Low-resistivity vertical current transport across AlInN/GaN interfaces

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Effects of n-type doping of Al0.82In0.18N/GaN heterostructures on the conduction band (CB) profile have been investigated. Doping concentrations well above 1019 cm−3 are required to reduce the large barriers in the CB. Experimentally, Si- and Ge donor species are compared for n-type doping during metalorganic vapor phase epitaxy. For Si doping, we find substantial interface resistivity that will strongly contribute to total device resistivity. Doping of AlInN is limited by either the onset of a self-compensation mechanism (Si) or structural degradation of the AlInN (Ge). Only by Ge doping, purely ohmic behavior of periodic AlInN/GaN layer stacks could be realized.

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Semiconductor microcavities in the group-III nitride material system find application in laser diodes and light-emitting diodes.1,2 Lattice-matched Al0.82In0.18N (LM-AlInN) can be grown on GaN with high quality allowing for a virtually unlimited number of layer pairs as mechanical stress may not be incorporated during growth.3–5 The refractive index contrast between these layers of about 7% is very attractive for edge emitting laser diodes and vertical cavity semiconductor lasers.6,7 Yet, the conduction band (CB) offset between LM-AlInN and GaN amounts to 1.0 eV.8,9 Also, the wurtzite nature of these materials gives rise to spontaneous polarization charges at each interface. As a consequence, large barriers build up in the CB of AlInN/GaN heterostructures which increases electrical losses in electrical injection lasers and LEDs. With the conventional growth direction in (0002) positive net polarization charges are located at the lower interface of AlInN/GaN heterostructures while negative charges exist at the upper interface. A comprehensive analysis is required on how impurity doping has to be applied to reduce such barriers.

In GaN bulk material, Si and Ge are known shallow donors with similar activation energies of about 17–19 meV for a doping level near 1017 cm−3.10 Reports about n-type doping of AlInN are scarce since uniform layer growth, as needed to perform reliable Hall-effect measurements, is limited to near lattice-matched composition and to about 100–200 nm thickness.9 A theoretical calculation of Ge substitutional donors in AlxGa1−xN presented by Gordon et al., predicts DX center formation for xAl > 0.5.10 Blasco et al. however demonstrated n-type conductivity in strained AlxGa1−xN/Ge/GaN layers grown by molecular beam epitaxy up to xAl = 0.66 with activation energies between 5 and 40 meV.11 Since lattice-matched Al0.82In0.18N compares to unstrained Al0.34Ga0.66N in terms of bandgap energy, Ge in Al0.82In0.18N could behave as an effective donor.

Furthermore, doping-related issues with the growth of GaN and AlInN have to be considered. Here, our focus is on metalorganic vapor phase epitaxy (MOVPE) as the dominant growth technology for semiconductor nitride materials. It is however noted that doping-related growth issues are less prominent in molecular beam epitaxy. Silicon doping in GaN during MOVPE growth is limited to about [Si]~2 × 1019 cm−3 by the onset of 3D growth which is caused by SiN formation.12 Still, conductive AlInN/GaN Distributed Bragg Reflector (DBR) structures grown by MOVPE with ohmic characteristics were demonstrated by the application of delta-like Si doping (6.0 × 1019 cm−3) and implementation of graded DBR interfaces. However, graded AlInN/GaN interfaces require a complex growth process and the achieved resistance of 17 Ωms is still relatively high.13,14 On the other hand, Germanium (Ge) doping of GaN during MOVPE growth is only limited by the onset of self-compensation at around [Ge]~3–4 × 1020 cm−3.15,16 Ge doping of AlInN has not been reported previously. The generally weak stability of AlInN during MOVPE growth may put limits on the doping with Ge atoms. In addition, Ge doping in MOVPE is known for a memory effect leading to non-uniform incorporation during growth.17 In this report, we investigate doping and growth of periodic AlInN/GaN layer stacks that can serve as one-dimensional photonic crystals for mode confinement in optical waveguides and cavities. As the simulation of the CB profile suggests, heavy n-type doping of AlInN and GaN is beneficial for electrical transport. Accordingly, the limits of n-type doping of AlInN by either Si- or Ge-atoms in MOVPE deserve investigation. For both species, very high doping levels may not be achievable due to either growth chemistry or incorporation issues. This report, therefore, explores Si- and Ge doping in AlInN with the goal to achieve low-resistive vertical current transport across AlInN/GaN heterostructures.

