THE INTRINSIC THERMAL CONDUCTIVITY OF AIN†

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Abstract—The thermal conductivity, λ , of high purity single crystals of AlN has been measured from 0.4 to 1800 K. These results show that the heat conduction is by phonons and that at the lowest temperatures the phonon mean-free-path, l, is limited by boundary scattering. The maximum $\lambda = 23$ W/cmK is at 52 K. Above 150 K the value of l is determined by phonon-phonon scattering and at 300 K the value of λ for pure AlN is 3.19 W/cmK (80% of that of pure copper). Above 500 K the value of λ decreases as $T^{-1.25}$.

Keywords: Aluminum nitride, thermal conductivity, phonon scattering, adamantine compounds.

1. INTRODUCTION

Aluminum nitride is one of the few high thermal conductivity non-metallic solids. It was pointed out by Borom et al. [1] that AlN should be a very good phonon heat conductor, but that phonon scattering by residual impurities limited its thermal conductivity to values considerable smaller than those predicted by theory. A general review of non-metallic crystals with high thermal conductivity by Slack [2] showed how pure AlN should compare with diamond. silicon, germanium and other high thermal conductivity solids. At that time [2] an impure single crystal of AlN was found which had a value of $\lambda = 2.0 \text{ W/cmK}$ at 300 K. The other literature data on the λ of AlN was also summarized [2]. Since then measurements have been reported by Slack and McNelly [3] on a single crystal with $\lambda = 2.53$ W/cmK at 301 K. Some of the results in this present paper on single crystal and polycrystalline AlN for temperatures in the range 100-1800 K were reported by Slack [4] in 1979. Data for pyrolytic, polycrystalline AlN have been reported by Fridlender et al. [5] for 1300-1830 K; while data on ceramic AlN have been reported by Sakai et al. [6], Inomata [7], Rafaniello et al. [8], Koshchenko et al. [9], Takeda et al. [10], Bentsen and Hasselman [11], and Kuramoto and Taniguchi [12]. The highest λ value at room temperature of 2.7 W/cmK was reported by Takeda et al. [10] for a two-phase mixture of 98.4 mol% AlN + 1.6 mole% BeO. The next-highest λ value reported was 0.95 W/cmK by Kuramoto and Taniguchi [12] for ceramic containing 0.6 wt.% oxygen in solid solution. The high λ of AlN has importance in its technological use as a heat-sink substrate for semiconductor devices [12].

In addition to the work on AlN, results have been reported [8, 11] for the λ of AlN-SiC solid solutions at room temperature and above.

2. SAMPLES AND APPARATUS

The preceeding work [1, 2, 7, 8, 11, 12] on the λ of AlN demonstrated that impurities, particularly oxygen, decrease the λ considerably below its intrinsic value at all temperatures. Thus high purity, singlecrystal samples are needed to obtain the intrinsic λ . A technique for growing single crystals of AlN by sublimation has been devised by Slack and McNelly [3, 14]. In this technique crystals are grown from high-purity AlN powder by vapor-phase transport in a sealed tungsten crucible. One such single crystal weighing 1.00 g, 1.41 cm long, and with an average diameter of 0.54 cm was used for the present measurements from 0.4-300 K. No other samples of comparable size were available, so the data are limited to one sample. The heat flow was along the c-axis. From 370 to 1800 K oligocrystalline samples of high-purity sublimed AlN in the form of flat discs 1.2 cm in diameter and 0.3 cm thick were used. The average crystallite size in these discs was about 0.2 cm diameter. Again the heat flow was predominantly along the c-axis. The λ of the single-crystal sample was measured by using three different steady-state, longitudinal-heat-flow apparatuses. From 0.4 to 1.7 K a He³ cryostat [15] was used, from 1 to 135 K

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a He⁴ cryostat [16] was used. Both of these are located at Cornell University. From 145 to 300 K a cryostat cooled by gaseous, cold nitrogen [17] was employed. In this third cryostat the sample is placed in a vacuum; it is surrounded by a close fitting thermal shield whose temperature distribution matches that in the sample, and the temperatures are measured with thermocouples. Heat is suppled at one end of the sample by a wire-wound resistor. Values of λ for these steady-state methods are estimated to be accurate to +5%. The results from 370 to 1800 K were measured using a laser-flash diffusivity technique [18]. This technique has been described by Danielson and Sidles [19]. The laser was a neodymium-doped glass unit operating at 1.06 microns with pulse lengths of 10⁻⁴-10⁻³ s, the infrared detector was InSb, and the sample surfaces were coated with colloidal graphite during the test to avoid laser shine-through. The sample was held in an inert atmosphere in a graphite resistance furnace in order to control its temperature. Values of λ are estimated to be accurate to $\pm 10\%$.

