Discrepancies in the Optical and Magneto-Optical Spectra of Eu:GaN: What is the Nature of the Majority Site?

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Introduction:

Eu has consistently been the most promising candidate as a dopant in the active layer for a red LED in GaN. Despite the abundance of experimental and theoretical studies performed on this system, there is ongoing debate over the structure of the majority incorporation environment of Eu, in GaN[1-4]. This is due to the complicated spectra, which do not agree with what is predicted for a single center.

Results and Discussion:

In the past, the majority center in Eu:GaN (Eu1), was assumed to be a relatively unperturbed center, with a symmetry close to C_{3V} symmetry. For a center with perfect C_{3y} symmetry, group theory predicts that three peaks should exist, where two have both π and σ allowed dipole transitions and one which only exhibits a π dipole transition. Two of these peaks should split under a magnetic field applied parallel to the c-axis. This would mean that the majority center should only show two peaks, when σ -polarized emission is detected, however, three peaks are observed. From sample to sample, the relative intensities of two of the peaks remain constant; however, the third peak weakens and broadens. If we take π -polarized emission into account as well, we find a total of 6 peaks, where we should only see, at most 5[Fig. 1]. This indicates that we are dealing with at least two centers that are similar in nature, and hence can be excited at the same resonant energy.

Magneto-optical studies were performed with the field applied both parallel and perpendicular to the c-axis. It is expected that two of the peaks will split for the C_{3v} configuration, for a field applied parallel to the c-axis. What is observed is an apparent splitting for two peaks, suggesting g-factors of 1.79 and 1.83 respectively, but it appears that the third peak also splits. Closer inspection of field dependent Zeeman data for different field directions reveals a more complex picture. However, after closer inspection, it was deduced that the peak shifts to the left, and a new peak appears and shifts, while gaining strength with increasing magnetic field. For the field applied perpendicular to the c-axis, no splitting is expected. As the magnetic field is increased in this geometry, we observe, for σ -polarized emission, the appearance of a peak corresponding to the strongest π transition. This emission line increases in intensity, and shifts as the magnetic field is ramped up.

For the ${}^{5}D_{0}-{}^{7}F_{1}$ transition at 2.016eV, similar discrepancies in the number of peaks are observed. Furthermore, when a magnetic field is applied, the resonantly excited emission is reduced in intensity, and never returns to its original intensity, even when the field is removed.



Fig. 1 Optical emission spectra taken in the π and σ orientation. A single center should only have 5 peaks, but we see at least 6.

Open Questions:

- 1) Why are there more peaks, without a magnetic field, than group theory predicts?
- Does the polarization mixing upon application of a magnetic field, depend on the substrate, or is it inherent to the Eu:GaN system?
- 3) How does the application of a magnetic field modify the emission intensity of the Eu ions (even after removal)?
- 4) Are the observed discrepancies a result of the coupling of the magnetic states to the host??

Conclusion: In summary, the emission from the majority center in Eu:GaN, has been seen to have too many peaks for a single center, even without the application of a magnetic field. This becomes even more apparent, when a magnetic field is applied and two of the peaks split. To explain these observations, we must consider that the emission from the majority center actually result s from, at least, two separate centers which are perturbed away from C_{3v} symmetry, possibly by the presence of a nitrogen vacancy, for which several distinct relative locations are possible.

References:

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