The analysis starts with the simulation of conduction and valence band profiles according to different doping levels in GaN and AlInN. These simulations are based on a self-consistent solution of the Schrödinger–Poisson-equations. Material parameters of GaN and AlInN to calculate the bands were taken according to Ref. 18. Respective results are shown in the top and middle graph of Fig. 1. Assuming donor activation energies of 19 meV (GaN) and 27 meV (AlInN). The latter value has been arbitrarily taken between 5 and 40 meV according to the work of Blasco et al. on Ge doping of Al0.34Ga0.66N since no activation energy for AlInN:Ge is known. Therefore, Fig. 1(c) shows band profiles for different Ge activation energies of AlInN. Our simulated structure consists of 35 nm Al0.82In0.18N embedded in GaN:Ge.

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Uniform doping profiles were assumed in each layer. The embedding GaN sections have always the same doping level. For doping concentrations below $10^{19} \text{ cm}^{-3}$ large barriers exist in the CB which originate from the upper AlInN/GaN interface as seen in Fig. 1(a). These barriers extend across the whole AlInN layer but also into the upper GaN layer. At the lower GaN/AlInN interface electron accumulation pins the whole AlInN layer but also into the upper GaN layer. In AlInN layers, doping levels are difficult to extract by Hall-effect measurements for reasons already mentioned. Optimum Ge doping in AlInN was evaluated for the vertical electrical resistance of single and stacked AlInN/GaN layers. In order to determine the onset of structural degradation for Ge doping within AlInN, we assessed structural properties by high-resolution X-ray diffraction and atomic force microscopy. Vertical electrical transport behavior through the AlInN/GaN stack was investigated by recording current–voltage ($I–V$) characteristics on $1 \times 1 \text{ mm}^2$ large mesa structures with top metal electrodes of $200 \times 200 \mu \text{m}^2$. These mesa structures were processed by dry etching and photolithography methods followed by deposition of a Ti (14 nm)/Al(33 nm)/Ni(10 nm)/Au(60 nm) metal stack by evaporation. In order to extract the interfacial resistances of the Al$_{0.82}$Ga$_{0.18}$N/GaN stack a modified transmission line measurement (TLM) method was used. Thereby, the current transport through pairs of $200 \times 100 \mu \text{m}^2$ sized mesa structures with varying distances was measured. Temperature-dependent $I–V$ measurements were done in a liquid-nitrogen cooled cryostat. Electrical four-point-probe measurements were performed to obtain precise resistance values.

From secondary ion mass spectroscopy (SIMS) analysis of a series of Ge-doped AlInN layers sandwiched between steadily Ge-doped GaN layers a linear incorporation behavior is found at $T_{\text{growth}} = 780 \, ^\circ \text{C}$. In this series, the molar flow rate of IBGe was varied between 0.6 and 6 $\mu\text{mol min}^{-1}$, which corresponds to a vapor phase concentration relative to TMIn.
plus TMAl of 0.9%–9%. The Ge concentration in AlInN has been roughly estimated by SIMS using the Ge signal in the surrounding GaN:Ge layers as a reference to the free carrier concentration found in above mentioned Hall-effect measurements. The accuracy of the Ge concentration levels in AlInN shown in Fig. 2 may therefore be only within an order of magnitude. A representative SIMS profile of the elemental distribution for Ge in the upper right inset of Fig. 2 reveals sharp doping profiles. Note, that the Ga signal indicates significant amounts of the order of a few percent at the lower AlInN/GaN interface suggesting the formation of a graded AlGaN interface. The lower left inset shows the surface morphology of the GaN layer at the end of the GaN:Ge/AlInN:Ge/GaN:Ge stack sequence. For the 10 $\times$ 10 $\mu$m$^2$ area a reasonable rms roughness of 0.47 nm is measured.

