OPTICAL-GAIN MEASUREMENTS ON GaN AND $Al_xGa_{1-x}N$ HETEROSTRUCTURES

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ABSTRACT

Optical gain processes in thin GaN and AlGaN are compared by means of gain spectroscopy using the stripe length method and high-excitation photoluminescence, both performed at various densities and temperatures. We find that inelastic excitonic scattering processes and biexciton decay are important at low temperatures and low excitation densities Both materials are similar in that increasing the excitation density results in gain spectra dominated by the electron-hole plasma and phonon-assisted band-to-band recombination. These also prevail at high temperatures.

INTRODUCTION

The physical processes causing stimulated emission in GaN-based laser structures are still subject to discussion. Localized biexcitons [1], the electron-hole plasma [2] and band-to-band recombination [3] have been claimed responsible for the laser mechanism in group-III-nitride heterostructures. Meanwhile, the properties of GaN and especially AlGaN at high excitation levels are not understood in detail. Most investigations have been limited to room temperature not allowing an unambiguous identification of the electronic processes involved. With increasing temperature the contribution of phonon-assisted processes becomes larger resulting typically in a broad spectral region of optical amplification. In our previous work [4] we studied the gain spectra of thick quasi-bulk GaN between 2 K and room temperature and found that excitonic processes add to the gain at high temperatures. The purpose of the present paper is to analyze optical gain spectra obtained from thin GaN epilayers at very high excitation levels and to study the influence of increasing Al content on the gain in AlGaN epilayers. For the identification of the mechanisms providing optical amplification in GaN and AlGaN useful information is obtained from high-density effects on the spontaneous and stimulated photoluminescence (PL).

EXPERIMENTAL

The results presented here were obtained from a MOCVD-grown GaN/SiC epilayer of 3 μm thickness [5] whose low-density free-exciton resonance energy is 3.468 eV at 1.8 K, and from a series $Al_xGa_{1-x}N$ samples grown by MBE on (0001) sapphire with a thickness of about 1 μm [6]. To obtain the high excitation density necessary for our investigations we used a dye laser pumped by an excimer laser, providing pulses with a duration of 15 ns at a rate of 30 Hz and a total energy of up to 20 μJ at 340 nm. AlGaN samples were pumped by the excimer

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laser at 308 nm using similar pulse energies. The samples were either mounted in a bath cryostat at 1.8 K or in a helium flow cryostat at temperatures varied between 4 K and 300 K. Gain measurements were performed using the stripe length method [e.g., 7].

RESULTS

Dependence of High-Excitation PL from Thin GaN Epilayers on Intensity and Temperature

In a previous paper [8] we demonstrated that the radiative decay of biexcitons (M-band) can be the dominating low-temperature emission and gain process in GaN at excitation densities up to 5 MW/cm^2. In order to study the properties of the electron-hole plasma (EHP) in GaN we further increased the excitation density. The dependence of the PL of GaN on excitation density up to 50 MW/cm^2 is displayed in Fig. 1. The most striking observation is the appearance of a new emission band around 3.43 eV at 10 MW/cm^2. It exhibits a superlinear growth of emission intensity while strongly shifting its peak position to lower energies. This behavior is typical for stimulated emission from an electron-hole plasma. Similar observations in GaN at low temperatures were made before by Cingolani *et al.* [9]. Wiesmann *et al.* recently showed by room temperature measurements that the observation of this strong peak from the surface of the epilayer is due to scattering of in-plane stimulated emission [10]. We ascribe the high-energy maximum around 3.46 eV to a superposition of various spontaneous and stimulated emission processes. With the fast decay of the density of the plasma after the excitation pulse band-to-band recombination (B), inelastic scattering processes between excitons (P), and the formation of biexcitons (M) become possible and all give rise to luminescence in the same energy range. The gain measurements presented below will prove this interpretation.

