

Mechanisms of optical gain in cubic gallium nitride

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We report on the mechanisms of optical gain in cubic GaN. Intensity-dependent gain spectra allow a distinction of the processes involved in providing optical amplification. For moderate excitation levels, the biexciton decay is responsible for a gain structure at 3.265 eV. With increasing excitation densities, gain is observed on the high energy side of the cubic band gap due to band filling processes. For the highest pump intensities, the electron-hole plasma is the dominant gain process. Gain values up to 210 cm^{-1} were obtained, indicating the high potential of cubic GaN for device applications. The observed gain mechanisms are similar to those of hexagonal GaN. © 1998 American Institute of Physics. [S0003-6951(98)03012-5]

Heterostructures based on wurtzite GaN (*h*-GaN) are now used for the production of prototype blue light-emitting laser diodes with dry-etched¹ or cleaved laser cavities.² The epitaxial growth of GaN with zinc blende (cubic) crystal structure (*c*-GaN) on substrates like GaAs with a common cleavage plane and readily high-quality, low-cost wafers may be considered as an alternative approach to the future realization of cleaved laser cavities. For laser applications, the optical gain is the most important parameter to be known. Although the epitaxy of *c*-GaN has been performed for many years,^{3–10} only one experiment on optical gain data has been reported so far.¹¹ This may be due to the fact that the molecular beam epitaxy (MBE) of homogeneous layers free of micron-sized crystals and hexagonal inclusions has been reported only recently.¹² Epitaxial layers of *c*-GaN on (001) GaAs were grown under carefully controlled stoichiometric growth conditions exploiting reflection high energy diffraction (RHEED) measurements of the surface reconstruction as an *in situ* control of the composition of the layer surface during growth.⁹

The properties of *c*-GaN at high excitation levels are not understood in detail. In our previous work¹³ we studied the gain spectra of thick quasibulk *h*-GaN between 2 K and room temperature and found that excitonic processes add to the gain at high temperatures. The purpose of the present letter is to analyze optical gain spectra obtained from thin *c*-GaN epilayers at high excitation levels and to study the mechanisms providing optical amplification. The results will be compared with the involved gain mechanisms of hexagonal GaN.

Cubic GaN films with a phase purity better than 99.9% were grown on semi-insulating GaAs (001) substrates by rf-plasma assisted MBE at a substrate temperature of 720 °C. The nucleation process and the layer growth were controlled using RHEED. Details of the growth procedure were reported in Ref. 9. The thickness of all the GaN layers was about 1.5 μm .

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. The sample was mounted in a bath cryostat at 1.8 K. Gain measurements were performed using the variable-stripe-length method.¹⁴ The excitation spot was focused onto a $l \times 50 \mu\text{m}^2$ stripe, where l denotes the excitation length. The photoluminescence (PL) spectra were recorded from the top of the sample with a continuous-wave (cw) helium–cadmium laser.

The typical cw PL spectrum of the *c*-GaN sample investigated is displayed in the lower part of Fig. 1 (semilogarithmic plot). A broadband at 3.144 eV dominates the spectrum and from time-resolved measurements a donor-acceptor-pair transition (DAP₁) is found to be responsible for this luminescence. A multiexponential decay is observed as expected for this transition.¹⁵ The lifetime of the DAP₁ is between 5 and 20 ns. The LO replica of the DAP band is indicated by arrows. At 2.880 eV a second donor-acceptor-pair (DAP₂) transition with its LO-replica at 2.780 eV is clearly observable. The luminescence at 3.26 eV with a full width at half maximum (FWHM) of 19 meV is a superposition of the free-exciton- and the donor-bound-exciton-recombination. The excitonic origin of the band is confirmed by time-resolved measurements where a decay time of 15 ps was observed. In Fig. 1(a) the stimulated emission of the sample at an excitation density of 5 MW/cm² is shown on a linear scale. An unstructured, broad peak at 3.250 eV dominates the spectrum and no distinction of the involved processes providing stimulated emission is possible.

