Optical Materials 33 (2011) 1059–1062

Contents lists available at ScienceDirect

Optical Materials

journal homepage: www.elsevier.com/locate/optmat

Optical and magneto-optical properties of erbium doped InGaN and GaN epilayers

N. Woodward [a,*], V. Dierolf [a], J.Y. Lin [b], H.X. Jiang [b], J.M. Zavada [c]

[a] Physics Department, Lehigh University, Bethlehem, Pennsylvania 18015, USA
[b] Department of Electrical and Computer Engineering and Nano Tech Center, Texas Tech University, Lubbock, Texas 79409, USA
[c] Department of Electrical and Computer Engineering, North Carolina State University, Raleigh, North Carolina 27695, USA

ARTICLE INFO

Article history:
Received 18 June 2010
Received in revised form 7 July 2010
Accepted 13 July 2010
Available online 8 August 2010

Keywords:
Rare earth
Luminescence
Zeeman splitting

ABSTRACT

Using combined excitation–emission spectroscopy we have studied the erbium incorporation into GaN and InGaN for in situ doped MOCVD-grown layers and compared them to samples grown by MBE. A smaller up-conversion efficiency for the main site is observed compared to minority sites in the same sample as well as versus all sites from MBE grown samples. Furthermore, we show that the 1.54 μm emission efficiency is reduced with increasing In-content both under excitation above the bandgap in the UV as well as under resonant excitation at around 980 nm. This indicates that non-radiative decay channels for the Er ion are the largest contributing factor for this behavior. Finally, the Zeeman splitting of the excitation and emission lines of Er:GaN under application of magnetic fields up to 6.6 T with B||*c*-axis is shown.

Following the commercial availability of high power III-nitride light emitting diodes in the 370–540 nm range, a strong desire for efficient Er-doped InGaN epilayers has developed. Recently Er-doped InGaN epilayers have been grown by MOCVD with good crystal quality. Nevertheless they show a relatively weak 1.54 μm emission efficiency when compared to Er-doped GaN [1]. Aside from application in novel light sources, there has been an increasing interest for some time in the magnetic properties of the dilute Er-dopants, since ferromagnetism of Er:GaN has been reported [2]. To contribute to the clarification of the underlying coupling mechanism, we have investigated the effects of applied magnetic fields on Er-doped GaN epilayers as grown by MOCVD in order to determine the effective *g*-values of the ions excited and ground state. In this paper, we want to particularly address the following questions: (1) are the multitude of excitation and emission lines related to a single site as shown in Ref. [3] or do additional incorporation sites appear as found for MBE grown samples in Ref. [4]. (2) What is the cause of the strong reduction in emission efficiency that has been observed for increased In-content under UV excitation [1]. Is this caused by a less efficient excitation channel or by a more pronounced non-radiative decay channel from excited Er ions. (3) What are the Zeeman splittings of the ground and excited levels and how do they behave for magnetic fields for which the Zeeman splitting is comparable to the crystal field splittings. Samples used in these studies were MOCVD-grown Er:GaN epilayers that were grown on sapphire and in situ doped with Er up to 10^{21} cm^{-3}. Details of these samples are described in Ref. [1].

As a first step, we show in Fig. 1 the combined excitation–emission spectroscopy (CEES) data (for details of the technique see e.g. Ref. [3]) for the technologically important erbium transition at around 1.54 μm. We excite this transition using a tunable semiconductor laser at around 980 nm. At first sight a large number of peaks appear in the image plot even in the rather small energetic window that is depicted. This may suggest a large number of sites. However, thermally activated levels, electron–phonon coupling, and energy transfer will increase the number of transitions excepted in the ideal case of a single site at zero temperature without coupling to lattice vibrations.

In the assignments, we follow the following guidelines:

- The emission spectra of a single site must be identical in spectral position and relative heights for all its excitation transitions and accordingly for excitation spectra. If this is not the case, we are dealing with multiple centers.
- Excitation starting from thermally excited levels (such as A2) of the ground state must be reduced in energy (b) by the same amount than seen in the emission (a). As a consequence, we will see transitions with lower energy, in the excitation spectra. The inverse is true when we consider emission starting from thermally excited states (such as B2).

Applying these rules to our Er:GaN system, it turns out that all transitions can be explained with the inclusion of thermally excited transitions. The measurement temperature is about 10 K and hence not only the lowest states (i.e. A1, B1, as indicated in Fig. 2) of the ground and excited state multiplets are populated. We can explain all transition energies when we consider excitation

* Corresponding author. Tel.: +1 860 480 2208; fax: +1 610 758 5730.
E-mail address: naw205@lehigh.edu (N. Woodward).

