A comparison of the Hall-effect and secondary ion mass spectroscopy on the shallow oxygen donor in unintentionally doped GaN films

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We report on temperature-dependent Hall-effect measurements and secondary ion mass spectroscopy on unintentionally doped, *n*-type conducting GaN epitaxial films. Over a wide range of free carrier concentrations we find a good correlation between the Hall measurements and the atomic oxygen concentration. We observe an increase of the oxygen concentration close to the interface between the film and the sapphire substrate, which is typical for the growth technique used (synthesis from galliumtrichloride and ammonia). It produces a degenerate *n*-type layer of ≈ 1.5 μm thickness and results in a temperature-independent mobility and Hall concentration at low temperatures (<50 K). The gradient in free carrier concentration can also be seen in spatially resolved Raman and cathodoluminescence experiments. Based on the temperature dependence of the Hall-effect, Fourier transform infrared absorption experiments, and photoluminescence we come to the conclusion that oxygen produces a shallow donor level with a binding energy comparable to the shallow Si donor. © *2000 American Institute of Physics.* [S0021-8979(00)00616-2]

I. INTRODUCTION

The source of the *n*-type conductivity in unintentionally doped GaN and the role of oxygen is still a matter of debate. According to calculations,[1–5] native defects, such as the nitrogen vacancy, cannot be responsible for the high background *n*-type conductivity with carrier concentrations in the range between high 10^{19} cm^{-3} and low 10^{17} cm^{-3}. Already in 1983 Seifert *et al.*[6] presented experimental evidence for oxygen as a potential candidate. Since the epitaxial growth of GaN from the vapor phase takes place at temperatures of 1000 °C and above, in hot wall reactors, in particular, there might be significant incorporation of silicon from the quartz reactor walls into the films. It was in 1994 that Neugebauer and Van de Walle, based on first principles calculations could show that O and Si have low formation energies and hence could be the source of the *n*-type conductivity.[2] The theoretical predictions were confirmed by experiment two years later by Götz *et al.* and other groups[7–14] on Si-doped as well as on unintentionally doped samples using the Hall-effect and secondary ion mass spectroscopy (SIMS). Either Si or O could account for the free carrier concentrations in hydride vapor phase epitaxy (HVPE) material.

In 1999 Chen *et al.*[15] reported on optical studies on films grown by different techniques with free carrier concentrations from highly *n* type to highly *p* type (before electron irradiation). The samples were then irradiated with 2 MeV electrons. A photoluminescence emission at 0.88 eV is enhanced by the electron irradiation. This emission of a deep center has a rich vibrational structure of lattice and local phonons, and is strikingly similar to the emission caused by oxygen on a phosphorus site in GaP. The authors concluded that O_N in GaN is most likely a deep donor (also from an energetic point of view). Hence, the O_N defect could not be responsible for the residual *n*-type conductivity.[15]

In 1998 we gave a preliminary report on Hall and SIMS measurements[16] on films grown by low pressure chemical vapor deposition (LPCVD). The films showed high free carrier concentrations between 7×10^{18} and 5×10^{19} cm^{-3}. The films were grown without a buffer layer on sapphire substrates and should have a highly conducting layer (influencing the Hall-effect) close to the interface, similar to the reported results of Götz *et al.*[10] At very high carrier concentrations, however, the morphology also degraded (cracks and large three-dimensional islands occurred). Although oxygen was found by SIMS in high enough concentrations to account for the carrier densities, oxygen might be incorporated in grain boundaries, dislocated areas, or be present as a Ga_2O_3 phase. We therefore extended our investigations on samples with a wider range of free carrier concentrations from low 10^{18} cm^{-3} to high 10^{19} cm^{-3}, we included Si in the SIMS analysis and measured depth profiles of O and Si by SIMS to conclude about the presence of a highly conductive layer close to the interface. Moreover, we used spatially resolved cathodoluminescence (CL) along a cleavage plane to observe the influence of the carrier density on the CL-transition energy. The energy position is known to redshift (in the absence of strain) due to renormalization ef-

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fects and blue shifts by the Burstein–Moss effect at high carrier densities. The Hall-effect integrates over the entire sample. In order to get information about density gradients we used micro-Raman monitoring of the phonon plasmon modes.

