

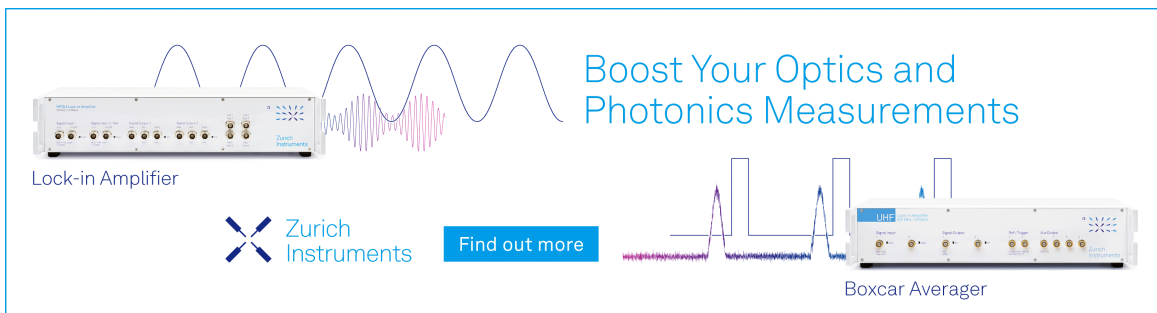
RESEARCH ARTICLE | APRIL 11 2023

High p-conductivity in AlGa_N enabled by polarization field engineering

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*Appl. Phys. Lett.* 122, 152105 (2023)<https://doi.org/10.1063/5.0143427>

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Cite as: Appl. Phys. Lett. **122**, 152105 (2023); doi: [10.1063/5.0143427](https://doi.org/10.1063/5.0143427)

Submitted: 23 January 2023 · Accepted: 29 March 2023 ·

Published Online: 11 April 2023



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ABSTRACT

High p-conductivity ($0.7 \Omega^{-1} \text{ cm}^{-1}$) was achieved in high-Al content AlGa_N via Mg doping and compositional grading. A clear transition between the valence band and impurity band conduction mechanisms was observed. The transition temperature depended strongly on the compositional gradient and to some degree on the Mg doping level. A model is proposed to explain the role of the polarization field in enhancing the conductivity in Mg-doped graded AlGa_N films and the transition between the two conduction types. This study offers a viable path to technologically useful p-conductivity in AlGa_N.

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The high degree of ionicity of the Al(Ga)-N bond and the non-centrosymmetric nature of the AlGa_N wurtzite crystal structure lead to a dipole moment pointing from the N atom toward the metal (Al or Ga) atom.¹ The magnitude of this dipole in AlN is about threefold of that in GaN. Therefore, a linear grading in Al composition along the thickness in a c-oriented AlGa_N film leads to a non-vanishing divergence of polarization along the growth axis, resulting in a bound volumetric charge.¹ The sign of this charge is determined by the polar orientation of the film and the direction of grading. A metal-polar, compositionally graded AlGa_N layer grown with decreasing Al composition (increasing Ga composition) leads to a net negative volumetric polarization charge. Although this charge is bound¹ and does not contribute to the electrical conductivity directly, interestingly, Mg doping of such graded AlGa_N layers has been shown to exhibit a significant improvement in p-type conductivity as compared to bulk Mg-doped AlGa_N films.^{2–6} p-type, compositionally graded AlGa_N films have, therefore, garnered significant interest as hole injection layers in UV lasers and light-emitting diodes and as hole collection layers in photodetectors.^{3,7} However, the mechanism behind the improvement in conductivity remains controversial. Of particular interest is the observation of reduced thermal dependence for conduction in the Mg-doped graded layers,^{2–6} despite the large expected Mg ionization energy of 260–780 meV for AlGa_N.⁸ This observation has

attracted several questions regarding the mechanism for hole generation and hole conduction in Mg-doped, graded AlGa_N structures.