Fig. 2 compares the temperature dependencies of the observed luminescence peak positions at 3 MW/cm^2 and at 50 MW/cm^2. The biexciton luminescence M is quenched at higher lattice temperatures. From the Arrhenius plot of the integrated M-band intensity up to 80 K (shown in the insert) we deduce an activation energy of 3.5 meV which in spite of its high tolerance of 40% confirms the interpretation of this band as well as the previously determined biexciton binding energy of 3.7 meV [8]. Above 60 K the observed peak position of the dominating luminescence at 3 MW/cm^2 agrees with that expected for inelastic exciton-exciton scattering (P-band) as indicated by the dashed line labeled P. Longitudinal optical (LO) phonon assisted exciton decay appears throughout the measured temperature range up to 250 K at the energy expected. Neither a screening of the exciton

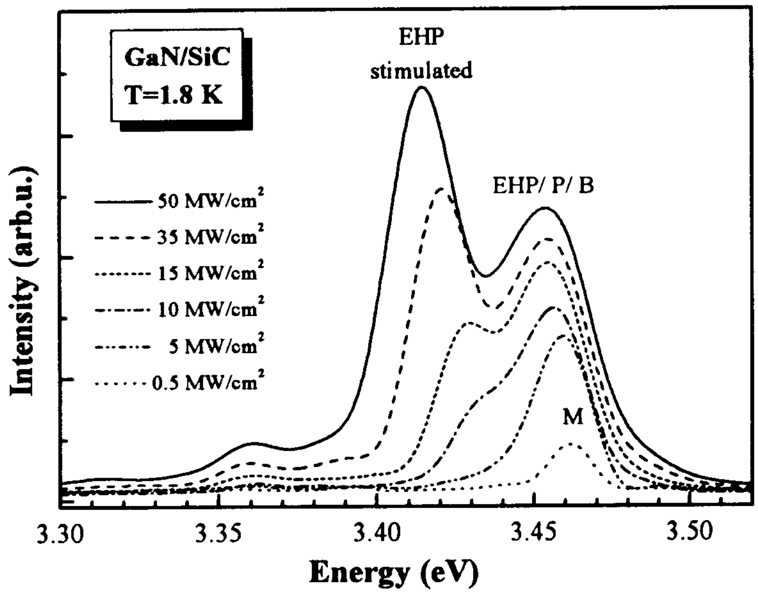


Fig. 1: Low-temperature surface PL from a 3 μm thick GaN/SiC epilayer at excitation densities between 0.5 and 50 MW/cm^2.

binding nor a band gap renormalization can be seen at this excitation level. In contrast, at 50 MW/cm^2 no LO replica of free-exciton emission is detected. The stimulated EHP peak exhibits a strikingly strong red shift with temperature. Above 200 K its position is near that of X_A-LO at lower densities. However, up to room temperature the integrated intensity of the EHP peak follows the predicted T^{-3}-rule [11]. This observation indicates that also at room temperature the emission is caused by an EHP. At 270 K the measured value of 3.221 eV is virtually equal to those often reported in the literature for stimulated emission from GaN at room temperature [e.g.,12]. The high-energy band seen in the luminescence spectra at 50 MW/cm^2 exhibits a red shift with temperature that is weaker than that of the stimulated EHP peak but stronger than the low-density excitonic band gap given by the dashed line labeled X_A. At higher temperatures it appears at the energy position expected for the P-Band. Summarizing the main results of this section, at very high excitation densities the PL of the thin GaN epilayer is dominated by the spontaneous and scattered stimulated emission of the EHP. In addition, a high-energy broad emission is attributed to a superposition of band-to-band recombination and excitonic processes. In the following section we will investigate the processes contributing to the optical gain at different excitation levels and lattice temperatures.

