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Diffusion lengths in GaN obtained from steady state photocarrier gratings (SSPG)

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We present diffusion length measurements in gallium nitride (GaN) and $Al_XGa_{1-X}N$/GaN multilayers with aluminium contents up to X_{max} = 37 at.%. The opto-electrical method employed is the steady-state photocarrier grating (SSPG), which uses two interfering laser beams to induce a periodic resistivity modulation normal to the sense current lines. We experimentally refined the SSPG, using a double beamsplitter technique and introducing a variable ND filter. We measured diffusion lengths of single GaN and $Al_XGa_{1-X}N$/GaN double layers, and analysed the obtained diffusion lengths as function of the aluminium concentration, comparing them with the respective drift lengths. Finally, we critically discuss the common interpretation in terms of ambipolar transport. We suggest a new model in which minority carriers are rapidly trapped in acceptor-like valence band tails and participate in the grating blurring passively, as spatially localized recombination centres.

1 Introduction

The steady-state photocarrier grating (SSPG) technique was introduced by Ritter et al. [1] to measure diffusion lengths on the micrometer scale, and was first applied to gallium nitride (GaN) thin films by Duboz et al. [2]. They found ambipolar diffusion lengths around a tenth of a micron, which is very similar to diffusion lengths previously deduced from electron beam induced current measurements [3]. In both [2] and [3], grain boundary recombination was evoked, as the typical columns widths were of the same order of magnitude.

In this work, we experimentally refined the SSPG, using the double beamsplitter technique and introducing a variable ND filter. We measured single GaN and $Al_XGa_{1-X}N$/GaN double layers, analysed the obtained diffusion lengths as function of the aluminium concentration, and compare them with the respective drift lengths. Finally, we critically discuss the interpretation.

2 Experimental details

The diffusion length L is obtained by measuring the experimental contrast of coherent and non-coherent photoresponse, defined by the experimental parameter β,

$$\beta = \frac{(I_{SIGNAL} \oplus I_{BIAS}) - I_{BIAS}}{(I_{SIGNAL} + I_{BIAS}) - I_{BIAS}} \tag{1}$$

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where I_{BIAS} is a strong (bias) beam induced photocurrent, and I_{SIGNAL} a (signal) beam induced photocurrent in the small signal approximation. Plus (+) in the current notation means addition under non-coherent condition, while circle plus ($\oplus$) means addition under coherent conditions, i.e. when the intersecting beams interfere constructively, building up an intensity grating. The distinction corresponds experimentally to the rotation of the polarization plane of the small signal beam. The parameter β is measured for different grating periods Λ, through the variation of the beam intersection angle Θ.
Under proper experimental conditions, the ambipolar diffusion length L can be obtained by fitting the experimental data points $\beta(\Lambda)$ to the simple analytic expression [1]

$$\beta = 1 - 2\gamma\gamma_0^2 \left[\frac{1}{1 + \left(2\pi L / \Lambda\right)^2} \right]^2 . \tag{2}$$

Here γ is the "Rose coefficient", i.e. the power law exponent in the photocurrent amplitude vs. light intensity dependence ($0.5 \le \gamma \le 1$), and γ_0 the grating quality factor ($0 \le \gamma_0 \le 1$).

A scheme of the experimental setup is shown in Fig. 1. We used a multimode HeCd laser operating at 325 nm with a maximum output power of about 10 mW. Allowing only the inner 1 mm diameter, we have a lateral uniformity to about 20 % of a mean intensity of approximately $P \approx 300$ mW/cm^2, which is above the limit of 200 mW/cm^2 invoked by [2].

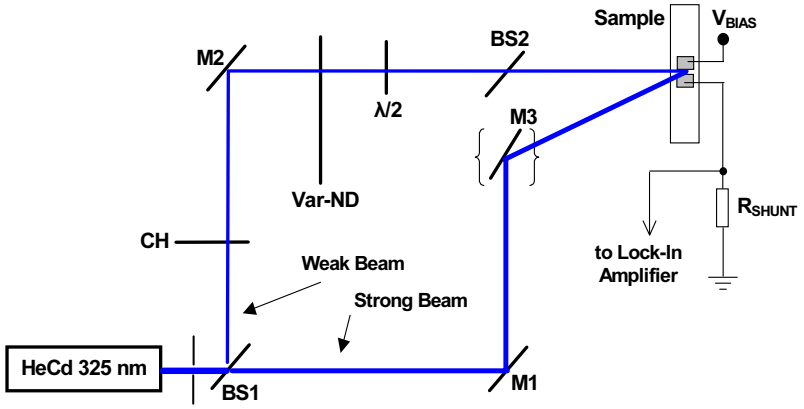

Fig. 1 Experimental setup: *BS1, BS2* - 50 % beamsplitter; *CH* - mechanical chopper; *M1, M2, M3* - aluminium mirrors; *Var-ND* - variable UV neutral density filter; *λ/2* - λ/2 plate.

