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Using Mosaicity to Tune Thermal Transport in Polycrystalline AlN Thin Films

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Abstract

The effect of controlling the c-axis alignment (mosaicity) to the cross-plane thermal transport in textured polycrystalline aluminum nitride (AlN) thin films is experimentally and theoretically investigated. We show that by controlling the sputtering conditions we are able to deposit AlN thin films with varying c-axis grain tilt (mosaicity) from 10° to 0°. Microstructural characterization shows that the films are nearly identical in thickness and grain size, and the difference in mosaicity alters the grain interface quality. This has a significant effect to thermal transport where a thermal conductivity of 4.22 W/mK vs. 8.09 W/mK are measured for samples with tilt angles of 10° vs. 0° respectively. The modified Callaway model was used to fit the theoretical curves to the experimental results using various phonon scattering mechanisms at the grain interface. It was found that using a non-gray model gives an overview of the phonon scattering at the grain boundaries, whereas treating the grain boundary as an array of dislocation lines with varying angle relative to the heat flow, best describes the mechanism of the thermal transport. Lastly, our results show that controlling the quality of the grain interface provides a tuning knob to control thermal transport in polycrystalline materials.

Keywords: Thermal conductivity Tuning, Mosaicity, AlN, Dislocations, Grain Interface

Introduction

Manipulating thermal conductivity of solids is of critical importance in a wide range of applications from heat management, power generation, to solid-state energy conversion, etc. Thermal transport of various single-crystalline materials is well understood and studied.¹⁻³ However, their application is limited to high-performance circumstances which renders them commercially challenging due to the high expenses associated with producing single-crystalline materials. Polycrystalline materials, on the other hand, are much more cost-effective and increasingly gain importance for various heat sensitive applications, such as thermal

management of high power density flexible electronic devices,⁴ solid-state devices as well as a thermoelectric generator. This even further accentuated with the advancement in nanotechnology, where there is a tremendous push to commercialize newly discovered methods to produce nanostructured bulk materials for various technologies.

Heat in solids is predominantly transferred by acoustic phonons over a spectrum of wavelengths and hence, structures of different length scales will scatter phonons at different rates.⁵ For single crystal dielectrics, point defects⁶ scatter phonons proportional to $\sim \omega^4$, thereby effectively scattering high-frequency phonons. Dislocations^{5, 7} and stacking faults⁸ have frequency dependence of $\sim \omega^{1-3}$ and hence are able to scatter phonon of mid-high frequency range effectively. However, at low temperatures, phonon scattering at the grain boundary has always been treated as a frequency independent parameter, $\tau_B \sim d$, where d is the sample characteristic dimensions. In polycrystalline materials, it is traditionally thought that such boundary scattering dominates heat transport where now d is the grain size. Thus, by varying this grain size one can tune thermal transport in polycrystalline materials accordingly.⁹⁻¹² This has always been the practice, even though experimental results show that the thermal conductivity of polycrystalline materials has T^2 dependence whereas the theoretical modeling without frequency dependent boundary scattering rate showed T^3 dependence. Therefore, there is a lack of physical accuracy in treating the grain boundary scattering as frequency independent. Recent work by Wang *et al.*¹³ showed that phonon scattering at the grain interface is better suited if it is treated as frequency dependent parameter by using a phonon scattering parameter at the grain boundaries as $\tau = \beta D_{avg} \omega_0/\omega$. This captured the temperature dependent thermal conductivity accurately, but unfortunately this description lacked the physical insight into the phonon scattering at the grain boundaries. To address this issue, Kim *et al.*¹⁴ treated the low angle grain boundaries as an array of sessile dislocation whose scattering was formulated by Klemens⁵ and analytically described by dislocation strain and dislocation core parameters. This approach showed both better understanding of the physical processes at the grain boundary and captured the low temperature thermal conductivity trend accurately.

