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Gain Spectroscopy of HVPE-Grown GaN

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Abstract

We report on photoluminescence and optical gain measurements of highly excited GaN crystals grown by hydride vapor physe epitaxy (HVPE). Inelastic scattering processes of excitons dominate the spontaneous emission spectrum under high excitation up to temperatures of 180 K. Towards room temperature phonon-assisted recombination of excitons and free carriers begins to dominate the spectrum. Similar characteristics are observed in temperature-dependent gain measurements.

1. Introduction

Tremendous successes 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]. Despite this progress in technology there is a need for the basic understanding of the physical mechanisms providing optical gain in GaN and its alloys with InN and AlN. We therefore performed photoluminescence and gain spectroscopy at high excitation densities on bulk GaN grown by hydride vapor phase epitaxy (HVPE).

2. Experimental

The samples used for this study were grown on (0001) sapphire substrates at 1090 C [2]. Their epilayer thickness ranges between 200µm and 400µm. The typical room temperature carrier concentration is less than $n = 10^{17}$ cm^{-3}.

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 [3].

3. Results

3.1. Photoluminescence at High Excitation Levels

At low excitation densities the HVPE samples investigated typically exhibit a very strong emission I_2 at 3.472 eV caused by the annihilation of excitons bound to neutral donors. This luminescence has a linewidth of less than 1meV in the sample presented here. The energy of the free A-exciton in these quasi-bulk samples is 3.4800 eV as determined in high-resolution reflection measurements [4]. Figure 1 shows low-temperature emission spectra obtained from a 300µm thick GaN crystal under various excitation densities. To avoid the detection of stimulated emission as much as possible the sample surface was at an angle of 45 degrees with respect to the optical axis of excitation for these measurements. Increasing the excitation density at low temperatures one observes a broadening of the spectral features. The maximum intensity is still observed around 3.473 eV for excitation densities below 17 MW/cm^2. Luminescence from excitonic molecules is expected in the same energy range. However, the

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lineshape of the observed emission is not typical for a biexciton decay. The assignment to an emission of donor-bound excitons in deeper, less excited regions of the 300μm thick sample seems more likely even though at this point no proof for the nature of this line can be given.

On the low-energy side of this feature a broad luminescence band (P) grows with increasing excitation density, eventually dominating the luminescence above 17 MW/cm^2. It is caused by inelastic exciton-exciton scattering [5]. The measured peak energy of the P-Band is 3.460 eV at 17 MW/cm^2. It agrees very well with the value expected for scattering of an exciton into the $A_{n=2}$ state, given the exciton binding energy of 26 meV [4]. We observe a red shift and broadening of the P-Band at higher excitation levels which can be explained by the fact that scattering into higher excited states becomes more important with increasing exciton density. The peak energy of the P-Band reaches 3.456 eV at 50 MW/cm^2. This value corresponds to scattering of an exciton into the $A_{n=3}$ state assuming an exciton binding energy of 26 meV.

3.2. Temperature Dependence of HE-Luminescence and Gain Spectra

Reports of the HE PL of GaN to date were restricted to 1.8 K [5] or only gave room-temperature and 77 K spectra without identifying the observed emissions [6]. In Figure 2 HE luminescence spectra taken at various temperatures between 4 K and 270 K are shown for an excitation density of 8 MW/cm^2. It is clearly seen that with increasing temperature the contribution of phonon-assisted recombination processes to the luminescence intensity becomes larger. Above 150 K the annihilation of free excitons accompanied by the creation of an LO phonon dominates the spectrum. There are further structures in the spectra which can be identified by their characteristic energy shift with increasing temperature. A plot of the peak energies as a function of temperature is given in Figure 3. The temperature dependence of the A-exciton energy as determined from low-density cw luminescence is given by the dashed line marked 'X_A'. It serves as a reference for calculations of other energy levels as a function of temperature, given by the other (solid) lines in Figure 3. For instance, subtracting the energy of 92 meV of the LO phonon [7] results in the curve marked X_{LO} which starts at 3.388 eV at low temperatures. In this case however, the fit to our experimental data (filled diamonds in Figure 3) is rather poor. There is a systematic blue shift of this phonon-assisted exciton annihilation process under high excitation. It can be explained by the high density of excitons shifting the maximum of the distribution function to higher energies.