The existing model proposes that the built-in electric field produced due to the volumetric polarization charge (polarization field) in the graded layer is large enough to generate carriers from the available sources of charge such that a three-dimensional (3D) mobile hole slab is formed.^{1,2} This model predicts the formation of a 3D hole slab even in an unintentionally doped, linearly graded layer. Contrary to this prediction, all the literature reports on Hall-effect studies^{2–5,9} of compositionally graded p-type AlGa_N layers have employed not only intentional Mg doping but also the Mg doping concentrations exceeding several-fold the volumetric polarization charge. It was speculated that the need for Mg doping to serve as the necessary source of holes in the polarization-graded layers arises due to the presence of deep-level trap-states that localize holes,^{2,10} although no evidence of such trap states was provided. It was proposed that if the graded layer is doped with Mg, the polarization field assists the ionization of Mg acceptors such that the generation of free holes from the field-ionized Mg acceptors screens the polarization field by neutralizing the fixed negative polarization charges.² In such a case, the polarization field should vanish as the free hole concentration becomes comparable to the volumetric polarization charge and the ionization of the remaining Mg atoms would rely on thermal activation. Based on this reasoning,

the need for Mg doping concentrations far exceeding the volumetric polarization charge cannot be understood. A study with Mg doping concentrations comparable to the volumetric polarization charge is expected to shed further light on the involved conduction mechanism.

Recently, we proposed an alternate mechanism leading to conductivity improvement in Mg-doped, graded AlGaN layers where it was hypothesized that the compositional-grading significantly enhances the probability of phonon-assisted transitions (hopping conduction) between the Mg atoms.¹¹ This conclusion was based on the temperature-dependent resistivity and Hall coefficient measurements that revealed a two-band transport mechanism where the free hole conduction in the valence band was favored at high temperatures and impurity hopping conduction dominated at room and low temperatures. The hopping conduction is associated with the overlap of the atomic wavefunctions of initial and final atomic configurations¹² and is assisted by the carrier-phonon interaction.¹³ In elemental semiconductors, such as Si and Ge, this interaction occurs via the deformation potential. In piezoelectric semiconductors, such as GaAs and CdS, this interaction is dominated by the piezoelectric phonons due to their long-range nature.^{13,14} While a similar behavior can be expected for AlGaN, the impact of the polarization field in the compositionally graded AlGaN films on the acceptor wavefunction and the carrier-phonon interaction needs a comprehensive investigation. In this Letter, we propose a model to understand this mechanism and validate it by performing a set of experiments to study the impact of composition gradient and Mg doping on the electrical characteristics of Mg-doped, graded AlGaN epilayers.

The compositionally graded AlGaN structures were grown in a vertical, low-pressure, rf-heated, cold-walled metalorganic chemical vapor deposition (MOCVD) reactor. Trimethylaluminum (TMA), triethylgallium (TEG), and ammonia (NH₃) were used as aluminum, gallium, and nitrogen precursors, respectively, while H₂ was used as the carrier gas, and Cp₂Mg was used as the Mg source. Three sets of compositionally graded AlGaN samples were investigated as shown in Fig. 1. It is noted that the direction of grading governs the sign of the volumetric polarization charge. In this Letter, we focus on the gradient direction from high to low Al mole fractions along the growth direction corresponding to a negative sign of the polarization charge. The sample in the first set [Fig. 1(a)] was intended for the largest composition gradient and was grown on a 1-in. single-crystal AlN substrate at a temperature and pressure of 1355 K and 20 Torr. The composition was graded from AlN to Al_{0.36}Ga_{0.64}N over a thickness of 70 nm (gradient of 0.9% nm⁻¹). The Mg doping concentration of 5×10^{18} cm⁻³

was introduced during the graded AlGaN growth. Next, a set of two samples was grown on the c-plane sapphire substrate at a Mg doping level of 5×10^{18} cm⁻³ [Fig. 1(b)]. The start and end compositions of these AlGaN layers were 40% and 10% of Al, respectively. The grading was realized over two thicknesses, 100 and 200 nm, corresponding to gradients of 0.30 and 0.15% nm⁻¹, respectively. In the third set of three samples [Fig. 1(c)], 200-nm-thick samples grown on sapphire with a constant compositional gradient of 0.15% nm⁻¹ were doped with Mg in the range of 2×10^{18} to 10^{19} cm⁻³. Compositional grading was characterized (not shown) using secondary ion mass spectroscopy (SIMS) and x-ray diffraction (XRD).

For Hall measurements, e-beam-evaporated Ni/Au (20/40 nm) contacts in the van der Pauw geometry were used on 1×1 cm² samples. The contacts were annealed at 875 K for 10 min under O₂ ambient for Ohmic contact formation. Temperature-dependent conductivity and Hall measurements were performed in a temperature range of 200–700 K using a Lake Shore 8400 series AC/DC Hall measurement system. For an accurate determination of the Hall coefficients, a 1.18 T AC magnetic field with a lock-in amplifier was used.

