RADIATIVE AND NONRADIATIVE RELAXATION MECHANISMS OF GROUP-III NITRIDES

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To improve the optical material properties of group-III nitrides investigations of the nonradiative processes which reduce the lifetime of corresponding applications in dependence on different material parameters for GaN and AlGaN were performed. The results show a strong correlation of the quantum efficiency with defects present in the epilayers. Additionally, with the calorimetric absorption spectroscopy we determined from absorption measurements a bowing of the band gap of AlGaN with increasing Al-content.

1 Introduction

GaN and its correlated ternary systems with Al and In are the most important materials for optoelectronical applications (e.g., LEDs, lasers) in the blue and blue-green spectral range. The optical quality of these materials is influenced by the nonradiative relaxations involved which are detrimental to the radiative ones and limit the lifetimes of such applications. Optical spectroscopy as for instance photoluminescence and reflection measurements only yield information about the existence of nonradiative processes but a qualitative or a quantitative determination of these processes cannot be realised. As a result we are using the calorimetric absorption spectroscopy [1] to obtain detailed information about the nonradiative and radiative relaxation mechanisms. For the first time the absorption resonance of AlGaN epilayers as a function of the Al-content is shown. Furthermore, a systematic investigation concerning the dependence of quantum efficiencies of GaN and AlGaN on different material parameters is presented.

2 Experimental Setup

The investigated samples are a series of GaN and one of AlGaN epilayers grown by MOCVD on sapphire. The buffer layer of GaN is either AlN or GaN with a thickness of 300 to 500 Å. The AlGaN epilayers were grown on 30 nm AlN and 2 µm GaN. The thicknesses of the GaN samples varied in the range of 1 to 2 µm whereas the thickness of the AlGaN epilayers differs from 230 to 740 nm.

The experimental method used to determine information about the nonradiative recombination and relaxation mechanisms is the calorimetric absorption spectroscopy (CAS) at mK temperatures. With this setup the nonradiative relaxation, i.e. the heating of the sample (CAS), its transmission (CTS), and its reflection (CRS) is detected simultaneously under monochromatic excitation of the crystal by thermal detectors (calorimeters). To calculate from these data the whole absorbed light (ABS) which contains as well the radiative as the nonradiative relaxation the power of the exciting light (P_{IN}) is also measured. As a result we determine from the calorimetric data the quantum efficiency (qe) according to

$$qe=1-P_{CAS}/P_{ABS}$$

All calorimetric data are calibrated in units of power by the CAS setup with a sensitivity of a few hundred pW. Additionally, the CAS allows to measure absorptions of actually not transparent thick crystals but also of very thin samples where the transmission spectra does not exhibit a resonance[2].

3 Experimental Results and Discussion

3.1 GaN

Typical data of a CAS measurement of a GaN epilayer is depicted in the lower part of Fig. 1. The resonances of the free A-exciton (X_A) and the free B-exciton (X_B) can be observed in the CAS and the CRS spectrum. In the lower energy range of the CRS spectrum Fabry-Perot interferences occur. The difference between the CAS and the ABS spectrum corresponds to the amount of radiative relaxations. In the upper part of Fig. 1 the result of the calculation of the quantum efficiency (qe) is shown. It is noticeable that the qe is below 20 % in the whole regarded spectral range what seems to be unexpected considering the Rashba-theory[3], especially for bound excitons. However, the determination of the radiative relaxation with the CAS takes only the luminescence light into account which can leave the crystal but not the re-absorbed light which leads to a heating of the sample. Therefore, the presented qe is either equal or lower than the qe of the isolated resonance. Nevertheless, the calculated qe reflects the most important value for the optical application. In Fig. 2 the qe of the free A- and the free B-exciton as a function of different parameters are shown. The qe increases with rising buffer layer and sample thicknesses but decreasing residual oxygen content in the sample. Additionally, the material of the buffer layer influences the qe. From these results a homoepitaxial growth of the GaN epilayers should be preferred. The data indicate a reduction of the excitonic qe mainly due to impurities present in the samples[4].

