Supplementary Material

Experimental Observation of High Intrinsic Thermal Conductivity of AlN

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The supporting information includes the characterization of dislocation density by TEM and X-ray topography study. The details of data fitting and error calculation of TDTR measurements are also included.

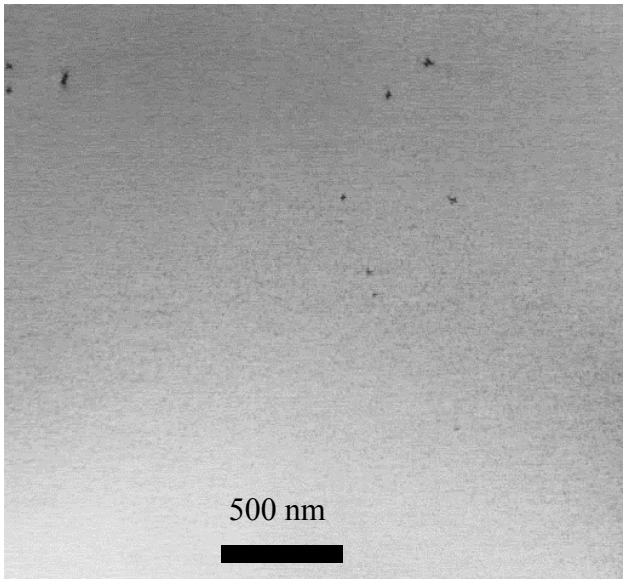
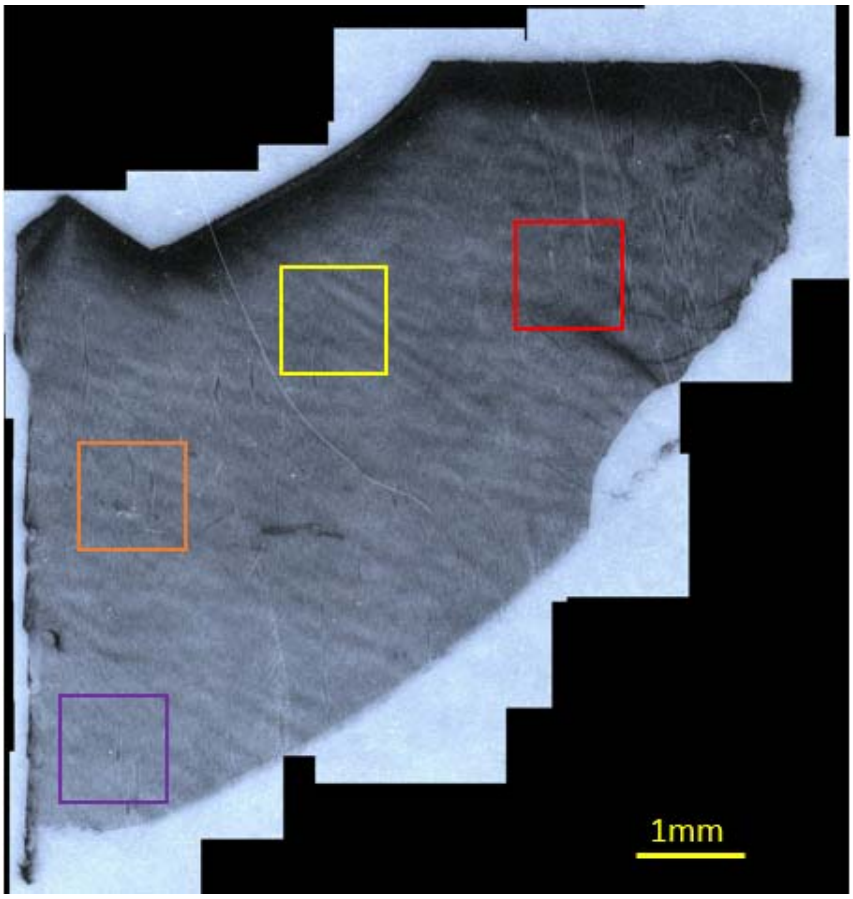


Figure S1. Near-surface plan-view (11$\underline{2}$0) two-beam condition STEM image of Samp_1. The black dots in this image are dislocations. The dislocation density is ~1.6 x 10^8/cm^2.