X-ray diffraction measurements on the sample series with increasing IBGe molar flow reveal compositional changes in the AlInN layers for IBGe flows $\geq$0.1 sccm ($\geq$0.6 $\mu$mol min$^{-1}$), as shown in Fig. 3. GaN and AlN as well as the shift of the AlInN (0002) reflections are marked in the figure, respectively. The smaller, more frequent oscillations are due to the finite thickness of the AlInN and their presence proves sharp interfaces between AlInN and GaN. They have been used to confirm the thickness of the AlInN layer. The shift of the AlInN diffraction peak towards larger angles marks a significant loss of In, leading to strained layer growth. This may also lead to the formation of the graded AlInGaN interface seen in the SIMS profiles. Such graded interfaces, as well as increasing surface roughness, causes a damping of the thickness oscillations towards the low-angle side of the diffraction diagram. Regarding the polarization charges, the substitution of In with Ga may lead to a still unstrained quaternary Al$_{0.82}$Ga$_{0.18}$In$_{1-x}$N$_{0.18}$N layer. One would then expect an ever higher spontaneous polarization which would increase the band bending. Compensation of the stronger band bending would require even higher n-type doping levels in the AlInN layers.

Indium desorption from InGaN and InAlN layers due to the presence of hydrogen in the gas phase is commonly known for In containing nitrides.19-22 When similar experiments were done with hydrogen-diluted GeH$_4$ (10% in gas mixture) dopant source we noticed an even more dramatic loss of In. However, AlInN layers doped with a SiH$_4$/H$_2$ mixture (100 ppm SiH$_4$) at a SiH$_4$/TMAI ratio of 10$^{-3}$ exhibited virtually no change in composition (not shown), although the amount of hydrogen introduced in the reactor chamber was similar. We conclude that hydrogen released during pyrolysis of the precursors reduces the In content of the AlInN. A size effect due to Ge atoms (similar size as Ga) can also have an effect on the In content but the change of the In content by a few percent is hardly explained by Ge doping levels of around 10$^{20}$ cm$^{-3}$. Another cause may exist by surface segregation of Ge atoms hindering In atoms to attach to the surface during growth. Through the reduction of the AlInN growth temperature by around 30 °C this precursor-related In loss is compensated and single AlInN layers with unchanged nominal In content with regard to undoped AlInN reference samples have been obtained at an IBGe input molar flow of 0.6 $\mu$mol min$^{-1}$.

In the following, we compare Si- and Ge-doping of GaN/AlInN stacks with regard to current transport across the GaN/AlInN interface. For that purpose, $I$–$V$ curves were recorded from mesa structures with top-side contacts on the mesa surface and bottom contacts on the n-doped GaN buffer below the stack as shown in the insets of Fig. 4(b). In a first series we varied the type of dopant (Si or Ge) in AlInN and GaN while in a second series the number of layer pairs was changed (1, 3, and 10 pairs). Figure 4(a) shows $I$–$V$ curves of samples where the flow of hydrogen-diluted silane (100 ppm in gas mixture) during growth of AlInN was increased from 0 nmol min$^{-1}$ to 200 nmol min$^{-1}$ while surrounding GaN layers were steadily doped at [Si]$\sim$1.1 $\times$ 10$^{19}$ cm$^{-3}$.
The current–voltage measurements on single GaN:Si/AlInN:Si/GaAs stacks exhibit a linear current–voltage dependence with a total series resistance (including setup resistance) of 10 Ω. The samples with undoped and lowly doped AlInN are very resistive (> 1 kΩ) and exhibit nonlinear I–V curves. While the resistivity across the AlInN/GaN stack reduces significantly with increasing doping levels, even at the optimum SiH₄ flow of 0.1 μmol min⁻¹ the nonlinear trend around 0 V bias evidently proves non-ohmic characteristics which is assigned to residual interface charges between AlInN and GaN. At SiH₄-flows of 0.2 μmol min⁻¹, the resistivity and nonlinearity eventually increase again which hints to a self-compensation mechanism as structural degradation is not revealed by X-ray diffraction analysis. The doping series is concluded with a sample where the free electron concentration in the surrounding GaN layers is increased to [n] = 4 × 10¹⁹ cm⁻³ now using Ge as the dopant. An ohmic I–V characteristic, similar to the reference GaN:Si-only sample, is observed. All these trends are in qualitative agreement with our initial simulations. Please note that the presence of a Ge-related memory effect may lead to auto-doping of the AlInN layer when grown successively after a GaN:Ge layer. In Fig. 4(b), I–V curves of the second series consisting of 1, 3, and 10 pairs of AlInN:Ge/GaN:Ge layers are shown. Here, large 1 × 1 mm² mesas were processed for the I–V measurements. The free electron concentration level in GaN:Ge is around [n] = 2 × 10¹⁹ cm⁻³ according to calibration samples while the molar flow of IBGe within the AlInN was set to 0.32 μmol min⁻¹ (0.4% IBGe/TMIn + TMAI) where the structural quality of the AlInN layer is still preserved. All stacks show ohmic characteristics and low series resistances. While an exact value of the Ge concentration in AlInN remains to be determined, the free electron concentration in AlInN must reach levels above 10¹⁵ cm⁻³ according to our simulation in order to eliminate the upper barrier in the CB.

