

How Isotopes Obscure the Anisotropic Phonon Umklapp Scattering in Dielectrics

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Abstract

Calculations of the thermal conductivity of h.c.p. ⁴He, doped with up to 1% ³He, are presented using a Debye relaxation-time approach. The results are analysed to extract information on the exponents in the anisotropic phonon Umklapp relaxation rate. The results show that even if 1% of ³He is present the exponents may be measured to within 7% of their undoped value. Consequently, anisotropic phonon Umklapp parameters may be determined in crystals where natural isotopes exist.

Introduction

The purpose of this paper is to investigate the effect that phonon isotope scattering can have in obscuring the inherent anisotropic phonon Umklapp-dominated thermal conductivity. Interest was stimulated in Umklapp anisotropy when a conductivity ratio of ~ 20 was observed in single crystals of solid h.c.p. ⁴He at temperatures above that of the conductivity maximum (Hogan *et al.* 1969). The consequences of such anisotropy are obvious from the simple Debye phonon-gas formula (equation 1 below) which relates the thermal conductivity K with the phonon specific heat C , phonon velocity v and phonon mean free path l :

$$K = \frac{1}{3}Cvl. \quad (1)$$

Umklapp anisotropy is very different from elastic anisotropy, which introduces an effect into the phonon velocity. Umklapp anisotropy, however, introduces an anisotropy into the phonon mean free path, and is only evident in temperature regions in which phonon Umklapp scattering mechanisms dominate the thermal conductivity. Simons (1960) has considered the implications of anisotropic phonon Umklapp scattering mechanisms and has shown that at very low temperatures the thermal resistance anisotropy is dependent on the geometry of the lattice unit cell.

Berman *et al.* (1972) applied Simons's (1960) theory to h.c.p. ⁴He. Other crystal structures have also been investigated and Simons's theory has been applied with varying degrees of success (Hudson 1976). To obtain information on Umklapp anisotropy the following criteria must apply: Single crystals of noncubic dielectrics are required which (i) are extremely pure, (ii) are isotopically pure, (iii) possess a simple primitive cell (Slack 1973), (iv) do not possess irregular phonon dispersion relations, and (v) are defect free. Criterion (ii) is extremely restrictive and it would be most interesting to know to what degree it needs to be satisfied in order to obtain

quantitative information on the anisotropic phonon Umklapp scattering rate which is determined by the crystal Brillouin zone. In fact if one assumes that no isotopes ought to be present at all then the number of possible systems on which meaningful measurements may be taken is very limited indeed.

To investigate the validity of criterion (ii), this work considers the effect of adding ^3He isotopes (up to 1% atomic) to h.c.p. ^4He in the temperature region where the thermal conductivity is determined predominantly by phonon Umklapp scattering. HCP ^4He is the most suitable material with which to work, due to the large effect the phonon Umklapp anisotropy has on the thermal conductivity. Thus the sensitivity of the result as a function of isotopic concentration is higher than for any other system which has been measured. Also, to clarify the interpretation of the results, the material must have a simple lattice structure, i.e. it must be either tetragonal or hexagonal. Helium-4 is the only crystal which has been measured and analysed and which satisfies all the criteria mentioned above. However, ^4He does have some disadvantages. It has been found that pure phonon isotopic scattering does not occur in h.c.p. ^4He . Some strain field scattering has been observed due to the atomic differences between ^3He and ^4He at very low temperatures. Also Normal (N) processes have been shown to play a very important part in the thermal conductivity of solid helium (Berman *et al.* 1965).

Berman *et al.* (1965) have made measurements on crystals of h.c.p. ^4He of unknown orientation and with concentrations of ^3He ranging from 1.22% to 11.1%. The results have given no information on Umklapp anisotropy, and the main purpose of the work was to investigate Callaway's (1959) relaxation-time theory and the general effect of isotopes on lattice heat conduction.

Theory

The theoretical details of phonon Umklapp anisotropy are covered extensively elsewhere (Simons 1960; Hudson 1976) as are the details of thermal conduction in dielectrics (Callaway 1959). Isotopic phonon scattering is a special case of mass-defect phonon scattering, which is analogous to Rayleigh scattering in optics. The phenomenon has been discussed by many authors (Klemens 1955; Carruthers 1961; Klein 1966). The isotopic phonon scattering rate τ_{iso}^{-1} is given by

$$\tau_{\text{iso}}^{-1} = \frac{V}{4\pi v^3} \sum_i f_i \left(\frac{\Delta M_i}{M} \right)^2 \omega^4, \quad (2)$$

where V is the average atomic volume, v is the average phonon velocity ignoring polarization and dispersion, M is the mean atomic mass and f_i is the fraction of atoms of mass $M_i = M + \Delta M_i$. Turk and Klemens (1974) have considered phonon scattering by strain fields which are introduced by point defects. Isotopes generally should not introduce strain fields (Berman and Brock 1965). However, due to differences in the zero point energy of the helium isotopes, small strain fields can be introduced (Klemens *et al.* 1964). The effect is small (Berman *et al.* 1965) and has been ignored in the present paper as it is not essential to the argument.