Figure 2 shows the schematic of the proposed model. For any constant composition AlGaN film, at relatively low Mg doping concentrations ($< 5 \times 10^{18}$ cm⁻³), the Mg dopants are isolated and non-interacting (Fig. 2, top-left). In this case, the only viable route for conduction is via ionization of Mg impurities followed by the hole transport in the valence band (Fig. 3, left). Grading of AlGaN composition generates a polarization field in the growing film. Following the approach by Anderson,¹⁵ we hypothesize an insulator-to-metallic transition via impurity band transport (Fig. 2, top-right) occurring through the Anderson transition at relatively low Mg concentrations (5×10^{18} cm⁻³). This is in contrast to our recent report on Mott-type transition in constant composition AlGaN films requiring heavy or degenerate doping.¹⁶ The Anderson transition requires a distribution of impurity (Mg) ionization energies in the material (graded AlGaN). The distribution does not occur due to the orbital overlap (the Mg atoms may be separated by distances much larger than the Bohr radius associated with an ionized Mg ion and a bound hole) as in the Mott transition¹⁶ but due to the dependence of ionization energy on the alloy composition in the graded region.¹⁷ The transition does demand a minimum overlap integral associated with the orbitals of neighboring Mg defect levels. We hypothesize that the polarization field acts as a perturbation to the electronic state of Mg, thereby spreading its wavefunction. The carrier transport then occurs through the delocalization of the carrier wavefunction via a cluster of resonant states.^{18,19}

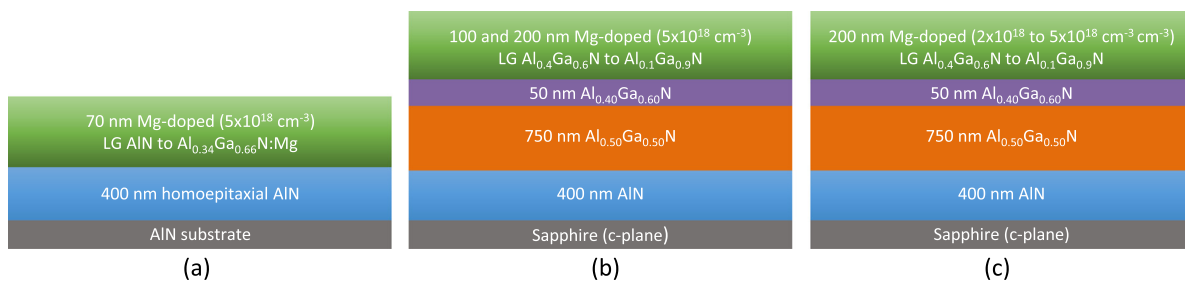


FIG. 1. Schematic showing the epitaxial stack of the three sets of linearly graded (LG) AlGaN samples. Note: In all samples, the composition was graded from high to low Al

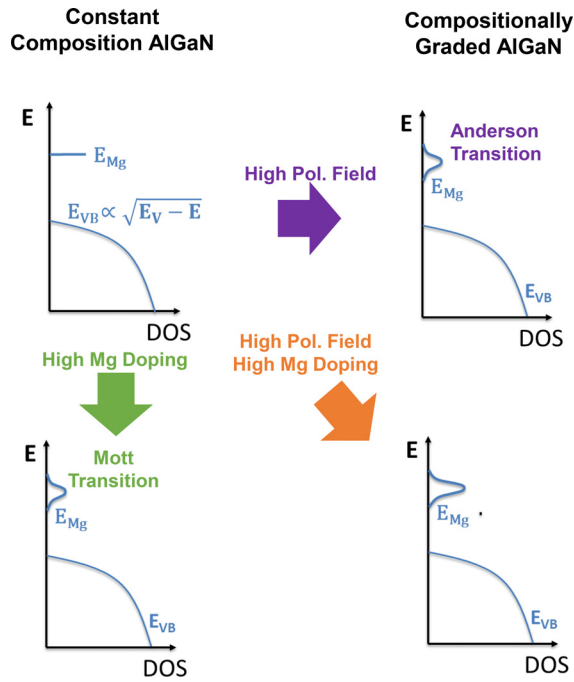


FIG. 2. A schematic of the density of states showing the evolution of the Mg acceptor level to a band on the application of polarization field via Anderson transition or by heavy doping via Mott transition.

The resulting conduction, i.e., impurity band conduction, is, therefore, more likely with increasing overlap integral i.e., larger polarization field in the graded AlGaIn layer (Fig. 2, top-right) or higher Mg doping concentration (Fig. 2, bottom-left). Thus, the model suggests that for Mg-doped, graded AlGaIn, the impurity band transport is favored through Anderson transition even at relatively low dopant concentrations ($5 \times 10^{18} \text{ cm}^{-3}$). In contrast, in uniform systems,¹⁶ a heavily doped material ($\sim 5 \times 10^{19} \text{ cm}^{-3}$) is required for impurity band transport via Mott transition (Fig. 2, bottom-left).