An impurity analysis of the AlN was conducted. Neutron activation [20] showed 343 ± 17 ppm oxygen by weight. The tungsten content was about 50 ppm by weight [14]. The oxygen contamination came from the starting powder; the tungsten contamination came from the crucible. Other impurities were at the 10 ppm level or less. The deviation from stoichiometry can be judged from the optical absorption curves in the $15,000-45,000 \, \text{cm}^{-1}$ wavenumber region [3]. The absorption was similar to sample W-154 (see Fig. 5 of Ref. [3]). Hence we estimate that the Al to N ratio was unity to 1 part in 10^4 or higher.

3. RESULTS

The composite λ vs T results are shown in Fig. 1. Data at representative temperatures taken from Fig. 1 are given in Table 1. From 0.4 to 300 K the results are for heat flow along the c-axis of the hexagonal structure crystal, above 300 K the crystallites in the disc samples were primarily oriented with the c-axis parallel to the heat flow. Since AlN has the hexagonal wurtzite structure one would like to know the λ perpendicular to the c-axis. However, no samples suitable for this measurement were available. Thus, one can make some estimate of the anisotropy as follows. The anisotropy in the sound velocity in AlN is about 5% based on the elastic constants measured by Tsubouchi et al. [21]. Comparing the data [22] on the λ of BeO with the present data on AlN it is expected that the anisotropy in the thermal conductivity of AlN should be less than 20% between values parallel and perpendicular to the c-axis at low temperaures and less than 5% at room temperature and above. The structures of AlN and BeO are identical and the anisotropy in their elastic constants [21, 23] is very similar. The λ anisotropy in ZnO, which possesses the same structure, has also been measured [24] and is about 20% from 30 to 300 K.

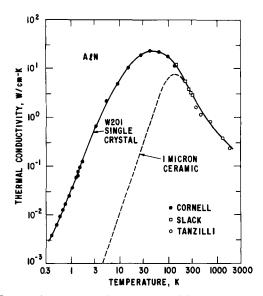


Fig. 1. The measured thermal conductivity of high purity AlN vs temperature (solid curve). The dashed curve shows the calculated behavior for AlN ceramic with a 1 micron grain diameter. Sample W201 was measured from 0.4 to 300 K, solid circles and open squares. The open circles are for an oligocrystalline sample measured by laser flash.

Because AlN is technologically useful as a high-thermal-conductivity non-metal both the absolute value and the temperature dependence of λ are important. We have found that $\lambda=2.85$ W/cmK at 300 K in sample W-201 (see Fig. 1 and Table 1). This value is higher than any previously reported values, and suggests that the present sample is purer than the others measured to date. The temperature dependences found are, we believe, closer to those of

Table 1. Thermal conductivity vs temperature for AlN

T	W-201	Pure AlN†
(K)	(W/cmK)	(W/cmK)
0.4	0.0038	0.0038
0.6	0.0097	0.013
1.0	0.032	0.058
2.0	0.21	0.48
4.0	1.08	3.8
6.0	2.4	20
10.0	5.7	58
15	10.2	195
20	15.0	450
30	20	700
45	23	460
60	22	205
100	17.5	47
150	11.0	15.7
200	6.5	7.8
300	2.85	3.19
400	1.80	1.95
600	0.96	1.00
1000	0.48	0.49
1800	0.24	0.24

[†] Corrected for the residual oxygen content.

pure AlN than the previous literature. From Fig. 1 the exponent, e, in $\lambda = aT^e$ has values of:

$$e = +2.54$$
 for $0.4 < T < 3$ K,

$$e = -1.25$$
 for $500 < T < 1800$ K. (1)

The maximum λ occurs at 52 K and is $\lambda = 23 \text{ W/cmK}$.

4. ANALYSIS OF THE RESULTS

4.1. Sound velocity and Debye temperature

In AlN there are no free electrons so that the dominant mechanism of heat transport is phonons. Two important parameters [25] for phonon heat transport are the sound velocity, v, and the Debye temperature, θ .

