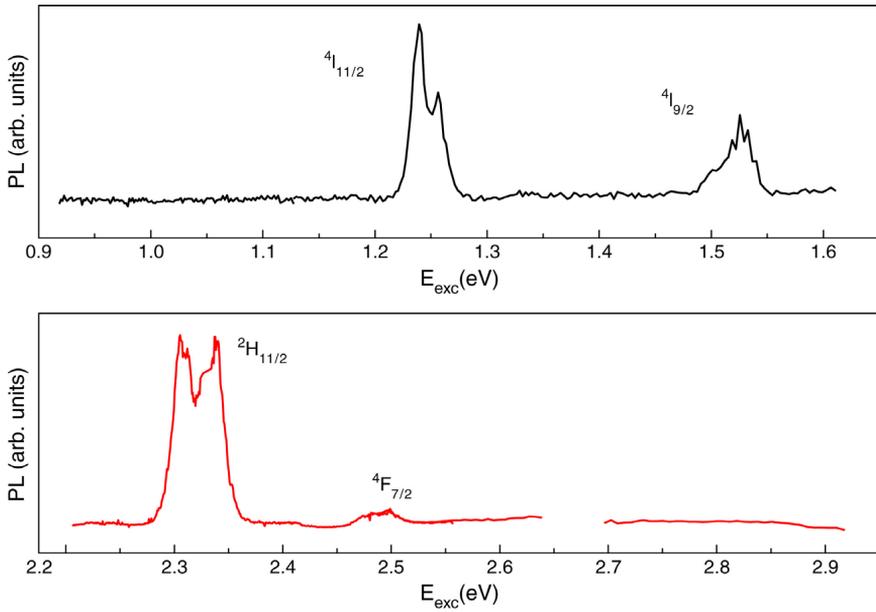
Fig. 2. The PLE spectrum at 1537 nm (0.807 eV) taken at room temperature. Excitation is provided by a pulsed OPO. Individual bands are identified with the corresponding intra-4f-shell transitions.
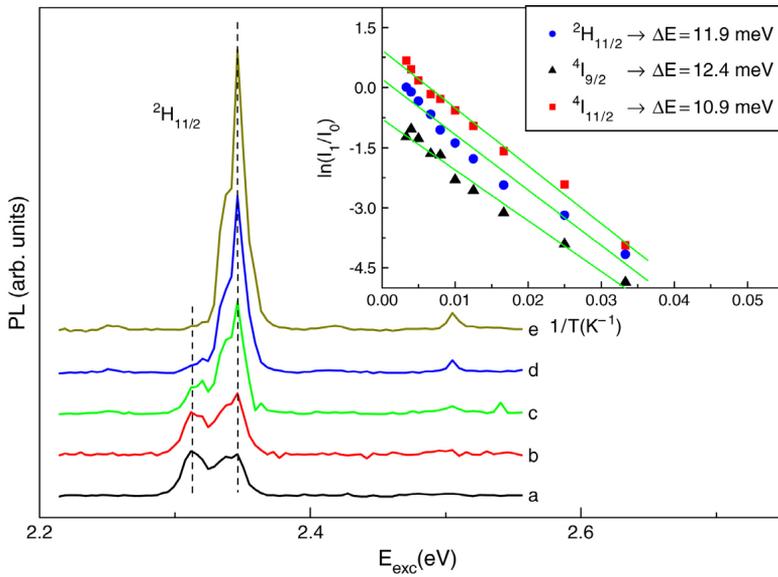

Fig. 3. The temperature dependence of the $^2H_{11/2}$ excitation band, monitored with the spectrometer set at 1537 nm. $T = 300, 200, 100, 50$ and 10 K for a, b, c, d, and e respectively. In the inset to the figure, Arrhenius plots of the intensity ratio of the two components of the individual PLE bands are given.

Table 1
Splitting of Er^{3+} states as deduced from the temperature dependence of the PLE bands

Er^{3+} level	E position (eV)	ΔE splitting (meV)
$^4I_{15/2}$	0	12
$^4I_{11/2}$	1.25	3.8
$^4I_{9/2}$	1.5	11
$^2H_{11/2}$	2.3	23.6

populate upper levels, leading to the appearance of new components in the PLE band. Since not only the ground state, but also the excited states of Er^{3+} ions will split due to the crystal field, the energy distance between the components can be different for specific PLE bands and also not coincident with the ground state splitting.

In this simple model, for each excitation wavelength, the luminescence intensity is proportional to the population of the ground state level. If we consider splitting of the ground state into two levels, the upper one being populated by thermalization from the lower one, the intensity ratio of the PLE band components I_1/I_0 will be given as

$$I_1/I_0 \propto N_1/N_0 = A \exp(-\Delta E/k_B T), \tag{1}$$

where N_1, N_0, ΔE and k_B are populations in the upper and the lower ground state levels, the ground state splitting and the Boltzmann constant, respectively. In the inset to Fig. 3, the intensity ratio of the two components of the three PLE bands investigated is here plotted as a function of inverse temperature. A similar activation energy of $\Delta E \approx 12$ meV is obtained for all cases. Following the assumed model, this value corresponds to the energy separation within the ground state. On the basis of this, we can estimate the excited state splitting—see Table 1.

We can conclude that the experimental data obtained thus far for PL of the 1.5 μm band can be given a satisfactory interpretation involving only one type of optically active Er^{3+} center in the host. This provisional conclusion needs to be corroborated by further studies; these are currently under way. The new measurements will include also Zeeman experiments, which will directly address the question of multiplicity of Er-related optically active centers in GaN.

References

[1] R.G. Wilson, R.N. Schwartz, C.R. Abernathy, S.J. Pearton, N. Newman, M. Rubin, T. Fu, J.M. Zavada, Appl. Phys. Lett. 65 (1994) 22.
[2] J.M. Zavada, D. Zhang, Solid-State Electron. 38 (1995) 1285.
[3] P.N. Favennec, H. L'Haridon, M. Salvi, D. Moutennet, Y. Le Guillou, Electron. Lett. 25 (1989) 718.
[4] A.J. Steckl, J.C. Heikenfeld, D.S. Lee, M.J. Garter, C.C. Baker, Y.Q. Wang, R. Jones, IEEE J. Sel. Top. Quantum Electron. 8 (2002) 749.
[5] D.S. Lee, J. Heikenfeld, A.J. Steckl, U. Hommerich, J.T. Seo, A. Braud, J. Zavada, Appl. Phys. Lett. 79 (2001) 791.