II. EXPERIMENT

Standard photoluminescence (PL) measurements were carried out at 300 and 4.2 K in a helium-bath cryostat. We used the 325 nm line of a HeCd laser (Omnichrom type series 74, power 23 mW) for excitation. The signal was detected by a 22.7 cm Spex monochromator (type 1681) and a photomultiplier (type Hamamatsu R375) with an operation range between 160 up to 850 nm. To achieve a better signal to noise ratio the photomultiplier was connected to an EG&G 5210 look-in amplifier.

The variable temperature Hall-effect measurements were conducted with a computer controlled system. A Keithley source measurement unit as a constant current source and a Keithley Hall-effect card (type 7065) were connected to the system for impedance changing. The Hall voltage was measured by a Keithley electrometer (type 617). Samples of 5×5 mm^2 size were contacted with In dots in the four corners and annealed at 380 °C to achieve ohmic contacts in the Van der Pauw geometry. The contacts showed excellent ohmic character over the whole temperature range. All layers were measured from 4.2 up to 500 K in an Oxford He cryostat.

Standard CL measurements were carried out between 5 and 500 K in a helium cryostat with a modified electron microscope. The maximum lateral resolution, i.e., excitation depth, depends on the applied voltage. It is possible to vary this voltage between 500 V and 40 kV. The maximum lateral resolution is limited by the microscope at 50 nm. In our experiments the effective resolution was restricted by the diffusion length in the GaN samples.

For hexagonal GaN samples Raman spectroscopy shows the following phonon modes: A_1(TO), A_1(LO), E_1(TO), E_1(LO), and E_2. The line position of the E_2 mode is independent of the carrier density. However, the position significantly shifts with σ_{xx} stress in GaN crystals.[17] Equation (1) shows the link between the line position $\omega(\sigma_{xx})$ and the stress σ_{xx} in GaN,

$$\omega(\sigma_{xx}) = 570 \, \text{cm}^{-1} + 4.2 \, \frac{\text{cm}^{-1}}{\text{GPa}} \sigma_{xx}. \tag{1}$$

With Raman spectroscopy it is also possible to measure the free carrier density, since the A_1(LO) phonon can couple to the plasmon to two new LPP$^+$ and LPP$^-$ modes (LPP =longitudinal optical phonon plasmon mode). From the energy position of the LPP$^+$ mode it is possible to estimate the carrier density in GaN. The frequencies of the LPP modes are shown as follows:

$$\omega(\text{LPP}\pm) = \tfrac{1}{2}\sqrt{(\omega_L^2 + \omega_P^2 \pm \sqrt{[(\omega_L^2 + \omega_P^2)^2 - 4\omega_P^2\omega_T^2]})}, \tag{2}$$

where ω_L and ω_T are the frequencies of the longitudinal and transversal optical modes. The plasma frequency depends only on the carrier density (electrons) and the materials constants (e, m_e^*, ϵ have their usual meanings) as shown in the following:

$$\omega_P = \sqrt{\frac{ne^2}{m^*\epsilon\epsilon_0}}. \tag{3}$$

Using Eqs. (2) and (3) allows us to link the carrier density to the measured LPP modes. Spatially resolved measurements were performed with the micro-Raman system along the edge of a cleaved sample to estimate the carrier density depth profile from the GaN layer to the sapphire/GaN interface. For excitation we used the 514 nm line of a Ar ion laser, the scattered light was analyzed with a triple spectrometer with an effective resolution of 0.1 cm^{-1}. Quantitative impurity analysis (based on implanted standards) on H, O, C, and Si was performed by secondary ion mass spectroscopy using a 14.5 keV Cs$^+$ ion beam. For O both isotopes ^{16}O and ^{18}O were analyzed to avoid any confusion with CH$_4$. The concentrations for both isotopes were identical.

The samples were grown by low pressure chemical vapor deposition at growth temperatures between 980 and 1060 °C without buffer layer on sapphire substrates. NH$_3$ and GaCl$_3$ were used as precursors. Typical growth rates were 3–4 μm/h. In contrast to hydride vapor phase epitaxy (HVPE) where the precursor GaCl is synthesized from metallic Ga and HCl at temperatures around 850 °C, the LPCVD technique uses a solid state Ga-precursor GaCl$_3$, which is a salt. As for metalorganic precursors it can be stored in a bubbler and upon heating the bubbler to temperatures between 40 and 140 °C it can brought into the gas phase and is transported by hydrogen or nitrogen carrier gases into the reaction chamber.[18]