Impurity band conduction provides an alternate route for conduction where the carriers can propagate with much-reduced activation energy as compared to the relatively large Mg ionization energy for the valence band transport. Therefore, impurity band conduction

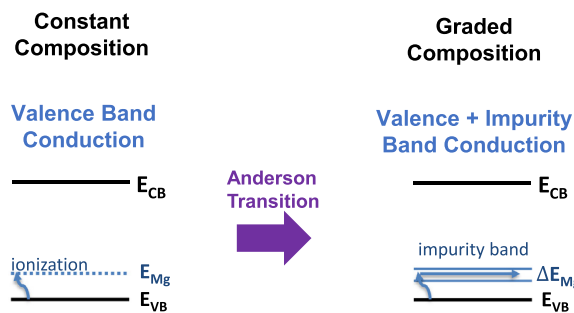


FIG. 3. Simplistic band diagram illustrating the viable conduction routes for non-degenerately doped constant composition and graded composition AlGaIn films.

would dominate at low temperatures where the valence band conduction is limited. Based on the model, we expect that for a graded AlGaIn film, the magnitude of the polarization field plays a more significant role than the Mg doping concentration itself and the impurity band formation is expected to scale with the composition gradient. Therefore, for a graded AlGaIn film grown with a relatively steep composition gradient, we predict the dominance of impurity band conduction at much lower Mg doping concentrations than for the constant composition films.

To validate this model, we first start with a Mg-doped graded AlGaIn sample grown with a relatively low Mg doping concentration of $5 \times 10^{18} \text{ cm}^{-3}$ and a large composition gradient of $0.90\% \text{ nm}^{-1}$ where the AlGaIn composition was varied from AlN to $\text{Al}_{0.36}\text{Ga}_{0.64}\text{N}$ over a 70 nm thickness. An estimate of the volumetric polarization charge for this graded AlGaIn structure is $\sim 5 \times 10^{18} \text{ cm}^{-3}$, a value comparable to the Mg doping concentration. Figure 4(a) shows the temperature-dependent conductivity (ρ) data obtained for this sample. The sample exhibited a rather low thermal dependence of the conductivity with an activation energy of only $\sim 24 \text{ meV}$.

Next, Hall-effect measurements were performed on this sample. It is noted that the interpretation of Hall-effect data on samples exhibiting impurity band conduction is not straightforward.¹¹ The contributions of Hall coefficients of the valence band (R_H^{VB}) and impurity band conduction (R_H^{IB}) to the combined Hall coefficient (R_H) measured by the Hall system depend on the respective conductivities of the two conduction mechanisms ($\sigma = \frac{1}{\rho} = \sigma_{\text{VB}} + \sigma_{\text{IB}}$),

$$R_H = R_H^{\text{VB}} \left(\frac{\sigma_{\text{VB}}}{\sigma} \right)^2 + R_H^{\text{IB}} \left(\frac{\sigma_{\text{IB}}}{\sigma} \right)^2. \quad (1)$$

Here, R_H^{VB} is positive as the free hole conduction with a positive Hall scattering factor generates a positive Hall voltage. Following the discussion by Emin^{20,21} and Holstein,²² the sign of the Hall scattering factor in impurity hopping conduction depends not only on the number of hopping sites but also the nature and relative orientations of the local orbitals between which the carrier hops. A negative Hall scattering factor has been reported for acceptor-related impurity band conduction mechanism in p-type GaN,²³ graded AlGaIn,¹¹ and SiC.²⁴ The negative Hall scattering factor would result in a negative Hall voltage, and, therefore, R_H^{IB} is negative. Figure 4(b) shows the temperature-dependent Hall coefficient (R_H) data. Interestingly, the measured Hall coefficient exhibited negative values across the entire temperature

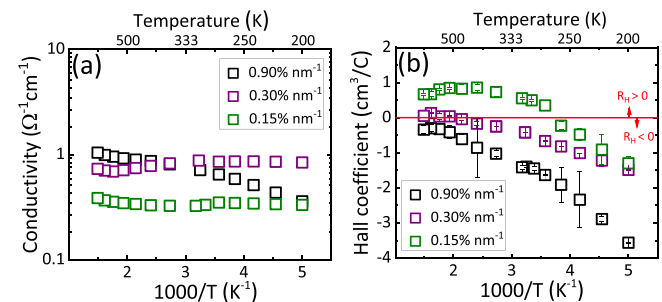


FIG. 4. Temperature-dependent (a) conductivity and (b) Hall coefficient for the Mg-doped ($5 \times 10^{18} \text{ cm}^{-3}$) graded AlGaIn structure with 0.90, 0.30, and $0.15\% \text{ nm}^{-1}$ composition gradients.

range. This implies the dominance of impurity band conduction in the graded AlGa_{0.36}N sample despite the relatively low Mg doping concentration, as predicted by the model.