This illustrates that, instead of treating the grain boundary as a frequency independent single interface, we could treat it as an interface consisting of an array of defects. This opens up the opportunity for finding new avenues to manipulate heat transport in polycrystalline materials by tailoring the quality of the grain interface and thus depending on the application, tailor thermal conductivity in both extremes. For example, in thermoelectric devices where low thermal conductivity below 1 W/mK is required, material systems such as PbTe, Bi_xSb_{2-x}Te₃, NaPb_mSbTe_{2+m} (SALT-m) achieved high thermoelectric performance by nanostructuring and increasing the defect densities of these granular materials.¹⁵⁻¹⁸ On the other spectrum for example in high power density applications, high single crystalline thermal conductivity values are required such as diamond and silicon carbide (SiC) with thermal conductivity of 2400

W/mK¹ and 350 W/mK,^{19, 20} respectively. However, polycrystalline diamond and SiC have much lower thermal conductivity values around 1800 W/mK²¹ and 270 W/mK²⁰ respectively. Therefore by tuning the grain interface quality it could be possible to enhance phonon transport in the material and achieve single crystalline values. Traditionally, polycrystalline materials are randomly oriented, and there is a lack of research in quantifying the quality of the grain beyond size and number to tune thermal transport. In this work, we show that by controlling the deposition temperature of DC sputtered AlN thin films, we can control the grain alignment (mosaicity) perpendicular to the thickness (c-axis) and increase the thermal conductivity by 91%. Structural and thermal characterization using the 3 ω method showed that mosaicity has a direct effect to the quality of the grain interface and consequently to the thermal conductivity. Specifically, we showed that there is an increase in the thermal conductivity with better c-axis alignment that was not simply due to grain size, but rather due to the better-quality grain interface. Finally, we fitted our temperature-dependent results using modified Callaway model where the grain interface scattering parameters were defined and studied individually using gray, non-gray, dislocation core/strain and stacking faults to best describe the thermal transport. This work offers an insight into looking at the grain boundaries quality as another knob to tune the thermal transport in the polycrystalline material.

Sample Preparation

AlN thin films were deposited on Si (100) substrate by DC reactive magnetron sputtering with unbalanced magnetic configuration at a base chamber pressure of 8.5 $\times 10^{-10}$ torr. Al target of diameter 1.5 inches and sample size of a quarter of 2 inches wafers were in an Ar-N₂ mixture plasma. A native oxide of Si substrate was not etched before the deposition. The AlN thin films were deposited at 400 $^{\circ}$ C and 600 $^{\circ}$ C while keeping other parameters same. Table 1 shows the different deposition parameters used in AlN thin film growth.

Table 1 Parameters for DC reactive magnetron sputtering.

Sample ID	Power (W)	Temperature ($^{\circ}$ C)	N ₂ /Ar flow ratio	Pressure (mTorr)
A	100	400	0.8	2
B	100	600	0.8	2

**Target Al (99.99% purity)*

Microstructural properties including plane texture and grain size were characterized by transmission electron microscopy (TEM) and X-ray diffraction (XRD).

Thermal Conductivity Measurement using the Differential 3 ω method

We measured the thermal conductivity by modifying our recent 3ω setup used for measuring the thermal conductivity of the flexible dielectric⁹ and employed a differential 3ω scheme, developed by Cahill *et al.*²² for dielectric thin films.^{23, 24}

Using microfabrication, a thin metallic bilayer of Cr/Au (10/250 nm thick) was deposited on both samples by e-beam evaporation with heater length (L) and width ($2b$) of 430 and 10 μm , respectively. The metallic heater acts as heater and thermometer which was heated by driving AC sine voltage using an SRS 850 lock-in-amplifier at an angular frequency of ω , which induced joule heating at an angular frequency of 2ω (Fig. 1a) shows the electrical circuit used in the 3ω measurement. In the differential 3ω technique, the thermal conductivity of the film of interest is extracted by measuring the temperature rise in the film with the substrate by detecting the third harmonic of the voltage in the metallic heater. The substrate temperature rise is determined by²⁵

$$\Delta T_{\text{substrate}} = \frac{P}{\pi l \kappa_{\text{subs}}} \left(0.5 \ln \left(\frac{4D}{b^2} \right) + \ln 2 + \eta - 0.5 \ln(2\omega) - \frac{i\pi}{4} \right), \quad (1)$$