Results

The relative importance of isotope scattering to Umklapp scattering in a given crystal is dependent on the temperature and the phonon frequencies involved. An

estimate may be made of that isotopic concentration above which more phonon scattering is attributable to isotopes than to the inherent Umklapp phonon scattering. In this case ($\tau_{\text{iso}} \approx \tau_{\text{u}}$, where τ_{u}^{-1} is the Umklapp scattering rate) the Umklapp character of the thermal conductivity must be largely destroyed. Using a dominant phonon approximation (Hudson and Gibbs 1976) and the data from Berman *et al.* (1965) it may be shown that this critical concentration is $\sim 30\%$. To be able to obtain accurate quantitative data for anisotropic phonon scattering, the isotope concentration must be considerably less than 30% . It was decided to investigate quantitatively the effect of increasing the ^3He isotope concentration from zero to 1.0% .

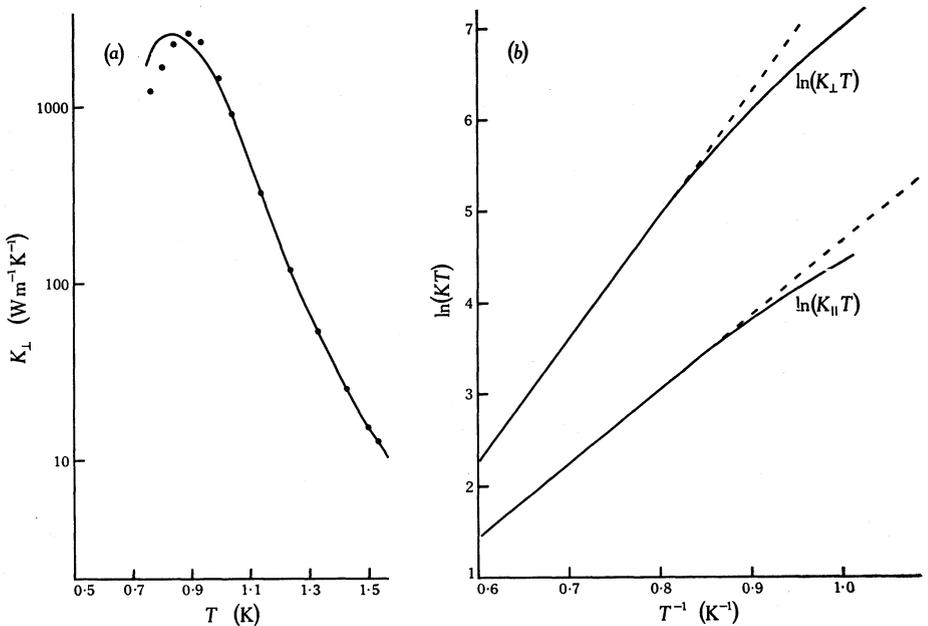


Fig. 1. Temperature dependence of the thermal conductivity of pure ($c = 0$) h.c.p. ^4He , as exhibited by plots of:

- (a) the present computed form (curve) of K versus T compared with the experimental data (points) of Hogan *et al.* (1969);
- (b) $\ln(K_{\perp} T)$ and $\ln(K_{\parallel} T)$ versus T^{-1} .

The experimental data on h.c.p. ^4He of Hogan *et al.* (1969) were used as the basis for the calculation. Their data on the two principal conductivities which determine the conductivity tensor for h.c.p. helium were fitted using a Debye-phonon-relaxation-time approach. Three scattering mechanisms were considered: boundary, isotope and Umklapp. Although N processes are undoubtedly important near the conductivity maximum, no data are available on how the crystal orientation affects N processes. Consequently it was decided to ignore N processes, and this was felt justified for the following reason: in the region where Umklapp processes dominate the thermal conductivity, N processes are playing a relatively small role and should not affect the result as resistive mechanisms like isotope scattering do. Consequently, as the region above the conductivity maximum is the region from which the data on

Umklapp anisotropy are extracted, the result will not depend significantly on whether N processes are considered or not. The temperature dependence of the conductivity shown in Fig. 1a is a typical result of the data fitting, and it is seen to be excellent in the Umklapp dominated region. In this region the conductivity may be expressed in terms of the phonon Umklapp relaxation time (Hudson 1976). The conductivities K_{\perp} and K_{\parallel} perpendicular to and parallel to the c axis respectively are related to the crystal Debye temperature θ and the absolute temperature T through

$$K_{\perp} = A_{\perp} T^{-1} \exp(\theta/b_{\perp} T) \quad \text{and} \quad K_{\parallel} = A_{\parallel} T^{-1} \exp(\theta/b_{\parallel} T), \quad (3)$$

where A , b_{\perp} and b_{\parallel} are constants, the latter two being related to the geometry of the Brillouin zone and thereby to the crystal symmetry. The ratio b_{\parallel}/b_{\perp} is determined exactly by the h.c.p. helium Brillouin zone and the anisotropic phonon velocity parameter γ (Hudson 1976). A plot of $\ln(KT)$ against T^{-1} reveals the relevant information concerning the b parameter without the need to consider a detailed Debye-relaxation-time analysis. Such a plot is given in Fig. 1b for the pure ${}^4\text{He}$ case. However, the introduction of small concentrations of isotopes or small defects will obscure the Umklapp dominance to some degree. A simple plot as detailed above would still give a straight line and a value for b_{\parallel}/b_{\perp} but the accuracy of the result would be diminished.