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doi:10.1016/j.optmat.2010.07.007

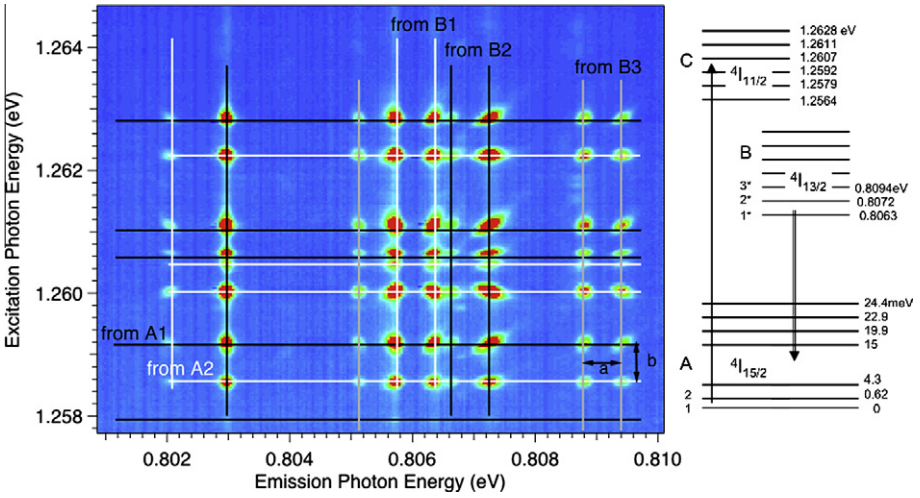

Fig. 1. Combined emission excitation spectral image plot of Er:GaN. The horizontal lines represent transitions from the ground level to the $^4I_{11/2}$ state, while the vertical lines represent emission peaks from the $^4I_{13/2}$ state to the $^4I_{15/2}$ ground level.

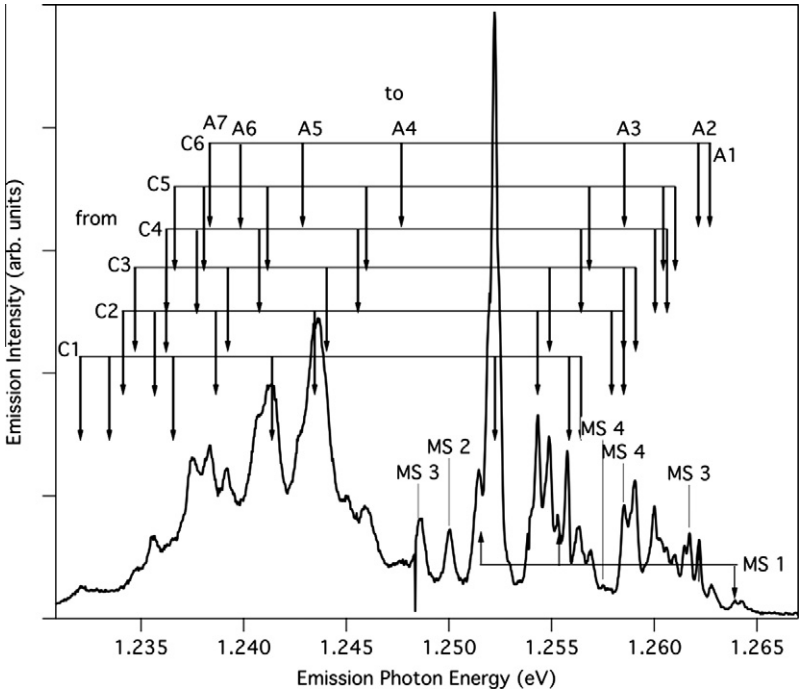

Fig. 2. Complete assignment of the emission peaks due to the $^4I_{11/2}$ to $^4I_{15/2}$ transition at around 980 nm observed under excitation in the 1.54 μm spectral region.

transition starting from the 2nd level (A2) of the ground state multiplet as well as emission transitions from the 2nd (B2) and 3rd level (B3) of the $^4I_{13/2}$ excited state. In both cases, this will lead to groups of transitions that are shifted relative to each other by an identical amount (i.e. the energy of the thermally excited state). In Fig. 1, these groups are indicated by lines of the same color/grayscale, with black lines indicating transitions from the respective lowest level, and the white and grey lines correspond to the groups from the 2nd and 3rd lowest levels. In the presented case, the original and final level is identical such that we can test the assignments for consistency. The separation indicated as *a* and *b* both represent the splitting of the two levels of the ground state and must hence be equal. This consistency condition is fulfilled with great accuracy, such that we are confident that the majority of lines can be assigned to a majority site. Using conventional excitation–emission spectroscopy, similar conclusions have been drawn independently for the same type of samples by Makarova et al. [4].