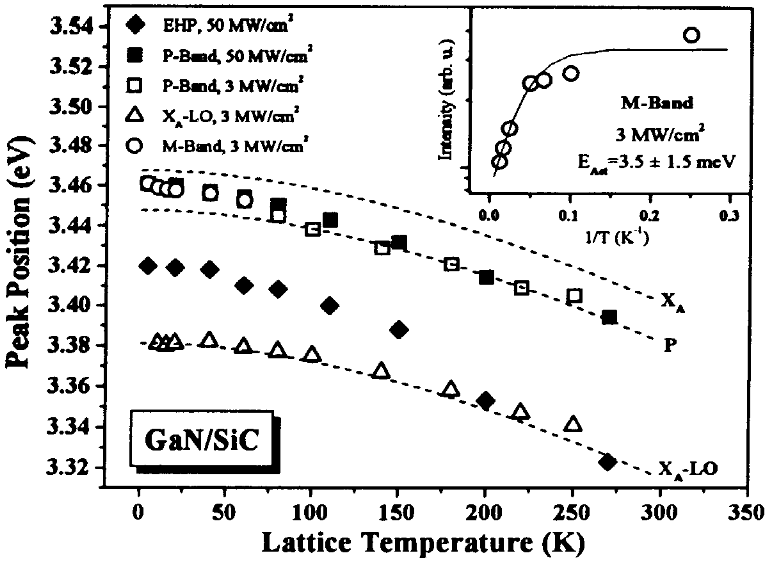
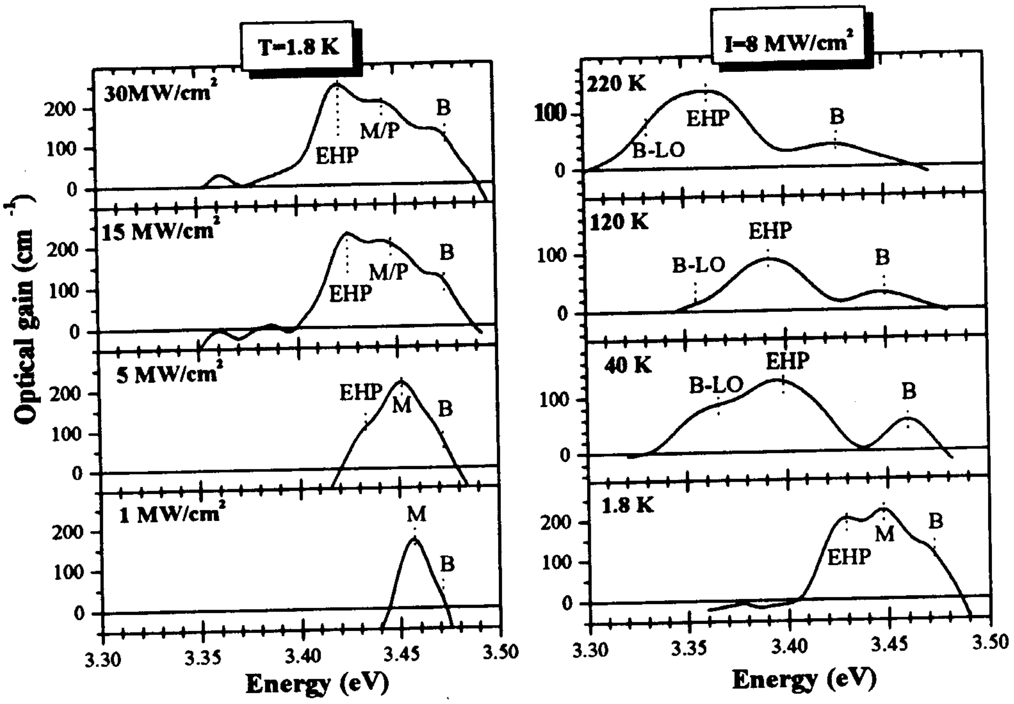


Fig. 2: Peak positions vs. lattice temperature for the PL of the GaN/SiC sample of Fig. 1 at excitation densities of 3 MW/cm^2 (open symbols) and 50 MW/cm^2 (full symbols). Dotted lines indicate expected peak positions for the respective emissions. The insert displays the integrated intensity of the M-Band as a function of lattice temperature.