The gain spectra for various excitation densities up to 10 MW/cm² are displayed in Fig. 2. The spectra presented here are smoothed to enhance the visibility of the observed spectral features. The threshold to obtain optical gain is 150 kW/cm², resulting in a small structure at 3.265 eV with a gain value of 10 cm^{-1} . From the spectral position only a biexciton decay—denoted as M in the Fig. 2—is likely to be responsible for this gain structure. Therefore, the biexciton binding energy in cubic GaN can be roughly estimated as 3 meV. In *h*-GaN a value for the biexciton binding energy of 3.7 meV was observed.¹⁶ No gain due to an exciton–exciton

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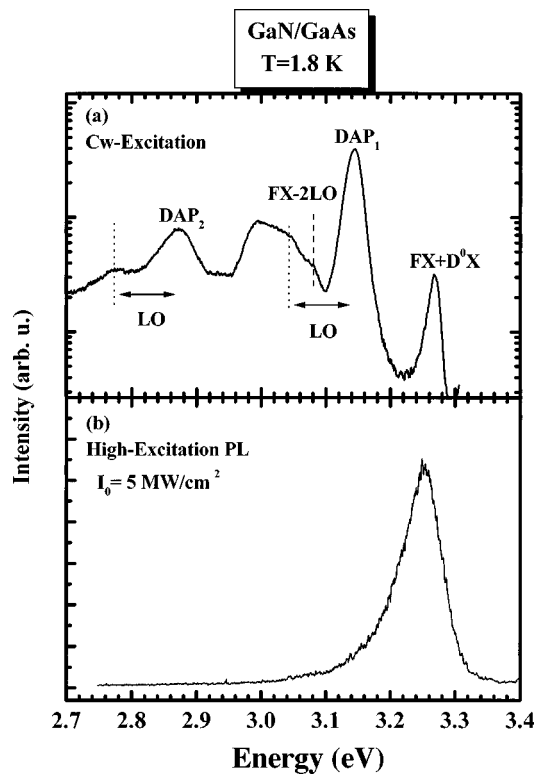


FIG. 1. (a) Photoluminescence of *c*-GaN/GaAs at cw excitation with a He-Cd laser at 1.8 K. (b) Stimulated emission of a *c*-GaN/GaAs sample at an excitation density of 5 MW/cm² at 1.8 K.

scattering—expected at around 3.24 eV—is found. For increasing excitation densities of 500 kW/cm² and more the gain peak shifts above the band gap of *c*-GaN. The peak position is 3.274 eV at 500 kW/cm² and 3.280 eV at 1.5 MW/cm². The gain above the GaN band edge originates from band filling processes, where the blueshift of the crossover gain absorption is caused by a shift of the respective quasi-Fermi levels of holes and electrons into the valence and conduction bands.¹⁷ At an energy of 3.219 eV another gain peak is observed—labeled EHP. With increasing excitation density this gain peak dominates the spectrum. Its peak position shifts to lower energies with increasing excitation density (3.210 eV at 5 MW/cm²). From the energy position and the intensity dependent behavior we attribute this gain structure to an electron-hole plasma. This process provides the highest gain values up to 210 cm⁻¹. To compare the processes responsible for optical amplification in cubic GaN with those of hexagonal GaN the gain spectra of a 3 μm thick hexagonal sample are shown in Fig. 2(b) (see details in Ref. 18). A broadening of the region of optical gain similar to that of *c*-GaN is observed with increasing pump intensity. 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/cm². For higher intensities inelastic exciton-exciton scattering—labeled P—contributes 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 characteristic shift which are identical to those of the stimulated emission.¹⁸ At 15 MW/cm² the EHP represents the dominating low-temperature gain mechanism with

