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Properties of the Biexciton and the Electron-Hole-Plasma in Highly Excited GaN

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Abstract

High-excitation processes like biexciton decay and recombination of an electron-hole-plasma are discussed as efficient mechanisms for lasing in blue laser diodes [1]. Therefore, the investigation of these processes is of fundamental importance to the understanding of the properties of GaN as a basic material for optoelectronical applications. We report on comprehensive photoluminescence and gain measurements of highly excited GaN epilayers grown by metal-organic chemical vapor deposition (MOCVD) over a wide range of excitation densities and temperatures. For low temperatures the decay of biexcitons and the electron-hole-plasma dominate the spontaneous-emission and gain spectra. A spectral analysis of the lineshape of these emissions is performed and the properties of the biexciton and the electron-hole-plasma in GaN will be disscused in comparison to other wide-gap materials. At increased temperatures up to 300 K exciton-exciton-scattering and band-to-band recombination are the most efficient processes in the gain spectra beside the electron-hole-plasma.

1. Introduction

Tremendous success in the growth and processing technologies of group-III nitrides have recently led to a first GaN-based laser structure operating in the ultraviolet spectral range [1]. For this structure the decay of localized biexcitons [2] and the recombination of an electron-hole-plasma are discussed as possible mechanisms responsible for lasing [3]. This indicates the need for a basic understanding of their properties in GaN. We therefore performed photoluminescence and gain spectroscopy at high excitation densities on GaN-epilayers grown by metal organic chemical vapor phase deposition (MOCVD).

2. Experimental

The sample used for this study was grown on a 6H-SiC substrate [4] with an epilayer thickness of 3µm. To obtain the high excitation (HE) density necessary for our investigations we used a dye laser pumped by an excimer laser, providing pulses at a rate of 30Hz with a typical duration of 15ns and an energy of up to 20µJ at 340nm. The sample was kept in a cryostat at temperatures varied between 2 K and 300 K. Gain measurements were performed using the stripe length method [5].

3. Results

3.1. Photoluminescence at High Excitation Levels

At low excitation densities the MOCVD sample investigated exhibits a very strong emission I_2 at 3.461 eV caused by the annihilation of excitons bound to neutral donors. This luminescence has a linewidth of 5 meV in the sample presented here. The energy of the free A-exciton in this sample is 3.467 eV as determined in high-resolution PL measurements. Figure 1 shows the emission spectra obtained from this epilayer for various temperatures up to 250 K and an excitation intensity of 3 MW/cm^2. For low temperatures up to 40 K the dominating emission is observed around 3.46 eV. Luminescence from excitonic molecules is expected in the same energy range and as shown in previous papers [6] an emission- here called M-band- occurs which we ascribe to the decay of biexcitons. Lineshape fits for the biexciton emission employing a simple model described in [6] reproduce the broadening in the low-energy tail and the red shift of the M-band. The intensity increase of the M-Band with excitation density has a slope of 1.5, typical for a high excitation effect. From the fitted parameter of the emission threshold the binding energy of the biexciton in GaN can be determined. It amounts to 3.7±0.5 meV. This is a smaller value than the reported binding energies of 5.3 meV [7] and 5.7 meV [8] for epitaxial GaN deduced from the difference of the peak positions of the free exciton and the biexciton. But it is important to note that the exact value of the binding energy of a biexciton can only be deduced from a lineshape analysis since the distribution of the kinetic energy of the biexcitons influences the lineshape, shifting the peak position to lower energies with increasing intensity while the binding energy, of course, is constant. The given value of the biexciton binding energy of 3.7 meV is be confirmed by results from temperature dependent measurements as well. From the Arrhenius plot- displayed in the insert of Figure 1- we deduce an activation energy of 4.1 meV which confirms the biexciton binding energy determined previously.

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

Above 60 K the observed peak position agrees with the predicted temperature dependence for inelastic exciton-exciton-scattering processes (P-band) as indicated by a dashed line in Figure 2 The LO-assisted exciton decay is observed up to 250 K as well at the expected energy position.

