Significant Phonon Drag at Room Temperature in the AlGaN/GaN 2D Electron Gas Revealed by Varying GaN Thickness

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In thermoelectric devices, the Seebeck coefficient is composed of a thermal diffusive component and a drag component, which arises from momentum exchange between charge carriers and lattice phonons. It is however hard to separate these components, which makes it challenging to understand whether drag is useful for thermoelectric power conversion. Here, we present the first study of the effect of phonon confinement on drag in the AlGaN/GaN two-dimensional electron gas by varying the underlying GaN layer thickness. We show that phonon drag does not contribute significantly to the thermoelectric performance in devices with a GaN thickness of ~100 nm, due to a suppression of the phonon mean free path. However, when the thickness is increased to ~1.2 μm, we uncover that 32% of the total Seebeck coefficient at room temperature can be attributed to the drag component. At 50 K, the drag component increases significantly to 88%. Further, by measuring the thermal conductivity in these AlGaN/GaN films, we show that the magnitude of the phonon drag can increase even when the thermal conductivity decreases. This decoupling of thermal conductivity and Seebeck coefficient could enable unprecedented advancements in thermoelectric power conversion.

Keywords: Thermoelectrics, phonon drag, 2D electron gas, electron-phonon interaction
In modern semiconductor devices, the scattering of charge carriers by lattice vibrations, known as phonons, is an important bottleneck in the design of energy-efficient devices.\textsuperscript{[11]} Yet, under a thermal gradient, commonplace in thermoelectric devices, these phonons can generate usable energy when they couple to charge carriers via an enhancement of the Seebeck coefficient ($S$).\textsuperscript{[2–4]} This momentum transfer to the charge carriers via non-equilibrium phonons, known as phonon drag ($S_{\text{ph}}$), supplements the thermal diffusion of charge carriers ($S_d$). Despite the potential gains in thermoelectric efficiency, understanding the contribution of phonon drag to the overall Seebeck coefficient has not received much consideration, largely due to early work which suggested that: (1) $S_{\text{ph}}$ is only significant at low temperatures (below $T \approx 50$ K), where the thermoelectric power conversion efficiency ($zT$) is low;\textsuperscript{[5]} (2) $S_{\text{ph}}$ is small relative to $S$ for degenerate semiconductors;\textsuperscript{[6,7]} which are common thermoelectric materials due to their large $zT$; and (3) an increase in $S_{\text{ph}}$ coincides with a corresponding increase in the thermal conductivity ($k$);\textsuperscript{[8–10]} and thus has little benefit for power generation, since $zT \propto S^2/k$.

Contrary to these observations, recent experiments show that $S_{\text{ph}}$ is almost 34% of the total $S$ at room temperature in degenerate, bulk Si (doping on the order of $10^{19}$ cm$^{-3}$).\textsuperscript{[11]} Further, recent first-principles calculations show that different portions of the phonon mean free path spectrum (MFP, denoted by $\lambda$) contribute to thermal conductivity and phonon drag. Remarkably, this decoupling makes it possible to reduce $k$ while preserving $S_{\text{ph}}$\textsuperscript{[4]} Though a detailed understanding of phonon drag can lead to advancements in solid-state thermoelectric power conversion, even at room temperature, there remain significant gaps in the literature. In particular, experiments which accurately quantify $S_{\text{ph}}$ are yet lacking for low-dimensional systems. Degenerate 2D semiconductor quantum wells often have Seebeck coefficients ($S$) and electronic conductivities ($\sigma$) which are enhanced relative to bulk,\textsuperscript{[12–15]} increasing $zT$. Further improvements in thermoelectric performance necessitate understanding the contribution of $S_{\text{ph}}$.

Previous measurements of $S_{\text{ph}}$ have been done in 2D electron gas systems by measuring the total Seebeck coefficient, theoretically estimating $S_d$, and calculating $S_{\text{ph}} = S - S_d$.\textsuperscript{[16]} However, estimating $S_d$ is difficult to do because it needs precise knowledge of the scattering mechanisms for the charge carriers, in addition to the subband energies of the 2D quantum well. In the simple model by Herring,\textsuperscript{[2]} $S_{\text{ph}} \propto \lambda_{\text{ph}}$, where $\lambda_{\text{ph}}$ is the MFP of the “representative” phonon that participates in drag. Thus, as shown in recent work on bulk Si,\textsuperscript{[11]} we can experimentally determine $S$ and $S_{\text{ph}}$ separately by varying dimensions,\textsuperscript{[17]} since this allows us to directly control the phonon MFP spectrum, and hence $S_{\text{ph}}$. In particular, as the sample
dimension is reduced below a critical value, \( S_{ph} \) disappears meaning that in these samples \( S \approx S_d \).\(^{[11]} \) \( S_{ph} \) in samples with a larger dimension can thus be estimated by subtracting out the Seebeck coefficient of these smaller samples. Because this method does not rely on the estimation of \( S_d \) from theory, it allows for a true extraction of \( S_{ph} \).

In this work, we extend the concept of dimension scaling to extract \( S_{ph} \) in the 2DEG that is formed when a nanometer-thick layer of unintentionally doped aluminum gallium nitride (AlGaN) is deposited on an underlying GaN buffer layer for the first time. This is an appealing heterostructure for use in space environments,\(^{[18]} \) where low-temperature thermoelectric power sources\(^{[19]} \) are necessary. We note here that the \( S_{ph} \) is due to the interaction of 3D phonons in the underlying GaN layer with the electrons at the AlGaN/GaN interface. Both GaN and AlN have relatively high Debye temperatures (600 K and 1150 K),\(^{[20]} \) suggesting that non-equilibrium phonons should be present in these materials below this temperature. This suggests that phonon drag could be observed, even at room temperature in this material system.

Using a combination of experiments and analytical modelling across 50 to 300 K, we experimentally separate out \( S_{ph} \) by comparing the thermoelectric properties of devices with various GaN thicknesses (100 nm to \(~1.2 \mu m\)). Further evidence for phonon drag is provided by examining the gate voltage dependency of the Seebeck coefficient. Our findings show that while \( S_{ph} \) is \(~32\%\) of the total \( S \) at room temperature in a thick \(~1.2 \mu m\) device, \( S_{ph} \) is almost completely suppressed when \( t_{GaN} \) is reduced to \(~100 \) nm. By measuring the thermal conductivity in these samples, we show that the magnitude of the phonon drag can increase even when the thermal conductivity is decreasing. This decoupling of \( k \) and \( S_{ph} \) could enable significant advancements in thermoelectric power conversion. These first measurements of the thermoelectric properties of the AlGaN/GaN 2DEG in the low temperature range, are an important step toward understanding drag in such low-dimensional systems.