The line at 3.473 eV can only be observed below 40 K. The P-Band also disappears in a broadened line at 40 K (down triangles in Figure 3, cf. also Figure 2) that exhibits a stronger red shift with increasing temperature than the excitonic band gap. This behaviour is well known from exciton-electron (X-e) scattering in other wide-gap materials such as CdS and ZnSe [8]. The red shift ΔE with respect to the excitonic gap as a function of temperature was calculated according to [9]

$$\Delta E = \frac{kT}{2}\frac{M}{m_e} \qquad (1)$$

M denotes the exciton mass, m_e the electron mass, and T the temperature. k is Boltzmann's constant. Using the effective mass values given by [10] a good agreement between the measured and calculated temperature dependencies is obtained. X-e scattering remains the dominating emission process until above 100 K the X_{LO} process becomes most intensive. Above 150 K a further structure is resolved (up triangles in Figure 3), whose energy shift suggests the identification as exciton-hole (X-h) scattering. For the calculation of the fit curve for this process in Figure 3 comparable simplifications were made as in the model given in Ref. [9] for X-e scattering. The result is that in this approximation equation 1 can be used replacing the electron effective mass by that of the hole. The fit obtained supports the assignment to inelastic X-h scattering. In the higher temperature range the structures are broadened considerably. Some data plotted for this range are tentative and are represented by the respective open symbols.

The first dedicated optical-gain investigations on epitaxial GaN date back to 1982 [11]. State-of-the-art HVPE material has not been studied by gain spectroscopy before. Gain spectra taken at low, intermediate and high temperatures are shown in Figure 4. At 4 K the dominating gain band is due to inelastic exciton-exciton scattering. The temperature dependence of the gain maximum is shown in Figure 3 by open circles. With increasing temperature the gain maximum shifts to lower energies in a manner which clearly shows that excitonic processes are dominating the gain spectrum up to 180 K. From the experimental data it cannot be decided whether exciton-exciton scattering or exciton-electron scattering is responsible for the gain peak. With increasing temperature phonon-assisted recombination processes play an important role. Above 180 K the gain spectrum is broad and extends over several hundred meV as shown in Figure 4 for room temperature. Gain is also observed at energies as high as 3.5 eV. This

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indicates that an electron-hole plasma may be involved in the process. Further detailed measurements and lineshape fits are under way to unambiguously determine the luminescence and gain properties of GaN.

4. Conclusion

In conclusion, we reported on the temperature dependence of spontaneous and gain spectra of GaN at high excitation levels for the first time. Inelastic scattering processes of excitons play a dominant role in the optical gain spectra up to 180 K. At higher temperatures, phonon-assisted excitonic recombination and the formation of an electron-hole plasma broaden the luminescence and gain spectra at these high excitation levels.

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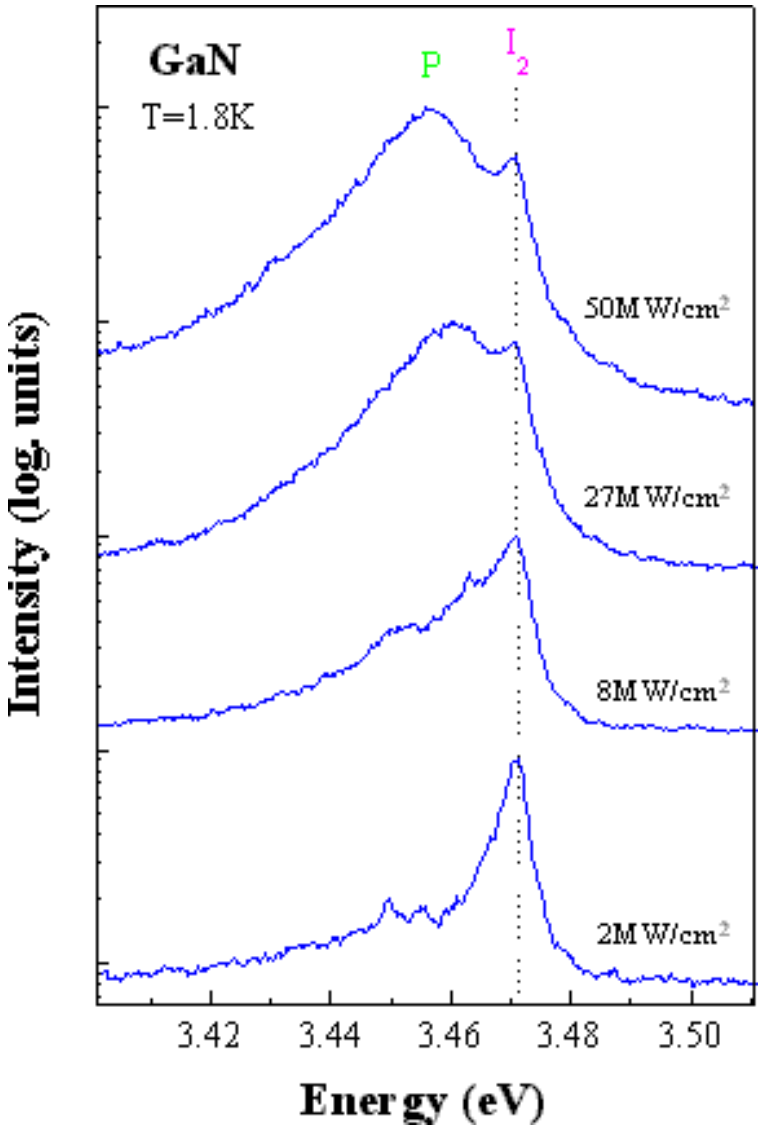