Temperature and Intensity Dependent Gain Properties of Thin GaN Epilayers

Fig. 3 displays two series of gain spectra taken at various excitation densities (a) and temperatures (b), respectively. The spectra presented here were smoothed to enhance the visibility of the observed spectral features. In the density series of Fig. 3 (a) a broadening of the region of optical gain with increasing pump intensity is observed. Additional structures B and EHP appear on both the high- and low-energy shoulders of the main peak M. Their relative strength with respect to M increases with excitation. This main peak is due to biexciton decay below 8 MW/cm^2. Above, inelastic exciton-exciton scattering P is likely to contribute in the same energy range. The low-energy peak EHP can be identified as due to the electron-hole plasma by its energy position and shift characteristics which are identical to those of the stimulated EHP emission shown in Fig. 1. At 30 MW/cm^2 the EHP represents the dominating low-temperature gain mechanism with a peak gain value of 250 cm^{-1}. The high-energy gain peak B appears at energies near or above the band gap of the sample. This peak



(a) (b)

Fig. 3: Optical gain spectra of a 3μm GaN/SiC epilayer (a) at 1.8 K and various excitation densities, (b) at a fixed excitation density of 8 MW/cm^2 and various temperatures. Labels are explained in the text.

does not correlate to a pronounced spontaneous emission peak. The zero-crossing energy due to this process shifts to higher values at increased pump intensities. We ascribe this gain peak to band-to-band recombination. The blue shift with increasing pump intensity is typical for this gain mechanism and is caused by a shift of the respective quasi-Fermi levels of holes and electrons into the valence and conduction bands [13].

Our observation of gain caused by plasma, free carriers, and excitonic processes at highest excitation levels does not imply that they all occur simultaneously. In our time-integrated measurements we detect all processes which produce optical gain after the excitation pulse. Time-resolved measurements of the optical gain in CdS showed that carrier diffusion as well as the formation of biexcitons and excitons contribute to the ultrashort decay of the plasma within 100-200 ps [14]. It is well known that free carriers at high densities as well as biexcitons and inelastic excitonic scattering processes give rise to optical gain themselves. Thus, our observations are in perfect agreement with the typical behavior observed in other direct wide-gap semiconductors. The temperature-dependent gain measurements of Fig. 3 (b) taken at a fixed excitation density of 8 MW/cm^2 show that below the near-gap gain band a second broad low-energy band appears and grows with increasing temperature. It is this structure that is responsible for the stimulated emission at room temperature. In the high-energy band only band-to-band recombination B can be identified without doubt due to its zero-crossing energy above the band gap of the sample at the respective temperatures. Since the shape of this gain band is untypical for pure band-to-band transitions excitonic processes probably also contribute to a small extent to the gain in this region at higher temperatures. Inelastic scattering processes between excitons or between excitons and free carriers are typical gain processes at higher temperatures and were observed in thick GaN epilayers before

