Supporting Information

Enhanced Thermal Boundary Conductance across GaN/SiC Interfaces with AlN Transition Layers

Ruiyang Li,1 Kamal Hussain,2 Michael E. Liao,3 Kenny Huynh,3 Md Shafkat Bin Hoque,4 Spencer Wyant,5 Yee Rui Koh,4 Zhihao Xu,1 Yekan Wang,3 Dorian P. Luccioni,3 Zhe Cheng,6 Jingjing Shi,6 Eungkyu Lee,7 Samuel Graham,6 Asegun Henry,8 Patrick E. Hopkins,4,9,10 Mark S. Goorsky,3 M. Asif Khan,2 Tengfei Luo1,11,12,*

1 Department of Aerospace and Mechanical Engineering, University of Notre Dame, Notre Dame, Indiana 46556, United States
2 Department of Electrical Engineering, University of South Carolina, Columbia, South Carolina 29208, United States
3 Materials Science and Engineering, University of California, Los Angeles, Los Angeles, California 90095, United States
4 Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, United States
5 Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, United States
6 George W. Woodruff School of Mechanical Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332, United States
7 Department of Electronic Engineering, Kyung Hee University, Yongin-si, Gyeonggi-do 17104, South Korea
8 Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, United States
9 Department of Materials Science and Engineering, University of Virginia, Charlottesville, Virginia 22904, United States
10 Department of Physics, University of Virginia, Charlottesville, Virginia 22904, United States
11 Department of Chemical and Biomolecular Engineering, University of Notre Dame, Notre Dame, Indiana 46556, United States
12 Center for Sustainable Energy of Notre Dame (ND Energy), University of Notre Dame, Notre Dame, Indiana 46556, United States

* Corresponding author. Email: tluo@nd.edu (T. Luo)
Figure S1. Cross-sectional STEM image of (a-f) GaN/SiC interfaces (samples I-VI) with AlN transition layers of varying thicknesses (0-73 nm).

Supplementary Note 1: TDTR data fitting

An additional bare SiC substrate was prepared to measure its thermal conductivity, which is used as a known parameter in the data fitting process. Three unknown parameters, $G_{\text{Al/GaN}}$, $\kappa_{\text{GaN}}$, and $G_{\text{GaN/SiC}}$, are extracted by TDTR with a modulation frequency of 3.0 MHz at 300 K and of 8.8 MHz at 500 K. The pump and probe beam sizes are 20.5 and 10.0 μm (diameters), respectively. TDTR measurements return phase signals (defined by $\tan^{-1}(Y/X)$, where $Y$ and $X$ are out-of-phase and in-phase signals, respectively) as a function of delay time, which are fitted by a heat conduction model to extract thermal properties. The TBCs are measured by three TDTR systems in University of Notre Dame, University of Virginia, and Georgia Institute of Technology to ensure that the observed trends can be reproduced. As for the known properties in the model, the heat capacities
of the three layers are taken from literature,\textsuperscript{1-2} and the thermal conductivity of the Al film is calculated from the Wiedemann-Franz law and its electrical resistance.\textsuperscript{3} The fitting is relatively insensitive to the Al transducer thermal conductivity.\textsuperscript{4} The cross-plane thermal conductivities of the SiC substrate are measured using the bare SiC substrate (\~312 W m\textsuperscript{-1} K\textsuperscript{-1} at 300 K and \~140 W m\textsuperscript{-1} K\textsuperscript{-1} at 500 K), which are close to the literature values.\textsuperscript{2, 5} The fitting errors are determined by the Monte Carlo method accounting for all the possible error sources.\textsuperscript{6-7} The heat capacity values obtained from the literature are assumed to have a standard deviation of 2%, while the standard deviations of SiC and Al thermal conductivity are 5% and 10%, respectively. The thicknesses of Al and GaN films are determined by TEM, with an error of 5% and 10% in fitting, respectively. These errors in Monte Carlo calculations are defined in consistence with prior TDTR works.\textsuperscript{6-7} During data fitting, we fit the phase signals by searching parameter spaces within the range of 10% to 1000\% of the initial guess for each unknown parameter.