III. EXPERIMENTAL RESULTS

A. Mass spectroscopy

In the SIMS experiments we focused on the role of O, Si, and C. C might be a potential acceptor in GaN. Hydrogen was detected in all films in the high 10^{19} cm^{-3} range, but seems to have no influence on the electrical properties. For Cl (which comes from the precursor) no standard is available, and we cannot state anything about its concentration. However, considering the situation in other III–V semiconductors, we assume that Cl induces no electrical active defect. In Fig. 1(a) we show the SIMS depth profile of a 9-μm-thick film with a free carrier concentration at room temperature of 2.8×10^{18} cm^{-3}. The Si concentration with 6×10^{17} cm^{-3} is constant throughout the film, C decreases slightly from a value of 2×10^{18} cm^{-3} at the surface toward the interface. Oxygen starts at a level of 3×10^{18} cm^{-3} at the surface, is rather constant over a distance of 8 μm, but piles up in the last 1.5 μm toward the interface and ends with a concentration of mid 10^{19} cm^{-3}. A second example is given in Fig. 1(b). The hydrogen and oxygen concentrations are between 5 and 6×10^{19} cm^{-3}, C shows a strong gradient decreasing from around 3×10^{19} to 1×10^{18} cm^{-3} toward the interface, Si is at a constant level of 1.5×10^{18} cm^{-3}. Again, in the last 0.5 μm there is a strong increase in the O concentration. The Hall-effect determined the free carrier concen-
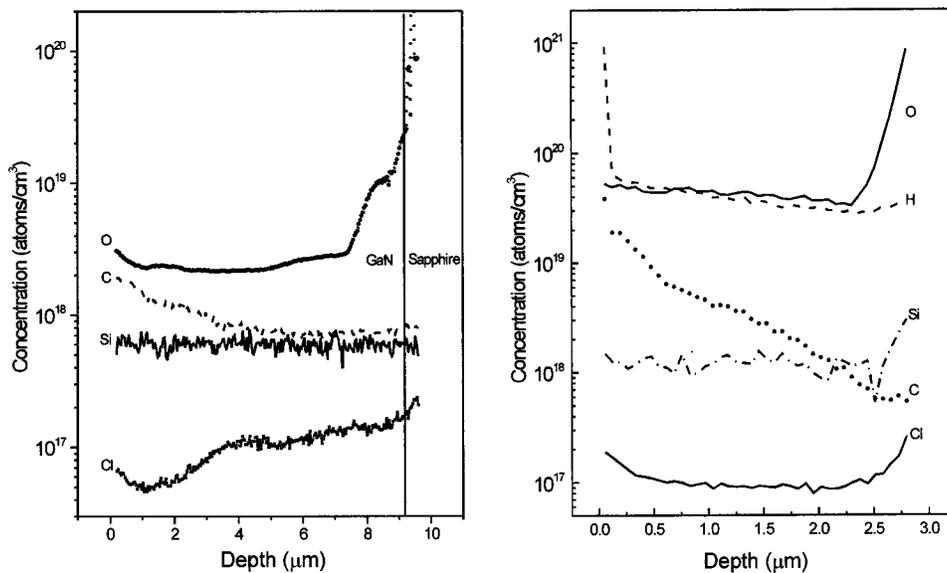
FIG. 1. SIMS depth profiles showing the atomic concentrations of O, Si, H, and C (Cl is only shown for comparison and is not quantitative). (a) For a sample with free carrier concentration 2.8×10^{18} cm^{-3} and (b) 4.5×10^{19} cm^{-3}.

tration at RT to 4.5×10^{19} cm^{-3}. The strong O gradient close to the interface is typical for HVPE grown GaN films (the LPCVD technique is just a variant of this method) where a low temperature buffer layer is not used. But even a GaCl pretreated sapphire substrate shows a highly conductive, ~200-nm-thick layer at the GaN/sapphire interface which can be detected by the Hall-effect and C–V measurements.[8,10] Look *et al.* derived similar conclusions.[11] They concluded that the presence of a degenerate *n*-type layer, which they found in all HVPE grown films, does not affect the electrical and optical properties of the bulk. This is certainly true for photoluminescence experiments (see the following) where the excitation is from the surface site and the diffusion length of the photogenerated carriers is much less than the sample thickness. However, the layer causes a current shunt and thus affects the conductivity and the Hall coefficient.

B. Optical investigation

Independent confirmation of the gradient in the free carrier concentration comes from spatially resolved Raman experiments. From the position of the LPP$^+$ mode taken along the cleavage plane of a GaN film one notices an increase in the free carrier concentration [using Eq. (2)] from the surface to the interface by a factor of 3 (see Fig. 2).