Experimental samples were fabricated via metal organic chemical vapor deposition (MOCVD) on an Si (111) wafer. A buffer stack consisting of Al\(_{1-x}\)Ga\(_{x}\)N was grown, followed by a GaN layer whose thickness was chosen to tune phonon confinement. Two variants were grown: (i) a “thin” sample with \( t_{GaN} \approx 100 \) nm and (ii) a “thick” sample with \( t_{GaN} \approx 1.2 \mu m \). Once the GaN layer was deposited, the 2DEG was formed by depositing 1 nm/30 nm/3 nm AlN/Al\(_{0.25}\)Ga\(_{0.75}\)N/GaN on top of the GaN layer, a standard stack for achieving high electron mobility (1500-2000 cm\(^2\)V\(^{-1}\)s\(^{-1}\) at room temperature).\(^{[21]} \) The 2DEG, which forms in GaN at the AlGaN/GaN interface, has a nominal sheet density, \( n_{2D} \approx 10^{13} \) cm\(^{-2}\) and a characteristic quantum well width of \(~5 \) nm.\(^{[14]} \) The buffer layers (Al\(_{1-x}\)Ga\(_x\)N, 0 \( \leq x \leq \) 1) and the GaN layer are
unintentionally doped below $10^{16}$ cm$^{-3}$, ensuring that the measured Seebeck coefficient is exclusively from the 2DEG.$^{[22]}$

Since the 2DEG is lateral, extraction of its thermoelectric properties ($S$ and $k_{\text{GaN}}$) is facilitated by setting up an in-plane temperature gradient. We accomplished this by etching the Si from the backside to create suspended AlGaN/GaN diaphragms, as depicted in Figures 1a and 1b. A 2DEG mesa was defined by etching the AlGaN on top of the membrane, except in a rectangular strip across which we extracted the Seebeck coefficient. Platinum (Pt) heater electrodes were deposited on top of a $\sim$47 nm atomic layer deposited (ALD) alumina layer to provide electrical isolation from the 2DEG (see Supplementary Note 1) and to create an in-plane temperature gradient across the 2DEG mesa. The mesa has a gate electrode (Au) on top of the alumina to modulate the charge density in the 2DEG, as seen in Figures 1a and 1c.

Figure 1 | Measurement platform to probe phonon drag. (a) Schematic of suspended device to measure Seebeck coefficient, showing the heater metal, the AlGaN/GaN mesa, and the gate. (b) Cross-sectional SEM image of the suspended region, showing Si, the buffer and the GaN layer. This image is for the thick GaN sample, with $t_{\text{GaN}} \approx 1.2$ μm. (c) 2D schematic of the suspended mesa region, showing the drag and diffusive components of the Seebeck voltage. The phonon wave vector is marked by the symbol $Q$. (d) Flowchart showing the numerical procedure to extract the phonon drag component of the Seebeck coefficient, $S_{\text{ph}}$.

Upon application of a temperature gradient via the Pt heater, a Seebeck voltage is measured across the mesa due to the thermal diffusion of the 2DEG electrons ($V_d$) and the drag imparted to them by phonons in the GaN layer ($V_{\text{ph}}$), as seen in Figure 1c. Using the heater as
a thermometer, we extracted the Seebeck coefficient from the voltage across the 2DEG mesa, after accounting for the thermal losses in the alumina layer and the various interfaces (see Supplementary Note 2). A similar structure with two metal electrodes (heater and sensor) on the suspended AlGaN/GaN diaphragm was used to extract the thermal conductivity of the GaN and the underlying buffer layers. Further details of the measurement process can be found in Supplementary Note 2 and our former work.\textsuperscript{[14]} The flowchart in Figure 1d details our numerical procedure to extract $S_{\text{ph}}$. Measurements of the 2DEG sheet density, $n_s$ and mobility, $\mu$ were taken and compared with an analytical model to obtain the scattering times, $\tau(E)$ for the 2DEG as a function of energy. The obtained $\tau(E)$ is used to calculate the diffusive component of the Seebeck coefficient, $S_d$. The thermal conductivity measurements are used to extract the MFP spectrum in the GaN layer, which is combined with $\tau(E)$ to calculate $S_{\text{ph}}$. This modeled $S_{\text{ph}}$, along with the calculated $S_d$, can be compared with the experimental values of the Seebeck coefficient for both the thick and thin GaN samples to shed light on the relative contribution of $S_{\text{ph}}$.

**Figure 2 | Measurements for un-gated devices.** (a) Temperature dependent sheet density the thick and thin GaN sample. The dotted lines show the simulated values obtained from a commercial solver. (b,c) Mobility for the thick and thin GaN sample, with the dotted lines showing the simulated components. (d) Measured Seebeck coefficient. The dotted lines show the calculated diffusive components, which are similar for the thick and thin GaN samples. (e) Measured and calculated thermal conductivities for the thick and thin GaN samples. (f) Simulated values of
the phonon drag component of the Seebeck coefficient obtained by sweeping the effective thickness of the GaN layer. The red markers show the estimated drag component for the thick GaN sample extracted from the experimental data. A clear suppression of phonon drag is observed with smaller GaN layer thickness.

