Letter

Comment on Witek's paper on thermal conductivity of some nitrides

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Abstract

Witek [Diamond Relat. Mater. 7 (1998) 962] has shown that the measured thermal conductivities of the adamantine nitrides AlN, GaN and cubic BN are well below the values calculated from a simple expression he derived for the ideal crystals. He ascribed these discrepancies to lattice defects in the cases of AlN and GaN, but to isotopes for BN. It is shown here, using a modicum of theory that these explanations are almost certainly correct. © 1999 Elsevier Science S.A. All rights reserved.

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1. Introduction

Witek [1] derived a simple expression for predicting the thermal conductivity of perfect dielectric crystals at 'high temperatures' in terms of simple observable properties for the crystal. He showed that this predicts quite well the conductivity of isotope-free diamond over a wide range of temperature, from 300 to 1000 K, but overestimates the conductivities of GaN, AlN and BN at room temperature. He attributed this discrepancy to crystal imperfections or isotopes in the constituent elements. He concluded, without numerical proof, that for GaN and AlN lattice imperfections are responsible for the extra resistivity and that for BN the high proportions of the two boron isotopes are responsible.

2. Theory

We calculate here the maximum value that the isotopic resistivity could have for the three nitrides, which would only occur if the 'Ziman limit' held (three-phonon wave-vector conserving normal (N) processes being dominant in the scattering of phonons). This is certainly true for diamond at room temperature with ^{13}C present up to about 1%, but would not hold for the greater concentrations of isotopes that occur in boron and gallium, when the isotope resistivity would be less than that calculated assuming that N-processes are dominant. Lattice defects of small dimensions would affect the conductivity in a way similar to isotopes. If N-processes are not dominant their scattering rate must be known in order to calculate the isotope resistivity. This rate has been deduced in a few cases by analysing the conductivity of crystals with varying isotope concentrations [2–4]. The method of calculating the 'Ziman limit' resistivity, which has been described by Nepsha et al. [5,6] and by Berman [7] and applied to diamond, will not be discussed here. The results of the calculation for the nitrides are shown in Table 1, where Witek's conductivities have been converted into resistivities since when N-processes are dominant resistivities are additive. It should be noted that the calculated resistivities of the ideal crystals, W_0, and the calculated isotope resistivities for the Ziman limit, W_Z^I are not given to great accuracy since (a) it is not known to what extent Witek's expression will give accurate values of the ideal conductivities for these nitrides and (b) although Witek gives the values of the Debye characteristic temperatures, θ, of each nitride, he does not give the phonon velocities which have been estimated here very crudely from the expression $v = 2a(k\theta/h)$ where a^3 is the average volume occupied by an atom, derived from the density and the molecular weight for AlN and BN, but given directly by Sichel and Pankove [8] for GaN; k and h are the Boltzmann and Planck constants (see Ref. [9]).

Table 1
Thermal resistivities at 300 K in units of 10^{-3} m K W^{-1}

Crystal	W_0^a	W_{exp}^b	$W_D = W_{exp} - W_0^c$	$W_Z^{I\,d}$
GaN	2.4	8.3	5.9	0.4
AlN	1.7	3.5	1.8	0.15
BN	0.3	1.3	1.0	1.7

[a] W_0 = resistivity of a perfect crystal as calculated by Witek [1].

[b] W_{exp} = measured resistivity.

[c] W_D = extra resistivity due to lattice defects and isotopes.

[d] W_Z^I = resistivity due to isotopes calculated as if the Ziman limit for N-processes dominant (see text) held true for all concentrations; it certainly does not and then the actual isotope resistivity must be less than W_Z^I.

It can be seen from Table 1 that for GaN and AlN $W_Z^I \ll W_D$. Since the true isotope resistivity $W^I < W_Z^I$ then $W^I \ll W_D$ and there can be only a small contribution of isotopes to the lattice defect resistivity. This is immediately understandable for AlN since the only isotopic impurity is 0.38% ^{15}N in the nitrogen. Less obvious is the fact that the large concentration, 38.5%, of ^{71}Ga in the gallium is offset by the small relative mass difference (the square of $\Delta M/M$ is important) The concentration of nitrogen vacancies given by Witek as 10^{19} cm^{-3} would roughly double the resistivity due to isotopes alone.

In BN, however, ^{10}B comprises 20% of the boron but the relative mass difference, when squared, is much greater than that for Ga so that the isotope resistivity could well outweigh that due to lattice defects. Since W_Z^I is nearly twice W_D for BN, the actual isotope resistivity W^I, although less than W_Z^I, could well make an appreciable contribution to W_D.

3. Conclusions

We have thus shown (in a slightly quantitative way) that Witek's assignment of lattice defects and isotopes as the main causes of the measured resistivities being greater than those calculated for ideal crystals is almost certainly correct. In the cases of GaN and AlN lattice defects and/or impurities are largely responsible for a reduction in conductivity, while for BN the isotope resistivity is important, so that its conductivity could only be improved appreciably by growing crystals with nearer mono-isotopic boron.

Acknowledgement

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