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Excitonic Quantum Efficiency of GaN

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Abstract

We report on investigations of the excitonic quantum efficiency in GaN in dependence of different buffer layer thicknesses and residual oxygen content in the crystal. The quantum efficiency of the free excitons rises with increasing buffer layer thickness and decreasing residual oxygen content. The influence of oxygen on the quantum efficiency is stronger than that of the buffer layer thickness. In general, the observed quantum efficiencies are below 20% indicating the strong impact of nonradiative relaxation and recombination processes in the excitonic range.

Introduction

Excitonic processes are likely to be important for the gain mechanisms in GaN based devices [1,2]. To date however, a high number of defects and impurities present even in high-quality material limits the lifetime of lasers due to the creation of nonradiative relaxation and recombination channels. The quantum efficiency determined is a measure for the role of nonradiative processes caused by these imperfections in calorimetric absorption spectroscopy [3]. We present a systematic study of the quantum efficiency in GaN samples with different buffer layer thicknesses and different amounts of residual oxygen present in the epilayer.

Experimental Setup

The investigated GaN epilayers were grown by metal organic chemical vapor deposition (MOCVD) on sapphire with an AlN buffer layer. The thickness of the buffer layers was varied in the range of 300 to 500 Å. Additionally, samples with different amounts of residual oxygen were investigated. Three samples had a residual oxygen content in the range of 10^{18} cm^{-3} and the fourth in the range of 10^{16} cm^{-3}. Details were given in refs. [4,5].
To obtain information about the radiative relaxation in GaN epilayers time-integrated and time-resolved photoluminescence (PL) measurements were performed. For the investigation of the nonradiative recombination the calorimetric absorption spectroscopy (CAS) at mK temperatures was used. With this experimental setup using thermal detectors (calorimeters) it is possible to measure simultaneously the nonradiative relaxation (CAS spectrum), the transmission (CTS spectrum), and the reflection (CRS spectrum) of a crystal. Additionally, we measured the power of the exciting light so it was possible to calculate the whole absorbed light (ABS spectrum). Beside the heating of the crystal (CAS spectrum) the ABS spectrum also includes the part which relaxes radiatively.

Taking into account all calorimetric data we determine the quantum efficiency (qe) according to

$$qe = 1 - \frac{P_{CAS}}{P_{ABS}}. \qquad \text{Eq. 1}$$

The heating of the calorimeters is measured in units of power with a sensitivity of a few hundred fW.

The CAS gives us the possibility to measure the absorption of a thick sample where a transmission signal cannot be detected but also the absorption of very thin crystals where resonances in the transmission spectrum do not occur [6]. For a detailed description of the CAS-method see [3].

Experimental Results and Discussion

A typical photoluminescence (PL) spectrum of a GaN epilayer grown by MOCVD is shown in Fig. 1. Similar to GaN bulk crystals it is dominated by the (D^0,X) complex at 3.480 eV but also the X_A and the X_B resonances at 3.486 eV and 3.493 eV, respectively, can be observed in the spectrum. In comparison to the excitonic spectrum of bulk GaN the X_A peak is blue shifted by 6 meV which indicates compressive strain. The structure of the PL spectrum is typical for the excitonic luminescence of a GaN epilayer. The FWHM of the (D^0,X) complex in Fig. 1 is 4 meV. In the lower energy range of the PL spectrum appears a peak at 3.267 eV which can be assigned to the donor-acceptor-pair (DAP) luminescence. The yellow luminescence at 2.34 eV is very weak.

To obtain more information about the properties of the radiative excitonic processes we investigated the samples with time resolved PL spectroscopy. The measurements yield lifetimes of the free excitons below 20 ps and of the bound excitons in the range of 50 ps. These short lifetimes indicate the strong influence of nonradiative relaxation channels for the excitonic recombination caused by defects. To investigate these nonradiative processes we performed CAS measurements in the excitonic region. In Fig. 2 the results of the CAS investigations are depicted. In the lower part the CAS, the CTS, and the CRS spectra can be seen. From the CAS and the CRS spectra the resonances of the free A- and B-exciton can be assigned. Additionally, Fabry-Perot interferences occur in the lower energy range in the CRS signal. The fourth spectrum (ABS) reflects the absorption of the crystal. In the upper part of Fig. 2 the quantum efficiency calculated from Eq. 1 is shown which is below 10 % in the excitonic region.

Important for the value of the quantum efficiency is the incorporated oxygen which cannot be measured with usual spectroscopic techniques. The PL spectrum of a sample with more residual oxygen (10^{18} cm^{-3}) does not differ from that with less residual oxygen (10^{16} cm^{-3}). But the CAS allows us to investigate the influence of the residual oxygen on the relaxation processes in the sample. Fig 3. gives CAS data for a sample with less oxygen content. The main difference between these

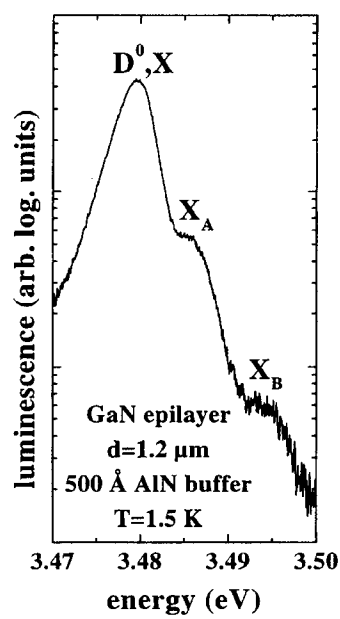

Fig. 1: PL spectrum of a GaN epilayer.