We first discuss the measurements of these parameters under no gate bias. Figure 2a shows measurements of $n_s$ for the thick and thin GaN sample, extracted via Hall effect and Van der Pauw measurements. We obtain a roughly constant $n_{2D} \approx 10^{13}$ cm$^{-2}$ from 50 K to 300 K, consistent with the weak dependence on temperature of the piezoelectric constants of both AlN and GaN.[23] The thin and thick GaN samples have a similar charge density,[14] verified using a commercially available Schrödinger-Poisson solver[24] as seen in Figure 2a. For simplicity, we set $n_{2D} = 10^{13}$ cm$^{-2}$ for both the thin and thick GaN samples in the models for the thermoelectric transport properties. Using the expression for the 2D density of states, assuming that all the sheet density is from a single subband, $g_{2D} = \frac{m^*}{\pi\hbar^2}$, we get $E_F - E_1 \approx 110$ meV, where $E_F$ is the 2DEG Fermi level and $E_1$ denotes the energy at the bottom of the first subband. This is consistent with the energies obtained from the solver (Supporting Note 3), and indicates that only the bottom subband contributes significantly to charge density. For the rest of this work, only this subband is considered in the calculation of the Seebeck coefficient, because the contribution to $S$ from each subband is weighted by the charge density in it.[25]

Next, we turn to measurements of the 2DEG mobility, plotted in Figure 2b and Figure 2c for the thick and thin GaN samples, respectively. The dotted lines show the calculated contributions to the mobility from scattering mechanisms that are dominant in AlGaN/GaN 2DEGs.[26] Other scattering mechanisms (e.g. dislocation, ionized impurity and piezoelectric scattering) are neglected. Rigorous justification of this approximation is found in Supplementary Note 2. For both thick and thin GaN, polar optical phonon (POP) scattering is the dominant scattering mechanism at room temperature, due to the large optical phonon energy ($\hbar\omega_{OP} = 91.2$ meV),[27] and the polar nature[28] of the GaN wurtzite crystal. As the optical phonon population decreases exponentially at lower temperatures, electrons in the lower subband scatter against the AlGaN/GaN interface (which we denote as roughness scattering). In order to calculate the roughness scattering, we set the average displacement of the interface, $\Delta$, to be 1 and 2 nm for the thick and thin GaN sample respectively (an AFM image of the sample surface can be found in Supplementary Figure S4). The good agreement between the model and experimental data allows us to extract the scattering time, $\tau(E)$ for the electrons in the bottom subband of the 2DEG.
Using these scattering times, we can calculate the diffusive component of the Seebeck coefficient for the bottom subband, \( S_d \), as:

\[
S_d = -\frac{1}{eT} \int \frac{f_0(E) \partial f_0(E)}{\partial E} \frac{(E - E_F - E_1) \tau(E) dE}{\int E \frac{\partial f_0(E)}{\partial E} \tau(E) dE},
\]

where \( f_0(E) \) is the Fermi function, and \( e \) is the magnitude of the electronic charge. These are plotted against the experimental data for the magnitude of the Seebeck coefficient (the actual sign is negative) in Figure 2d. The theoretical curves deviate slightly from a linear dependence on temperature, typical for a degenerate semiconductor with \( \tau(E) \propto E^\alpha \), where \( \alpha \) is a scattering exponent.\(^{[25]}\) This deviation is due to POP scattering, which forbids electrons with energies smaller than \( h\omega_{\text{OP}} \) from emitting optical phonons.\(^{[29]}\) The slight difference in the calculated values of \( S_d \) for the thick and thin GaN sample is attributed to differences in roughness scattering. We observe that the Seebeck coefficient for the thin GaN sample agrees well with the calculated \( S_d \), however this model cannot describe the thick GaN sample (Figure 2d). In addition, the magnitude of the Seebeck coefficient in the thick GaN sample exhibits a prominent upturn at low temperatures, hinting at phonon drag.\(^{[16]}\)

In our device, 3D phonons, represented by the wave vector \( \mathbf{Q} = (q, q_z) \), which represent the in-plane (of the 2DEG) and out-of-plane component, scatter with 2D electrons in the bottom subband, giving rise to \( S_{\text{ph}} \). To calculate this drag, we follow the approach introduced by Cantrell and Butcher\(^{[3]}\) and later modified by Smith.\(^{[30,31]}\) We additionally add a correction to include the phonon scattering time (\( \tau_{\text{ph}} \)) as a function of the phonon wave vector explicitly:

\[
S_{\text{ph}} = -\frac{(2m^*)^2 \nu_{\text{av}}^2}{4(2\pi)^3 k_B T^2 n_{2D} \rho} \int_0^\infty dq \int_{-\infty}^\infty dq_z \frac{\Xi^2(Q)q^2Q^2|l(q_z)|^2 G(Q)\tau_{\text{ph}}(Q)}{S^2(q, T) \sinh\left(\frac{h\omega_Q}{2k_BT}\right)},
\]

In Equation 2, \( \nu_{\text{av}} \) is the average phonon velocity over the different modes, \( k_B \) is the Boltzmann constant, \( m^* \) is the electron effective mass and \( \rho \) is the density of GaN. The values of the parameters used for our calculations are in Supplementary Table S1. The phonon frequency, \( \omega_Q \) is approximated as \( \nu_{\text{av}} \sqrt{q^2 + q_z^2} \) assuming a linear dispersion. The term \( l(q_z) = \int \psi(z)^2 e^{i\mathbf{q} \cdot \mathbf{z}} \) describes the electron-phonon momentum conservation in the \( z \) direction, where \( \psi(z) \) is the wave function of the electrons in the bottom subband. \( \Xi(Q) \) represents the strength of the electron-phonon coupling. The terms \( S(q, T) \) and \( G(Q) \) represent a screening function for the electrons and an energy integral, respectively (the detailed explanation of these terms is discussed in Supplementary Note 4). Of particular interest to this work is \( \tau_{\text{ph}}(Q) \), the term
representing the relaxation time of the phonons. This term describes the scaling dependence of $S_{\text{ph}}$ on sample thickness, because $\tau_{\text{ph}}(Q) \propto t_{\text{GaN}}$ due to boundary scattering.

To calculate $\tau_{\text{ph}}(Q)$ accurately, we measured the thermal conductivity, $k$, of the suspended diaphragms, presented in Figure 2e. Since our suspended film is a composite consisting of an AlN layer, Al$_x$Ga$_{1-x}$N transition layers and a GaN layer, the overall thermal conductivity must be estimated from a weighted average of the thermal conductivities and thicknesses of individual layers. For each layer, we used a Boltzmann Transport Equation (BTE) model to quantify its thermal conductivity. The dotted lines in Figure 2e show the modeled $k$ for the entire stack, taking into account phonon-phonon, dislocation, alloy and boundary scattering (details in Supplementary Note 5). The alloy and dislocation scattering terms are challenging to estimate analytically,$^{[32,33]}$ which could explain the disagreement between the model and the data at the lower temperatures. Yet, this model will suffice to explain the observed trends in the phonon drag behavior. Assuming that only the phonons in the GaN layer contribute to drag, the modelled $\tau_{\text{ph}}$ for this layer is combined with Equation 2 to calculate $S_{\text{ph}}$.