A strength of the CEES technique compared to traditional excitation–emission spectroscopy lies in the ability to identify weaker spectral features by exploiting the dynamic range of the CCD array. By overexposing the main site, a minority site becomes visible which exhibits an emission intensity that is weaker by a factor of about 40.

These additional sites become more apparent when we consider up-conversion excitation. In the simplest case, we use a fiber-amplified tunable 1.54 μm laser to excite the ions in two steps to the $^4I_{9/2}$ state and observe the emission from the $^4I_{11/2}$ state at around 980 nm. This case is essentially the inverse to the direct excitation and hence identification of the sites is easiest. We show emission spectra under these conditions in Fig. 2 in which the

Table 1
Characteristic transition energies of the observed minority sites of Er in GaN. Transition energies for which the sites are most distinct are chosen.

	MS 1	MS 2	MS 3	MS 4	MS 5	MS 6
$^4I_{15/2} \leftrightarrow {}^4I_{13/2}$	0.8040	0.8090	0.8087	0.8083	0.8066	
	0.8068	0.8071		0.8076		
	0.8085	0.8050		0.8068		
				0.8056		
				0.8042		
				0.8035		
$^4I_{15/2} \leftrightarrow {}^4I_{11/2}$	1.2640					
	1.2625					
	1.2619					
	1.2598					
	1.2590					
	1.2583					
$^4F_{9/2} \rightarrow {}^4I_{15/2}$		1.8524	1.8660	1.8146	1.8057	
			1.8495			
$^4S_{3/2} \rightarrow {}^4I_{15/2}$		2.2328		2.2436		
				2.2412		
				2.2360		
$^4I_{15/2} \rightarrow {}^2H_{11/2}$		2.3462	2.3308	2.3521	2.3545	2.3533
		2.3392		2.3503	2.3516	
		2.3345		2.3450	2.3462	
				2.3408		
				2.3378		
				2.3324		
				2.3286		

transitions for the main site are identified. We clearly see emission from all of the crystal field levels of the $^4I_{11/2}$ state (labeled as from C1, C2, ..., C6) due to a larger thermal excitation from increased excitation laser power. The remaining transitions are related to minority sites (MS1...6). Transition energies of these sites are combined in Table 1. The fact that the minority sites become increasingly apparent for up-conversion excitation in two and (even more so) in three steps is an indication that either the up-conversion efficiency is very low for the main site or that efficient de-excitation channels for the main site exist for higher excitation levels. The observation of a relatively weak green emission in the MOCVD samples under UV excitation suggests the second assumption. This is supported by the observation (see Ref. [3]) that for MBE grown samples the main site exhibits reasonably high up-conversion efficiency even though these samples are of lower crystalline quality and have larger numbers of minority sites.

Turning our attention now the resonant excitation of the Er ions in InGaN with increasing In-content, we find as expected a significant broadening of the lines due to the disorder in the Ga sublattice (see Fig. 3). Neglecting this broadening effect, we find almost identical transition energies suggesting that the majority of the Er ions are still surrounded by Ga. No additional minority sites become apparent but they may be hidden under the broadened transition lines. Even if the whole emission intensity across the broadened excitation transitions is integrated, the intensity is still much weaker than in the Er-doped GaN; similar to the situation under UV excitation. This indicates that an increase in non-radiative decay after Er excitation is the largest origin of the decrease in 1.54 μm intensities when In is introduced in Er:GaN.

