Rare Earth doped GaN for photonic devices

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Abstract:

We report excitation mechanisms, quantum efficiency as well as the thermal quenching of photoluminescence processes in rare earth ions (Er and Nd) in GaN grown by metal organic chemical vapor deposition.

1. Introduction

The incorporation of rare earth (RE) into wide bandgap semiconductors is of significant interest for optoelectronic devices, because of their temperature independent, atomic-like and stable emission together with the optical and electrical excitation.^{1, 2} RE elements such as Er and Nd doped GaN materials have attracted much attention due to their capability to provide highly thermal stable optical emission in the impressive developments in this area, the GaN:Er and GaN:Nd systems remain poorly understood and even controversial in regard to the d microscopic structure of RE optical centers and the relevant energy transfer mechanisms, which constitute a barrier to further increases of device emission efficiency and thermal stability.

The excitation of RE ions can take several steps before reaching the luminescence of the ions.¹ A common way of exciting RE ions which has been done in most RE-based lasers is to employ an optical pumping source with a photon energy that matches a higher-lying inner 4*f* shell transition. This type of excitation scheme is therefore called direct or resonant excitation. The method needs a high optical pumping power due to a low excitation cross-section for RE ions in insulators with a typical value of 10⁻²⁰ cm².³ Other types of excitation mechanisms involve host excitation first and then Er³⁺ ions using a band-toband excitation. The common feature to these excitation processes involves the recombination of electrons and holes with a non-radiative energy



Figure 1. PL spectra of the 0.5 μ m GaN:Er epilayer at 1.54 μ m within the ${}^{4}I_{13/2} \rightarrow {}^{4}I_{15/2}$ transition at 15 K under (**a**) ${}^{4}I_{15/2} \rightarrow {}^{4}I_{9/2}$ resonant (1.533 eV or 809 nm), (inset) energy level diagram of the isolated Er optical center, and (**b**) non-resonant or band-to-band (3.531 eV or 351.1 nm) excitation, (inset) energy level diagram of the defect-related Er optical center.

transfer to nearby Er³⁺ ions. These processes are called indirect or non-resonant band-to-band excitation with an efficiency about three to five orders of magnitudes higher than the resonant process. Both electron and hole carriers can be bound to each other forming either a free exciton or a bound exciton (BE) trapped by RE ions or an impurity/local defect nearby RE ions. In this presentation, we will discuss different mechanisms, quantum efficiency and the thermal quenching of RE optical centers in GaN epilayers prepared by metal-organic chemical vapor deposition (MOCVD) using direct and indirect excitation processes.

2. Samples and experimental results

The RE ions (Er and Nd) doped GaN epilayer samples were prepared by MOCVD method in a horizontal reactor.^{4, 5} The concentration of RE in GaN epilayer varing from 0.02 % to 6%, in 0.5 μm thickness, were grown on a thin un-doped GaN template of 1.2 μm on top of (1000) *c*-plane sapphire substrate. The growth temperature of RE-doped GaN layer was 1040 °C. The X-ray diffraction spectra indicated that GaN:RE epilayers have high crystallinity and no second phase formation.^{4, 5} The high resolution PL spectra were conducted using a Horiba iHR550 spectrometer equipped with a 900 grooves/mm grating blazed at 1500 nm and detected by a high sensitivity liquid nitrogen InGaAs detector. The PL experiments were carried out in a variable temperature closed-cycle optical cryostat (Janis) providing a temperature range from 10 K to 300 K. Both resonant excitation and the non-resonant excitation were employed to investigate the optical properties of RE in GaN epilayers using a tunable wavelength Ti:Sapphire laser and Ar UV laser.⁶