**Figure S2.** Effective thermal resistance of GaN and GaN/SiC interface at (a) 300 K and (b) 500 K ($R_{\text{GaN}} = d_{\text{GaN}}/\kappa_{\text{GaN}}$; $R_{\text{GaN/SiC}} = 1/\kappa_{\text{GaN/SiC}}$).
Supplementary Note 2: Material characterization

For structural characterization in this work, an FEI Nova 600 Nanolab Dual Beam SEM/FIB was used to prepare cross section TEM samples roughly 100 nm thick using a Ga source and transferred to a TEM grid using a standard lift out procedure. High-resolution TEM images were taken using an FEI Titan and double tilt stage at 300 kV to study the crystallinity at the interfaces. STEM and EDS measurements were done on an FEI Talos and double tilt stage at 200kV. Triple-axis X-ray measurements and reciprocal space mapping were performed using a Bruker D1 diffractometer with an incident beam mirror producing a parallel beam and a Si (220) channel cut collimator (Cu Kα1 radiation). The scattered beam optics include a Si (220) channel cut crystal.

![Image of STEM images of samples I, III, and V](image_url)

**Figure S3.** Low magnification STEM images of samples I, III and V.
**Figure S4.** Inverse Fast Fourier Transforms (IFFT) taken across the GaN/AlN and AlN/SiC interface in (a) sample I, (b) sample III and (c) sample V reveal local distortions in the lattice caused by misfit dislocations.
**Figure S5.** EDS map of sample III (with 15-nm-thick AlN)

**Figure S6.** EDS map of sample V (with 65-nm-thick AlN)
Supplementary Note 3: Triple-axis XRD for samples IV, V and VI

To characterize the AlN structure, we measured the lattice parameters of AlN in samples IV, V and VI which have relatively thick AlN interlayers. We performed triple-axis X-ray diffraction (TAXRD) measurements. Figure S7 shows the symmetric $\omega$:2$\theta$ line scan results, and the peaks are relative to the (0006) 6H-SiC. This can give us out-of-plane lattice parameter, denoted as $c$. We were unable to measure the AlN peak for samples II and III due to too low signal not visible with typical laboratory diffractometers (e.g., higher flux at a synchrotron would be needed). While the (0002) GaN peak and the (0006) 6H-SiC peak overlap well among three samples, we observe that the (0002) AlN peak shifts with different AlN thicknesses, which indicates different amounts of relaxation. In Fig. S7, the fully relaxed AlN is marked with the dashed line at ~780 arcsec from the (0006) 6H-SiC peak. We find that thin AlN layers are strained, and thicker layers are increasingly relaxed to the bulk AlN lattice parameter. This can be confirmed with reciprocal space maps (RSMs) determining both in-plane and out-of-plane lattice parameters (see Fig. 6).

Figure S7. Triple-axis ($\omega$:2$\theta$) scans of samples IV, V and VI. The black dashed line denotes the fully relaxed AlN.
Supplementary Note 4: Development of neural network potentials (NNPs)

To build NNPs for GaN/SiC and GaN/AlN/SiC interfaces, we first generated two datasets from \textit{ab initio} molecular dynamics (AIMD) simulations. Each data snapshot contains potential energy, atomic forces, atomic coordinates, and supercell lattice vectors. For the GaN/SiC model construction, we first relaxed the $2 \times 2 \times 3$ SiC and GaN supercells, after which we rescaled the dimensions of the SiC supercell in the $x$ and $y$ directions to align with GaN. We then flipped the SiC supercell in the $z$-direction and layered it with the GaN supercell, resulting in the interface model. The method for constructing the GaN/AlN/SiC interface model closely resembles this process. Each interface model is relaxed only in the $z$-direction to minimize the energy at the interface. The relaxed interfaces are then simulated in the canonical ensemble (NVT) at temperatures ranging from 300 to 500 K to generate data snapshots with all the atomic information mentioned above. Concurrently, we also incorporated AIMD data for bulk materials into the dataset to ensure the stability and accuracy of NNP-MD simulations. All \textit{ab initio} data was subsampled to ensure the independence of each snapshot in the dataset. \textit{Ab initio} calculations are performed using the Vienna Ab initio Simulation Package (VASP)\textsuperscript{8} under the local density approximation (LDA). The Brillouin zone was sampled using a k-point mesh generated by a gamma-centered Monkhorst-Pack scheme and the reciprocal spacing is 0.03 Å$^{-1}$. Finally, each of the training datasets contains ~4000 snapshots for either GaN/SiC or GaN/AlN/SiC interface and ~2000 snapshots for the bulk materials.