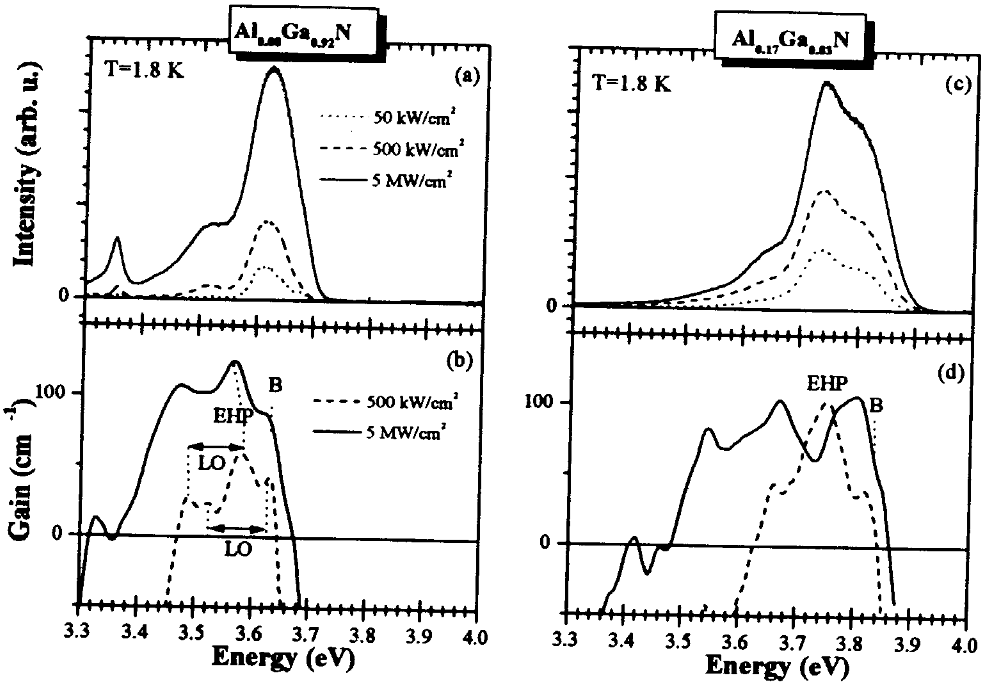


Fig. 4: Comparison of low-temperature high-excitation luminescence (a), (c) and optical gain spectra (b), (d) from two $Al_xGa_{1-x}N$ samples. Legends on the left-hand side also apply to the plots on the right-hand side.

[4]. However, in the thin epilayer investigated here excitonic processes do not play an important role for the gain at high excitation densities and high temperatures. Instead, the electron-hole plasma and LO-assisted band-to-band recombination are the dominating processes causing the low-energy gain band at temperatures above 40 K.

High-Excitation PL and Gain in Thin AlGaN Epilayers

Fig. 4 displays a comparison of high-excitation photoluminescence and gain spectra taken at 1.8 K from two $Al_xGa_{1-x}N$ samples with x equal 0.08 on the left-hand side and 0.17 on the right-hand side. The excitation density was varied between 50 kW/cm^2 and 5 MW/cm^2. From low-density PL measurements the excitonic band edge of these samples is found at 3.63 eV (x=0.08) and 3.83 eV (x=0.17) at 1.8 K. Both the luminescence and gain spectra of the two samples extend to energies well above these values, shifting further to higher energies with increasing excitation density. This behavior gives strong evidence for band-to-band recombination. The gain spectrum of $Al_{0.08}Ga_{0.92}N$ in Fig. 4 (b) also exhibits a pronounced peak around 3.57 eV strongly shifting to lower energies with increasing excitation density. This allows an attribution of this band to the electron-hole plasma. No indication of a stimulated peak is observed in the luminescence spectra up to 5 MW/cm^2. The two weak low-energy gain structures around 3.5 eV are found one LO phonon energy below the dominating lines and strongly gain intensity with increasing pump power. The role of phonon-assisted processes is stronger here than in the experiments on GaN because the excitation energy (4 eV) is higher above the band gap producing a plasma with higher effective temperature whose relaxation towards thermal equilibrium creates a larger number of phonons available for a stimulated emission process. Unlike the results for GaN even at low temperatures excitonic processes do not seem to contribute much to the optical gain at the given excitation densities in this AlGaN sample. We ascribe this to the higher density of excited carriers due to the small thickness of 1 µm which is a factor of three lower than that of the GaN sample studied. Similar considerations apply to the gain spectra of the $Al_{0.17}Ga_{0.83}N$ sample in Fig. 4 (d). It is obvious by the spectrally broad region of optical amplification that phonon-assisted processes play the dominant role at 5 MW/cm^2. From the results obtained to this point we conclude that the AlGaN samples investigated show gain mechanisms very similar to those of high-quality GaN. The electron-hole plasma and band-to-band recombination are the main processes providing optical gain at high excitation densities. While the absolute values are somewhat lower our results show that the fabrication of laser structures with active AlGaN layers operating in the ultraviolet spectral range is possible.

CONCLUSIONS

In conclusion, we compared the luminescence and optical gain properties of thin GaN and AlGaN epilayers at high excitation densities. The GaN and AlGaN samples investigated exhibit a similar behavior. Excitonic processes play an important role only at relatively low densitiy and at low temperatures. With increasing pump intensity the electron-hole plasma and band-to-band recombination exhibit the highest gain reaching values of 250 cm^{-1} in GaN and 150 cm^{-1} in AlGaN. Towards room temperature phonon-assisted band-to-band recombination and electron-hole-plasma create a gain structure roughly 100 meV wide giving rise to the often reported stimulated emission peak.

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