where κ_{subs} and D are the thermal conductivity and thermal diffusivity of the substrate respectively, b is the heater half width, P power per unit length of heater and η is fitting parameter usually ranges from 0.9 – 1.05.²⁴ As the thermal conductivity of silicon is 20 times higher than the thin films under investigation, the increase in temperature rise due to thin film is given by²⁴

$$\Delta T_f = \frac{P t_f}{l \kappa_f (2b)}, \quad (2)$$

where κ_f and t_f are the thermal conductivity and film thickness respectively. Figure 1b shows the temperature oscillation as a function of second harmonic frequency, where the red solid line indicates the temperature oscillation caused by the 240 nm thin AlN films and blue dashed line depicts the temperature rise due to the substrate which is simulated using Equation 1.

The temperature-dependent thermal conductivity of AlN thin films was measured in the high vacuum environment of 5×10^{-5} mbar in the temperature range of 450 – 300 K. The samples were annealed overnight at 450 K in high vacuum to remove any impurity in the metallic heaters.²⁶ The frequency range was selected so that the heat wave penetration depth ranges from 13 to 470 μm and the linear regime where the in-phase temperature oscillation decays logarithmically.²²

Figure 1: Shows the schematic 3ω method. (a) the electrical circuit design for the 3ω setup (b) Temperature oscillation vs. logarithm second angular frequency of AlN thin films. Power of the heater was fixed to be 7.8 mW. The red solid line indicates the data obtained for the 240 nm thin AlN film (T_h) and blue dashed line shows the simulated temperature oscillation of a bare Si substrate (T_s). Inset shows the temperature distribution.

Result and Discussion

Microstructural Analysis

To quantify the grain quality of the samples we carried out detailed analysis using XRD and cross-sectional TEM. Figure 2 shows XRD $\theta - 2\theta$ scan patterns of the AlN films with peak near $2\theta = 36^\circ$. We observe increase in the intensity of peak and slight decrease in the average mosaicity (out-of-plane) disorientation. The mosaicity change is evident from the change in full width half maximum (FWHM) of the (0002) AlN peak of rocking curves from 6.5° to 5.5° as a function of the film deposition temperature (inset of Fig. 2). This increase in the intensity and decrease in the mosaicity is attributed to the increased out-of-plane (c-axis) crystallite alignment.

Figure 2: Depicts the $\theta - 2\theta$ scan for the AlN films deposited on Si (100) substrate via. DC magnetron reactive sputtering at two growth temperatures 400 °C (black) and 600 °C (blue). Inset shows the rocking curves for the samples A, B.

Figures 3&4 show the cross-sectional TEM images for samples A and B, respectively. The conventional TEM imaging (Fig. 3(a) & 4(a)) was performed and weak beam dark field images were acquired under two beam conditions at an acceleration voltage of 300 kV using a FEI Titan ST microscope system. High resolution TEM imaging ((Fig. 3(b-f) & 4(b-f)) was performed using a FEI Titan Cubed microscope equipped with a spherical corrector system for the objective lens. The thin TEM lamellae of the AlN on Si samples were prepared via. focused ion beam (FIB) milling by using a FEI Helios G4 FIB-SEM system with a Ga ion source. From Fig. 3(a) and 4(a) we can clearly see that both samples have the same thickness of ~240 nm as well as the same grain size of ~30 nm. Furthermore, sample A clearly shows that the grains are grown at an angle whereas sample B clearly shows that the grains are almost perfectly aligned with no angle validating the difference in the mosaicity as indicated by the XRD. This was further quantified by dark field TEM discussed in detail later, which shows that the tilt angle for sample A is ~10° vs. 0° for sample B.