The NNPs are trained using a deep neural network potential scheme developed by Zhang \textit{et al.},\textsuperscript{9} which has been proved to be capable of simulating solid interfaces. Within this scheme, chemical species and atomic coordinates are first mapped to embedded features, which will be fed into a deep neural network. This fitting network is trained to learn the potential energies and forces
by minimizing the mean squared error training loss. In this study, our potential model consists of a three-layer embedding network (with 20 × 40 × 80 nodes) and a two-layer fitting network (100 × 100 nodes), and it uses an embedding submatrix size of 16 (i.e., the “axis_neuron” parameter in the DeePMD configuration⁹). The interaction cutoff for our potentials is set to 5.5752 Å. Two potentials are developed for GaN/SiC and GaN/AlN/SiC, respectively, as interactions are different at these two interfaces. For both potentials, accuracy was assessed on a held-out test set of representative configurations. The GaN/AlN/SiC potential gives a force component mean absolute error (MAE) of 22.5 meV/Å and a corresponding energy MAE of 0.068 meV/atom. The GaN/SiC potential has a force MAE of 35.2 meV/Å and a corresponding energy MAE of 0.134 meV/atom. A parity plot for our GaN/SiC potential is shown in Fig. S8(a). We can see that both atomic force and potential energy achieve great agreement between ab initio benchmark and NNP predictions.

**Supplementary Note 5: Non-equilibrium MD (NEMD) simulations**

NEMD simulations were performed using the LAMMPS package with our NNPs for the target interfaces. For the atomic structures, the c-axis of the wurtzite crystal is aligned with the z-direction. To avoid lattice mismatch in MD simulations, we enforce the cross-sectional lattice size (along the x and y direction) to be identical to the lattice size of SiC. The numbers of unit cells for GaN and SiC are 9 × 6 × 21 and 9 × 6 × 7, respectively. For the GaN/AlN/SiC interface, a 2.8-nm-long AlN layer sandwiched by GaN and SiC layers. At the interfaces, SiC is Si-terminated, and GaN is N-terminated. AlN is N-terminated at the AlN/SiC interface as well. In MD simulations, periodic boundary conditions are applied to all three spatial directions, and a time step of 1.0 fs is employed. After relaxing the structure under isobaric–isothermal conditions (NPT) and NVT for a total of 4.0 ns, we freeze the two ends of the system and apply a temperature difference using
Langevin thermostats in the microcanonical ensemble (NVE) for at least 8.0 ns. The temperatures applied on the two thermostats are ±25 K of the target mean temperature. Based on NEMD simulations, TBC is obtained by $G = Q/(A\Delta T)$, where $Q$ is the steady-state heat flux along the $z$-direction, $A$ is the cross-sectional area, and $\Delta T$ is the temperature difference at the interface determined by extrapolating linear fits of the temperature profiles of two sides (i.e., GaN and SiC) to the interface and calculating the difference. In the case of the GaN/AlN/SiC structure, linear fits are extended individually to the GaN/AlN and AlN/SiC interfaces, then $\Delta T$ is determined by computing the difference between the edge temperatures at these two interfaces. Example temperature profiles in NEMD simulations are plotted in Fig. S8 (b-c).

**Figure S8.** Non-equilibrium molecular dynamics (NEMD) simulations of GaN/SiC and GaN/AlN/SiC interfaces using neural network potentials (NNPs). (a) Standardized potential energies and atomic forces obtained from GaN/SiC NNP and *ab initio* calculations for the testing dataset. Energies and forces are standardized by subtracting the average from them and then divided by the standard deviation. (b) Schematic of the GaN/SiC structure and steady-state temperature profiles at 300 K along the direction perpendicular to the interface. (c) Schematic of the GaN/AlN/SiC structure and steady-state temperature profiles at 300 K along the direction perpendicular to the interface.
To affirm the accuracy of our NNP and eliminate concerns regarding size effects in our simulations, we conduct a comparison of the phonon density of states (PDOS) derived from MD simulations and Density Functional Theory (DFT). In MD simulations, the PDOS is determined through the Fourier transform of the velocity autocorrelation function, while DFT phonon calculations utilize a density functional perturbation theory approach. We compute the MD PDOS for each layer within the GaN/AlN/SiC interface structure at 300 K. Concurrently, DFT phonon calculations are performed for each lattice at 0 K. The results of these calculations are presented in Fig. S9. Despite the potential volume expansion in MD, the resulting DFT and MD PDOS are in good agreement. They effectively capture the main features in the density of states, providing assurance that vibrations in different layers are accurately represented in our NNP-MD simulations.

![Phonon density of states](image)

**Figure S9.** Phonon density of states (PDOS) for each layer of the GaN/AlN/SiC interface in MD simulations, compared with PDOS for each lattice from DFT.


