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Journal of Luminescence 72–74 (1997) 59–61

JOURNAL OF LUMINESCENCE

Photoluminescence and optical gain in highly excited GaN

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Abstract

A systematic study of the near-band-edge photoluminescence of epitaxially grown GaN as a function of excitation density has been carried out. While at low densities free and bound-exciton emission lines govern the spectrum, new luminescence bands are detected at densities above 1 MW/cm^2. Gain measurements and temperature-dependent investigations indicate that nonlinear processes like biexciton annihilation, exciton–exciton-scattering and stimulated A–LO-emission are dominant at low temperatures, while exciton–free-carrier scattering occurs at temperatures above 200 K.

Keywords: GaN; High-density excitation; Photoluminescence; Stimulated emission

In contrast to the tremendous progress of the device technology for group-III nitrides in recent years, our knowledge on basic physical properties of this semiconducting material is rather poor. Neither are all the fundamental parameters of excitonic polaritons known, nor do detailed investigations on high-excitation phenomena exist which are important for laser devices. Therefore, we present luminescence and optical gain investigations of two different GaN epilayers as a function of excitation intensity in the range of 10 kW/cm^2 to 5 MW/cm^2. One of the samples (300 μm thick) was grown by hydride vapor phase epitaxy (HVPE) on (0 0 0 1) sapphire, the other one (3 μm thick) by metalorganic chemical vapor deposition (MOCVD) on 6H-SiC.

From a careful analysis of reflectivity and luminescence spectra at low excitation intensities we received important parameters which we need in order to discuss our results at high excitation intensities: exciton energy $E_{ex} = 3.480$ eV, A–exciton binding energy $E_{ex}^{b} = 26.1$ meV, parallel component of the exciton mass $M_{ex\|} = 0.98 m_0$ and the effective masses of the electron and the hole $m_{e\|} = 0.23 m_0$ and $m_{p\|} = 0.75 m_0$, respectively. The ratio $\sigma = m_{e\|}/m_{p\|}$ is 0.31.

At increasing excitation densities the two samples behave quite differently. For the thick, bulk-like epilayer, superimposed to an intensive donor-bound-exciton luminescence $I_2 = 3.4728$ eV with a binding energy of 7.2 meV and a half-width of only 1 meV [1], a broader peak develops at about $E_p = 3.453$ eV. This position corresponds precisely to that expected for an inelastic scattering process of two excitons in which the second exciton is scattered to the continuum ($n = \infty$). This P-band is already known in II–VI semiconductors for a long time and has been described for GaN by Hvam and Ejder [2] as well.

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PII S0022-2313(97)00215-3

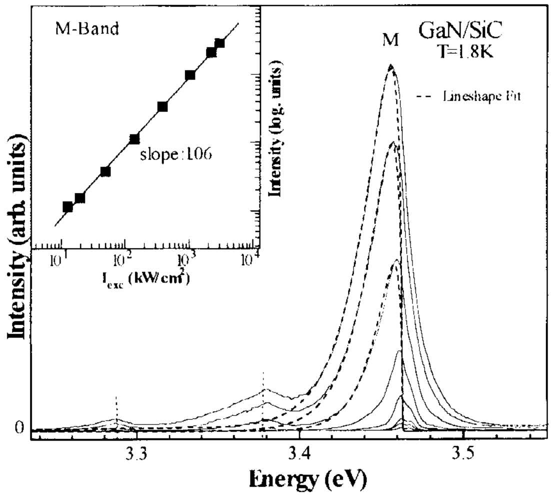

Fig. 1. M-Band emission for different excitation densities. The inset shows the integrated intensities independence on excitation density.

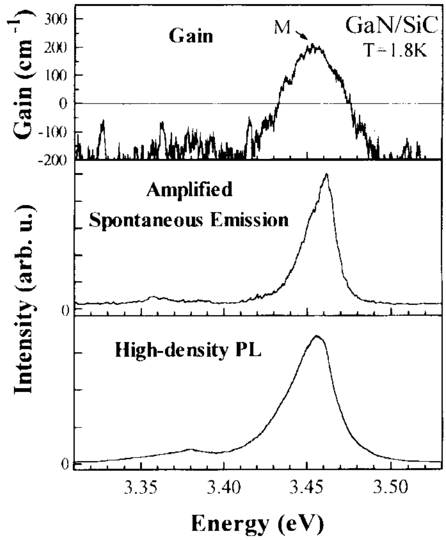

Fig. 2. High-density photoluminescence, stimulated emission and gain spectrum of a 3 μm thick GaN/SiC-epilayer.

The 3 μm thick layer shows at low excitation densities a donor bound exciton emission which has almost the same intensity as the free-exciton luminescence. The peak positions of both lines are shifted to lower energies by 12.5 meV with respect to the corresponding bulk peak energies, due to the tensile strain in thin epilayers. At increased excitation levels (Fig. 1) these lines give way to an intensive broadened emission, which we ascribe to the decay of biexcitons. The dashed curves represent line shape fits for the biexciton emission, reproducing the broadening in the low-energy tail and the red shift of this so-called M-band. The analysis is based on a simple, well-known theoretical model [3]: Biexcitons with momentum k_i, created by two-step or two-photon excitation, annihilate under conservation of energy and momentum into two excitonic polaritons, one in the photonic branch and one in the excitonic branch, longitudinal or transversal. In a simplified model the energy of the emitted photon is given by

$$h\nu = E_{ex} - E^{b}_{biex} - (\hbar^2 k_i^2 / 4M_{ex}).$$

If one assumes a Boltzmann distribution for the biexcitons, one expects an emission band which resembles an inverted Maxwell distribution, extending from $E_{ex} - E^{b}_{biex}$ to lower photon energies. The line shape analysis of the emission band allows to determine the binding energy of the biexciton E^{b}_{biex} and the temperature of the biexciton gas. In our case we receive $E^{b}_{biex} = 3.7$ meV and a biexciton energy of $E_M = 6.931$ eV. If one compares the value of E^{b}_{biex} with that of the II–VI compound CdS it fits well into existing theories [3]. For the higher value of σ_{GaN} ($\sigma_{CdS} = 0.14$), one expects a smaller binding energy. The observation of luminescence above the threshold energy $E_{ex} - E^{b}_{ex}$ is a sign of lifetime broadening of the momentum states due to scattering processes with excitons or biexcitons. This process is also evident by the observed almost linear dependence of the M-band intensity on excitation density as seen in the inset of Fig. 1. Under the equilibrium conditions created in our experiment by excitation pulses of 15 ns duration the interaction with excitons is capable to reduce the logarithmic slope of the integrated intensity from the expected value of 2 to unity.

Highly excited semiconductors are known to produce stimulated emission, resulting in laser action [4]. Indeed, as can be seen from Fig. 2, optical gain can be observed. The gain spectrum was obtained using the stripe length method [5]. It is interesting to note that for both samples the maxima of the gain spectra coincide nicely with the maxima of the high-density luminescence for the M-, P- and the A–LO-transitions.

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