Furthermore, the fast Fourier transforms (FFTs) illustrate that there was no transition region between Si and AlN layers in our samples, unlike the earlier reported AlN thin films^{27, 28} where they distinguish three distinct structural domains namely the Al-bearing amorphous region between Si/AlN interface, the transition region between the amorphous layer and the columnar microcrystals and lastly the columnar grain structure with preferred c-axis alignment. The absence of transition layer in our samples is due to the unique film deposition conditions and high vacuum pressure. The amorphous-like layer at the Si/AlN interface is a native oxide of the Si substrate.

Figure 3: (a) Cross-sectional transmission electron microscopy (TEM) images of 240-nm thick AlN thin film for samples grown at 300 °C Sample A, (b,c) is the columnar region at the location marked with numbers 1-2 for the sample. (d,e) is the corresponding indexed FFTs taken at the region 1-2 and, (f) is the near-interface region with ~ 2 nm amorphous-like layer at the Si/AlN interface marked by 5.

Figure 4: (a) Cross-sectional transmission electron microscope (TEM) images of 240 nm thick AlN thin film for samples grown at 600°C Sample B, (b,c) is the columnar region at the location marked with

numbers 3-4 for the sample. (d,e) is the corresponding indexed FFTs taken at the region 3-4 and, (f) is the near-interface region with ~ 2 nm amorphous-like layer at the Si/AlN interface marked by 6.

Energy dispersive X-ray (EDS) and Secondary Ion Mass Spectrometry (SIMS) was carried out to investigate the amount of oxygen content present in the films. The effect of oxygen content on the thermal transport of AlN thin films has been widely studied²⁹⁻³¹ and it has been reported that having higher content of oxygen impurities reduces the thermal conductivity in these films. Hence to ascertain that we have minimal oxygen content in our deposited films, we maintained superior vacuum condition during film growth with an oxygen partial pressure of 2×10^{-10} Torr according to the in-situ Residual Gas Analysis (RGA), thereby avoiding any oxidizing agent in the chamber.

The SIMS results depicted in Fig. 5, shows nearly identical in-depth elemental distribution of oxygen (O) and carbon (C) for both the samples, indicating that there is negligible difference in the oxygen content between the two samples; and the EDS results showed an oxygen concentration of only around 0.9 at %, indicating that our deposition at low vacuum resulted in minimal presence of oxygen impurities.

Figure 5: SIMS depth distribution of oxygen (O) and carbon (C) shown in Sample A and B.

Thermal Conductivity and Callaway Model

The lattice thermal conductivity (κ_l) of the AlN films were calculated using Equation 3 and fitted with modified Callaway model as described below

$$\frac{t_{tot}}{\kappa_{eff}} = \frac{t_f}{\kappa_l} + R_{Si/AlN}, \quad (3)$$

where t_{tot} is the thickness of AlN film and amorphous like layer between the interface of Si/AlN, κ_l – lattice thermal conductivity, κ_{eff} – the effective thermal conductivity determined from the differential 3ω measurement for the two AlN samples, $R_{Si/AlN}$ – the thermal boundary resistance between the Si and AlN thin film. The thermal boundary resistance of $1 \times 10^{-8} \text{ m}^2\text{K/W}$ was used to calculate the intrinsic thermal conductivity of our films.^{31, 32}

The reduced thermal conductivity of AlN thin films, is due to increased phonon scattering rate at the grain boundaries among others, impurity scattering, and micro-structural defects. To quantify the true contribution of the various phonon scattering parameters we used the Callaway/Klemens' formulation^{5, 33} derived from Boltzmann transport equation used previously^{34, 35} which was modified and discussed in detail below.

This formulation entails the description of various phonon scattering mechanism. The lattice thermal conductivity in above model was given by two terms $\kappa_l = \kappa_1 + \kappa_2$, where κ_1 and κ_2 are calculated by.