In Figure 3 the PL measurements for higher excitation densities up to 50 MW/cm^2 are displayed. At 10 MW/cm^2 a new emission band appears at 3.43 eV. This band exhibits a superlinear growth and strongly shifts its peak position to lower energies. The linewidth amounts to 20 meV for the highest intensities. These are typical properties for the recombination of an electron-hole-plasma: Similar observations in GaN at low temperatures were made by Dai *et al*. [9] and Cingolani *et al.* [10].

The temperature dependence of this band is displayed in Figure 4 for a range of 4 to 250 K and an excitation intensity of 30 MW/cm^2 . The energy position of the peak exhibits a strong red shift with temperature due to the renormalization of the band gap by a strong screening of the coulomb interaction at the high carrier densities given.

From the lineshape analysis the effective temperature and the carrier density of the plasma were deduced. As an example of the nonequilibrium behaviour of the EHP the effective temperature is plotted versus the lattice temperature in the insert in Figure 4. The external variation of the lattice temperature has only little effect on the plasma carrier. This is known from other materials like CdS as well [11]. At high excitation densities the relaxation time for carriers in the bands is limited by the lifetime of the plasma. For CdS some ten ps as relaxation time for hot carriers are observed [12], which is in the range of the lifetime of the EHP. Therefore, the carrier-phonon interaction is not an effective relaxation channel. Only the carrier-carrier interaction determines the effective temperature of the EHP, which is largely due to the high excitation energy.

It is interesting to note, that the often reported stimulated emission peak [13], [14], agrees very well in energy position and spectral features to the EHP band observed in our measurements. It is important to take into account that the spectral features of an EHP luminescence are strongly influenced by the superposition of the stimulated emission due to high gain values. Therefore, despite the different excitation conditions the often reported stimulated-emission peak is due to the recombination of an electron-hole-plasma.

On the high energy side of the EHP-band another band- here called B- is observed up to 250 K. The energy of the peak position agrees with the temperature dependence of an exciton-exciton-scattering process, as shown in Figure 5. The high-energy tail of the luminescence exhibits a “blueshift» with increasing temperature, if one considers the temperature dependence of the bandgap. This indicates band-to-band transitions as another emission process. Therefore, we assign this luminescence band B to a superposition of several recombination processes. This will be discussed in detail in the next section where intensity-and temperature-dependent gain measurements will be presented.

3.2. Temperature and Intensity Dependence of the Gain Spectra

In Figure 6 gain spectra taken at helium temperature for various intensities are shown. The spectra presented here were smoothed to enhance the visibility of the observed spectral features. In the density series of Figure

6 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. This main peak is due to biexciton decay for densities up to 5 MW/cm^2 . For higher intensities inelastic exciton-exciton scattering is likely to contribute to the optical gain in this energy range.

The low-energy peak can be identified as due to the recombination of an electron-hole plasma by its energy position and shift characteristics which are identical to those of the spontaneous emission shown in Figure 2. At 30 MW/cm^2 the EHP represents the dominating low-temperature gain mechanism with a gain value of 250 cm^{-1} . On the high-energy side of the M- and B-band occurs a broadening occurs at energies near or above the band gap of the sample and the crossover from gain to absorption shifts to higher energy at increased pump intensities. Due to its spectral position and the typical blue shift with intensity we ascribe the high-energy gain to band-to-band (here called BBT) transitions. This typical blue shift with increasing pumping intensity is caused by a shift of the quasi Fermi levels into the valence and conduction bands due to band filling processes. It is interesting to note that electron-hole-plasma, free-carrier and excitonic processes coexist at high excitation levels. This is well known from other materials like CdS [12] and is explained by a spatial and temporal seperation of the excitonic and many-particle processes. After the excitation a particle diffusion to lower excited regions occurs and there the formation and recombination processes of biexcitons as well as exciton-exciton-scattering are effective as gain mechanisms. In our time-integrated measurements all these processes contribute to the observed optical gain as well. The lifetime of the EHP can be roughly estimated to 350 ps by comparing the number of excited carriers and the carrier densitiy from the line shape analysis. Thus, our observations are in good agreement to the behaviour of other direct-gap semiconductors like CdS or GaAs.