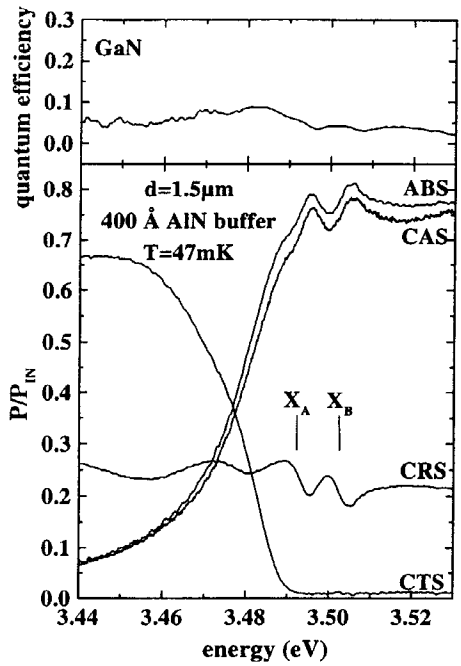

Fig. 2: CAS, CTS, CRS, and ABS spectrum of a GaN epilayer; the quantum efficiency is shown in the upper part.

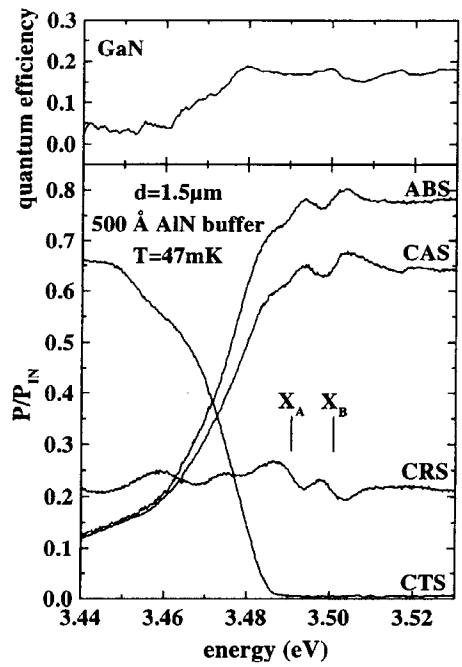

Fig. 3: CAS, CTS, CRS, and ABS spectrum of a GaN epilayer with less residual oxygen; the quantum efficiency is shown in the upper part.

two samples is the lower residual oxygen content of the crystal presented in Fig. 3. In the energy range of the free excitons and at higher energies the absorbed light spectrum (ABS) is much higher than the heating of the sample (CAS spectrum). This means that more luminescence light can leave the crystal and so the quantum efficiency increases. The quantum efficiency in the excitonic range for this crystal is about 18 %.

Two possible explanations can be given for this result. On the one hand the lower residual oxygen content could be the reason for this rising quantum efficiency but also the buffer layer thickness could influence the radiative relaxation. To obtain detailed information we measured crystals with different buffer layer thicknesses, but same residual oxygen content (10^{18} cm^{-3}). The values for the quantum efficiency of the X_A and the X_B

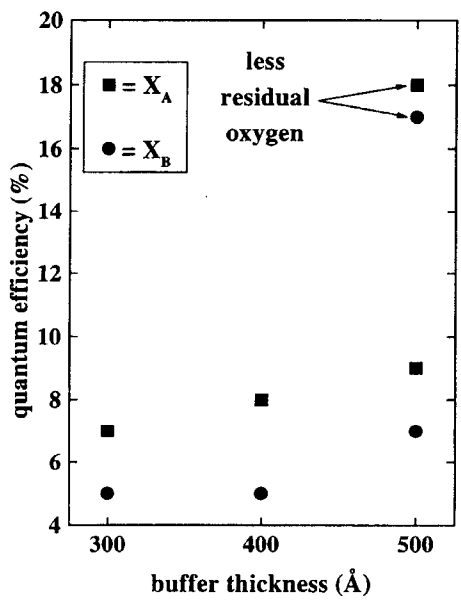

Fig. 4: Quantum efficiency of the GaN epilayers as a function of the buffer layer thickness for different oxygen contents.

resonance for these samples in dependence of the buffer layer thickness are shown in Fig. 4. It can be seen that the quantum efficiency increases with rising buffer layer thickness. This is due to the reduction of interface defects through the buffer layer [7]. Increasing the buffer layer thickness by 100 Å however, the quantum efficiency rises by one percent only. That indicates a small influence of the buffer layer thickness on the quantum efficiency. The quantum efficiency values for the sample with lower residual oxygen content (10^{16} cm^{-3}) are also presented in Fig. 4. They are 18 % for the X_A and 17 % for the X_B exciton indicating a stronger impact of the residual oxygen content on the quantum efficiency compared to the buffer layer thickness.
It is interesting to note that the observed quantum efficiencies are below 20% for all samples studied. A high density of structural defects and impurities is responsible for these low values. Especially for bound excitons higher quantum efficiencies are expected due to the breakdown of the k-selection rule [8].

In conclusion, we showed that an increasing buffer layer thickness enhances the quantum efficiency of the radiative excitonic decay. This is caused by a reduction of structural defects in samples with a higher buffer layer thickness. Samples with a lower oxygen concentration (10^{16} cm^{-3}) also exhibit a significant increase of the quantum efficiency. We demonstrated that the calorimetric absorption spectroscopy is a unique and extremely sensitive method (some hundred fW) to investigate the influence of defects and impurities on nonradiative processes and to determine quantum efficiencies in GaN.

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