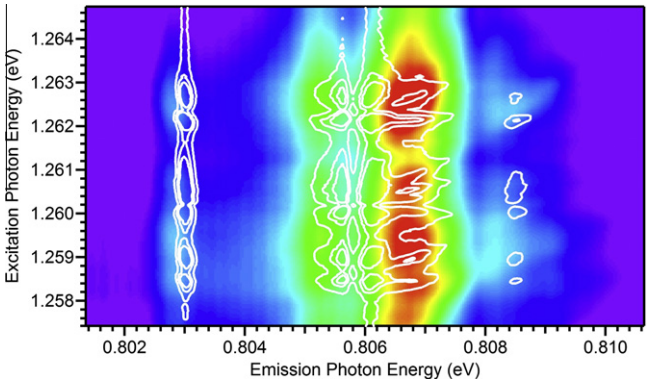

Fig. 3. Image plot of Er:InGaN with 5% In-content. The contour plot in white shows the CEES of Er:GaN.

Finally, we consider again the main incorporation site of Er ions in GaN and study the splitting of the transitions under an application of magnetic fields, parallel to the c-axis, up to 6.6 T. Similar studies have been performed by Vinh et al. for the Er ion in silicon [5]. The presence of many transitions and the fact that there is a splitting of both the emission and excitation lines contributes to the complicated analysis of Fig. 4. The latter contribution leads to an inability to directly assign *g*-factors to splittings seen in the data unless the *g*-factors of one of the involved levels are nearly zero. For the ground state, a *g*-value of 2.861 has been measured by EPR for the magnetic field aligned parallel to the hexagonal axis of the crystal [6,7]. We are able to find splittings that correspond to this *g*-factor as indicated in Fig. 4 by lines labeled as transitions from B1, B2, and B3 suggesting that the splittings of the excited states are small. However, for some levels, the crystal field

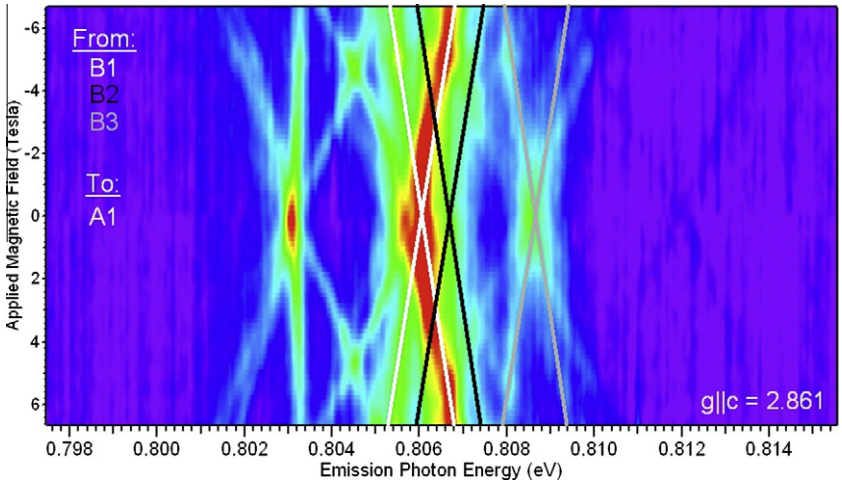

Fig. 4. Application of magnetic fields applied in the + and −*c*-axis directions up to 6.6 T. The added lines are a projection of the splitting of a level due to a *g*-factor of 2.861 from levels B1, B2, and B3 to A1.

splittings are small compared to the Zeeman splittings and hence we have to take the mixing of levels with equal symmetry into account. Considering all these complications it is no longer surprising that we see asymmetric splitting, level crossings of transitions as well as nonlinear behavior of the shift as function of magnetic field. A reliable assignment and analysis of the *g*-factor will require a better signal to noise ratio, the consideration of additional transitions, and additionally a full angular dependence measurement of the PL.

In summary we have used combined excitation–emission spectroscopy to study the erbium incorporation into GaN and InGaN for in situ doped MOCVD-grown layers and compared them to samples grown by MBE. A decrease in up-conversion efficiency for the main site is observed compared to the up-conversion efficiency of both minority sites in the same sample and MBE grown samples. Furthermore, we show that the 1.54 μm emission efficiency is reduced with increasing In-content not only under excitation above the bandgap in the UV but also under resonant excitation at around 980 nm. This indicates that non-radiative decay channels for the Er ion are the largest contributing factor for this behavior. Finally we report on initial investigation of the Zeeman splitting of the excitation and emission lines of Er:GaN under application of magnetic fields up to 6.6 T.

Acknowledgements

This work is supported by NSF (Grant No: DMR 0705217 and ECCS-0823894) and ARO (Grant No: W911NF-09–1-0277). H.X. Jiang and J.Y. Lin would like to acknowledge the support of Edward Whitacre and Linda Whitacre endowed chairs through the AT&T foundation.

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