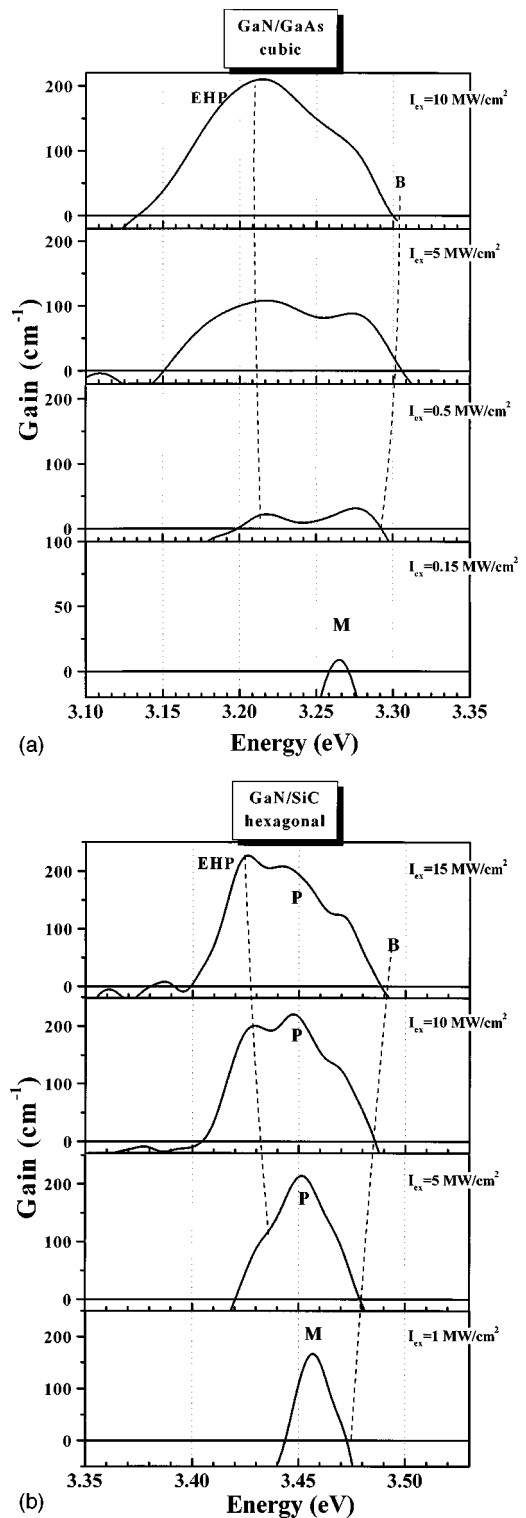


FIG. 2. (a) Gain spectra of cubic GaN for various excitation densities up to 10 MW/cm² at 1.8 K (note that the lowest gain spectrum is represented in a different Y scale to facilitate the visibility). (b) Gain spectra of hexagonal GaN for excitation densities up to 15 MW/cm² at 1.8 K (see Ref. 16) (the dotted lines indicate the energy shift of the peak position and the crossover gain absorption, respectively).

a gain value of 250 cm⁻¹ for *h*-GaN. On the high-energy side of the *M* band a gain structure—labeled B—occurs at energies near or above the band gap of the sample and the crossover from gain to absorption shifts to higher energies at increased pump intensities. Due to its spectral position and the typical blueshift with intensity we ascribe the high-energy gain to band filling processes.

By comparison of the involved gain processes a very similar behavior of the cubic and hexagonal phase is observed. For lower excitation densities excitonic processes like the biexciton decay or exciton–exciton scattering are responsible for optical gain. With increasing excitation density the band filling results in a strong blueshift of the crossover gain absorption. At highest excitation levels an electron-hole plasma dominates the gain spectra providing high gain values up to 250 cm^{-1} . As the main difference the contribution of excitonic processes like the biexciton decay or the exciton–exciton scattering to optical gain up to high excitation densities in *h*-GaN is observed while it is not detected in *c*-GaN. For an excitation density of 10 MW/cm^2 these excitonic processes provide optical gain in hexagonal GaN with gain values of 220 cm^{-1} , whereas in cubic GaN many-particle effects are the dominant mechanisms for optical amplification.

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 CdS¹⁹ and is explained by a spatial and temporal separation 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 optical properties of highly excited cubic and hexagonal GaN are in good agreement with the observed behavior of other wide-gap materials.^{19,20}

In conclusion, we reported on the mechanisms of optical gain for the cubic phase of GaN. Excitonic processes seem to play a role only at relatively low densities. With increasing pump intensity, band filling processes become more dominant and the electron-hole plasma exhibit the highest gain reaching values of 210 cm^{-1} for an excitation density of 10 MW/cm^2 . In comparison the hexagonal and cubic GaN epilayers exhibit similar gain mechanisms at comparable excitation densities.

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