Region	Area	Dislocation density
Red	1 x 1 mm^2	2 x $10^4/cm^2$
Yellow	1 x 1 mm^2	1.8 x $10^4/cm^2$
Orange	1 x 1 mm^2	3.4 x $10^4/cm^2$
Purple	1 x 1 mm^2	1.7 x $10^4/cm^2$

Figure S2. X-ray topography exposed at the (11$\underline{2}$4) AlN asymmetric reflection using a wavelength of 0.12984 nm for Samp_4. The estimated average dislocation density of Samp_4 from the four areas indicated on the topography image is 2.2 x $10^4/cm^2$. The dislocation densities of Samp_3 and Samp_4 are several orders of magnitude lower than Samp_1 and Samp_2.

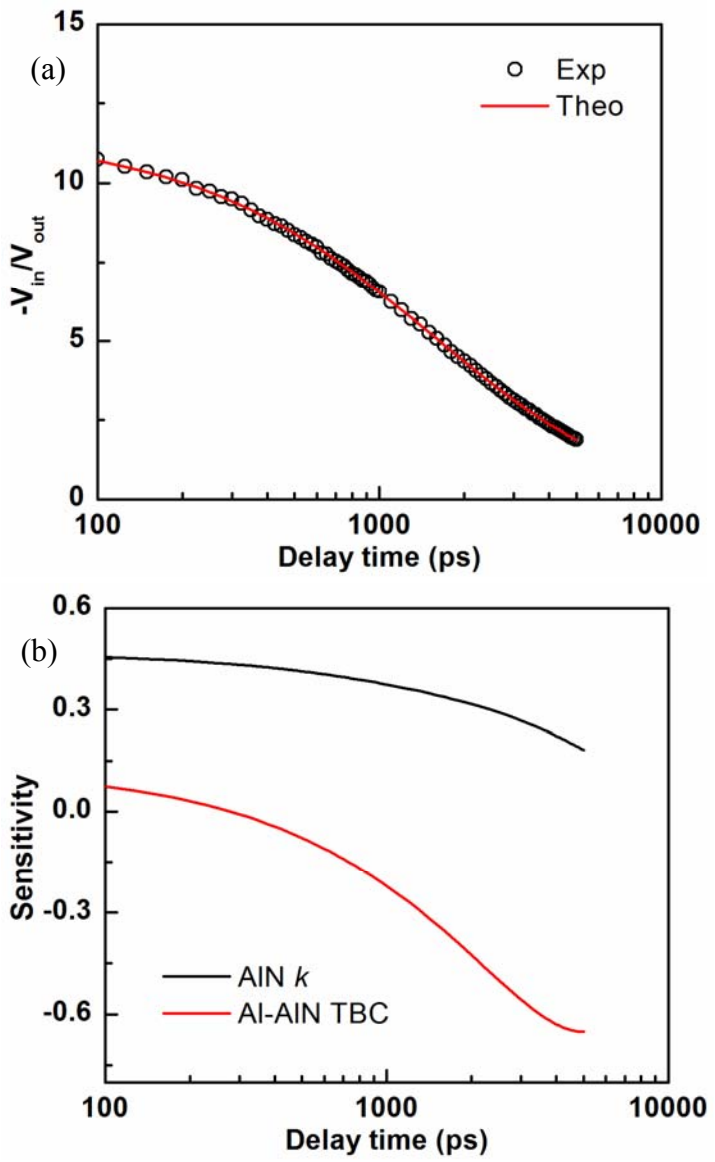


Figure S3. (a) TDTR data fitting of Samp_4 at room temperature with 3.6 MHz. (b) TDTR sensitivity of AlN thermal conductivity (AlN *k*) and Al-AlN thermal boundary conductance (Al-AlN TBC). High sensitivity here results in accurate measurements.

Error bar calculation

A Monte Carlo method is used to calculate the error bar of TDTR measurements.[1] The error of pump and probe beam spot sizes are ± 0.5 μm. The errors of the heat capacity of Al and AlN

are ± 2%. The error bar of Al thermal conductivity is ± 10% and that of Al thickness is ± 3 nm. An example is shown in Figure S4. 1000 times is fitted to calculate the error bar of AlN thermal conductivity. The 90% percentile confidence interval of the AlN thermal conductivity is 161.1-18.3/+20.7 W $m^{-1}K^{-1}$.