The temperature-dependent gain measurements are displayed in Figure 7. For a fixed excitation intensity of 8 MW/cm^2 a structured gain spectrum is observed up to high temperatures. On the high- energy side of the peak B a “blueshift» with temperature can be observed, considering the stronger red shift of the bandgap. This strongly indicates that band-to-band transitions are responsible for gain as well up to 220 K. The B peak shifts its energy position with temperatures exactly like exciton-exciton-scattering, which indicates that excitonic contributions are effective gain processes up to high temperatures. This was shown for thick GaN epilayers as well [6]. From 40 K on phonon-assisted processes like the A-LO decay become important. For higher temperatures the dominating gain peak is a superpositon of the EHP and phonon-assisted transitions, reaching gain values up to 130 cm^{-1} for 220 K. This indicates the importance of these processes as highly efficent mechanisms for lasing in GaN.

4. Conclusion

In conclusion, we reported on the temperature and intensity dependence of spontaneous-emission and gain spectra of thin GaN epilayers at high excitation levels. The biexciton decay plays a dominant role in the optical gain spectra for lower temperatures up to 40 K. At higher temperatures, phonon-assisted and excitonic interactions, the formation of an electron-hole plasma and band-to-band transitions are responsible for gain values up to 130 cm^{-1} indicating their importance as effective mechanisms of optical amplification for device applications.

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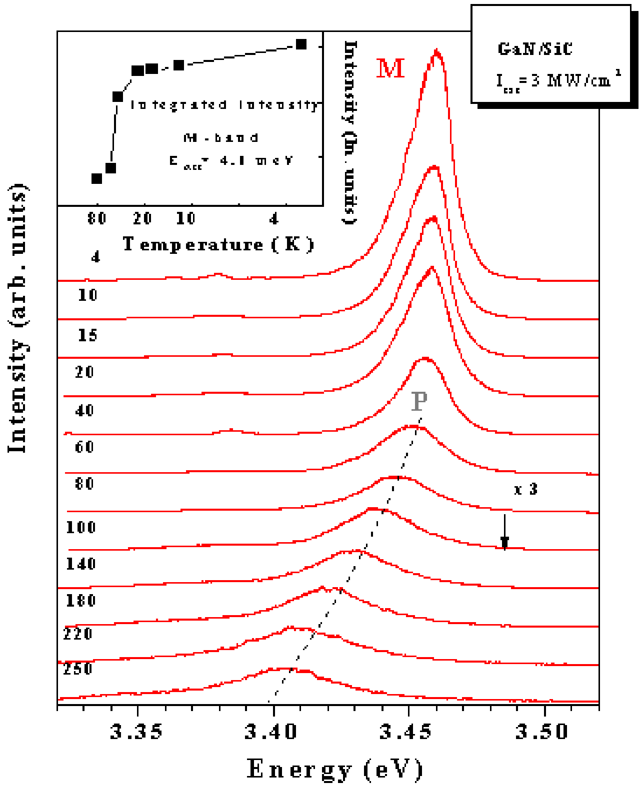

Figure 1. Temperature dependent PL spectra for an excitation density of 3 MW/cm^2 . The insert displayes the integrated intensity of the M-band versus temperature.

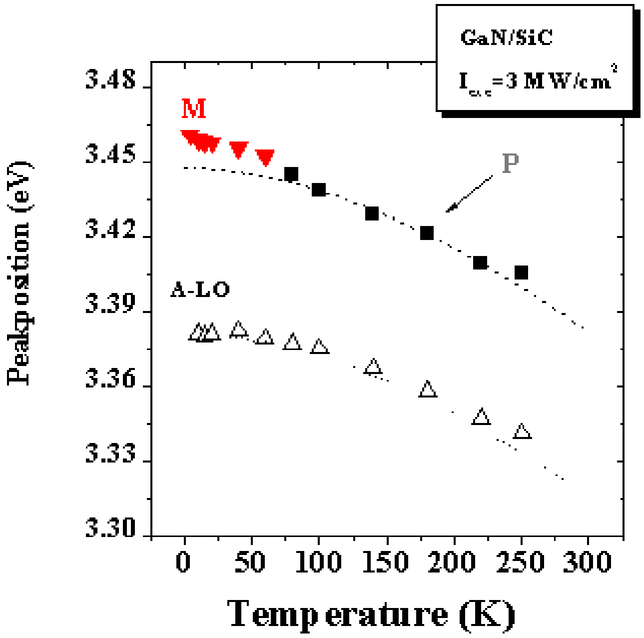

Figure 2. Temperature dependence of the observed peak position for an excitation density of 3 MW/cm^2