Figure 1. Low-temperature photoluminescence spectra of HVPE GaN at various excitation densities.

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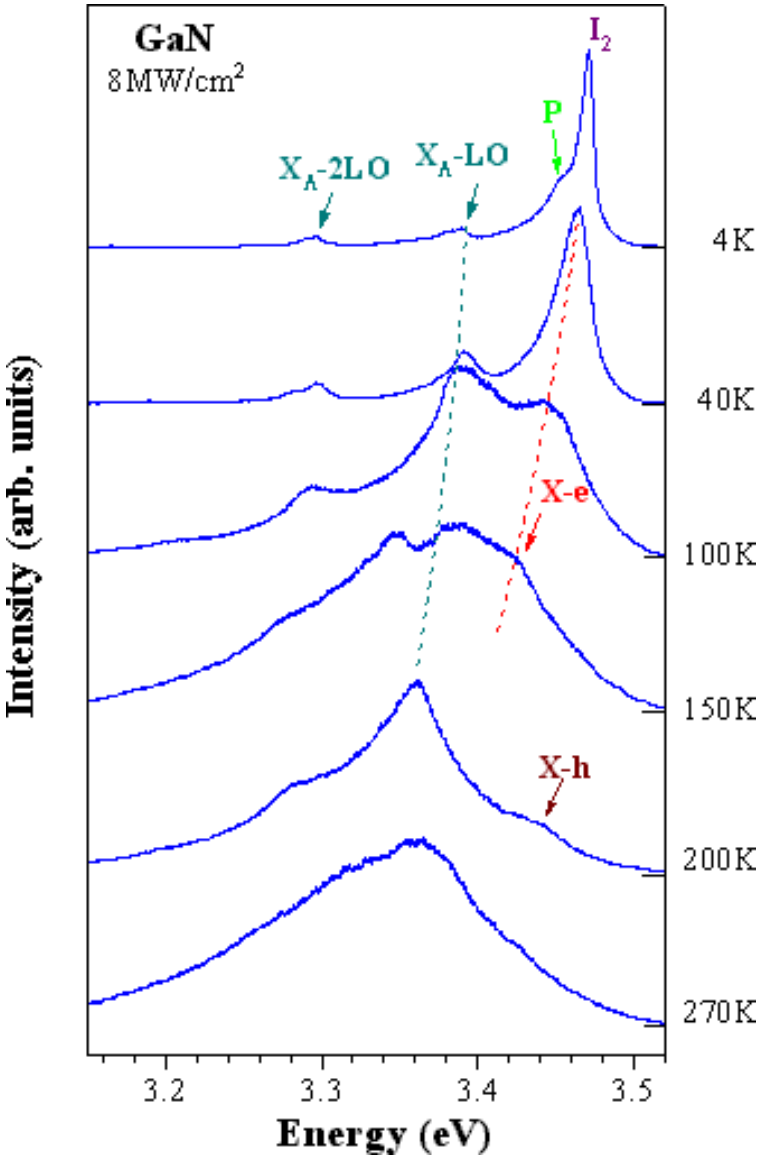

Figure 2. Temperature dependence of the HE luminescence of GaN at an excitation density of 8 MW/cm^2.

