ELSEVIER

Diamond and Related Materials 00 (2001) 00–00

DIAMOND AND RELATED MATERIALS

www.elsevier.com/locate/diamond

Subbandgap absorption from photocurrent spectra in $Al_{0.18}GaN/GaN$ heterostructures

M. Niehus[a,*], R. Schwarz[a], S. Koynov[a], M. Heuken[b], D. Meister[c], B.K. Meyer[c]

[a] *Instituto Superior Técnico, Departamento de Física, Lisboa, Portugal*
[b] *Aixtron GmbH, Aachen, Germany*
[c] *I. Physikalisches Institut, Universität Giessen, Giessen, Germany*

Received 6 September 2000; accepted 8 December 2000

Abstract

We have measured optical absorption in $Al_{0.18}GaN/GaN$ heterostructures, prepared by MOCVD on sapphire substrates, by the constant photocurrent method (CPM) between 300 and 500 K. The constant photocurrent mode is appropriate since the response time decreases when the incident light energy crosses from below to above the bandgap energy. The films were further characterized by temperature-dependent dark conductivity and Hall measurements. We have found exponential band tails exceeding thermal broadening in the absorption spectra below the bandgap energy indicating the presence of disorder and/or deep defects. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Gallium nitride (GaN); UV photocurrent spectroscopy; Urbach tails; Temperature dependence

1. Introduction

AlGaN alloys with bandgaps between 3.4 and 6.2 eV are currently explored for application in laser systems as well as for UV detection and high temperature electronics. However, few studies on the photocurrent features in the temperature range up to 500 K are available. Assuming the presence of deep defects in GaN films one should expect a broad subbandgap shoulder in their optical absorption spectra. In addition, disorder will result in spectral broadening, which is usually described by an Urbach tail parameter E_u that exhibits a particular temperature dependence as predicted by an Einstein oscillator model.

In a previous work, the secondary photocurrent transient after pulses of a Nd:YAG laser system showed a slow decay reaching the millisecond time region after both green and UV laser excitation [1]. The exponents in the power law decay range from −0.1 to −0.3. As an explanation for such long times the presence of deep centers was invoked [2].

In this contribution we report about spectroscopic photoconductivity studies near the bandgap in a double $Al_{0.18}GaN/GaN$ heterostructure. The temperature range lies between 300 and 500 K. Since the response time is varying with energy we have used the constant photocurrent method (CPM) where the photocurrent spectrum is taken at an appropriately chosen constant photocurrent level [3]. We found the usual bandgap shift from approximately 3.43 to 3.36 eV when increasing the temperature from 300 to 500 K. The values for the tail slope E_u range from approximately 30 to 50 meV for energies near the bandgap. This first tail is followed by a flat region with characteristic energies of

* Corresponding author. Tel.: +351-21-841-7775; fax: +351-21-841-9118.
E-mail address: manfredo@fisica.ist.utl.pt (M. Niehus).

PII: S0925-9635(01)00369-7

approximately 350 meV. We discuss the different spectral regimes and especially the exponential (Urbach) tails of the GaN layer.

2. Experimental and analytical procedures

The sample, an $Al_{0.18}GaN/GaN$ double heterostructure, was prepared by MOCVD on a sapphire substrate with thicknesses of 0.2 and 1.6 μm for the AlGaN and GaN layer, respectively. The films were characterized by Hall measurements and photoluminescence. The n-type carrier densities increase from 3×10^{16} cm^{-3} at $T = 300$ K to 1×10^{17} cm^{-3} at $T = 500$ K, accompanied by a decrease in mobility from 50 to 20 cm^2/Vs, respectively [1]. Hence, the dark conductivity shows a *weak* temperature dependence, rising *linearly* from 300 to 500 K [2]. The bandgaps obtained from photoluminescence spectroscopy at room temperature are 3.4 and 3.8 eV for the GaN and the $Al_{0.18}GaN$ layer, respectively [1].

For photoconductivity measurements light from a 75-W Xe lamp was dispersed with a 25-cm monochromator with a 3000 grooves/mm holographic grating and chopped at 7–500 Hertz. The GaN band tail was analysed by means of the temperature dependence of the photocurrent between $T = 300$ and 500 K. The light was focused to a spot size of approximately 2.5 mm diameter into the center of two soldered indium contacts with a spacing d of approximately 5 mm. In this geometry, no primary photocurrent was present, as shown by ohmic behaviour for applied bias voltage U_{Bias} up to ± 20 V, for the whole spectral range. The photocurrent was read with lock-in technique as the voltage drop across a shunt resistance. The acquisition speed was held as low as 0.5 nm/s, to minimise long time decay effects induced by persistent photoconductivity (PPC).