$$\kappa_1 = C \int_0^{\theta_D/T} \frac{\tau_C x^4 e^x}{(e^x - 1)^2} dx \quad \text{and} \quad (4)$$

$$\kappa_2 = C \frac{\left\{ \int_0^{\theta_D/T} \left(\frac{\tau_C}{\tau_N} \right) x^4 e^x (e^x - 1)^{-2} dx \right\}^2}{\int_0^{\theta_D/T} \frac{\tau_C}{\tau_N \tau_R} x^4 e^x (e^x - 1)^{-2} dx}, \quad (5)$$

where $x = \hbar\omega/k_B T$, ω is the phonon frequency. $C = \frac{k_B^4 T^3}{2 \hbar^3 \pi^2 v}$, k_B - Boltzmann's constant, $\hbar$ - is the Plank constant, v – average group velocity, τ_i - relaxation time, i – N, R and C are Normal, resistive and combined relaxation times respectively. The combined relaxation rates are given by Matthiessen rule $\tau_C^{-1} = \tau_R^{-1} + \tau_N^{-1}$.

Table 2 shows the various parameters used in calculating the lattice thermal conductivity of AlN film.

Sample	θ_D (K)	Thickness (nm)	Grüneisen Parameter	Thermal conductivity (Wm ⁻¹ K ⁻¹)	Ref.
Bulk AlN	950	7.3×10 ⁶	0.7	293	³⁶
Sample A	840	240	0.7	4.22	This work
Sample B	840	240	0.7	8.09	This work
Thin film	-	268	-	3.1	²⁷
Thin film	-	1000	-	9.9	²⁸
Thin film	-	299	-	5.4	³⁷

First, we consider, the lattice thermal conductivity modeling for the single crystal bulk AlN sample shown in Figure 6(a), the parameters used to fit the Callaway and Klemens' formulation are listed in Table 2. We found our parameter to be in line with the reported literature values for bulk single crystal AlN samples.^{30, 36} For a pure defect-free sample ($\tau_N \approx \tau_R$) the normal relaxation rate is comparable to the resistive process. However, in real sample the, resistive relaxation rate includes phonon-phonon Umklapp scattering, point defect scattering, boundary scattering to mention a few. We consider Umklapp (τ_U), boundary (τ_B), impurity (τ_I) and normal (τ_N) scattering rates. Hence the total combined scattering rate or relaxation time is given by the Matthiessen's rule (τ_C)

$$\frac{1}{\tau_C} = \frac{1}{\tau_U} + \frac{1}{\tau_I} + \frac{1}{\tau_B} + \frac{1}{\tau_N}. \quad (6)$$

The phonon-phonon Umklapp scattering rate given by Slack *et al.*³⁸ suggested the following form for the Umklapp rate

$$\tau_U^{-1}(x) = B_U \left(\frac{k_B}{h}\right)^2 x^2 T^3 e^{-\left(\frac{\theta_D}{3T}\right)} \text{ and} \quad (7)$$

$$B_U \approx \frac{\hbar \gamma^2}{M v^2 \theta_D}, \quad (8)$$

where M is the average atomic mass, γ - Grüneisen parameter varies from negative to positive for zone-center and zone boundary phonons modes³⁹ and was taken to be 0.8 for bulk single crystal AlN⁴⁰ and 1.162 for the thin films samples⁴¹.

The phonon scattering rate due to impurities in the Callaway model is given by

$$\tau_I^{-1} = A \omega^4, \quad (9)$$

where A ($5 \times 10^{-46} \text{ s}^3$) is an adjustable parameter that depends on impurity concentration in

the highly pure bulk single crystal AlN.⁴⁰ Polycrystalline samples on the other hand, can have a higher concentration of oxygen impurities and thus greater effect to the thermal conductivity even at higher temperatures. Yagi *et al.*²⁹ investigated the effect of oxygen content in polycrystalline AlN films with similar grain sizes. They report A values in the range of 3×10^{-44} for AlN samples with oxygen content of 1.1 at %, and 7×10^{-43} for samples with oxygen content of 13.7 at %. Fitting our experimental data to the grain boundary and impurity scattering alone we extracted an impurity fitting parameter A to be in the range from $0.1 - 3.0 \times 10^{-43}$ for sample A and B respectively which correspond to oxygen content higher than the 13.7 at %. As we showed earlier, we have an oxygen content less than 1 at %, therefore we took a worst-case scenario A value of 3×10^{-44} for both samples since there is no significant difference in the oxygen impurity concentration between them as evident from the SIMS analysis.