The electrical readout of the photocurrent was done using a shunt resistance R_{SHUNT} and conventional lock-in technique. R_{SHUNT} was chosen as about a tenth of the sample dark resistance, and the bias voltage U_{BIAS} was limited to values that avoided excessive sample heating and lock-in amplifier saturation. Typical values were $U_{BIAS} \approx 2$ V and $R_{SHUNT} \approx 50\ \Omega$.

The photosensitivity of our samples was moderate, with photo- to dark conductivity ratios of about 10^{-3} under the described conditions. We used coplanar aluminium contacts separated about 1 mm (cf. 15 μm used in [2]). The Rose coefficients varied from sample to sample between 0.5 and 1, but were approximately constant for each sample for the whole range of excitation densities applied.
Instead of single beam splitting we used a double beamsplitter technique (DBST), as shown in Fig. 1. The UV ray is split at the beamsplitter BS1. While the weak beam arrives after reflection at mirror M2 and passage through beamsplitter BS2 between the two coplanar aluminium contacts on the sample, the strong beam is reflected at mirror M1 and can then also be reflected at BS2, or can be alternatively reflected at a mirror M3 onto the sample surface. The advantage of this technique is that we obtain a much wider range of possible beam intersection angles and therefore of experimental grating periods. Note that

this is of tremendous importance, as the grating period in the SSPG technique should be about 10-100 times the diffusion length to obtain a reliable measurement. The DBST allows adjusting experimentally grating periods continuously between 100 nm ≤ Λ ≤ 0.1 mm.

The DBST has a second advantage: it allows to confirm *directly* the interplay of critical optical parameters, like linear polarization, λ/2-plate, and coherence length. The excitation grating and its blurring after the rotation of the λ/2-plate can be visualized optically for high grating periods (low beam intersection angles), projecting the interference pattern by a small focal length lens onto a distant paper sheet. As a disadvantage, the DBST introduces asymmetric path lengths for the interfering beams.

The transmissivity through the λ/2-plate and the second beamsplitter was significantly different for linear and perpendicular polarization. We compensated the difference by a variable UV ND filter that assured that the relative change in weak beam intensity was below 5 %. The same optical element assured also that the ratio of weak-to-strong beam intensity was about 1:20.

The grating period Λ was found by triangulation, assuming the small angle approximation $\Lambda \approx (h/b) \times 0.325$ μm , where h is the height and b the base length of the triangle which lateral sides are the interfering beams. The error in the angle measurements is negligible compared to that obtained for the β values.

The SSPG measurements in GaN are notoriously difficult due to the poor photosensitivity, as mentioned above. From theoretical considerations, it is expected that the grating quality parameter γ_0 would approach zero for low photosensitivities [1]. This was not the case. However, it was necessary to use extensive averaging, through the lock-in amplifier time constant (several seconds), acquisition moving averaging (hundreds of points) and the observer.

Fitting was done using standard non-linear least square fitting techniques.

3 Experimental results

In Fig. 2 we show a typical experimental result for a 3 μm thick single layer of gallium nitride, with a diffusion length is L = (0.12 ± 0.01) μm**,** and in Table 1 we resume several experimental results of measurements at single GaN and $Al_XGa_{1-X}N$/GaN double layers, with variable aluminium concentrations.

