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OPTICAL PROPERTIES OF HIGHLY EXCITED GaN

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Photoluminescence and optical-gain measurements at high excitation densities are presented. Luminescence and optical gain are found from excitonic molecules (biexcitons). The binding energy of the biexciton is deduced from a lineshape analysis of the biexciton luminescence band. It amounts to 3.7±0.5 meV. Thick epilayers exhibit inelastic exciton-exciton scattering as the dominant high-density luminescence and gain mechanism at low temperatures. At increased temperatures the photoluminescence is dominated by inelastic scattering processes of excitons with free carriers and LO phonons.

1 Introduction

Optical properties of wide-gap semiconductors have been studied in great detail over the last decades. Unfortunately, the technological relevance of most of the materials has been rather limited. The opposite holds today for the wide-gap III-V semiconductors. In relation to the recent device developments based on group-III nitrides and their key compound GaN the knowledge about this material system is rather poor. Especially, optical high-excitation phenomena which are important for laser devices have not been studied in detail. In this contribution we therefore focus on the optical properties of highly excited GaN studying the high-excitation photoluminescence (HEPL) and the optical gain at various temperatures.

2 Results

The study was carried out using two different samples. Sample 1 is a 300 µm thick GaN epilayer grown by hydride vapor phase epitaxy (HVPE) on (0001) sapphire.[1] The substrate was removed after growth. Sample 2 is a 3 µm thick GaN epilayer grown by metalorganic chemical vapor deposition (MOCVD) on 6H-SiC.[2] For high-density excitation above the band gap we used a pulsed dye laser pumped by an excimer laser. Gain measurements were performed using the stripe length method.[3] In Fig. 1 we compare the high-excitation photoluminescence (HEPL) of the two samples. The low-density emission from the quasi-bulk sample 1 exhibits

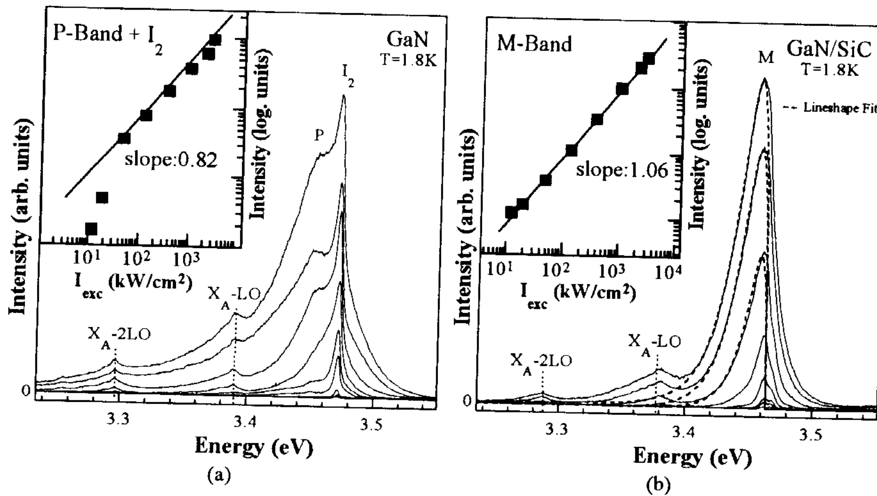

Fig. 1 Dependence of low-temperature photoluminescence from GaN on excitation density, (a) for a quasi-bulk sample, (b) for a 3μm thick GaN/SiC epilayer. The insets give the integrated intensity of the observed PL bands as a function of excitation density which was varied between 10kW/cm^2 and 3MW/cm^2.

an intensive donor-bound-exciton luminescence (I_2) at 3.4728 eV with a binding energy of 7.2 meV and a full width at half maximum of only 1 meV typical for the HVPE samples.[4] At increased excitation levels a broad P-band superimposing the I_2 is detected, Fig. 1 (a). Its peak shifts only slightly to lower energies with increasing density and is found at 3.453 eV at 3 MW/cm^2 and beyond. This position corresponds precisely to that expected for an inelastic scattering process of excitons in which the second exciton is scattered into the continuum.[5] On the very broad low-energy tail of this band we find two LO side bands of the free-exciton emission X_A whose zero-phonon line is obscured by the high-energy shoulder of the P-band. The integrated intensity of this band exhibits a sublinear increase with excitation density. The slope is 0.8. This picture changes for the thin epilayer, Fig. 1 (b). Here, at low excitation densities the free and donor-bound-exciton emissions are detected with almost equal intensities. Their peak positions are shifted to lower energies by 12.5 meV with respect to the corresponding bulk peak energies due to the tensile strain in the epilayer.[6] At increased excitation levels these lines give way to a broadening emission M which we ascribe to the decay of biexcitons. Lineshape fits for the biexciton emission employing a simple model described below are represented by dashed curves in Fig. 1 (b). They reproduce the broadening in the low-energy tail and the red shift of the M-band. From the fitted parameter of the emission threshold the as yet unknown binding energy of the biexciton in GaN can be determined for the first time. It amounts to 3.7±0.5 meV. The intensity increase of the M-Band with excitation density has a virtually linear slope of 1.06. We