The modeled $S_{\text{ph}}$ is plotted in Figure 2f for a range of $t_{\text{GaN}}$ values. It is seen that $S_{\text{ph}}$ for the thin GaN is between 4 and 8 $\mu$VK$^{-1}$ across all $T$, significantly less than the measured 40 to 80 $\mu$VK$^{-1}$ (Figure 2d), supporting the conclusion that $S \approx S_d$. The nearly constant $S_{\text{ph}}$ can be attributed to $k_{\text{GaN}}$ being limited by boundary scattering across the entire temperature range. $S_{\text{ph}}$ in the thick GaN film was estimated by subtracting a linear fit (including the origin) of the measured Seebeck coefficient in the thin GaN sample from the measured Seebeck coefficient of the thick GaN sample. We have used a linear fit to the origin of the thin GaN Seebeck coefficient to avoid overestimating the diffusive component of the Seebeck coefficient. This is because the measured Seebeck coefficient values of the thin GaN sample still includes a small phonon drag component, which is visible as a slight upturn at the lower temperatures (blue triangles in Figure 2d). The estimate of $S_{\text{ph}}$ for the thick GaN sample after subtraction from the linear fit is plotted in Figure 2f (red markers). The shaded region shows the calculated $S_{\text{ph}}$ for various effective GaN thicknesses ($t_{\text{GaN}}$) from 1 to 3 $\mu$m using Equation 2. We have swept the GaN thickness in the model since it under-predicts $S_{\text{ph}}$ if we use the actual thickness (1.2 $\mu$m). This inaccuracy may arise from the simple model for the thermal conductivity and phonon drag used here, in addition the 2DEG quantum well thickness, which is challenging to obtain experimentally. The data and model agree within the swept thickness range, and shows that ~32% of the total $S$ at room temperature can be attributed to drag, increasing to almost 88% of
$S$ at 50 K. The inverse temperature dependence of $S_{\text{ph}}$ is reflective of phonon-phonon scattering, from which the phonon MFP scales as $T^{-1}$.

The measurements of the Seebeck coefficient and the thermal conductivity in Figures 2e and 2f also suggest that the phonon drag continues to increase even when the thermal conductivity starts decreasing (below $\sim 90$ K), providing experimental demonstration that these two parameters can be decoupled. This is in agreement with previous theoretical work suggesting that these two parameters can be decoupled to increase $zT$.$^{[34,35]}$

The application of a gate voltage, $V_G$, can tune the thermoelectric power factor ($S^2\sigma$) without changing $k$, which can further optimize $zT$.\textsuperscript{[36,37]} While the effect of $V_G$ on $S_d$ is well known, only a few studies have attempted to quantify its effect on drag.\textsuperscript{[16,38,39]} In particular, application of $V_G$ tunes the quantum well width and 2DEG charge density ($n_{2D}$), simultaneously. $S_{\text{ph}}$ is inversely proportional to $n_{2D}$ giving it a strong dependency on this parameter, as seen in Equation 2. Quantum well width affects $S_{\text{ph}}$ through $I(q_z)$ which is strongly dependent on wave function shape $\psi(z)$. A more tightly confined wave function in real space (which corresponds to larger $n_{2D}$) is broader in Fourier space, increasing $I(q_z)$. These two effects compete against each other resulting in a complex gate voltage dependency.

![Figure 3](image-url)

**Figure 3**] Measurements with a gate bias. (a) Modulation of the sheet density in the 2DEG with applied gate bias at 300 K. The inset shows the simulated wave function in the bottom subband of the 2DEG for three different sheet densities. The coordinate $z=0$ corresponds to the AlGaN/GaN interface, as seen in the band diagram (black...
Measurements of the 2DEG sheet density as a function of gate voltage are presented in Figure 3a. These data demonstrate a depletion of the 2DEG sheet density by up to a factor of ~3x from its un-gated value as \( V_G \) is lowered to -12 V. The gating is similar at lower temperatures (Supplementary Figure S4c) and for the thin GaN sample. The inset of Figure 3a shows how depletion widens the quantum well at the AlGaN/GaN interface. Depletion also reduces the 2DEG mobility as seen in Figure 3b, similar to former work.[40,41] To study the effect of gating on \( S_{\text{ph}} \), we need to first estimate \( S_d \) as a function of gate voltage. This can be done by studying the effect of \( V_G \) on the thin GaN sample, presented in Figure 3c. For a degenerate 2D quantum well, we can roughly approximate the diffusive Seebeck coefficient as \( S_d \propto T/(E_F - E_1) \).[25] Since \( n_{2D} \propto (E_F - E_1) \), the magnitude of the diffusive Seebeck coefficient should increase with negative \( V_G \) as it depletes the 2DEG and decrease linearly with \( T \). Both features are visible in Figure 3c.

Figure 3d shows the effect of \( V_G \) on the thick GaN sample, where the upturn below ~150 K is apparent even after depletion. As in Figure 2f, we subtracted a linear fit of the thin GaN Seebeck coefficients (in Figure 3c) from the values for the thick GaN to estimate \( S_{\text{ph}} \) for different \( V_G \). This is plotted at 50 K for the thick GaN sample in Figure 3e. To validate these values, we also calculated \( S_{\text{ph}} \) for different \( n_{2D} \) using Equation 2, taking into account the shape of the quantum well. These data are plotted in Figure 3f at a temperature of 50 K. The good quantitative agreement allows us to conclude that in our samples, the magnitude of both \( S_{\text{ph}} \) and \( S_d \) increase with 2DEG depletion, indicating that the dependence of phonon drag on the quantum well width, via \( I(q_z) \), is less crucial. Further, the dependence of \( S_{\text{ph}} \) on \( V_G \) rigorously confirms that the Seebeck coefficient trend in the thick GaN sample is indeed due to phonon drag.