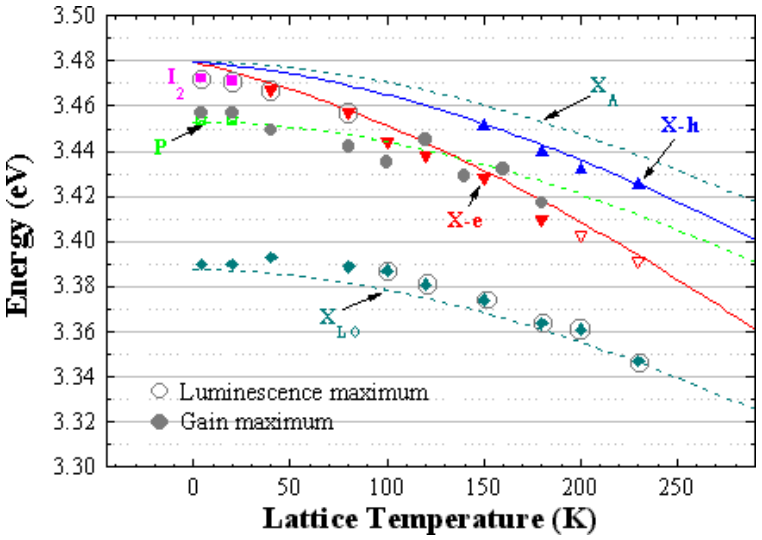

Figure 3. Peak positions of HE luminescences as a function of temperature. Symbols represent the experimental data. Circled symbols mark the energy of the maximum HE-luminescence intensity, full circles give the energy of the maximum of the optical gain at the respective temperature. Dashed lines give the temperature dependence of the free-exciton energy and, deduced from that, its LO-replica and inelastic exciton-exciton scattering (P). Solid lines are fits, cf. text.

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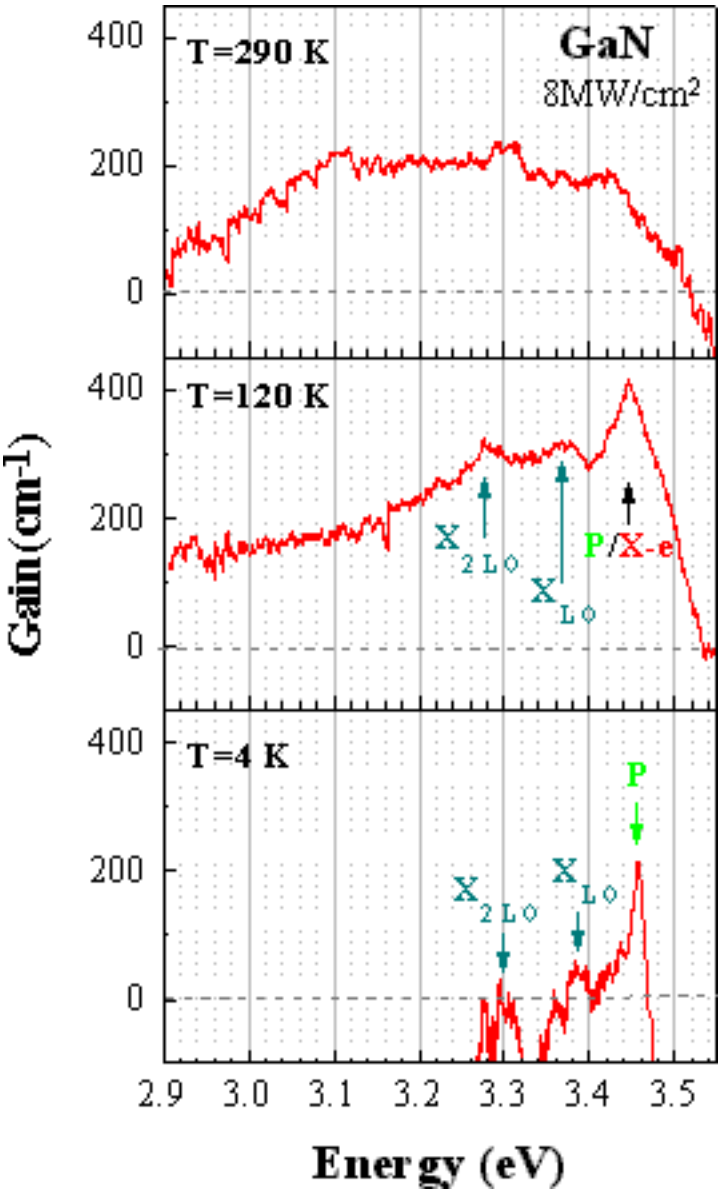

Figure 4. Gain spectra of GaN at three different temperatures.

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