We compared photocurrent spectra (PC) measured for constant illumination power with the one obtained by CPM [3]. Usually, the photocurrent density I_{ph} is proportional to the absorption coefficient α and the light flux Φ. In the low absorption region, the photocurrent can be expressed as (q and R are the electron charge and reflection coefficient, respectively):

$$I_{ph} = \alpha q \Phi \eta (\mu\tau)_n (1 - R) E \quad (1)$$

Here we assume that the mobility of electrons will largely exceed that of holes: $(\mu\tau)_n \gg (\mu\tau)_h$. This is justified by the mobility measurements mentioned above. The applied electric field E is given by $E = U_{Bias}/d$. The quantum efficiency η is assumed to be unity. The total change of the refractive index, as determined from reflectivity measurements, is of the order of 30%. The ampere–lux characteristics will be discussed by means of the Rose coefficient β: $I_{ph} \propto \Phi^{\beta}$.

We have calibrated the CPM spectra with an absolute measurement of the absorption coefficient obtained from optical transmission at light energy where αd is near to unity.

3. Results and discussion

3.1. Spectral photocurrent between $T = 300$–500 K

Fig. 1 shows photocurrent spectra in the GaN bandtail region, for different temperatures between 300 and 500 K. The maximum photocurrent is found for absorption inside the GaN layer, approximately one order of magnitude higher than for absorption inside the $Al_{0.18}GaN$ layer. The PC spectra indicate low optical absorption below the bandgap and show saturation due to high absorption above the bandgap energy. We have determined the GaN bandgap E_g and its temperature dependence from the onset of saturation in the photocurrent spectra. The GaN bandgap decreases from 3.44 eV at 300 K to 3.36 eV at 500 K. The thermal coefficient is -0.42 meV/K. These results are in good agreement with values taken from literature.

The GaN bandtail is composed of two different exponential decay regimes: at ambient temperature, we find a steep region (T1) reaching from the bandgap energy E_g down to approximately 3.3 eV into the bandgap. A second, less steep region (T2) extends to approximately 2.9 eV. Below this energy, no significant change in the slope can be observed. We characterize the shape of the absorption spectra by the characteristic tail energy E_u, according to $I_{ph} \propto \exp(E/E_u)$. This Urbach energy is conventionally used to describe the band tails of absorption coefficients in crystalline and amorphous semiconductors [5,6]. We found the values given in Table 1.

The decrease of the band tail energies with increas-

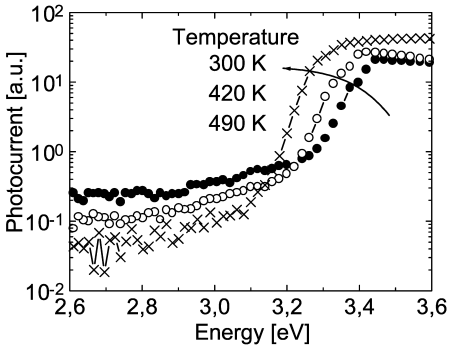

Fig. 1. Photocurrent spectra (PC) in the GaN band tail region between $T = 300$–500 K. High absorption above the bandgap energy leads to saturation.

Table 1
Measured Urbach tail energies and their thermal coefficients

	300 K [meV]	500 K [meV]	$\delta E_u/\delta T$ [meV/K]
E_u (T1)	48	30	-0.10 ± 0.01
E_u (T2)	350	260	-0.50 ± 0.10

ing temperature was essentially linear. For higher temperatures, the tail energies E_u started to increase again. Some uncertainty is due to the fact that for the slope values the photocurrent data rather than the CPM spectra were used. The tendencies as a function of temperature should, however, be the same.

3.2. Ampere–lux characteristics, frequency dependence, and decay time analysis

Fig. 2 shows the ampere–lux characteristics of the sample. We find a Rose coefficient $\beta = 1$ for low illumination power (as indicated by the dashed lines), and a transition to a sublinear behaviour for increasing power. β lies between 0.5 and 1. The sublinear regime indicates that the lifetime decreases with higher illumination power or higher photocurrent.