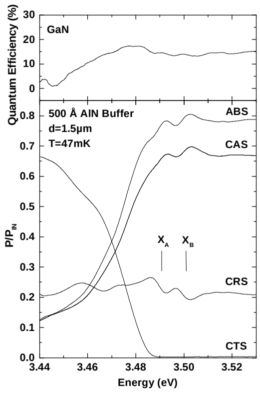

Figure 1. CAS/CTS/CRS/ABS-spectrum of a GaN epilayer; the calculated quantum efficiency is shown in the upper part.

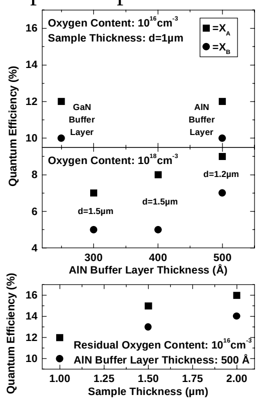

Figure 2. Dependence of the qe of the X_A and X_B resonances in GaN on the buffer layer thickness for different oxygen contents (↑$10^{16}cm^{-3}$ ↓$10^{18}cm^{-3}$) and on the sample thickness for an oxygen content of $10^{16}cm^{-3}$.

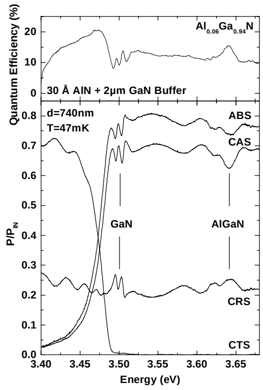

Figure 3. CAS/CTS/CRS/ABS-spectrum of a AlGaN epilayer (6 % Al); the calculated quantum efficiency is shown in the upper part.

3.2 AlGaN

The typical results of a CAS measurement of an AlGaN epilayer are presented in Fig. 3. The resonance of the free A-exciton of AlGaN in absorption is observed at 3.641 eV. Additionally, the X_A- and the X_B-resonance of the GaN buffer layer is detected. The qe shown in the upper part exhibits a peak at the AlGaN energy but is even lower than 20 % as in the whole excitonic region.

From the energy positions of the free excitons in GaN (Fig. 4 lower part) we estimate that the compressive strain of the buffer layer of the investigated samples is approximately the same ($\varepsilon_{zz} \approx 0.0012 - 0.0015$). Therefore, the resonance positions of the AlGaN epilayers should only be influenced by the Al-content (Fig. 4 upper part). Our investigations show a bowing of the AlGaN bandgap with increasing Al-content up to 22 % Al.

The qe of the free A-excitons of GaN and AlGaN are depicted in Fig. 5. With increasing Al-content we observe an enhancement of the qe of the X_A resonance in AlGaN. Nonetheless, the qe of the GaN buffer layer seems to have a strong impact on this of the AlGaN epilayer. Because of the different material parameters of the AlGaN crystals (e.g., epilayer thickness, buffer layer quality) but also due to their

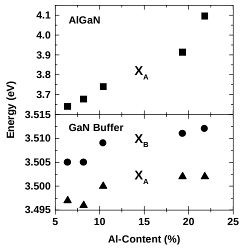

Figure 4. Dependence of the absorption energies of the X_A-resonance in AlGaN on the Al-content (upper part); in the lower part the X_A- and the X_B-resonance energies of the GaN buffer layer are shown.

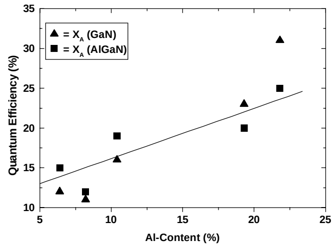

Figure 5. Quantum efficiency of the free A-exciton in the AlGaN epilayer and the GaN buffer layer as a function of the Al-content. The qe of X_A-resonance in the AlGaN epilayer increases with rising Al-content.

limited Al-content (max. 22 %) further investigations have to be performed to obtain more information about the dependence of the qe in AlGaN.

4 Summary

We presented for the first time absorption measurements of AlGaN from which we obtained a bowing of the bandgap for Al-contents up to 22 %. Furthermore, a systematic investigation of the dependence of the qe in GaN on different material parameters showed the importance of defect free growth of the samples to obtain higher qe. In AlGaN an increase of the qe with rising Al-content was found.

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