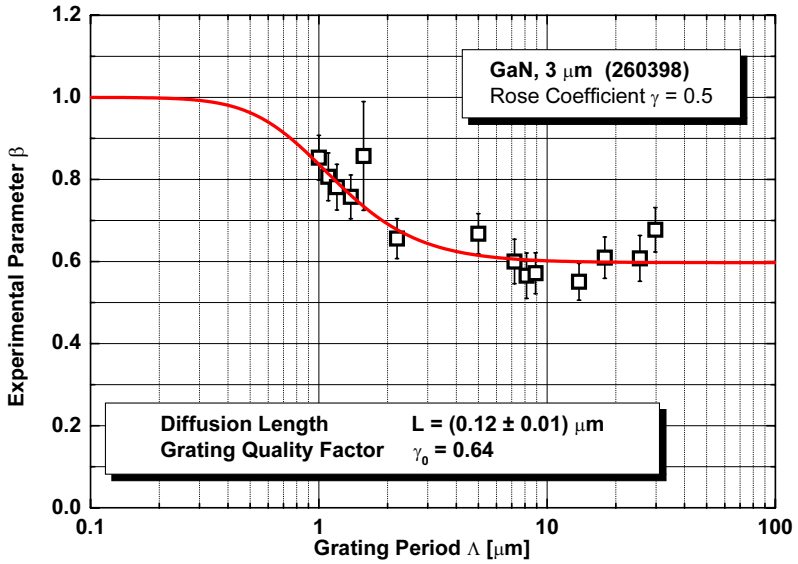

Fig. 2 Experimental result for a 3 μm thick GaN sample and best fits according to Eq. (2).

We find SSPG diffusion lengths varying between about 100 nm to 3 μm, with experimental errors which are typically of the order of 10 %, but may be as high as 50 %, reflecting strong scattering of experimental data points. Note that even in the more reliable measurements at single GaN layers as shown in Fig. 2 we obtain some data points which occasionally escape completely from the expected behaviour. In comparison, the scattering of data points in double layers is still more pronounced, and we attribute it to the limited sensitivity of our equipment. A second feature that appears generally in all experiments is an apparent up-bending of the grating parameter β at high grating periods. This is not expected from the idealized theory outlined above, but can be justified generically by a gradual decrease of the grating quality parameter as the grating period approaches the laser spot or mode size.
At the moment, it is not clear for what reason the measured SSPG diffusion lengths vary so much between single GaN layers, where similar values may be expected.

Table 1 SSPG diffusion lengths measurements of single GaN and $Al_XGa_{1-X}N$/GaN double layers.

Sample structure	Aluminium Conc. [at.%]	Thickness [μm]	SSPG Diffusion Length L_{SSPG} [μm]	Rose Coefficient	Grating quality factor
GaN	-	3	0.12 ± 0.01	0.5	0.64 ± 0.01
GaN	-	3	0.77 ± 0.05	0.51	0.56 ± 0.01
GaN	-	3	1.1 ± 0.5	0.6	0.26 ± 0.05
AlGaN / GaN	2.5	0.2 / 2	0.6 ± 0.2	0.70	0.49 ± 0.04
AlGaN / GaN	5	0.2 / 2	0.34 ± 0.04	0.68	0.59 ± 0.04
AlGaN / GaN	10	0.2 / 2	0.19 ± 0.04	0.95	0.40 ± 0.02
AlGaN / GaN	18	0.2 / 2	0.37 ± 0.08	0.99	0.56 ± 0.02
AlGaN / GaN	37	0.2 / 2	0.34 ± 0.16	0.63	0.49 ± 0.04

Neither the dependence of L_{SSPG} with aluminium concentration reveals a definite trend. Apparently, L_{SSPG} decreases up to aluminium alloying ratios of about 10 at.%, but one may expect much different values taking into account the influence of the spontaneously formed two dimensional electron gas at the AlGaN/GaN interface. Nevertheless, we think that the values are correct from an experimental point of view, and reliable taking into account the limitations of the SSPG technique. The SSPG technique described is essentially sensitive to the lateral diffusion of excess carriers in a slice within the penetration depth d_α of the excitation beam, which in our case is of the order $d \equiv \alpha^{-1} \approx (10^4\ cm^{-1})^{-1} \approx 1$ μm. Even in the double layer samples with a top aluminium layer of thickness 200 nm, we will therefore sense mainly the bottom GaN layer and explain the similar values of multilayer L_{SSPG} diffusion lengths compared to single layers.

In any case, the experimental values in Table 2 represent a reliable conjecture to be confirmed in further measurements under systematic variation of experimental conditions, and in particular operating with a monomode HeCd laser instead of the multimode that we used for the present study.

4 Discussion and conclusions

The values for the diffusion lengths are in general agreement with those measured by the authors mentioned above [2, 3], and are possibly a hint for a correlation between transport and morphology (columnar structure). The order-of-magnitude of grain sizes found with atomic force microscopy (AFM) or scanning electron microscopy (SEM) in our samples are typically in the range of several hundred nanometers.

However, we think that a direct correlation between structure and electronic properties has not been demonstrated unambiguously yet. From Setos theory [4] it should be expected that the electronic lengths scale depend strongly on the background electron concentration instead of the grain size. The fact that

several authors find the same diffusion lengths at different samples, with different morphologies and electron concentrations, suggest that there is a very fundamental mechanism that limits the diffusion length.