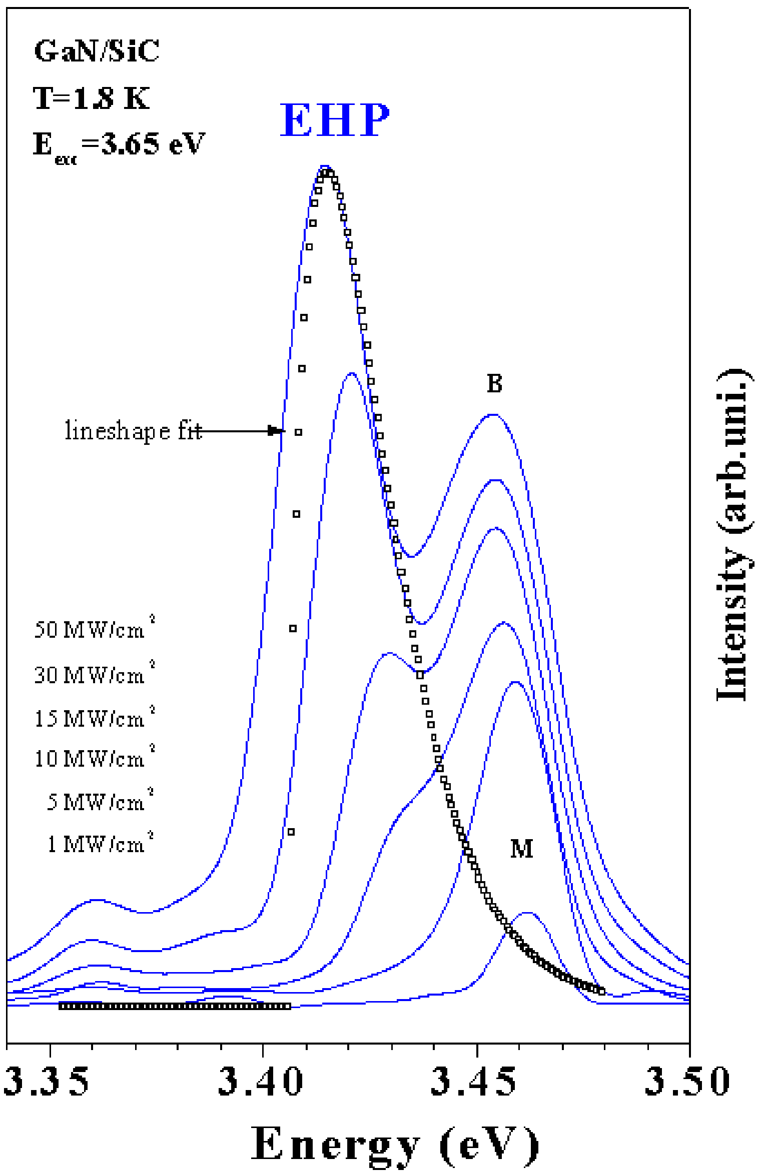

Figure 3. Intensity dependence of the PL at low temperatures

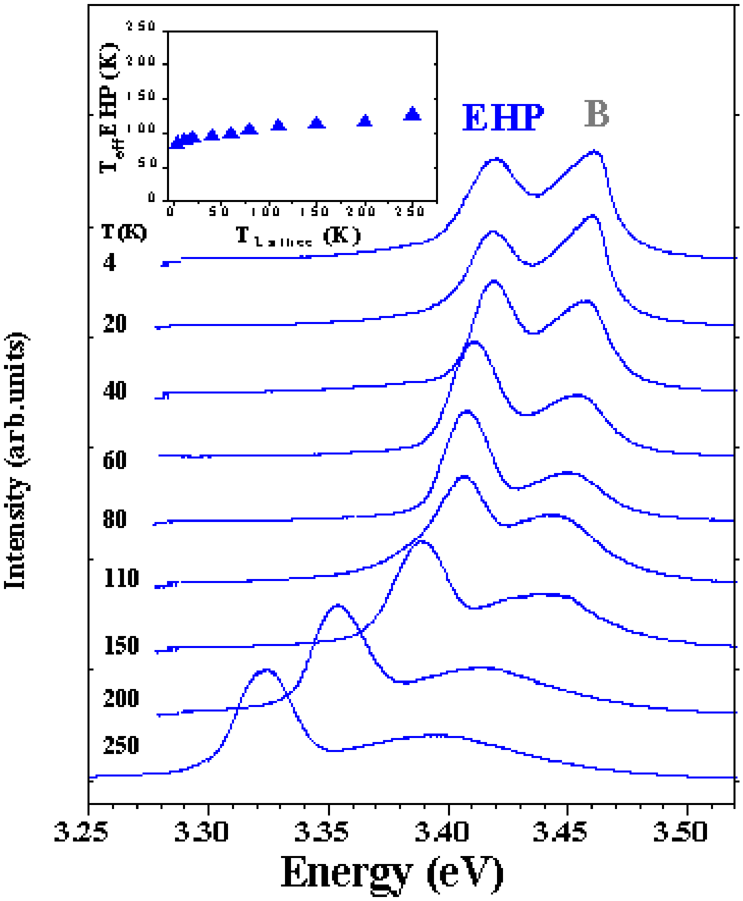

Figure 4. Temperature dependent PL spectra for an excitation density of 30 