The normal scattering rate

$$\tau_N^{-1} = B_N \omega^2 T^3 \text{ and} \quad (10)$$

$$B_N = \frac{k_B^4 \gamma^2 V}{M \hbar^3 v^5}, \quad (11)$$

where M is average atomic mass.

Gray Boundary Scattering

The phonon boundary scattering at the grain boundaries is given by Casimir⁴² as

$$\tau_B^{-1} = v/d, \quad (12)$$

where d is the experimentally determined grain size. This is the traditional way of estimating the effect of grain boundary phonon scattering, and Fig. 6(a) (dotted red line) illustrates the calculated thermal conductivity by taking this into account and using an average grain size $d = 30$ nm. It is evident by our calculations, that this treatment of the grain boundary scattering in the Callaway model failed to capture our experimental values. This indicates that either the gray approximation is not adequate, or a smaller length scale (Casimir length) is needed in the place of the grain diameter. More recently, Moraes *et al.*⁴³ used similar Callaway model to fit the thermal conductivity of the AlN ($\text{Al}_{0.90}\text{Cr}_{0.1}\text{N}$) films of thickness 1180 nm. To describe the suppression of thermal conductivity in the thin AlN film from bulk, a different boundary scattering parameter $\tau_B^{-1} = B$ was used, where B is a fitting parameter. Using Eq. 11 to obtain the boundary scattering relaxation time B of 5.64×10^{12} (1/s) and the average AlN phonon velocity of 6237 (m/s)³⁷ results in an average grain size d of 10 Angstrom. In the Casimir limit, having such a low grain size does not physically make sense. Thereby, simply fitting the parameter B to the thermal conductivity of thin film, results in erroneous contribution of boundary scattering to the phonon transport.

Figure 6: (a) Temperature dependent thermal conductivity for experimental bulk AlN (ref. ³⁶) and the Callaway model fitted for bulk as well as gray model. (b) Temperature dependent thermal conductivity for thin film AlN samples A&B, both experimental and Callaway model fitted with various grain boundary scattering rates such as non-gray boundary scattering, dislocation scattering and stacking fault scattering.

Non-Gray Boundary Scattering

Reduction in the thermal conductivity due to the grain boundary is widely considered as frequency-independent (gray) Casimir model. Wang *et al.*¹³ used frequency-dependent grain boundary (non-gray) model developed by Ziman⁴⁴ to define the grain boundary scattering and Hua *et al.*⁴⁵ used Monte Carlo simulation which showed the frequency-dependence transmissivity at the grain boundary to be consistent with $\tau_B^{-1} \approx \omega$. In Fig. 6(b), the Frequency-dependent A and B curves show the fitting by treating the grain boundary scattering as¹³

$$\tau_B \approx \alpha \beta \left(\frac{\omega_o}{\omega} \right) \left(\frac{d}{v_g} \right), \quad (13)$$

where α is the phonon transmission coefficient at the grain boundary, $\beta = 0.7097$ is a fitting parameter for most of Debye and Born-von Karman phonon dispersions, ω_o is the phonon maximum frequency of 17.9 THz which was calculated from the phonon density of state approximated by the Debye model and was found to be in line with the acoustic phonon frequency measured elsewhere.⁴⁶ Table 3 lists the parameter that fit the data for our samples. The only tunable parameter in our fitting is the phonon transmission coefficient α , which for our case showed sample B has higher phonon transmission than sample A. This is expected since the angle of tilt in sample A is greater than sample B which increases the resistance to the heat flow. Furthermore, this can also be due to the difference in the grain quality between the

two samples. The larger angle of sample A could cause a rougher interface at the grain or a higher density of defects such as stacking faults, which we investigate later on. Even though this model fits the data, it lacks the physical insight as to what transpires at the grain boundaries and how the phonons scatter.

Table 3 Shows fitting parameter used for non-gray boundary scattering.