In conclusion, we have experimentally shown that phonon drag can be a significant portion of the total Seebeck coefficient in a 2DEG, even at room temperature. By using thickness as a “knob” to control sample dimensions, we show that \( S_{\text{ph}} \) is suppressed in the AlGaN/GaN 2DEG at a film dimension of ~100 nm. From a thermoelectric power conversion perspective, we shed light on two important phenomena: First, the magnitude of the phonon
drag can increase even when the thermal conductivity is decreasing, which means that these could be tuned separately. Second, depleting a 2DEG can lead to an increase in both the phonon drag and diffusive contributions of the Seebeck coefficient. These findings enable a better understanding of the phonon drag effect, and can lead to advancements in thermoelectric power conversion across a wide range of temperatures.

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Supporting Information

Significant Phonon Drag at Room Temperature in the AlGaN/GaN 2D Electron Gas Revealed by Varying GaN Thickness

Table of Contents:

**Supplementary Note 1**: Fabrication Process
**Supplementary Note 2**: Test Setup and Measurement Notes
**Supplementary Note 3**: Mobility Model
**Supplementary Note 4**: Phonon Drag Model
**Supplementary Note 5**: Thermal Conductivity Model
**Supplementary Note 6**: Codes
Figure S1 | Fabrication process. The panels show the eight mask process to fabricate the suspended thermal conductivity and Seebeck coefficient measurement platforms.

Figure S2 | Cross-Sectional Images. (a) Schematic of grown AlGaN/GaN heterostructure, along with the different buffer layers. (b,c) SEM image of the suspended portion of the thick and thin GaN sample.
**Figure S3** Measurement Process. (a) Microscope image of the suspended thermal conductivity measurement structure. (b) Schematic of the thermal conductivity measurement platform, showing the different pathways for gate sinking. (c) Model for the thermal conductivity of alumina, extracted from experimental data in the literature. (d) Schematic of the Seebeck coefficient measurement platform. (e) Microscope image of Seebeck coefficient measurement platform, showing the gated 2DEG mesa region with Ohmic contacts to extract the Seebeck voltage.
Figure S4] (a) Energies of the bottom 2 subbands in the AlGaN/GaN quantum well for the thick and thin GaN samples. (b) AFM image of the surface of the thick GaN sample (with the alumina layer on top). The RMS roughness is estimated to be ~1.4 nm. (c) Gating of the sheet density in the thick GaN sample at a temperature of 50 K. (d) Electron-phonon momentum conservation in the out-of-plane direction for 2DEG sheet densities varying from 1-10 × 10^{12} cm^{-2}. The corresponding shape of the wave function, ψ(z), for the bottom subband at the 2DEG at the AlGaN/GaN interface is shown in the inset.

Table S1| Definitions of selected parameters.

| Parameter                               | Symbol (units) | Value          | Reference       |
|-----------------------------------------|----------------|----------------|-----------------|
| Effective electron mass                 | \( m^* \)      | 0.22m_e        | Gurusinghe et al.[1] |
| GaN dielectric constant                 | \( \epsilon \) (Fm^{-1}) | 10.4\( \epsilon_0 \) | Gurusinghe et al.[1] |
| GaN sheet density                       | \( n_{2D} \) (cm^{-2}) | ~1×10^{13} cm^{-2} | Our measurements |
| GaN deformation potential               | \( D \) (eV)   | 8.5            | Sztein et al.[2] |
| Optical phonon energy                   | \( h\omega_{OP} \) (meV) | 91.2          | Sztein et al.[2] |
| Density of GaN, AlN                     | \( \rho \) (kgm^{-3}) | 6150, 3266    | Sztein et al.[2] |
| Grüneisen parameter                     | \( \gamma_G \) | 0.5            | Sztein et al.[2] |
| Atomic mass of GaN, AlN                 | \( M \) (amu)   | 83.7, 40.99    | Sztein et al.[2] |
| Average phonon velocity in GaN, AlN     | \( v_{av} \) (ms^{-1}) | 5070.5, 7183.5 | Sztein et al.[2] |
| GaN, AlN Debye temperature             | \( \theta_D \) (K)   | 600, 1150      | Sztein et al.[2] |
| GaN Umklapp scattering constants       | \( P \) (eV), \( C_U \) (K) | 1.1375 eV, 132 K | Cho. et al.[3] |
| AlN Umklapp scattering constants        | \( P \) (eV), \( C_U \) (K) | 2.0625 eV, 382 K | Slack et al.[4] |
Supplementary Note 1: Fabrication Process

Figure S1 shows the eight-mask process to fabricate the fully-suspended AlGaN/GaN platform for thermal measurements. A schematic of the heterostructure showing the different buffer layers and the silicon substrate is illustrated in Figure S2a. The AlGaN/GaN/buffer heterostructure for the thin and bulk GaN samples was grown using an in-house metal organic chemical vapor deposition (MOCVD) chamber on a Si (111) substrate. In order to define the 2DEG mesa, we etched the AlGaN/GaN layers to a depth of ~50 nm using an inductive coupled plasma technique with BCl3/Cl2 gases as shown in Figure S1a. This was followed by the deposition of ~4 μm PECVD oxide on the backside and selectively patterned to define the Si removal region, as depicted in Figure S1b. The Ohmic contacts to the 2DEG were patterned by depositing Ti/Al/Pt/Au (20/100/40/80 nm) followed by a rapid thermal anneal (RTA) in N2 ambient at 850°C for 35 seconds (Figure S1c). Next, we deposited ~47 nm of atomic-layer deposited (ALD) Al2O3 followed by patterning Ti/Pt (10/100 nm) heater and sensor metal lines, as shown in Figure S1d. To deposit Ti/Au (20/200 nm) bondpad metal, we opened vias in the ALD film using a 20:1 buffered oxide etch for ~2 min (Figure S1e). The gate metal Ti/Au (20/200 nm) was deposited after the bondpad metal, as shown in Figure S1f. In order to release the AlGaN/GaN/buffer heterostructure, Si was finally etched from the backside using a deep reactive ion etching (DRIE) technique, stopping at the buffer/Si interface. SEM images of the suspended portion of the thick and thin GaN sample are shown in Figure S2b and Figure S2c, respectively. After suspension, the total thickness of the heterostructure layers was obtained as ~2.85 μm for the thick GaN heterostructure and ~1.695 μm for the thin GaN heterostructure and heterostructure from the SEM cross-section images, shown in Figure S2b and Figure S2b.