In undoped films with low free carrier concentrations the luminescence spectrum is dominated by the recombination of excitons bound to neutral donors ($D^O X$). Its full width at half-maximum can be taken as a measure of the quality of the films, since it is rather sensitive to the carrier density. The linewidth of the donor bound exciton [see Fig. 3(a)] in HVPE films with $n = 2 \times 10^{17}$ cm^{-3} was only 2.4 meV, best values have been reported as narrow as 0.8 meV.[19,20] With increasing carrier density the linewidth increases. For $n = 2.4 \times 10^{18}$ cm^{-3} (RT) we obtain 6.6 meV at 4.2 K (34 meV at RT), for $n = 5.2 \times 10^{18}$ cm^{-3}, 36 meV (66 meV at RT), and for $n = 4 \times 10^{19}$ cm^{-3} we measure 60 meV at 4.2 K (100 meV at RT). The linewidth at the highest concentration may be taken as an upper limit, since the 3.42 eV band

caused by structural disorder[21] may also contribute to the linewidth. The highly conductive layer at the interface can be seen in PL when exciting through the transparent substrate [see Fig. 3(b)]. We see a pronounced broadening indicative of the high carrier density together with a blueshift which originates from the Moss–Burstein effect. We are above the Mott transition, thus we deal with band to band (BB) transitions where electrons from the Fermi level which might be inside the conduction band recombine with holes. The donor acceptor pair band at around 3.28 eV followed by one and two LO phonon replicas, which at high carrier densities is replaced by an electron in the conduction band to acceptor transition (eA^O) involving a shallower acceptor with $E_A = 220$ meV, is also pronounced at the interface. This transition is hardly seen when exciting from the surface site [see Fig. 3(b)].

Apart from the Moss–Burstein effect, there are two other mechanisms[22] that influence the energetic position of the band edge excitonic luminescence. Due to the difference in thermal expansion coefficients between the GaN layer and
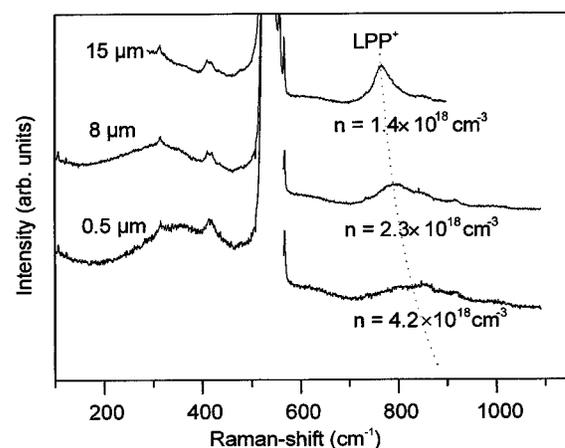

FIG. 2. Micro-Raman spectra taken along the cleavage plane at three different distances with respect to the interface. The position of the phonon plasmon mode (LPP$^+$) gives the free carrier concentration.
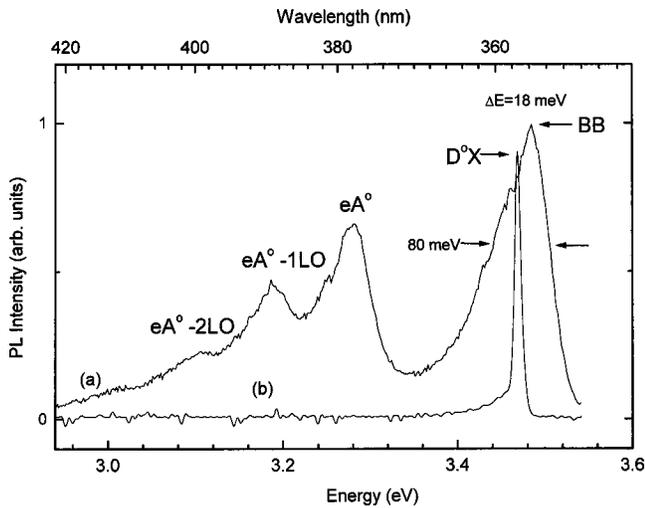
FIG. 3. Photoluminescence spectra of a GaN film with a free carrier concentration of 3×10^{18} cm^{-3} with excitation through the substrate (a) and of the GaN surface site (b).
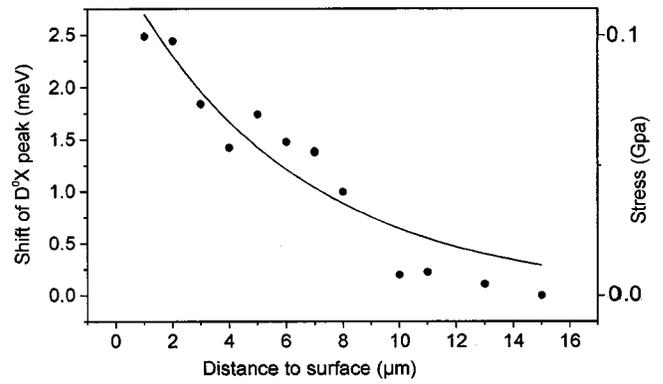