The frequency dependence of the photocurrent is shown in Fig. 3. From the lowest frequencies (7 Hz) the signal decreases, for both subgap and above-bandgap illumination. This is usually attributed to the PPC effect. Up to the highest observed frequencies (500 Hz), the decrease is slower than $I_{ph} \propto f^{-1}$, as expected from simple lifetime consideration.

We measured the decay times around the GaN bandgap for different temperatures. Values are taken from the oscilloscope when the decay of the voltage drop across a 1-kΩ shunt resistance reaches $1/e$ of its maximum. Due to the PPC effect [4], even for the lowest experimentally accessible frequency of 7 Hz, the photocurrent did not reach a steady-state upper or lower level upon photomodulation. This makes an accurate measurement of a response time rather difficult.

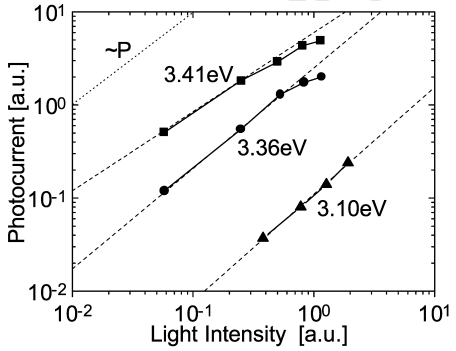

Fig. 2. Ampere–lux characteristics, showing the transition to a sublinear regime with a Rose coefficient β between 0.5 and 1. The dashed lines are linear fits to the low intensity range.

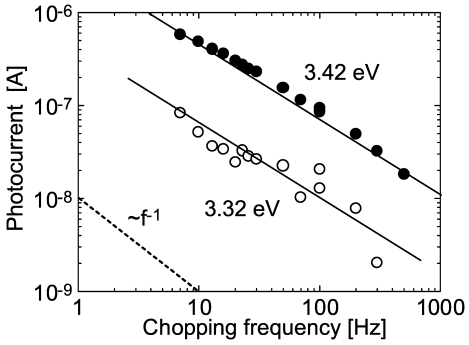

Fig. 3. Photocurrent as function of the chopping frequency between 7 and 500 Hz. The dark current level was approximately 1 mA. The decrease in photocurrent is usually attributed to the PPC effect.

However, it is certain that the decay for above-gap illumination is significantly faster than at low energy (see Fig. 4). The same is true for higher temperatures, at least above 400 K: in this regime, enhanced non-radiative recombination is expected to dominate.

3.3. CPM analysis of photocurrent

Fig. 5 compares the photocurrent as measured by conventional photocurrent spectroscopy (PC) with that measured by CPM. Both spectra are normalized with optical transmission measurements. We use the term apparent optical absorption coefficient to indicate that all changes of parameters in Eq. (1) will deform the spectral shape. The CPM values for the high (low) absorption region are higher (lower) than those of the conventional PC technique, as expected from the decay time measurements in Fig. 4. The qualitative features of the individual spectral regions, however, are unchanged.

3.4. Discussion

We will first address the question of the validity of the CPM approach. From Fig. 4 we found a decrease in

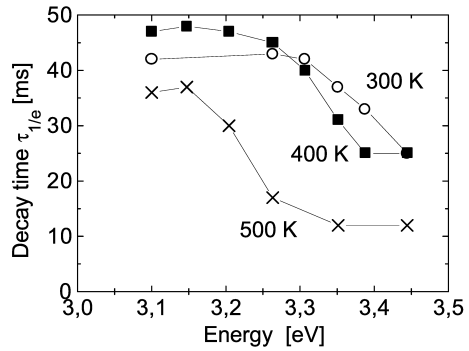

Fig. 4. Spectral photocurrent decay time around the GaN bandgap, between $T = 300$–500 K. The high decay times are experimentally limited by the chopping frequency. Above gap illumination has a clearly faster decay. Between 400 and 500 K the decay times are also reduced, presumably due to enhanced non-radiative recombination.