Supplementary Note 2: Test Setup and Measurement Notes

In order to obtain the thermal conductivity and Seebeck coefficient of the AlGaN/GaN 2DEG, a measurement procedure identical to our earlier work is used, which has all the experimental details.[5] The main distinction from our former work is that these experiments (from 50 to 300 K) were done in vacuum using a temperature controlled cryostat, while our former work (300 – 573 K) were done in air as the ambient. To obtain the gate and temperature dependent sheet density, we performed Hall effect and Van der Pauw measurements in a vacuum cryostat. The use of vacuum ensures that any errors in the extraction of the thermal conductivity and Seebeck coefficient due to thermal convection effects are eliminated. In our measurement structures, the metal lines are placed on top of an amorphous alumina film of
thickness $\sim 47$ nm (to avoid electrical conduction with the underlying 2DEG), which needs to be properly accounted for to extract the thermal conductivity of the suspended film and the Seebeck coefficient of the 2DEG.

We first focus on the thermal conductivity extraction procedure. In our device, the heater and sensor lines have a width ($W$) of 5 $\mu$m, and are spaced by a distance ($D_{HS}$) of 75 $\mu$m, as seen in Figure S3a. The location of the heater and sensor resistances on the suspended membrane ($R_H$ and $R_S$) are chosen such that the heat transfer can be well approximated as 1-D.$^{[5,6]}$ Figure S3b shows a cross-section schematic of the thermal resistance network with the different pathways for heat sinking when a current is applied to the heater metal. Since we have established that thermal conduction is the only heat transport mechanism that needs to be accounted for, the thermal resistance of the suspended film ($R_F$) can be written as:

$$ R_F = \frac{2(T_H - T_S)}{P_H} - 2R_{Al} - \frac{2(R_{max} + R_{oxg})}{A_H} $$

(S1)

where $T_H$ and $T_S$ are the heater and sensor line temperatures, $P_H$ is the input power to the heater and $R_{Al}$ is the thermal resistance of the Al$_2$O$_3$ layer, $A_H$ is the area projected area of the heater electrode (5 $\mu$m x 200 $\mu$m), $R_{max}$ is the thermal boundary resistance of the Heater/Al$_2$O$_3$ interface and $R_{oxg}$ is the thermal boundary resistance of the Al$_2$O$_3$/GaN interface. The thermal conductivity of the film can be extracted from $R_F$ and the known film dimensions. To calculate the thermal resistances, we denote $t_{Al}$ and $t_F$ are the thicknesses of the alumina and AlGaN/GaN/buffer film, respectively. We used a thermal boundary resistance of $2.8 \times 10^{-8}$ m$^2$K W$^{-1}$ for $R_{max}$.\[^7\] Although the data for the thermal boundary resistance across the Al$_2$O$_3$/GaN film interface is not available, we estimated $R_{oxg} \approx 1 \times 10^{-8}$ m$^2$K W$^{-1}$ based on measurements across amorphous dielectric/Si interfaces,\[^8\] since GaN and Si have similar Debye temperatures.\[^9\] The thermal resistance of the alumina layer can be estimated as $R_{Al} = t_{Al}/(k_{Al}A_H)$, where $k_{Al}$ is the temperature dependent thermal conductivity of the alumina layer. The measurements for the thermal conductivity of amorphous alumina films have been published in the literature before. It is worth noting that amorphous films are typically modeled by the differential effective-medium (DEM) approximation, where $k \propto n^2$, with $n$ denoting the atomic density of the film.\[^10\] Thus, the variation in the thermal conductivities between the different films is attributed to the different densities, which depends strongly on the growth technique and deposition temperature. Our film is prepared via atomic layer deposition (ALD) at a temperature of 200$^\circ$C, whose thermal conductivity has been previously measured by Gorham et al. under the same condition at room temperature.\[^10\] The temperature dependent thermal conductivities of alumina films prepared under different conditions have been reported
by a few other research groups,\textsuperscript{[7,10–12]} as seen in Figure S3c. Since we can attribute the difference exclusively to density, we fit the thermal conductivity obtained by Lee \textit{et al.} for different temperatures,\textsuperscript{[12]} and scale it to match the value obtained by Gorham \textit{et al.} at room temperature\textsuperscript{[10]} to obtain $k_{Al}$, marked by a red line in Figure S3c. In conclusion, since $R_{\text{mox}}, R_{\text{oxg}}$ and $R_{Al}$ are known from Equation S1, we can calculate $R_F$ which can be used to obtain the thermal conductivity ($k_F$) of the suspended film.

Next, we point out the sources of error that need to be accounted for to accurately measure the Seebeck coefficient. The Seebeck voltage of the 2DEG is given as $S = V_{2DEG}/(T_1 - T_2)$, as depicted in Figure S3d and Figure S3e. The temperature at the contact outside the suspended region ($T_2$) is assumed to be at the substrate temperature. The temperature drop in the silicon supported region is <1% of the total temperature drop ($T_1 - T_2$), thus, the contribution to the Seebeck coefficient from the supported region can be ignored. $T_1$ is related to the heater temperature $T_H$ as:

$$\frac{(T_H - T_1)}{P_H} = R_{Al} + \frac{R_F}{2} + \frac{(R_{\text{mox}} + R_{\text{oxg}})}{A_H}$$  \hspace{1cm} (S2)

where $R_F$ is calculated using the measured film thermal conductivity and a length of 30 μm ($D_S$, depicted in Figure S3d) and $R_{Al}$ is calculated as discussed earlier. Knowing $T_1, T_2$ and $V_{2DEG}$, the total Seebeck coefficient $S$ can be extracted.