A value for θ at absolute zero can be calculated from the measured elastic constants [21, 26] using the procedure developed by Wolcott [27]. The result, using room temperature elastic constants [21], is: $\theta_0 = 903 \text{ K}$. The polycrystalline elastic constants [26] give $\theta_0 = 950 \text{ K}$ using Anderson's method [28] of calculation. This is the preferred value. Some low temperature specific heat capacity measurements on AIN have been made by Koshchenko *et al.* [29]. These data give a θ value of $800 \pm 2 \text{ K}$ for temperatures below 5 K. It is believed by the present authors that this value is too low.

4.2. Absolute value of thermal conductivity

The absolute value of λ at a temperature equal to the Debye temperature can be calculated by the method used by Slack [25]. The equation is:

$$\lambda'(\widetilde{\theta}_{\infty}) = Bn^{1/3} \overline{M} \delta(\widetilde{\theta}_{\infty})^{2} (\widetilde{\gamma}_{\infty})^{-2}, \tag{2}$$

where

 $B = 3.04 \text{ W/g cm}^2\text{K}^3$

n = number of atoms in a primitive unit cell = 4,

 \overline{M} = average atomic mass = 20.49 g,

 $\delta = (average atomic volume)^{1/3} = 2.186 \text{ Å},$

 $\tilde{\theta}_{\infty}$ = acoustic branch Debye temperature in the high temperature limit

and

 $\tilde{\gamma}_{\infty}$ = Grüneisen parameter for acoustic phonons in the high temperature limit.

The thermal Grüneisen parameter at high temperatures derived from the average thermal expansion coefficient [30], the bulk modulus of Gerlich et al. [26] and the specific heat capacity [31, 32] is:

$$\gamma_{\infty} = 0.77. \tag{3}$$

It will be assumed that the acoustic phonon value of $\tilde{\gamma}_{\theta}$ is equal to 0.77 also. This value is quite similar to values tabulated [25] for other adamantine crystals.

The value of $\tilde{\theta_0}$ is given by:

$$\tilde{\theta}_0 = \theta_0 / n^{1/3}. \tag{4}$$

Based on a comparison of AlN with SiC, BP and other adamantine compounds [25] it is assumed that for AlN:

$$\tilde{\theta}_{\infty} = \tilde{\theta}_0 / 1.16 = 516 \text{ K}. \tag{5}$$

These values give a calculated value of

$$\lambda'(516 \text{ K}) = 0.97 \text{ W/cmK}.$$
 (6)

The experimental value of κ for pure AlN at 516 K from Table 1 is:

$$\lambda(516 \text{ K}) = 1.25 \text{ W/cmK}.$$
 (7)

The agreement between λ and λ' is comparable to that found [25] for AlAs and AlSb.

4.3. Temperature dependence of thermal conductivity

At $T > \tilde{\theta}_{\infty}$ the exponent e given by:

$$e = +\left(\frac{\partial \ln \lambda}{\partial \ln T}\right)_{P} \tag{8}$$

is:

e = -1.25 for sample W-201,

$$e = -1.69$$
 for pure AlN (Table 1). (9)

The subscript P denotes constant pressure conditions. This value of e is very similar to that found [25] for many other adamantine crystals, and is indicative [25] of a λ influenced by both acoustic-acoustic and

acoustic-optic phonon scattering. The temperature dependence of λ at the very lowest temperatures is indicative of thermal conductivity limited by phonon scattering from the crystal boundaries [22]. An average mean-free-path, l, for the phonons can be calculated from:

$$l = \frac{3\lambda}{vC},\tag{10}$$

where \bar{v} is the average sound velocity of:

$$\bar{v} = 6.98 \times 10^5 \,\mathrm{cm} \,\mathrm{s},$$
 (11)

and C is the specific heat capacity calculated from the Debye expression at low temperatures:

$$C/C_{\infty} = 77.92727(T/\theta_0)^3$$
. (12)

The specific heat capacity at $T\gg\theta_0$ is the Dulong-Petit value, C_∞ . From the results in Fig. 1

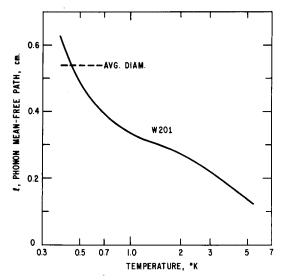


Fig. 2. The phonon mean-free-path for a single crystal (W201) of AlN calculated from eqn (10).

and eqn (10) the experimental l vs T plot in Fig. 2 is obtained. The value of l is equal to the average sample diameter of 0.54 cm at 0.45 K. Thus λ reaches the boundary scattering limit at the lowest temperatures. Whatever impurity scattering of phonons that may be present due to the residual tungsten, oxygen or nitrogen, the deficit has finally disappeared at these temperatures.