3. Results and discussion

The difference between resonant and non-resonant band-to-band spectra allows us to identify two types of optical centers with different local defect environments (Fig. 1). In our GaN:Er

materials we determine: (1) Er ions in an isolated local environment can be excited via the resonant excitation ${}^{4}I_{15/2} \rightarrow {}^{4}I_{9/2}$ transition and also the non-resonant excitation due to the recombination of electrons and holes with a non-radiative energy transfer. This optical center is referred to an isolated Er optical center labeled L with PL lines L_1 , L_2 , L_3 , L_4 , L_5 , L_6 , L_7 . (2) Er ions strongly associated with nearby defects or impurities can be excited indirectly via the host involving a trapped (bound) exciton. This center is referred to a defect-related Er optical center labeled L' with PL lines L'1, L'2, L'₃, L'₄, L'₅, L'₆, L'₇, L'₈. The spectrum under band-to-band excitation has more narrow PL lines. The PL lines come from all Er optical centers (isolated and defect-related Er optical centers) in GaN. The difference of PL spectra indicates a difference of the local defect environment for two Er optical centers under different excitation processes.

To determine the crystal field splitting of the first excited state (${}^{4}I_{13/2}$) of the isolated Er optical centers, the high resolution spectra as well as the temperature dependence of PL spectra, have been investigated under resonant excitation (Fig. 2).⁶ Comparing with the low temperature spectra, new PL lines, labeled



Figure 2. The PL spectra at 20 K and 120 K of the GaN:Er epilayer at 1.54 µm within the ${}^{4}I_{13/2}$ $\rightarrow {}^{4}I_{15/2}$ transition under the resonant excitation (${}^{4}I_{15/2} \rightarrow {}^{4}I_{9/2}$). At low temperature, the spectrum of the isolated Er optical center consists of a set of narrow and intense PL lines (L₁ to L₇). At higher temperatures, hotlines, L_{1,2}^h, L₄^h, L₅^h, L₆^h, appear and are displaced by about 12.6 meV. The intensities of hotlines rapidly increase with increasing temperature while the intensities of the main PL lines decrease.

hotlines $L_{1,2}^{h}$, L_{4}^{h} , L_{5}^{h} , L_{6}^{h} , appear at higher temperature (Fig. 2). These hotlines are displaced by about 12.6 meV. The integrated PL intensity measurements for the whole 1.54 µm band (not shown) under the resonant excitation ($\lambda = 809 \text{ nm}$) indicate a low thermal quenching of 20% from 10 K to room temperature from Er³⁺ ions in our GaN:Er epilayer. The thermal quenching shows a stronger effect with higher concentration of RE in GaN.

Decay dynamics of optical centers allows us to understand the energy transfer and recombination processes in the solid materials. Under resonant excitation and low concentration materials, the dynamics of all PL lines of the isolated RE optical centers appear as a single experiential decay dynamics of 3.3 ± 0.3 ms and 1.2 ± 0.2 ms for Er and Nd, respectively, at all temperature. The value is similar with previous reports on the RE ions in GaN.^{7, 8} It indicates that the isolated Er optical center has no quenching channels. Under band-to-band excitation, while again the isolated Er optical center reveals a similar decay time, the defect-related RE optical centers show a double-exponential decay dynamics that strongly depend on the temperature. The double-exponential decay process of optical centers indicates that there are non-radiative recombination processes taking place in PL signals. In addition, by integrating all PL intensities from the isolated RE optical centers in the PL spectrum under band-to-band excitation and taking into account the decay dynamics for optical centers in the materials, we find that there is more than 60% contribution of the isolated Er optical centers in GaN to the PL spectrum.⁹

In this presentation, we will discuss the excitation mechanisms of RE optical centers in GaN epilayer prepared by MOCVD. The PL spectra under resonant and non-resonant band-to-band excitation reveal an existence of two types of optical centers; the defect-related and the isolated Er optical centers. The defect-related optical centers appear a strong temperature dependence of PL intensity with a double-exponential decay dynamics and the highest excitation cross-section. The excitation cross-section of this optical center under band-to-band excitation is much larger than that using the resonant excitation process via the ${}^4I_{15/2} \rightarrow {}^4I_{9/2}$ transition. The high excitation cross-section combined with the temperature independence of decay dynamics and high percentage of optically active center of the isolated RE optical centers makes the GaN:REs materials synthesized by MOCVD interesting for GaN photonics. In particular, the material appears promising for realization of population inversion and, consequently, optical amplification at room temperature.

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