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Excitonic structure of GaN epitaxial films grown by hydride-vapor-phase epitaxy

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Abstract. We report on highly resolved exciton spectra of GaN films grown by hydride vapor phase epitaxy. Using calorimetric absorption and calorimetric reflection spectroscopy the excitonic transitions originating from the A-, B-, and C-valence bands are precisely determined and the crystal-field and spin-orbit-splitting energies are calculated. We also present the first magneto-optical experiments on the neutral-donor-bound exciton. Electron as well as the hole g-values are obtained by analyzing the Zeeman splittings at 12 T.

1. Introduction

GaN is a direct-band-gap semiconductor with an energy gap of 3.43 eV at room temperature. When deposited on (0001)-sapphire it exhibits wurtzite structure and the valence band is split by the combined impact of the crystal field and spin-orbit interaction. The valence band structure is of considerable importance, e.g., for the identification of the effective-mass-like acceptors. Due to the progress in growth technology during the last years epitaxial layers of good crystalline quality and relatively low carrier concentrations are now available. For epitaxial films grown by the hydride vapor phase epitaxy (HVPE), a linewidth of the donor-bound-exciton line as narrow as 1.4 meV was reported [1]. At this point it becomes possible to employ highly resolved optical spectroscopy and Zeeman investigations to study the electronic structure in greater detail. We therefore used very sensitive photo-thermal techniques as well as polarized-luminescence and magnetooptical experiments in the range of free and bound excitons to obtain accurate information on the electronic structure of wurtzite GaN.

2. Experimental

Details about the growth procedure and sample properties are given in [1] and related references therein. For the first photo-thermal experiments on GaN epilayers we used the highly sensitive calorimetric absorption and calorimetric reflection spectroscopy (CAS/CRS) at 43 mK [2]. In a CAS experiment we measure the temperature increase of the photo-excited sample as a function of excitation energy. It is effected by the generation of phonons during nonradiative relaxation to thermal equilibrium. In a CRS experiment the reflection of the sample is detected by the absorption-induced heating of a black body. CAS and CRS spectra are recorded simultaneously. We used an excimer-laser-pumped dye laser as quasimonochromatic light source giving a spectral resolution of 50 μeV. For photoluminescence experiments the 325 nm line of a HeCd laser was employed. In Zeeman experiments the sample was placed in the center of a superconducting solenoid-type magnet (maximum field 12 T) at a temperature of 5 K and both, excitation and emission were guided to and from the sample by an optical fiber. The orientation of the magnetic field was varied between parallel and perpendicular to the c-axis of the crystal.

3. Experimental results and discussion

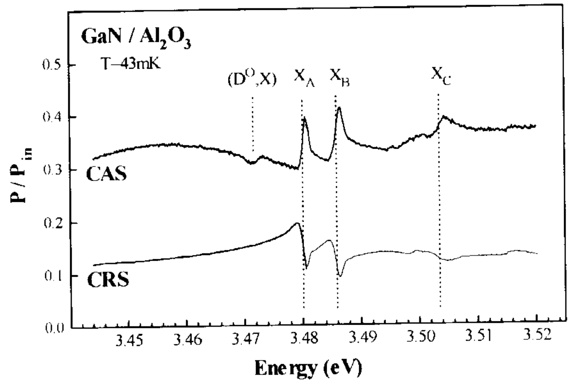
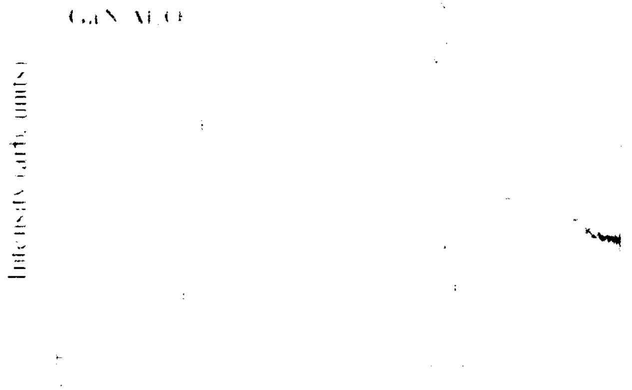


Figure 1. Calorimetric absorption and calorimetric reflection spectrum of excitons in epitaxial GaN.

Fig. 1 gives highly resolved calorimetric reflection and calorimetric absorption spectra of a n-type 400μm GaN/Al$_2$O$_3$ epilayer. In the CRS spectrum three structures are clearly resolved and attributed to the excitons X_A at 3.4800 eV, X_B at 3.4860 eV and X_C at 3.5025 eV, involving holes from the A, B, and C valence-bands, respectively. A minor structure is detected at 3.4995 eV and ascribed to the $A_{n=2}$-exciton in accordance with the position expected given the exciton binding energy of 26 meV. In the CAS spectrum the same structures are found since a reduction of the reflectivity implies an increase of light absorption in the crystal which in turn triggers non-radiative relaxation processes.


GaN/Al2O3
Intensity (arb. units)

$\theta = 90°$, i.e., the magnetic field perpendicular to the c-axis, the contribution of the hole vanishes and only the donor Zeeman-splitting remains (see below). If the magnetic field is parallel to the c-axis the hole and electron Zeeman contributions add (see Fig. 3).

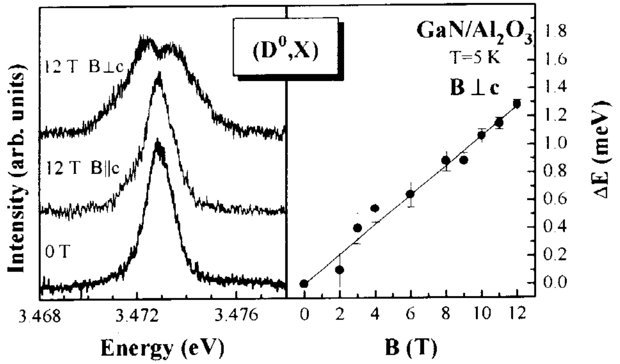


Figure 3. Spectra of I_2 at 0 and 12 T for different magnetic-field orientations (l.h.s.). Splitting in dependence on magnetic field for $B \perp c$ (r.h.s).

This should result in well resolved splittings at high magnetic fields $B \geq 10$ T. For B perpendicular to the c-axis only the electron Zeeman-contribution remains. The experimental results of the Zeeman investigation are summarized in Fig. 3. For B parallel to the c-axis, we observe neither a splitting nor a broadening. The same result was found in CdS [5]. It was explained by an accidental cancellation of the contributions from the electron and hole Zeeman-interactions - the hole g-value equals the electron g-value. It also holds for GaN. For B perpendicular to the c-axis, one should observe a broadening of the donor-bound-exciton line at fields above 6 T. It increases from 1.6 meV to 2.8 meV at 12 T and shows partially resolved splitting which amounts to 1.3 ± 0.1 meV at 12 T. The resulting g-value is 1.9 ± 0.1, very close to the free-electron g-value of 1.95 in GaN. In III-V and II-VI semiconductors the g-values of the hole range between 0.7 and 0.8[6]. For a $J = 3/2 \pm 3/2$ state the energy separation is given by $\Delta E = 3 g_h \mu_B B$. If it equals the Zeeman splitting of the neutral donor one obtains $3g_h = g_e$. With $g_e = 1.95$ one calculates $g_h = 0.65 \pm 0.1$, thus quite consistent with values from other compound semiconductors.

References

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