It is worth noting that constant composition AlGa_{0.36}N films grown with any composition in the range over which the graded AlGa_{0.36}N film was grown (AlN to Al_{0.36}Ga_{0.64}N) are typically insulating for Mg doping concentrations around $5 \times 10^{18} \text{ cm}^{-3}$. The impurity band conduction characterized by a significantly reduced activation energy of tens of meV as compared to the Mg ionization energy of hundreds of meV allows for a room temperature conductivity of $0.7 \Omega^{-1} \text{ cm}^{-1}$ in the graded AlGa_{0.36}N sample despite a relatively high average Al-mole fraction of 68%. This is a significant improvement in conductivity as compared to $0.01\text{--}0.1 \Omega^{-1} \text{ cm}^{-1}$ reported for constant composition Al_{0.7}Ga_{0.3}N films doped with much higher Mg concentrations of $3\text{--}5 \times 10^{19} \text{ cm}^{-3}$.^{16,25,26}

To understand the impact of the polarization field on the impurity band conduction, graded AlGa_{0.36}N samples [Fig. 1(b)] with reduced composition gradients of 0.30 and $0.15\% \text{ nm}^{-1}$ were investigated while keeping the Mg doping concentration constant at $5 \times 10^{18} \text{ cm}^{-3}$. These gradients correspond to a threefold and a sixfold reduction in the magnitude of the polarization field, respectively, compared to the previous sample. The temperature-dependent conductivity and Hall coefficient data obtained for these samples are shown in Figs. 4(a) and 4(b), respectively. For both samples, the conductivity remained constant across the entire temperature range with the larger gradient ($0.30\% \text{ nm}^{-1}$) sample exhibiting higher conductivity. For this sample, the sign of the Hall coefficient was negative below 500 K and transitioned to positive at higher temperatures. This transition is attributed to an increasing contribution of the valence band transport to the Hall coefficient at higher temperatures [Eq. (1)]. The smaller gradient sample ($0.15\% \text{ nm}^{-1}$) exhibited this transition at a much lower temperature of 280 K, pointing to a reduced contribution of impurity band conduction to the overall transport. This confirms that a smaller gradient, corresponding to a weaker polarization field, leads to a reduction in the impurity band conduction for the same doping level. It is noted that close to the transition temperature, the positive and negative contributions of the valence and impurity band conduction mechanisms cumulatively lead to the generation of very small Hall voltages, $<1 \text{ mV}$. The use of an alternating magnetic field with a lock-in amplifier helps to filter out most error contributions in the Hall setup to reliably measure small Hall voltages in the range of hundreds of μV , which is a challenge in the DC Hall setups.

Next, the impact of the Mg doping level was studied for the smallest composition gradient of $0.15\% \text{ nm}^{-1}$ [Fig. 1(c)]. Figure 5(a) shows the temperature-dependent conductivity data obtained for Mg doping concentrations varying from 2×10^{18} to 10^{19} cm^{-3} . While the low-temperature conductivity was invariant with temperature, the high-temperature behavior varied with the doping level. For a Mg doping level of $5 \times 10^{18} \text{ cm}^{-3}$, the conductivity remained constant across the entire temperature range. For the higher doping of 10^{19} cm^{-3} , the conductivity increased above 320 K with a 50 meV activation energy, suggesting an enhancement in the relative contribution of the valence band conduction. For the lower Mg doping of $2 \times 10^{18} \text{ cm}^{-3}$, the conductivity reduced above 375 K, which is possibly related to a decrease in the preexponent term in the hopping mobility, $\mu_{\text{hop}} = \mu_{\text{hop0}} \left(\frac{T_{\text{hop}}}{T} \right)^{3/2} \exp \left(-\frac{T_{\text{hop}}}{T} \right)$. The conductivity became constant