Sample	α	β	$\omega_o(\text{THz})$
Sample A	0.50	0.7097	17.95
Sample B	0.99	0.7097	17.95

Stacking Fault Scattering

As described earlier the potential difference of defect density at the grain boundary could dominate cross-plane thermal conduction beyond boundary scattering. The AlN crystal structure is characterized by the stacking sequence ...ABABABAB... along the direction perpendicular to the basal plane and the presence of these stacking faults will scatter phonons at the rate given by equation (13).^{8, 47}

$$\tau_{SF}^{-1} = 0.7 (6\pi^2)^{\frac{2}{3}} \gamma^2 N_S v \left(\frac{T}{\theta_D}\right)^2 x^2, \tag{14}$$

where N_S is the number of stacking faults per unit length and was used as a fitting parameter in the modified Callaway model. Table 4 shows the fitted values and the respective curves are shown in Fig. 6(b). Our fitting indicates sample A has higher density of stacking faults than sample B, which can be expected based on the crystal quality difference evident from the TEM. To the best of our knowledge, no other detailed microstructural studies on stacking fault density for sputtered AlN thin films are available for comparison to this date, but values of $5 \times 10^6 \text{ cm}^{-1}$ have been reported for wurtzite AlN thin films grown by plasma assisted molecular beam epitaxy.⁴⁸ In our case having a stacking fault density $\sim 10^6 \text{ cm}^{-1}$, indicates that the films were of high quality. We can see that the stacking faults scattering can capture the temperature dependent trend of thermal conductivity better than the Non-gray scattering discussed above indicating that the frequency dependence of the stacking fault scattering rate gives a better physical understanding of the phonon scattering mechanism than the Non-gray model.

Dislocation Scattering

To understand the scattering of phonon at the grain boundaries, Kim *et al.*¹⁴ considered grain boundary as an array of low angle sessile dislocation whose formulation was given by Klemens.⁵

Essentially the phonon scattering of dislocation consists of scattering by the dislocation cores (τ_{DC}) and by the dislocation strain (τ_{DS}).

$$\tau_{DC}^{-1} = N_D \left(\frac{v_L^4}{v_T^2} \right) \omega^3 \text{ and} \quad (15)$$

$$\tau_{DS}^{-1} = \eta B_{Def}^2 N_D (\gamma + \gamma_1)^2 \omega \left[\frac{1}{2} + \frac{1}{24} \left(\frac{1-2r}{1-r} \right)^2 \left\{ 1 + \sqrt{2} \left(\frac{v_L}{v_T} \right)^2 \right\}^2 \right], \quad (16)$$

where η is the weight factor 1, 0 and 0.55 for dislocation line perpendicular, parallel and random to the temperature gradient respectively, B is the Burgers vectors 2.54 Angstrom⁴⁹, N_D is dislocation density, r the Poisson's ratio and $v_{L(T)}$ is phonon longitudinal and transverse velocity for the thin film AlN.⁴³ The dislocation density for AlN film is reported to vary in the range of 10^8 - 10^{11} cm⁻² for various deposition methods such as molecular beam epitaxy (MBE),⁵⁰ metal-organic chemical vapor deposition (MOCVD)⁵¹ and sputtering.⁵² Unfortunately, we were unable to quantify the dislocation density in the films by structural characterization so we had to fit these values by taking some assumptions with regards to the weight factor. We first fitted the data by assuming the dislocations were randomly oriented with a weight factor $\eta=0.55$, since this is the traditional method of treating the dislocation scattering rate. By doing so, we were able to fit our data by assuming a dislocation density of 1.8×10^9 and 5×10^8 cm⁻² for sample A and B respectively. Having such a low dislocation density of $\sim 10^9$ cm⁻², is not expected in sputtered AlN films. Hence, we assumed a dislocation density of 1×10^{10} cm⁻² reported in literature^{53, 54} for sputtered thin AlN films on different substrates to fit the modified Callaway model for both our samples as shown in Fig. 6(b) and Table 4. To help quantify the effect of the mosaicity (c-axis tilt) to thermal transport we kept the dislocation density constant and varied the weight factor (η). Physically, the dislocation density of both the samples can be assumed to be similar, as the annihilation of defects in AlN thin films where shown to be around 1200 °C⁵⁵ and since the highest growth temperature of our films does not exceed 600 °C, our assumption of similar dislocation density for both the sample should suffice. Our fitting shows that η is 0.2 and 0.05 for samples A and B respectively. This indicates that the dislocation lines for sample A are at a higher tilt angle than sample B with respect to the temperature gradient. As it is evident from the TEM in Fig. 4 and also from the dark field TEM in Fig. 7, the tilt angle for sample A is $\sim 10^\circ$ vs. almost 0° for sample B validating that the mosaicity can indeed affect the thermal transport in these AlN films.