MW/cm^2 Insert: effective plasma temperature versus the external lattice temperature

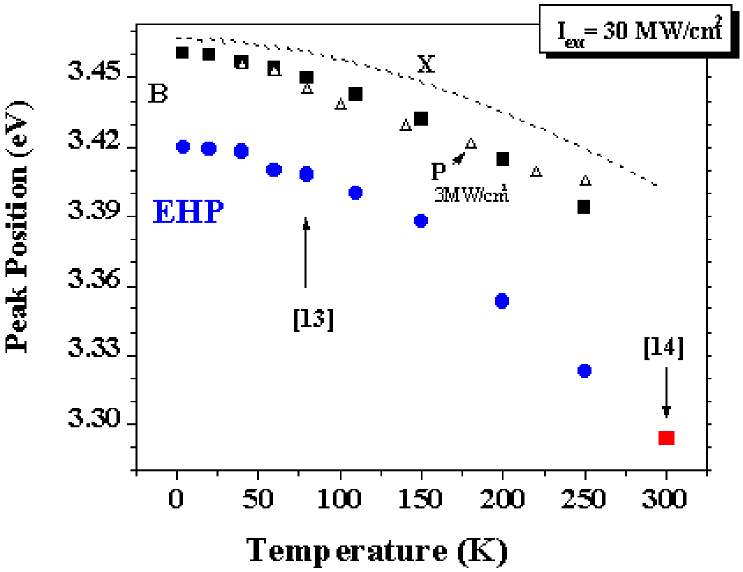

Figure 5. Temperature dependence of the peak position for an excitation intensity of 30 MW/cm ^{2}