FIG. 4. Shift of the neutral donor bound exciton line as a function of the distance from the substrate. The right axis shows the corresponding stress values as determined by micro-Raman.

the sapphire substrate, the films are under compressive strain. With increasing layer thickness the strain (given in gigapascals) decreases as shown by measurements along a cleavage plan with micro-Raman (Fig. 4). The stress was calculated using Eq. (1). With the lowering of the compressive strain the excitonic emission shifts to lower energy. For the 16-μm-thick epilayer this shift is 2.5 meV. Note that the energy difference in Fig. 3 is 18 meV. At carrier densities between 10^{18} and 10^{19} cm^{-3} renormalization of the band gap energy also leads to a blue shift.[22] The three contributions to the line position could be distinguished in spatially resolved cathodoluminescence (CL) experiments (for further details
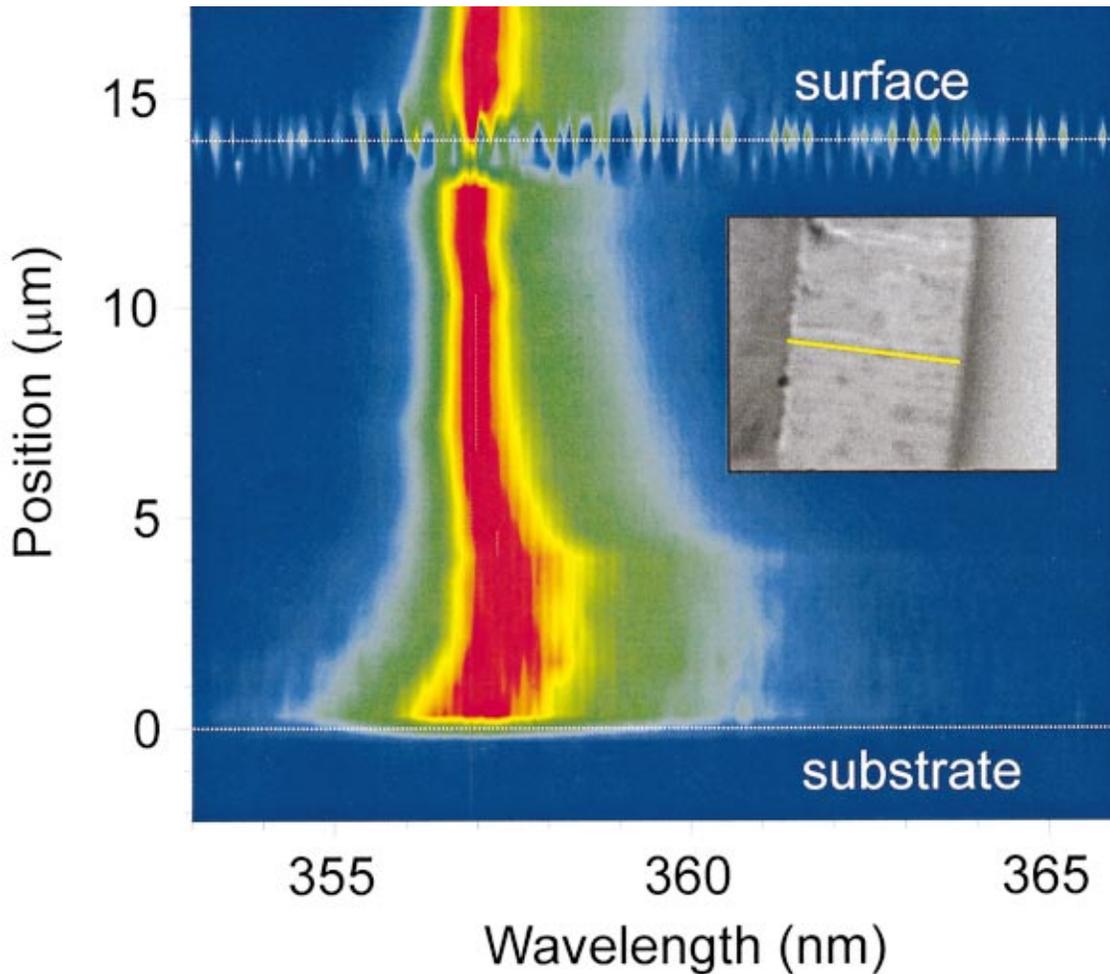