4.4. Thermal conductivity at room temperature

It was shown [2] in 1973 that pure AlN was expected to have a $\lambda > 1$ W/cmK at room temperature, and thus should be considered as a "high thermal conductivity" non-metallic crystal. In this study a Leibfried-Schlomann scaling parameter of $\overline{M}\delta\theta_0^3$ was used to order the increasing values for the expected thermal conductivity at room temperature of numerous crystals with an adamantine structure. This scaling parameter is derived from the numerator

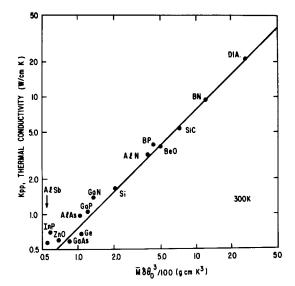


Fig. 3. The thermal conductivity at 300 K of several adamantine structure crystals as limited by intrinsic phonon-phonon scattering plotted vs a Leibfried-Schlömann scaling parameter (see Ref. [2]). The effect of isotope scattering has been subtracted.

in eqn (2) multiplied by $(\tilde{\theta}_{\infty}/T)$. Equations (4) and (5) are also applied to convert to θ_0 .

With the present new results a similar comparison with other high-thermal-conductivity crystals is given in Table 2 and Fig. 3. Since 1973 some more data have become available in the literature [33–59] on λ values at room temperature and the Debye temperatures at absolute zero, θ_0 .

4.4.1. Isotope correction. In Fig. 3 the measured λ has been corrected for isotope scattering in order to obtain the thermal conductivity, λ_{pp} , as determined only by phonon-phonon scattering. The isotope scattering has been estimated from the natural isotopic abundances of the elements and the additive resistivity approximation of Ambegaokar [60]. The added refinement of including the ratio of phonon-

Table 2. Thermal conductivity at room temperature for several adamantine crystals

	λ		100 W,	λ_{pp}	θ_0	$M\delta\theta^3 \times 10^{-2}$	R	ef.
Crystal	(W/cmK)	$\Gamma \times 10^6$	(cm K/W)	(W/cmK)	(K)	$(g \cdot cm \cdot K^3)$	λ	θ_0
Diamond	20.0	6.38	0.27	21.1	2240	24.07	[2]	[2]
Bn	7.6	43.95	2.40	9.3	1750	12.02	[33]	[34, 36]
SiC	4.9	17.57	1.70	5.4	1185	7.25	[2]	[37, 38]
BeO	3.7	2.24	0.18	3.7	1280	4.99	[2]	[38-40]
BP	3.6	15.16	1.86	3.9	975	4.39	[2]	[41, 42]
AlN	3.19	0.35	0.04	3.19	950	3.84	Ť	†
Si	1.56	16.74	3.70	1.66	648	2.07	[43]	[44]
GaN	1.3	22.89	5.17	1.39	525	1.360	[45]	[46]
GaP	1.0	15.77	5.11	1.05	444	1.201	[47, 48]	[49, 50]
AlAs	0.98	0	0	0.98	417	1.042	[51]	[52]
InP	0.70	1.29	0.62	0.70	301	0.583	[53]	[25]
Ge	0.60	48.95	19.56	0.68	374	1.074	[43]	[44]
ZnO	0.54	64.39	18.53	0.60	419	0.683	[24]	[58, 59]
GaAs	0.58	7.64	3.29	0.59	347	0.854	[54]	[55]
AlSb	0.56	7.38	4.14	0.57	289	0.551	[56]	[57]

[†] This work.

scattering rates due to normal and umklapp processes [56, 61] is omitted. This inclusion increases [56] the calculated isotope thermal resistivity, W_I . Thus [60]

$$\lambda^{-1} = \lambda_{pp}^{-1} + \Delta W_I. \tag{13}$$

For calculating ΔW_i , one has [56, 60]:

$$\Delta W_I = (6\pi^2)^{2/3} h \delta \Gamma / k^2 \theta, \qquad (14)$$

with h = Planck's constant, k = Boltzmann's constant and $\Gamma = \text{isotope}$ scattering parameter. For a single element with i different isotopes [62]:

$$\Gamma = (1/12) \sum_{i} c_{i} [(m_{i} - \bar{m})/\bar{m}]^{2},$$
 (15)

where c_i is the abundance of the *i*th isotope of mass \bar{m}_i , and \bar{m} is the average mass for all of the isotopes. For a compound AB composed of two elements A and B the Γ value is a weighted average [62] of those of the two elements. Note that in Table 2 the isotope correction is especially important for BN, Ge and ZnO. The room temperature thermal conductivity of cubic BN can be increased by 22% by using a single isotope of boron in the crystal. The isotope effect in AlN at room temperature is insignificant.