**Supplementary Note 3: Mobility Model**

To model the mobility in the AlGaN/GaN 2D electron gas, we need to understand the scattering rates for the electrons in the 2DEG quantum well with phonons (acoustic and optical), as well as the 2DEG interface. The electronic state for a 2D quantum well can be described by wave vector $\mathbf{k} = (k_x, k_y)$ in the plane of the AlGaN/GaN interface, and subband index $n$ to describe the wave function along the confinement direction ($z$). Under this assumption, we can write the wave function and electron energy for the electrons in the bottom subband as:

$$\Psi_{n,k} = \psi(z)e^{i\mathbf{k}\cdot\mathbf{r}}$$  \hspace{1cm} (S3)

$$E_n(\mathbf{k}) = E_n + \frac{\hbar^2k^2}{2m^*}$$  \hspace{1cm} (S4)

where $\mathbf{r} = (x, y)$ denotes the spatial coordinate in-the-plane of the 2DEG and $E_n$ is the energy at the bottom of the subband corresponding to index $n$.\textsuperscript{[13]} Figure S4a shows $E_n$ as a function of temperature (50 to 300 K) with respect to the Fermi level for the bottom two subbands for the thin and thick GaN sample. Since the majority of conduction electrons (> 90 %) are present in the lowermost subband, we only consider the bottom subband ($n = 1$) for evaluating all the
electronic transport properties (thus neglecting inter-subband scattering). To model the wave function along the confinement direction in Equation S3, we can use the Fang-Howard expression, where \( \psi(z) = \sqrt{\frac{b^3z^2}{2}} e^{-\frac{bz}{2}} \). Here, the parameter \( b = \left( \frac{12m^*e^2n_{\text{eff}}}{\epsilon \hbar^2} \right)^{\frac{1}{3}} \), where \( n_{\text{eff}} \approx \frac{11}{32} n_{2D} \), assuming that the barrier layer is un-doped and all the 2DEG electrons are a result of built-in polarization fields at the AlGaN/GaN interface.

The scattering rates for electrons can be evaluated using Fermi’s golden rule, for which we need to calculate the matrix elements with the correct scattering potentials for the different mechanisms. For the sake of brevity, we will skip the details, which can be found elsewhere.\(^{[1]}\) In our scattering picture, the 3D phonon can be represented by the wave vector \( \mathbf{Q} = (q_x, q_z) \), where \( q \) and \( q_z \) represent the in-plane and out-of-plane component. When an electron with initial wave vector \( \mathbf{k} = (k_x, k_y) \) scatters with \( \mathbf{Q} \), its final state can be written as \( \mathbf{k}' = \mathbf{k} + \mathbf{q} \) from conservation of momentum in-plane. If the collision is elastic, we can write \( |q| = \frac{2k}{\sin \left( \frac{\theta}{2} \right) } \), where \( \theta \) is the angle between \( \mathbf{k} \) and \( \mathbf{k}' \). The in-plane scattering matrix elements \( (M) \) are identical to the ones used in for scattering with 3D electrons. However, in this case, because we need to account for the out-of-plane phonon wave vector \( q_z \), the 2D matrix scattering element is modified as

\[
M^2_{2D} = \int M^2 |I(q_z)|^2 \, dq_z,
\]

where \( I(q_z) = \int \psi(z)^2 e^{i q z} \). Using the Fang-Howard form for \( \psi(z) \), \( |I(q_z)|^2 \) can be simplified as \( \frac{b^6}{(b^2+q_z^2)^3} \).\(^{[1]}\) For the purposes of calculating the AlGaN/GaN mobility, the mechanisms we consider here are scattering from acoustic phonons, optical phonons and roughness at the AlGaN/GaN quantum well interface. In particular, scattering by ionized impurities is neglected since the AlGaN barrier layer is assumed to be un-doped. Further, only acoustic scattering via the deformation potential is considered and piezoelectric scattering is neglected as it is found to be negligible.\(^{[1]}\)

Screening of the electron-phonon interaction by the carriers themselves is important to consider for the elastic processes (in our case, for acoustic phonon scattering and roughness scattering). This is often done by scaling the matrix scattering element \( M_{2D} \) by the screening function, defined as\(^{[1]}\)

\[
S(q, T) = 1 + \frac{e^2 F(q) \Pi(q, T)}{2\epsilon q},
\]

\[
\frac{1}{\Xi_{\text{local}}} = \prod_{\mathbf{Q}} \frac{1}{E_{\mathbf{Q}}^{\text{EL}}} - \frac{1}{E_{\mathbf{Q}}^{\text{NO}}}
\]
where \( q = |\mathbf{q}| \), \( F(q) \) and \( \Pi(q, T) \) are the form factor and the polarizability function whose definitions are well known in the literature.\(^{[13]}\) Once \( S(q) \) is known, we can calculate the scattering times \( \tau(E) \) for the 2DEG electrons as functions of electron kinetic energy \( (E) \). The integrated expressions for \( \tau(E) \) over the limits of the scattering angle \( \theta \) (from 0 to \( 2\pi \)) for acoustic deformation potential scattering, \( \tau_{ac}(E) \) and optical phonon scattering, \( \tau_{opt}(E) \) can be found in former work.\(^{[1]}\) For roughness scattering, we correct the expression found in former work\(^{[1]}\) (missing a factor of \( \pi \)), and is correctly written as

\[
\frac{1}{\tau_{ir}(E)} = \frac{m^* \Delta^2 \lambda^2 e^4 (n_{2D})^2}{8\pi \hbar^3 \epsilon^2} \int_0^{2\pi} e^{\frac{-q^2 \lambda^2}{4}} \left( 1 - \cos \theta \right) S(q, T)^2 d\theta, \tag{S7}
\]

where \( \Delta \) is the average displacement of the interface and \( \lambda \) is a parameter defined as the auto-correlation length.\(^{[1]}\) In our calculations, we have set \( \lambda = 7.5 \) nm. In order to accurately fit the mobility data over temperature, we set values of \( \Delta \) corresponding to 1 nm and 2 nm are used for the thick GaN and thin GaN sample, respectively. An AFM image of the sample surface is shown in Figure S4b. Once the values for the different scattering times are obtained, the total scattering time \( \tau(E) \) can be calculated by adding up the different scattering rates. Finally, we calculate the energy averaged scattering time as a function of temperature as

\[
\tau_{av}(T) = \int \frac{\tau(E) \frac{\partial f_0(E)}{\partial E}}{E \frac{\partial f_0(E)}{\partial E}} dE, \tag{S8}
\]

where \( f_0(E) \) is the Fermi function and the limits of integration are from the subband bottom \( E_1 \) to \( \infty \) (referenced to \( E_F \)). Since \( n_{2D} \approx \frac{m^*(E_F - E_1)}{\pi \hbar^2} \) when using only the bottom subband, we obtain \( E_1 \approx -108 \) meV, which is consistent with the Schrödinger–Poisson model (Figure S4a). Once \( \tau_{av}(T) \) is calculated from Equation S8, the 2DEG mobility for both the experimental samples can be obtained.