FIG. 5. (Color) Spatially resolved cathodoluminescence spectra taken along the cleavage plane of a film.
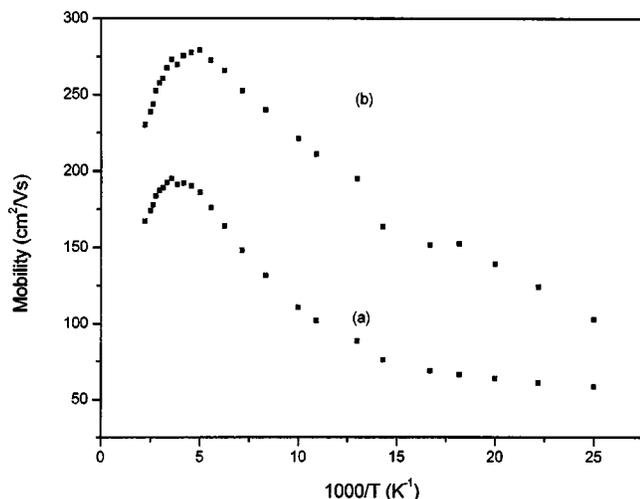
FIG. 6. Mobility as a function of the reciprocal temperature (a) as measured from the Hall-effect, (b) corrected for the influence of a degenerate *n*-type layer at the interface GaN/sapphire.
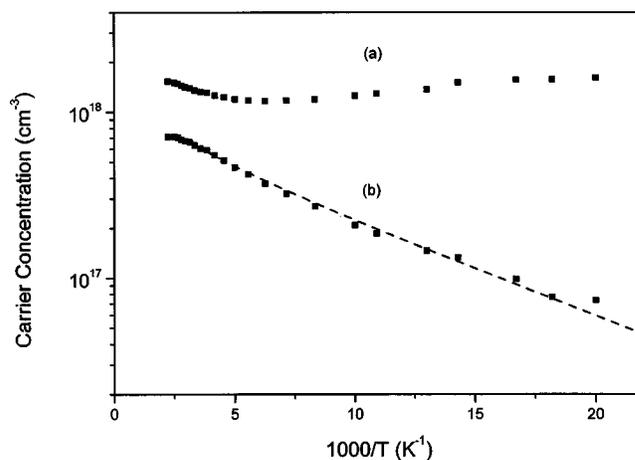

FIG. 7. Hall carrier density as a function of the reciprocal temperature (a) as measured, (b) corrected for the influence of a degenerate *n*-type layer at the interface GaN/sapphire. The dashed line is a theoretical fit giving an activation energy of 8 meV.

see Ref. 23). For the case discussed here it is more informative to look at the luminescence linewidth. In Fig. 5 we show the CL results. The wavelength spectra were taken along the cleavage plane starting from the substrate toward the surface. Different colors correspond to different intensities and range from blue (lowest intensity) over green to yellow up to red (highest intensity). One can directly see the reduction in linewidth from the interface toward the surface of the film. Thus, the results from SIMS, Raman, CL, and PL confirm the presence of a highly conductive layer at the interface GaN/sapphire.