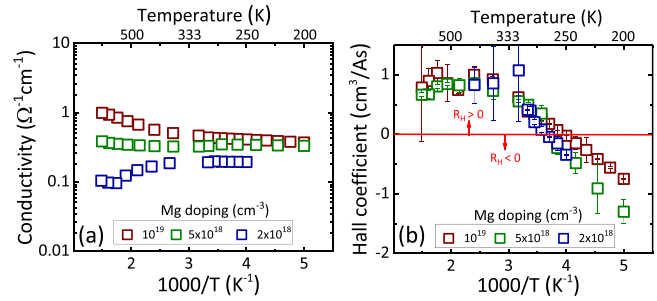


FIG. 5. Temperature-dependent (a) conductivity and (b) Hall coefficients for the Mg-doped graded AlGa_{0.36}N structure with $0.15\% \text{ nm}^{-1}$ composition gradient as a function of Mg doping concentration.

above 550 K, suggesting the enhancement in the relative contribution of the valence band conduction. A decrease in the Mg doping concentration is expected to suppress both conduction mechanisms: the valence band conduction due to a reduction in free hole concentration and the impurity band conduction due to an increase in the inter-Mg distance. These results suggest that for the doping range investigated in this study, a reduction in doping concentration has a stronger impact on the valence band conduction as compared to the impurity band conduction.

Interestingly, the Hall coefficient did not exhibit a significant variation with Mg doping, as seen in Fig. 5(b). Above room temperature, the Hall coefficient was positive for all samples and varied in the range of $0.5\text{--}1.1 \text{ cm}^3/\text{As}$. As the temperature was lowered, the Hall coefficient changed the sign in the temperature range of 250–300 K. The impurity band formation is, therefore, evident even for the lowest doping level of $2 \times 10^{18} \text{ cm}^{-3}$ and the lowest composition gradient of $0.15\% \text{ nm}^{-1}$.

Thus, the different sample sets exhibited salient features that are consistent with the predictions of the proposed model. As was shown in Fig. 4, the first sample with the largest composition gradient and doped with $5 \times 10^{18} \text{ cm}^{-3}$ Mg exhibited impurity-band-dominant conduction for the entire temperature range. In the next sample set (Fig. 4), reduced composition gradients revealed a distinct transition between the impurity band and valence band conduction mechanisms, validating that the impurity band conduction scales with the composition gradient and the induced polarization field. Finally, Fig. 5 shows that the presence of a polarization field plays a more significant role in conduction than the Mg doping concentration.

In summary, we propose a model that describes the role of the polarization field on the conduction in Mg-doped, compositionally graded AlGa_{0.36}N films. It is hypothesized that the polarization field in the graded AlGa_{0.36}N film acts as a perturbation to the electronic state of Mg, which spreads the carrier wavefunction and favors the impurity band transport via the Anderson transition. This hypothesis was validated by a systematic set of experiments involving a variation in the composition gradient and Mg doping levels in the AlGa_{0.36}N layers. It was shown that steeper gradients favor impurity band conduction regardless of the doping level and enable technologically useful p-conductivity in high Al-content AlGa_{0.36}N.

The authors gratefully acknowledge funding in part from AFOSR (Nos. FA9550-17-1-0225, FA9550-19-1-0114, and

FA9550-19-1-0358), NSF (Nos. ECCS-1610992, ECCS-1508854, ECCS-1916800, and ECCS-1653383), and ARO (Nos. W911NF-16-C-0101 and W911NF-22-20171).

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Shashwat Rathkanthiwar: Conceptualization (lead); Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (lead); Visualization (equal); Writing – original draft (lead); Writing – review & editing (equal). **Ramón Collazo:** Conceptualization (equal); Funding acquisition (lead); Project administration (equal); Supervision (lead); Validation (equal); Visualization (lead); Writing – review & editing (equal). **Zlatko Sitar:** Funding acquisition (lead); Project administration (equal); Supervision (lead); Validation (lead); Writing – review & editing (lead). **Pramod Reddy:** Conceptualization (equal); Formal analysis (equal); Visualization (equal); Writing – original draft (supporting). **Baxter Moody:** Formal analysis (supporting); Investigation (supporting); Validation (equal). **Cristyan Quiñones-García:** Investigation (supporting). **Pegah Bagheri:** Data curation (supporting); Investigation (supporting); Software (equal). **Dolar Khachariya:** Investigation (supporting). **Rafael Dalmau:** Investigation (supporting). **Seiji Mita:** Investigation (supporting); Resources (equal). **Ronny Kirste:** Resources (supporting); Validation (supporting).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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