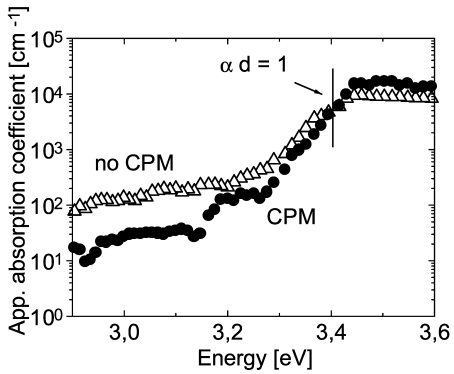

Fig. 5. The apparent absorption coefficient as measured by CPM (solid circles) and conventional photocurrent spectroscopy (open triangles). The CPM spectrum has been calibrated with optical transmission for $\alpha d = 1$ (as indicated).

response time of the photocurrent with increasing light energy. The measured decay time is not directly the recombination life time of photocarriers, if a high density of trap states or a broad distribution of such states is present. Then the occupation of those traps and thermal emission rates have to be taken into account and can only talk of an effective life time. As mentioned above, we assume that the main contribution of the photocurrent will come from electrons due to their higher mobility with respect to holes.

For the CPM approach to be valid, all parameters in Eq. (1) need to be independent of light energy. We have assured a constant mobility-lifetime product of electrons by choosing the CPM mode which keeps the quasi-Fermi levels and trap state occupation constant. In addition, we assume that carrier mobility and photoelectric conversion efficiency do not change with energy. Under these assumptions the CPM spectra will reflect all possible transitions of electrons from the valence band and the midgap region into the conduction band of GaN.

The optical absorption measured in regions T1 and T2 show an exponential decay into the bandgap. We can attribute this signal to some structural disorder present in the AlGaN/GaN heterostructure bulk material. We can also speculate that a highly conductive channel at the heterostructure interface might give a strong contribution due to strong band bending. Here the defect density might be particularly large. The low energy portion of the CPM spectra most probably stems from deep defects. In previous work [2], we found a broad defect distribution some 400 meV below the bandgap by means of transient photoconductivity (TPC).

The temperature dependence of the band tail slopes is in contrast to reports by other groups, which found a perfect Urbach–Martiennsen tail behaviour [5] from optical absorption measurements at low temperatures, i.e. the exponential tail *broadens* with *higher* temperature, and linear fits converge in *one* point on a semilogarithmic plot of the absorption coefficient as a function of the excitation energy, as predicted by theoretical arguments [6]. As a cause, exciton–phonon coupling has been evoked. In fact, photoluminescence measurements revealed the presence of excitons in GaN well above room temperature. In contrast to broadening at high temperatures, our data points to *less steep* tails for increasing temperatures.

4. Conclusion

We have demonstrated that the optical absorption spectra in GaN below the bandgap can best be measured by photocurrent spectroscopy in the constant photocurrent mode, CPM, since the lifetime of photogenerated carriers is substantially higher for the low absorption regime. For above-bandgap illumination the lifetime decreases, and, in addition, the Rose coefficient is smaller than 1. We measured the bandgap temperature coefficient by photocurrent spectroscopy in accordance with published values. We found, however, a negative linear temperature coefficient of the characteristic energy for the band tail slope deduced from the photocurrent spectra.

Acknowledgements

The work at IST is supported by the Foundation of Science and Technology FTC through project PRAXIS/P/FIS/10178/1998 and by the European Union through the COPERNICUS project IC15-CT98-0819. M.N. acknowledges travel support from the FCT. R.S. acknowledges financial support from the German Academic Exchange Service DAAD.

References

[1] D. Meister, M. Top, I. Dirnstorfer, B.K. Meyer, R. Schwarz, M. Heuken, Phys. Stat. Sol. (b) 216 (1999) 749.
[2] M. Niehus, R. Schwarz, S. Koynov, M. Heuken, B.K. Meyer, C. Main, and S. Reynolds, presented at the European Mat. Res. Soc. Conf., Strasbourg, May 2000; to be published in Sem. Sci. Technol. (2000).
[3] M. Vanecek, J. Kocka, J. Stucjlik, Z. Kozisek, O. Stika, A. Triska, Solar Energy Mater. 8 (1983) 411.
[4] C. Johnson, J.Y. Lin, H. Xiang, M. Asif-Khan, C.J. Sun, Appl. Phys. Lett. 68 (1996) 1808.
[5] S. Chichibu, T. Mizutani, T. Shioda et al., Appl. Phys. Lett. 70 (1997) 3340.
[6] G. Cody, T. Tiedje, B. Abeles, B. Brooks, Y. Goldstein, Phys. Rev. Lett. 47 (1981) 1480.