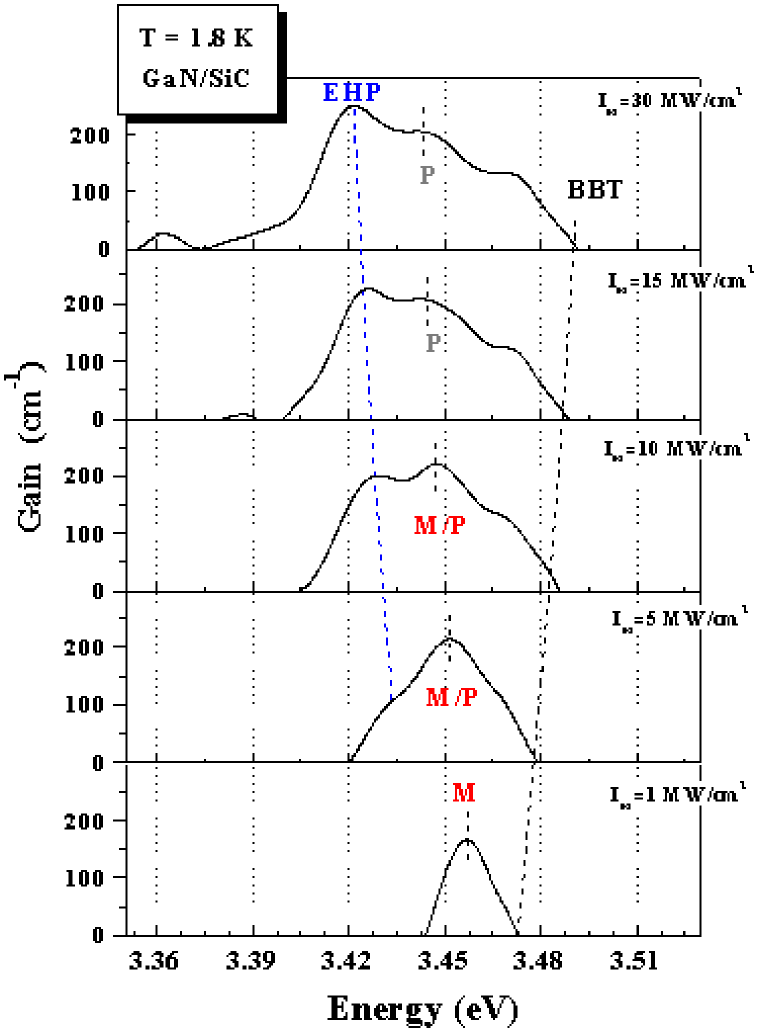

Figure 6. Intensity dependence of the gain spectra at 1.8 K

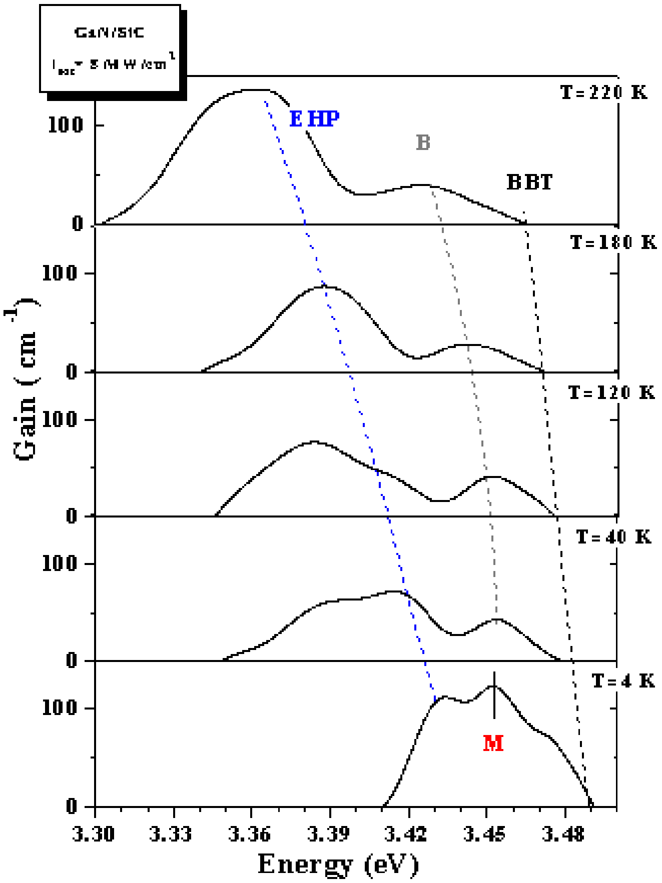

Figure 7. Temperature dependent gain spectra for an exciation intensity of 8 MW/cm^2