C. Electrical measurements

For the analysis of the temperature-dependent Hall-effect a two-layer model—layer I for the bulk and layer II for the interface layer—is used following the interpretation of Look.[11] Figure 6(a) shows the temperature dependence of the mobility from 450 to 40 K, which passes through a maximum at 150 K and is constant at temperatures below 60 K. The Hall concentration, likewise constant at low temperatures, goes through a minimum and increases for temperatures above 300 K [see Fig. 7(a)]. Correcting for the influence of the interlayer, as outlined in detail in the works of Look,[11] gives the corresponding data for mobility in Fig. 6(b) and Hall concentration in Fig. 7(b). A theoretical fit of the Hall concentration n_H versus reciprocal temperature $1/T$ is based on the charge neutrality condition, assuming a single donor type level.[11,12] The fitting parameters are the concentration N_A of compensating acceptors, the concentration N_D of donors, as well as the thermal activation energy E_D. As a result we obtained: $N_D = 1.4 \times 10^{18}$ cm^{-3}, $N_A = 2 \times 10^{17}$ cm^{-3}, and $E_D = 8$ meV. Considering screening, E_D is connected to the respective donor binding energy $E_D = E_{D0} - \beta N^{1/3}$ with $\beta = 2.1 \times 10^{-5}$ meV cm.[24] We obtained for $E_{D0} = 32 \pm 5$ meV, which can be compared with the value of 34.5 meV we calculated from Fourier transform infrared (FTIR) experiments using a simple effective mass theory approach.[24,25] In the FTIR experiments on HVPE films the

transitions from the neutral donor ground state to Rydberg-type excited states were observed. On the basis of the Hall-effect alone we cannot decide whether the residual donor is oxygen or silicon, since for Si similar activation energies were found in temperature-dependent Hall measurements.[12] From magneto-optical experiments for Si donor a binding energy of 29 meV was obtained.[26]

It is now generally accepted that in metalorganic vapor phase epitaxial grown GaN films the residual donor is Si, as long as the moisture in the ammonia is blocked by proper oxygen filters. In our growth technique we have an additional source for O, since the GaCl$_3$ salt is very hygroscopic and O contamination cannot be avoided. Considering the SIMS results, we feel confident that oxygen is the residual donor in our films. Si partially contributes to the free carrier concentration for $n < 4 \times 10^{18}$ cm^{-3}, but for films with $n > 10^{19}$ cm^{-3}, Si is in general more than one order of magnitude below the O concentration and hence too low to explain the free carrier concentrations (see Fig. 1). Within the set of samples we investigated we always find that n_H is $\approx 0.7–0.8$ times the oxygen concentration (see Fig. 8). O, which is not electrically active, could exist in other phases such as Ga$_2$O$_3$. Goetz recently reported[12] on Hall-effect measurements of Si, Ge, and O donors in GaN. The O donor concentration of 1×10^{18} cm^{-3} was only 10% of the total atomic O concentration for reasons that were unclear. Such a large discrepancy did not appear in our films. He obtained a 300 K mobility of 314 cm^2/V s, a value which comes close to our results for the same carrier concentrations (see Fig. 9). He also concluded that the O donor binding energy of 35 meV could be consistent with his Hall data.[12]

IV. DISCUSSION

Optical spectroscopy, especially photoluminescence, is commonly used to characterize the material properties of nitride epitaxial films. In nominally undoped layers with donor concentrations below 5×10^{17} cm^{-3} the dominant recombination is the neutral donor bound exciton line. Apparently
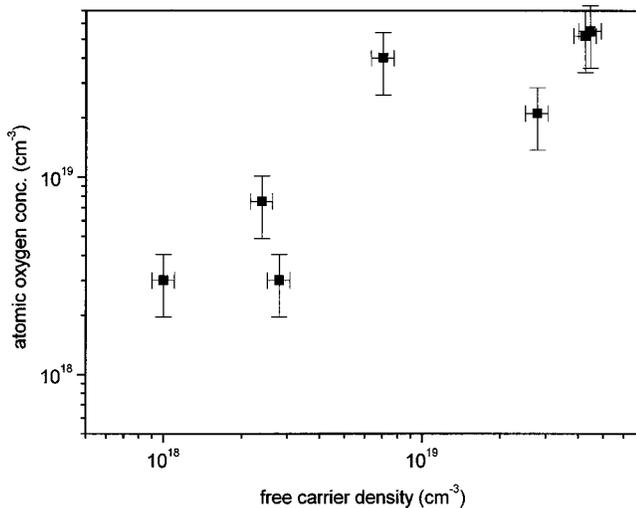
FIG. 8. Atomic oxygen concentration measured by SIMS vs the free electron concentration as measured by the Hall-effect.