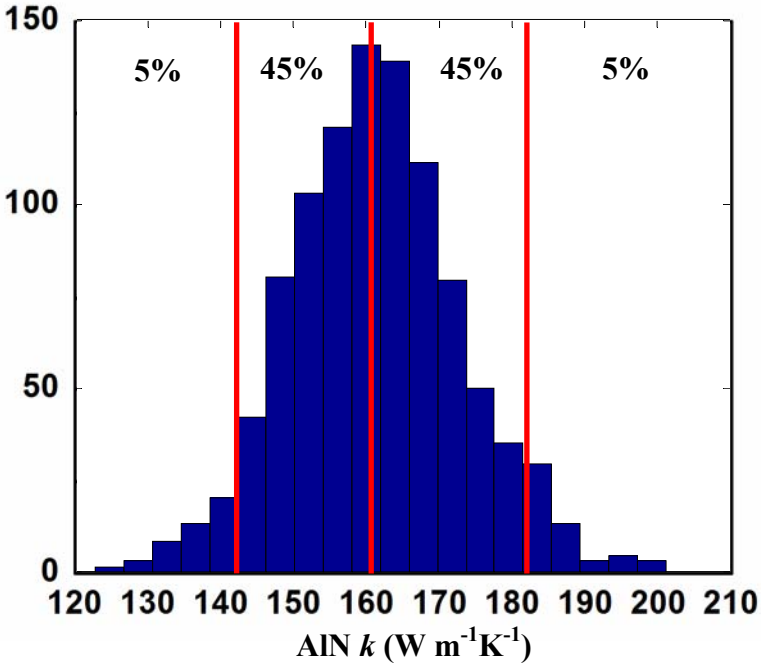


Figure S4. Monte Carlo error bar calculation with 1000 times of data fitting. The data is for Samp_4 at 380 K. 90% percentile confidence interval (AlN thermal conductivity =161.1-18.3/+20.7 W/m-K).

In our study, we performed a series of thermal conductivity calculation based on different discretization size of the first Brillouin zone. Below is a plot of the calculated bulk thermal conductivity at 300K as a function of the grid size we used. Considering the fact that the thermal conductivity has been saturated after the grid size of 12, the grid size of 12x12x12 is chosen for a balance of speed and accuracy. A locally adaptive Gaussian smearing implemented in ShengBTE

applied in our calculation for the energy conservation where the Gaussian smearing is depending on the group velocity and phonon wave vector involving in this three-phonon scattering process.

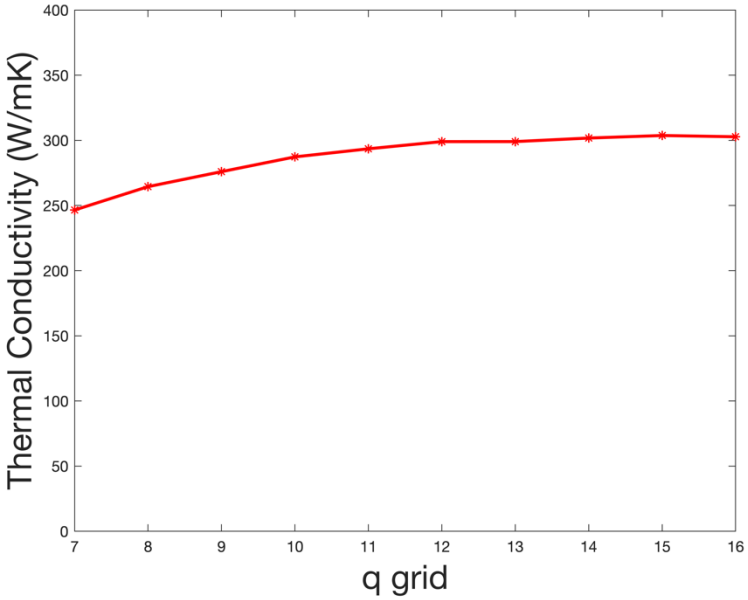


Figure S5. Thermal conductivity of AlN with *q* grid used in the DFT calculation.

References

[1] T. L. Bougher, L. Yates, C.-F. Lo, W. Johnson, S. Graham, B. A. Cola, *Nano. and Micro. Thermophys. Engineer.* **2016**, *20*, 22.