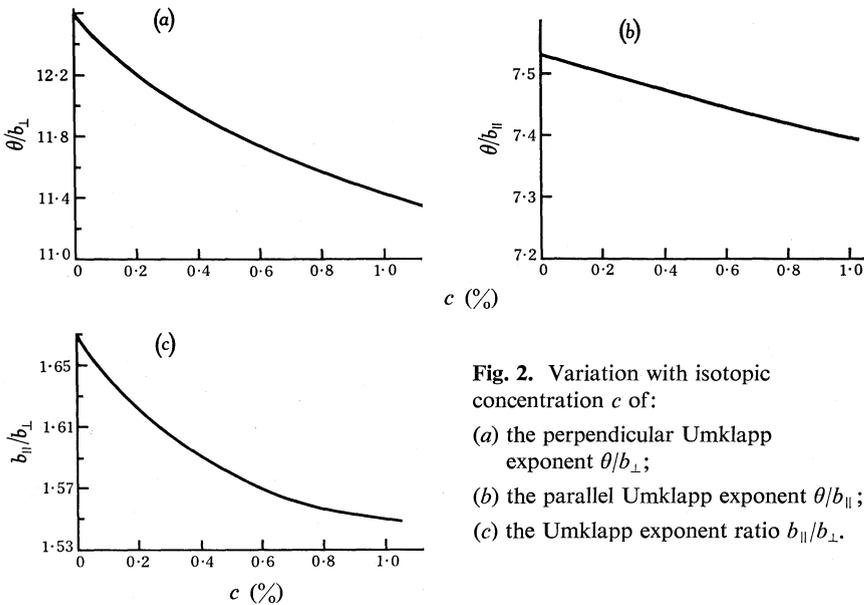


Fig. 2. Variation with isotopic concentration c of:
 (a) the perpendicular Umklapp exponent θ/b_{\perp} ;
 (b) the parallel Umklapp exponent θ/b_{\parallel} ;
 (c) the Umklapp exponent ratio b_{\parallel}/b_{\perp} .

Various concentrations of ${}^3\text{He}$ were added to contribute to the total phonon resistive relaxation rate. For a given concentration the boundary and Umklapp terms were maintained as for the 'pure' ${}^4\text{He}$ case, and the resultant conductivity was computed against temperature. The conductivities could still be represented as in equation (3), but with a slightly increased value of b . Figs 2a and 2b depict the variation of the parameter θ/b as a function of isotope concentration. Finally, Fig. 2c indicates how the ratio b_{\parallel}/b_{\perp} is dependent on isotope concentration.

Discussion

Figs 2a and 2b show that b_{\perp} is very much more sensitive to isotope content than b_{\parallel} , which is to be expected. The phonon Umklapp scattering rate τ_u^{-1} is considerably larger for the parallel direction than for the perpendicular direction. Consequently any additional isotropic scattering will affect the perpendicular conductivity much more severely than the parallel conductivity. It is apparent that the exponent sensitivity to isotopes $-d(\theta b^{-1})/dc$ decreases as the concentration increases. Even for the extreme 1% case, the measured ratio of b_{\parallel}/b_{\perp} is only reduced by 7% relative to the 'pure' case ($c = 0$).

Other Materials

The present results may be applicable to other systems which exhibit Umklapp anisotropy. Phonon isotope scattering is extremely strong in the case of helium due to the severe mass perturbation. Consideration of equation (2) reveals that 1% of ^3He in ^4He scatters phonons more than four times more strongly than the natural isotope content of magnesium in magnesium fluoride (Hudson 1976). The magnitude of the Umklapp scattering rate also influences the effect of isotopic scattering. The actual magnitude of this scattering rate at temperatures where it is dominating the conductivity is not too dependent on the crystal material. This is a consequence of the fact that thermal conductivity can be expressed generally in a reduced form (Cezairliyan and Nalbantyan 1971; Simons 1973; Mikhail and Simons 1974). Certainly the exponent is approximately constant, as the temperature region in which the interest lies is $\sim \theta/20$.

Conclusions

It was found that the phonon Umklapp parameter b as defined by equation (3) may still be measured to within 7% in the extreme case of h.c.p. ^4He doped with 1% isotopic concentration of ^3He . This result may be extended to other dielectric crystals to indicate that fairly accurate phonon Umklapp scattering data may be obtained even if the crystals possess large natural concentrations of isotopes. This result gives more weight to the argument that the low temperature thermal conductivity divergence in tetragonal MnF_2 and MgF_2 is a consequence of optical phonon Umklapp excitation (Hudson 1976).

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