FIG. 9. Mobility vs free carrier density (at room temperature).

the localization energies of the excitons bound to Si or O donors are very similar and the chemical nature of the donor cannot be determined from PL experiments alone. Our optical experiments using PL, CL, and Raman, tell us that high carrier densities are present, they resolve density gradients from close to the substrate up to some micrometers into the bulk of the sample, but they are not sensitive to the microscopic origin of the donor(s). In secondary ion mass spectroscopy the impurities with the highest concentration were H, C, O, and Si. Si and O are donors in GaN, other possible donors such as Ge, S, or Se were not detected by SIMS. For the interpretation of the Hall data we only have to take into account O and Si. In the films with high oxygen concentrations and high free carrier densities ($>10^{19}$ cm^{-3}) the Si concentration was a factor of 50 below the O concentration [see Fig. 1(b)] and the contribution of Si donors to the free carrier density is negligible. In our films with lower carrier concentration Si donors can contribute to the free carrier concentration, but on the basis of our data it is only a 10% effect [see Fig. 1(a)]. We therefore conclude that O induces a shallow donor level and is the source of the residual *n*-type conductivity in undoped GaN films, in agreement with theoretical predictions and experimental findings.[1,2,6,12] We now provide a final comment on the concentration and nature of acceptors in *n*-type GaN.

The number of compensating acceptors is usually small in a *n*-type material, Look[11] gave a value N_A/N_D of 0.25 and Goetz *et al.* of 0.1.[18] From our fitting procedure to the free carrier concentration the ratio of compensation is found to be $N_A/N_D = 0.14$. A potential candidate could be carbon on the nitrogen site, and C seems to be an omnipresent residual impurity (see also Fig. 1). *P*-type doping with C has been reported in Ref. 27 and the achieved hole densities were in the low 10^{17} cm^{-3} range. According to theoretical predictions[28] there is a solubility limit for C$_N$ at mid 10^{18} cm^{-3}.

Comparing our experimentally determined mobilities with the theoretical predictions of Chin *et al.*[29] would indicate a significantly higher degree of compensation (N_A/N_D
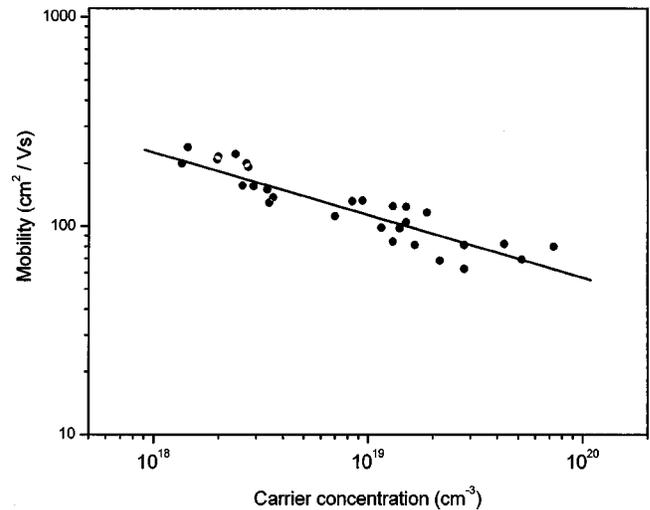
around 0.6). They included four scattering mechanism: ionized impurity, piezoelectric, acoustic deformation potential, and polar-mode optical phonon. GaN epitaxial films are grown on highly lattice mismatched substrates and the high numbers of dislocations directly affect the mobility as demonstrated recently.[30,31] Hence low mobilities are not a consequence of higher numbers of compensating acceptors but reflect the high number of dislocations—in our films typically around 10^{10} cm^2.

V. SUMMARY

We have reported on temperature-dependent Hall-effect measurements and secondary ion mass spectroscopy on unintentionally dopes, *n*-type conducting GaN epitaxial films. Over a wide range of free carrier concentrations we find a very good correlation between the Hall density and the atomic oxygen concentration. Si is commonly one to two orders of magnitude lower in concentration. We find an increase of the oxygen concentration close to the interface between the film and the sapphire substrate, which is typical for the growth technique—a variant of HVPE. It produces a degenerate *n*-type layer and the Hall-effect data have to be analyzed in a two-layer model. The gradient in free carrier concentration can also be seen in spatially resolved Raman via the shift of the phonon plasmon mode, and in the linewidth broadening and the energetic shift of the near band gap emission in cathodoluminescence experiments. Based on the temperature-dependent Hall-effect, recently performed Fourier transform infrared absorption and photoluminescence experiments we conclude that oxygen produces a shallow donor level with a binding energy comparable to silicon.

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