4.4.2. Intrinsic limit. The graph in Fig. 3 shows that the present results of 300 K for pure AlN are in good agreement with the data on other pure adamantine-structure crystals when $\overline{M}\delta\theta_0^3$ is used as a scaling parameter. Thus, the value of $\lambda = 3.19 \text{ W/cmK}$ is very close to the intrinsic limit for pure AlN.

4.5. Phonon scattering by defects in AlN

4.5.1. Oxygen. The oxygen in AlN appears [2] to be a substitutional impurity in the crystal. The oxygen is incorporated as dissolved Al₂O₃ according to:

$$3(1-\epsilon)AlN + \epsilon Al_2O_3 \rightarrow Al_{3-\epsilon} \square_{\epsilon} (N_{1-\epsilon}O_{\epsilon})_3.$$
 (16)

Here ϵ is defined as:

$$\epsilon = \Delta n/n_0, \tag{17}$$

where Δn is the number density of the oxygen impurities and n_0 is the number density of the nitrogen atoms in AlN; $n_0 = 4.79 \times 10^{22} \,\mathrm{cm}^{-3}$. The oxygen replaces nitrogen and the aluminum deficit produces vacancies in the aluminum sublattice. These defects produce [14] a thermal resistivity at room temperature of:

$$(\Delta W/\epsilon) \approx 43 \text{ cm K/W}.$$
 (18)

For our present crystal with 340 ppm by weight of oxygen $\epsilon = 8.71 \times 10^{-4}$. Thus,

$$\Delta W = 0.172 \text{ cm K/W}.$$
 (19)

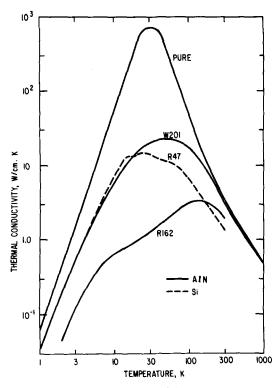


Fig. 4. The thermal conductivity of AIN vs temperature as influenced by oxygen impurities. The "PURE" curve has been estimated for a single crystal with no oxygen and a sample diameter of 0.54 cm. Curves W201 and R162 were measured for single crystals of AIN containing oxygen (see Table 3). Curve R47 was reported for a silicon single crystal containing oxygen (see Refs [63-65]).

Using eqn (13) and replacing ΔW_i , by ΔW we obtain for λ_{pure} at 300 K:

$$\lambda_{pure} = 3.19 \text{ W/cmK}. \tag{20}$$

This is very close to the value previously predicted [2] in 1973 for pure AlN of 3.2 W/cmK.

Let us now consider the data on sample R162 which is much less pure. In the single crystal sample of AlN of Ref. [2] the Δn is $300 \pm 200 \times 10^{18} / \text{cm}^3$ and $\epsilon = 63 \pm 40 \times 10^{-4}$. Its λ vs T curve is shown in Fig. 4. Its thermal conductivity is substantially lower than that of W-201, and there is a suggestion of a "resonance-dip" in the curve at about 30 K. Measurements of λ vs T for silicon by Holland [63, 64] and Holland and Neuringer [65] show signs of resonance scattering by oxygen impurities in silicon. The results for their sample R47 are given in Fig. 4. It contains 1.1×10^{18} /cm³ of oxygen and 9.7×10^{14} /cm³ of boron. It appears to show "resonance-dips" at 18 and 40 K. It is not intended to imply that all of the oxygen impurities in AlN have a local atomic structure identical to those in Si. However, it is clear that very small amounts of oxygen in an otherwise clean crystal can drastically reduce the thermal conductivity. Some fraction of the oxygen in AlN may act as resonance

Table 3. Oxygen content of various crystals

Material	Sample No.	Oxygen content (10 ¹⁸ atoms/cm ³)	
AIN	W201	42 ± 2	
AIN	R162	300 ± 200	
Si	R47	1.1	

scattering centers. The oxygen concentration in the AlN sample R162 is about 300 times that in the most heavily doped Si sample, R47 (see Table 3).