There is a non-monotonic increase of the series resistance with the number of layer pairs in this series but for 1 and 3 AlInN/GaN layers in the stack the setup-related resistance dominates the measurement. For quantitative evaluation of the series resistance, TLM measurements were applied to a 10-pair Ge-doped AlInN/GaN stack with two different current transport schemes [Fig. 4(c)]. In a lateral transport scheme, 100 × 200 μm² contacts with a set of increasing spacings were deposited on the surface, while for the vertical transport scheme, the semiconductor material between the contacts was etched down to the n-doped GaN buffer layer. These two TLM measurement configurations allow us to separate the resistivity of the stack structure (as a contact resistance value) from the contact resistivity value of the metal/semiconductor contacts. For the lateral transport, the contact resistance as calculated from the TLM I–V data was assumed to be dominated by the metal/semiconductor contact resistance. TLM analysis of the second configuration (with mesa etching) yields a contact resistance that is the sum of both metal/semiconductor contacts and resistance of the stack structure. With the specific contact resistance of ρc ∼ 3 × 10⁻⁶ Ω cm² a value of 0.15 × 10⁻⁶ Ω cm² per AlInN/GaN interface is determined, about two orders of magnitude lower than the contact resistance as deduced from the data given in Ref. 14.

Upon increasing the number of layer pairs to 30, as in typical application scenarios such as vertical optical micro-cavities, we note a significant loss of optical transparency in such Ge-doped structures which requires further optimization (will be published elsewhere).
Fig. 5. (Color online) Series resistance of a 10-fold AlInN:Ge/GaN:Ge layer stack between 77 and 400 K sample temperature as obtained from $I-V$ curves shown in the inset. Down to 77 K ohmic characteristics of the transport through the stack.

Temperature-dependent $I-V$ curves were recorded in a liquid-nitrogen cooled cryostat over a temperature range of 77–400 K in 20 K steps. Electrical connections were wire-bonded onto respective bond pads on the sample enabling vertical current transport measurements in four-point-probe geometry. As Fig. 5 shows only a weak temperature dependence with a 10% decreasing resistance towards lower temperatures is found. This is similar to metallic behavior and indicates the existence of a degenerate electron gas in the stack.

The impact of n-type doping with either Si or Ge during MOVPE of periodic AlInN/GaN stack structures has been investigated in terms of structural and electrical characteristics. A donor concentration well above $1 \times 10^{19}$ cm$^{-3}$ in both AlInN and GaN is advantageous to reduce polarization charge induced barriers at AlInN/GaN interfaces. According to our experiments, AlInN cannot be doped to yield such high free electron concentration due to a self-compensation effect in the case of Si and due to structural degradation in the case of Ge. Free electron concentrations of $[n] > 4 \times 10^{19}$ cm$^{-3}$ in the GaN layers which are easily achievable. Using Ge doping, we find a significant reduction of the series resistance of AlInN/GaN layers with ohmic current transport characteristics even at cryogenic temperatures (77 K). Therefore, Ge doping is favorable for periodic AlInN/GaN stack structures with many such interfaces. As an example, we have demonstrated 10xAlInN/GaN periodic layer stacks with specific contact resistances of $1.5 \times 10^{-7}$ Ω cm$^2$ per interface.

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