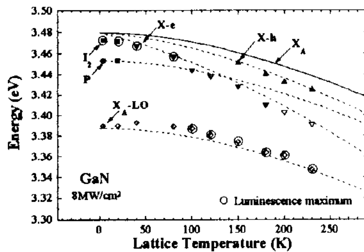

Fig. 2: Temperature dependence of the HEPL peaks of sample 1. Symbols represent experimen-tal values, dashed lines calculations. The solid line is the excitonic band gap measured at low-density cw excitation.

observe two LO sidebands of X_A but the P-band, expected around 3.441 eV, is not found in this sample.

In order to identify relevant emission mechanisms up to room temperature we also performed temperature-dependent HEPL measurements. The results are summarized in Fig. 2 which is a plot of the peak energies as a function of temperature for the quasi-bulk sample 1. The plot shows the experimental values represented by symbols as well as expected temperature shifts (dashed lines) which were calculated using specific models and the measured dependence of the excitonic gap (full line).[7] The energy of the overall luminescence maximum is marked by an open circle for each temperature. At low temperatures up to 40 K the HEPL is dominated by the I_2 (filled squares). Above 40 K inelastic scattering between excitons and free electrons becomes most prominent (down triangles). It is identified by its strong red shift with increasing temperature which can be followed up to 220 K. Above 100 K the LO-sideband of the free-exciton emission exhibits the maximum intensity (diamonds). In this temperature range we also observe another band whose energy shift points to inelastic exciton-hole scattering as the responsible mechanism (up triangles). Towards room temperature the luminescence structures become very broad but are still dominated by the LO processes.

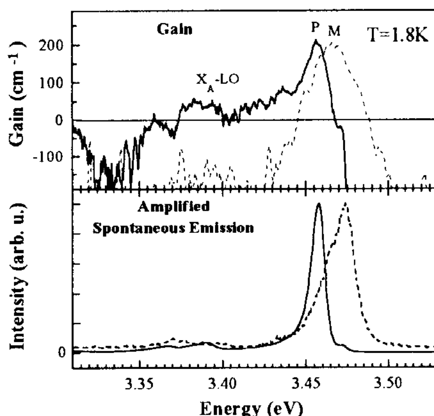

Fig. 3: Low-temperature amplified sponateous emission and gain spectra at excitation densities of ≈5 MW/cm². The spectra of sample 2 (dashed lines) are shifted by 12.5 meV to higher energies to compensate strain effects and allow a direct comparison.

Amplified-spontaneous-emission and gain spectra of both samples are compared in Fig. 3. The low-temperature gain of sample 1 reflects the corresponding HEPL spectrum of Fig. 1 (a). Inelastic exciton-exciton scattering (P) is the only mechanism producing substantial gain. A weak maximum is seen for the LO side band of the free exciton. For the thin epilayer the gain maximum at low tempera-tures is also seen at the position of the HEPL maximum, i.e., the M-band and its phonon replica.

3 Discussion

The decay of biexcitons and the resulting luminescence is well understood for most wide-gap semiconductors from both the experimental and theoretical points of view.[8] Two different excitonic polaritons are created under conservation of energy and momentum, one in the photonic branch and one in an excitonic branch, longitudinal or transversal. In a simplified model the luminescence spectrum exhibits the inverted distribution of the population of momentum states on its low-energy side, i.e., a Boltzmann tail multiplied by the density of states.[9] The lineshape fits of Fig. 1 (b) were calculated this way. The high-energy threshold E_{th} of the luminescence is expected for vanishing biexciton momentum and is therefore given by

$$E_{th} = E_X - E_M^b . \quad (1)$$

E_X denotes the exciton energy. Thus, by fitting the M-band the value of the biexciton binding energy E_M^b can be determined. With the value of 3.7 meV we obtain a biexciton energy for this GaN epilayer of $E_M = 6.930 \pm 0.001$ eV. The observation of luminescence above E_{th} is a sign of lifetime broadening of the momentum states due to scattering processes with excitons or biexcitons.[10] This process is also evidenced by the almost linear dependence of the M-band intensity on excitation density, cf. the inset of Fig. 1 (b). Under the equilibrium conditions created in our experiment by the comparatively long excitation pulses of 15 ns duration the interaction with excitons is capable of reducing the logarithmic slope of the intensity dependence from the expected value of 2 to a linear relation.[11] As to why the spectra of the two samples exhibit different high-excitation effects at low temperatures only speculations are possible at this point. More detailed investigations are under way.

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