Our work here illustrates that dislocations seem to have a significant effect to the thermal conductivity in the studied temperature range, and more importantly the mosaicity directly affects the magnitude of the effect of dislocations as predicted by Klemens. This was not shown till this date, since to the best of our knowledge the dislocations were at random directions with respect to the temperature gradient that always lead to the fixed $\eta = 0.55$. In our work by

controlling the mosaicity we were able to control the direction of the dislocation lines and thus η , and in essence transition the line between Rayleigh to Mie scattering (Fig. 7(a,b)).

Table 4 The various parameter used to fit the modified Callaway model which include dislocation scattering and stacking fault scattering mechanism.

AlN Thin Films	Sample A	Sample B
$N_D \times 10^{10} (cm^{-2})$	1	1
$\omega_o(THz)$	17.94	17.94
r (poisson's ratio)	0.23	0.23
v_L	6230	6230
v_T	5340	5340
η	0.20	0.05
$N_S \times 10^6 (cm^{-1})$	0.9	0.43
$A \times 10^{-44} (s^3)$	3.0	3.0

Figure 7: Illustrates the (a,b) phonon scattering at the grain boundaries parallel to the heat current. Two beam dark field images showing c-axis tilt varying from 10^0 to 0^0 for (c,d) A and (e,f) B samples respectively.

Fernandez *et al.*²⁷ recently studied the effect of structural inhomogeneity and defects in polycrystalline AlN thin films and developed a model based on the thermal conductivity of the three distinct regions interface, transition and columnar. They showed enhancement in the AlN film by increasing the thickness of the film and reducing the transition region for their samples.

For the ease of theoretical modeling, they assumed that the grain interface is rough hence all the phonon scatter diffusively. Along similar lines, Pan *et al.*²⁸ showed thermal transport enhancement for 1- μm -thick AlN films by varying the growth temperature which lead to reduced transition region as a function of growth temperature. The enhancement in the thermal conductivity for their respective sample was contributed to the transition layer thickness and no comparison of mosaicity was done in either study. However, we show an enhancement in the thermal transport of more than 90% for the same thickness of AlN film. We contribute the enhancement of thermal conductivity to the smoother grain interface obtained by controlling the mosaicity in our sample. The absence of transition layer in our samples helps us better understand the role of grain interface quality in the thermal transport and thus were able to clearly correlate the increase in the thermal conductivity due to the c-axis alignment for our thin AlN films and discuss the various phonon scattering mechanism which provides more insight in the thermo-microstructural relationships and thereby furnishing a tuning knob for adjusting the thermal properties in polycrystalline materials.

Conclusion

In this work, we studied the temperature-dependent thermal conductivity of AlN thin films and showed an enhancement in their thermal conductivity by varying the c-alignment axis (mosaicity) measured using the 3ω technique. The enhancement of the thermal transport was attributed to the better-quality grain interface which results in the Mie scattering of phonons parallel to the c-axis. Modified Callaway model was used to fit the theoretical curves to the experimental result using various phonon scattering mechanisms at the grain interface. It was found that treating grain boundaries as an array of sessile dislocation lines with varying angle relative to the heat flow, best describes the mechanism of the thermal transport. However, using non-gray model gives an overview of the phonon scattering at the grain boundaries. Lastly, our results show that controlling the quality of the grain interface provides a tuning knob to control the thermal transport in the polycrystalline materials.

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