**Supplementary Note 4: Phonon Drag Model**

As discussed in main paper, the expression for phonon drag for the case of 3D phonons interacting with 2D electrons is:

\[
S_{ph} = - \frac{(2m^*)^3 \nu_{ph}^2}{4(2\pi)^3 k_B T^2 n_{2D} \epsilon \rho} \int_0^{\infty} dq \int_{-\infty}^{\infty} dq_z \frac{\Xi(Q)q^2 Q^2 |I(q_z)|^2 G(Q) \tau_{ph}(Q)}{S^2(q, T) \sinh \left( \frac{\hbar \omega_Q}{2k_B T} \right)}, \tag{S9}
\]

The definitions for \( S(q, T) \) and \( I(q_z) \) follow from Supplementary Note 3. The explicit expression for \( \Xi(Q) \) is\(^{[14]}\)
23

\[ |\Xi|^2 = D^2 + \frac{8q^2_2 q^2 + q^4}{2(q^2 + q^2_2)^2}, \]  

where the first term represents the scattering via the deformation potential (with strength of the interaction described by constant \( D \)) and the second term accounts for piezoelectric scattering.

In Equation S9, \( G(Q) \) is the energy integral, which is written as:

\[ G(Q) = \frac{1 - e^{-\frac{\hbar \omega q}{k_B T}}}{\hbar \omega Q} \times \int_{\gamma}^{\infty} dE \frac{f_0(E)(1 - f_0(E + \hbar \omega q))}{\sqrt{E - \gamma}}. \]  

In Equation S11, \( \gamma = \frac{(\hbar \omega q - E_q^2)}{4E_q} \), with \( E_q = \frac{\hbar^2 q^2}{2m^*} \).

In Figure S4d, we show the form \( I(q_z) \) for a range for \( n_{2D} \) varying from \( 10^{12} \) to \( 10^{13} \) cm\(^{-2} \), where we note that for small values of \( q_z \), \( I(q_z) \approx 1 \), but around \( q_z \) corresponding to the Debye wavelength (about \( 1.55 \times 10^{10} \) m\(^{-1} \) in GaN), \( I(q_z) \approx 0 \). The physical interpretation is that for thinner quantum wells (smaller \( \Delta z \)), larger values of \( q_z \) are allowed to interact with the 2D electrons because the momentum conservation in the out-of-plane direction is less stringent. Finally, we also note that because of the specific shape of \( I(q_z) \), we can set the limits of the integration in Equation S9 to the Debye wave vector (instead of \( \infty \)).

### Supplementary Note 5: Thermal Conductivity Model

As seen in Supplementary Note 4, in order to accurately calculate \( S_{ph} \) via Equation S9, it is necessary to calculate the phonon scattering time, \( \tau_{ph}(Q) \) in the GaN layer. This can be estimated accurately from the in-plane thermal conductivity measurements of the suspended AlGaN/GaN film. Since we do not have thermal conductivity measurements of the GaN layer exclusively, we follow a more involved approach. In particular, we first model the thermal conductivity of the composite film and compare with experimental data. Then, we use the model for the GaN film to estimate the \( \tau_{ph}(Q) \) needed to calculate \( S_{ph} \).

Since our suspended film is a composite consisting of an AlN layer, Al\(_{x}\)Ga\(_{1-x}\)N transition layers and a GaN layer, the overall thermal conductivity (\( k \)) can be estimated as \( \Sigma k_i t_i / \Sigma t_i \), where \( k_i \) and \( t_i \) refer to the thermal conductivities and thicknesses of individual layers. For each multilayer, we use a Boltzmann Transport Equation (BTE) model to quantify \( k_i \) with layer thickness (\( t_i \)). Using a simple Debye approximation for the phonon dispersion with an average velocity over the acoustic phonon modes (\( v_{av} \)), the in-plane thermal conductivity for each layer can be written as[16]
\[ k_i = \frac{k_B T^3}{2\pi^2 \hbar^3 v_{av}} \int_0^{\theta_D/T} x^4 e^x \tau(x) \left( e^x - 1 \right)^2, \]  

where \( \theta_D \) is the Debye temperature for the multilayer, \( T \) is the temperature, and \( x = \hbar \omega / (k_B T) \). Here, \( \omega \) is the phonon frequency, which can be approximated as \( v_{av}\sqrt{q^2 + q_z^2} \) assuming a linear phonon dispersion. The total scattering time \( \tau \) is calculated by Mathiessen’s rule with contributions from Umklapp \( (\tau_U) \), alloy \( (\tau_A) \), boundary \( (\tau_B) \) and defect scattering \( (\tau_D) \), respectively. Phonon-phonon scattering is evaluated using via the relaxation term for Umklapp processes\[17\]

\[ \tau_U(x) = \frac{P k_B T^3 x^2 e^{-C_U \tau}}{x}, \]  

where the constants \( P \) and \( C_U \) for GaN and AlN are listed in Table S1. Scattering with impurities is neglected since its effect is found to be negligible for unintentionally doped films.\[18\] For the Al\(_x\)Ga\(_{1-x}\)N layers, all the material parameters (e.g. \( v_{ac}, \theta_D, P, C_U \)) are averaged over the AlN and GaN fractions, in context of the virtual crystal model.\[19\] Alloy scattering severely reduces the thermal conductivity of the transition layers and is evaluated as a point defect scattering term.\[20\] For the sake of brevity, we skip the details, which can be found in Liu et al.\[20\] The defect scattering term \( (\tau_D) \) included core, screw, edge and mixed dislocations with total density \( N_{dis} \), whose effect is to reduce the thermal conductivity.\[21\]

Although we have a composite film (and thus, the dislocation density is expected to vary for the different layers), we estimated an average dislocation density for the suspended film via X-Ray Diffraction (XRD) measurements. These values were estimated to be \( \approx 9 \times 10^8 \) cm\(^{-2}\) and \( \approx 2.5 \times 10^9 \) cm\(^{-2}\) for the thick and thin GaN samples, the details of which can be found in former work.\[5\] To evaluate the boundary scattering term, we used \( \tau_B \approx 2.38 t_i / v_{av} \), which is a model that is used for nanowires,\[18\] but will suffice to model the dependencies observed in the measured thermal conductivity with temperature.

**Supplementary Note 6: Codes**

The codes to simulate the diffusive Seebeck coefficient, the phonon drag component of the Seebeck coefficient and the thermal conductivity are available (as MATLAB files) at: https://github.com/ananthy/PhononDrag
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