If we interpret our results in terms of ambipolar transport, we can convert the SSPG diffusion length L into the minority carrier (hole) μτ-product [5],

$$(\mu\tau)_{SSPG} \approx \frac{1}{2}\frac{q_{el}}{k_B T} L^2 \tag{3}$$

with the electron charge q_{el} and the thermal energy $k_B T$. In this equation -as in Eq. (4) - we assume that the hole mobility μ_p is much smaller than the electron mobility μ_n.

We can compare the obtained values with the majority carrier (electron) μτ-product as obtained from the steady state photoconductivity σ_{ph}, measured in the same coplanar contact configuration:

$$(\mu\tau)_{SSPC} \approx \frac{\sigma_{ph}}{q_{el}}\frac{E_{h\nu}}{\alpha \cdot P} \tag{4}$$

where $E_{h\nu}$ is the photon energy, α the absorption coefficient and P the excitation intensity. The so-obtained values are visualized in Fig. 3, as function of the aluminium concentration.

We find about a order of magnitude difference for majority and minority μτ-products, with the majorities showing values of the order ($\mu\tau_n = 10^{-6}$ cm^2/V. The electron μτ-product obtained from the SSPC is order-of-magnitude consistent with typical Hall mobilities around μ_H = 50 cm^2/Vs and lifetimes in the microsecond time range, as determined in room temperature Hall measurements and transient photoconductivity, respectively.

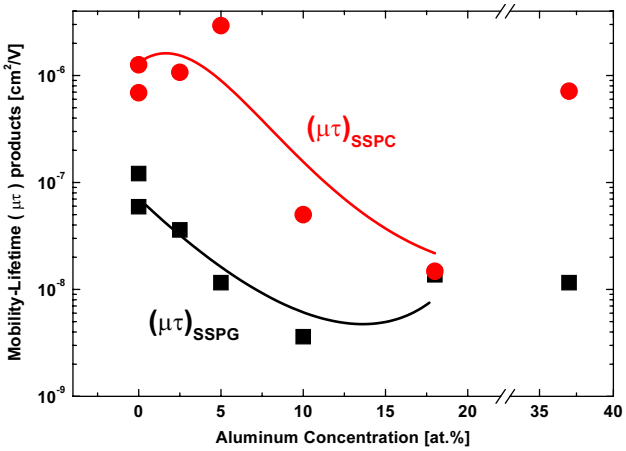

Fig. 3 μτ-products obtained from steady state photoconductivity (SSPC) and from the steady state photocarrier grating (SSPG) technique as function of the aluminium concentration. The lines are guides to the eye.

Both μτ products decrease continuously with increasing aluminium concentration, an order-of-magnitude at X = 10 at.%. The slight initial constancy of the SSPC-μτ may be attributed to the fact that we sense partly the bottom thick GaN layer. If we attribute the constant difference of a factor of ten to the difference in electron and hole mobilities, our data suggests that minority and majority carrier lifetimes are very similar. This is most likely associated to a common recombination mechanism.

The complete theoretical description, including drift and diffusion effects on several time scales and as function of electrical and optical bias, shows that the exact SSPG equations are quite complex [6], even without taking trapping into account. Drift becomes important at high bias voltage, and has the effect to

reduce the grating quality factor. Additionally, internal space charge field drift components may dominate the response at high intensities. Both cases can be excluded under the present experimental conditions.

An important question is if the system had time to pass from the lifetime regime [7], i.e. excess carriers move independently, to the ambipolar regime, with coupled dynamics of free carriers. As the high dark conductivity implies a short dielectric relaxation time, we should be in the ambipolar regime - unless recombination annihilates electrons and holes.

However, our main argument here is that fast trapping in extended acceptor-like valence band tails prevents the free excess minority carriers (holes) to recombine with the background electrons. This picture is in line with the slow response of transient photocurrents and transient photoluminescence, and we have recently developed a simple analytical model to describe its dynamics. Trapped holes will no longer participate *actively* in the grating blurring, but *passively,* as spatially localized recombination centres. In this sense, the transport is not ambipolar. We are instead in a second pos-trapping lifetime regime.

Though the quantitative consequences of this proposal have still to be elaborated in more detail, it is clear that it offers a valuable new organization principle, able to answer the two main questions that are directly connected to the two fitting parameters:

1. Why does the grating quality parameter not approach zero in spite of the moderate photoresponsivity ?
2. Why do all diffusion length measurements yield so similar results?

We observe also that the experimental beta coefficient does not become negative, as is expected from theory and found in amorphous semiconductors, but not in wide gap semiconductors.

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