4.5.2. BeO and SiC impurities. The thermal conductivity of a ceramic sample of AIN containing 3.0 mole% BeO in solid solution has been measured [17] in the present series of studies at 300 K as $\lambda = 0.18$ W/cmK. This translates into

$$(\Delta W/\epsilon) = 175 \text{ cm K/W}. \tag{21}$$

There are literature data [8, 11] available for ceramic AlN samples doped with SiC. These translate into, at low SiC concentrations,

$$(\Delta W/\epsilon) = 8.0 \text{ cm K/W}. \tag{22}$$

We can make a theoretical estimate of the impurity-induced thermal resistivity increase as follows. A value of Γ/ϵ can be calculated from eqn (5) of Ref. [2] as:

$$\Gamma/\epsilon(1-\epsilon) = 1/12[(\Delta M/\bar{M})^2 + 40(\Delta\delta/\delta_0)^2]. \tag{23}$$

The first term of eqn (23) is the mass difference, the second is the lattice misfit. For $\epsilon \to 0$ this is just Γ/ϵ . This value can be inserted into eqn (14) to yield the $\Delta W/\epsilon$ values in Table 4. The $\Delta \delta/\delta_0$ value for Al₂O₃ in AlN is taken from Ref. [2], where δ_0 is the average volume per atom in pure AlN. This calculation shows that the measured and calculated $\Delta W/\epsilon$ values are satisfactory for BeO and Al₂O₃ impurities, but that the measured value for SiC impurities is too high by a factor of about 5. It is probable that some oxygen contamination was present in the AlN-SiC mixes that were used [8, 11], and this is responsible for the high $\Delta W/\epsilon$ values measured. The calculated ΔW value for oxygen as Al_{0.67}O in the AlN lattice is 48 times larger than that of SiC in AlN. Thus, unless the samples are very low in oxygen, it is difficult to

Table 4. Comparison of measured and calculated $\Delta W/\epsilon$ values for impurities in AlN at 300 K

Impurity	$\Delta W/\epsilon$ (cm K/W) calc., eqn (23)	$\Delta W/\epsilon$ (cm K/W) meas., eqn (13)	δ (Å)	$\Delta\delta/\delta_0$ †
BeO	237	175	1.904	-0.129
Al_2O_3	70	43	2.165‡	-0.021
SiC	1.5	8.0	2.180	0.0027

 $[\]dagger \delta_0 = 2.18 \,\text{Å}$ for AlN.

measure the true effect of SiC as an additive by itself. We estimate that AlN containing 13 mole% of SiC in solution with no oxygen impurities would still have a high thermal conductivity of $\lambda = 2.0 \text{ W/cmK}$ at room temperature.

4.5.3. Ceramic samples. Many of the AlN samples whose thermal conductivity values have been reported in the literature have been polycrystalline ceramics. It is interesting to compute what the predicted thermal conductivity of a ceramic sample would be if the grain size were, say, 10^{-4} cm; a common grain size in such ceramics. The phonon λ , if limited only by boundary scattering with $l=10^{-4}$ cm, is given by:

$$\lambda_B = l\bar{v}C/3,\tag{24}$$

where \bar{v} is the average sound velocity (=6.98 × 10⁵ cm/s) and c is the specific heat capacity per unit volume. This can be combined with the intrinsic phonon-phonon scattering λ_{pp} by:

$$\lambda_{TOT}^{-1} = \lambda_B^{-1} + \lambda_{np}^{-1} \tag{25}$$

to give the dashed curve shown in Fig. 1 for a 1 micron-grain-size ceramic. Notice that for $T>200~\rm K$ the ceramic will have almost the intrinsic λ values. Thus, we could use λ values for ceramic samples at and above room temperature as being very close to the true values unmodified by grain-boundary scattering.

5. CONCLUSIONS

- 1. The thermal conductivity of single crystals of AlN has been measured over a wide temperature range. The crystals were sufficiently pure so that the extrapolated thermal conductivity of pure AlN could be determined.
- 2. The thermal conductivity of pure AlN at room temperature is about 80% of that of copper.
- The thermal conductivity of pure AlN is in good agreement with data on AlAs and AlSb, with other pure adamantine compounds, and with theoretical calculations.
- 4. It appears possible to make dense, ceramic samples of AlN with high thermal conductivity at and above room temperatures by careful control of the impurity content, particularly oxygen.

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