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The High Temperature Thermal Conductivity of Semiconductor Alloys

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Abstract. A theory of the high temperature lattice thermal conductivity of semiconducting alloys is described in which normal processes are taken into account by the use of Callaway's method. This theory is in good agreement with data on germanium-silicon alloys, published recently by Abeles *et al.*, but it is shown that the theory implies that the transport of heat is largely by transverse phonons, whilst direct investigation of the scattering processes suggests that longitudinal phonons should predominate. Some possible reasons for this discrepancy are put forward.

§ 1. INTRODUCTION

THE search for efficient materials for thermoelectric applications has resulted in a number of investigations into the thermal conductivity of semiconducting alloy systems. Much of this work has been reviewed by Drabble and Goldsmid (1961) and, in addition, there has recently been a careful investigation of the germanium-silicon system by Abeles, Beers, Cody and Dismukes (1962). Abeles *et al.* were unable to account satisfactorily for their results using the existing theory, due originally to Klemens (1960). This theory assumed that the phonons were scattered by the differences in mass in the unit cell produced by alloying and the three-phonon Umklapp (U) processes which arise from anharmonic interatomic forces. It is possible, using Klemens' formula, to compare the variation of thermal conductivity with alloy composition of many systems on a common graph and this was done by Drabble and Goldsmid. Using earlier results on Ge-Si alloys obtained by Steele and Rosi (1958) they found excellent agreement with theory, but, if the more refined results of Abeles *et al.* are used, there appears to be a discrepancy which increases as the alloy scattering becomes relatively stronger. The thermal resistance is greater than that predicted.

There are two main criticisms which can be directed at Klemens' theory. The first is that he completely neglects the three phonon normal (N) processes, and this paper attempts to take these into account in an appropriate manner. The second is that no account is taken of the effect of atomic misfit in alloys which might be expected to scatter phonons. This question will only be touched on here.

§ 2. THE KLEMENS-CALLAWAY THEORY OF THERMAL RESISTANCE DUE TO POINT IMPERFECTIONS

There are at present two distinct procedures being employed in the theoretical calculation of the thermal conductivity. One, described at length by Ziman (1960), uses a variational method, whilst the other, due to Callaway (1959), involves an ingenious method of taking the N-processes into account through a relaxation time. In

fact it is finding a correct way of incorporating the N-processes into the theory that constitutes the main problem.

In its simplest form, as used by Leibfried and Schlomann (1954), the variational method assumes that N-processes are completely dominant so that the phonon distribution function is a Planck function displaced from the centre of the Brillouin zone such that a heat current exists. However, this assumption is not essential to the method, which has the advantage of working in terms of transition probabilities rather than relaxation times.

Callaway uses relaxation times in his calculations (Callaway 1959, 1961, Callaway and von Baeyer 1960), generally employing the 'single-mode relaxation time' which is really all that can be calculated. However, whilst the non-momentum conserving U-processes and point defect scattering are made to relax the phonons towards the equilibrium distribution, the N-processes relax the system towards the displaced Planck function of the previous paragraph. In calculating the high temperature thermal resistance of a conductor with point defects Callaway neglected the N-processes and hence arrived at Klemens' equation.

Callaway found that the thermal conductivity is given by

$$\kappa = \frac{k}{2\pi^2 v} \left(\frac{kT}{\hbar} \right)^3 \left(I_1 + \frac{I_2^2}{I_3} \right) \quad (1)$$

where

$$I_1 = \int_0^{\theta/\tau} \frac{\tau_c x^4 e^x}{(e^x - 1)^2} dx \quad I_2 = \int_0^{\theta/\tau} \frac{\tau_c}{\tau_N} \frac{x^4 e^x}{(e^x - 1)^2} dx$$

$$I_3 = \int_0^{\theta/\tau} \frac{1}{\tau_N} \left(1 - \frac{\tau_c}{\tau_N} \right) \frac{x^4 e^x}{(e^x - 1)^2} dx \quad x = \frac{\hbar\omega}{kT}.$$

The effects of the N-processes are largely contained in the term I_2^2/I_3 . It is assumed that relaxation times exist, τ_N being that for normal processes. τ_c is a combined relaxation time obtained by reciprocal addition of all the relaxation times present including τ_N . Thus if τ_U is the relaxation time for U-processes and τ_D that due to point defects, we have

$$\tau_c^{-1} = \tau_N^{-1} + \tau_U^{-1} + \tau_D^{-1}. \quad (2)$$

The following assumptions are customarily made by Callaway about the relaxation times:

$$(i) \quad \tau_D^{-1} = A\omega^4 \quad \text{where} \quad A = \frac{\Omega_0}{4\pi v^3} \sum_i f_i \left(1 - \frac{M_i}{M} \right)^2$$

$$= \frac{\Omega_0 \Gamma}{4\pi v^3} \quad (3)$$

$$M = \sum_i f_i M_i.$$

Here Ω_0 is the volume per atom, M_i is the mass of the i th type of atom, these being a fraction f_i of the total number of atoms. This expression was derived by Klemens (1955) and assumes that the scattering is due entirely to mass difference. This will be true for isotopic disorder but in other cases the strain field due to atomic misfit should scatter phonons as well.

$$(ii) \quad \tau_N^{-1} = C_N T \omega^2 \quad (4)$$

C_N being independent of temperature. This is the form appropriate to longitudinal phonons at high temperatures and was obtained in the first place by Herring (1954).

$$(iii) \quad \tau_U^{-1} = C_U T \omega^2 \quad (5)$$

C_U being independent of temperature. This comes from Klemens' pioneer work on thermal conductivity (Klemens 1951).† Both this and the previous expressions are only plausible for long wavelength phonons.

Callaway makes the drastic assumption of a Debye spectrum for the lattice vibrations, making no distinction between different polarizations which are assumed to have the same sound velocity v . However, he adduces reasons why this use of a Debye spectrum should be better for thermal conductivity than for heat capacity calculations.

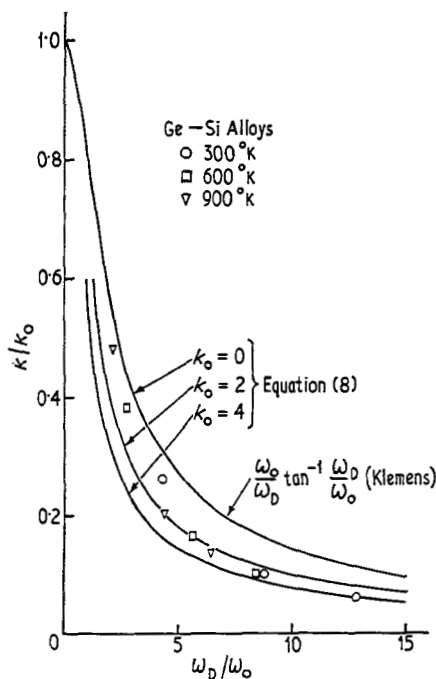


Figure 1. Comparison of experimental data with equation (8). At each temperature, the points correspond to 8%, 30% and 70% silicon, in order of increasing ω_D/ω_0 .

At high temperatures, $x^2 e^x / (e^x - 1)^2$ is approximately unity. Then if we neglect N-processes, the integral I_1 can easily be evaluated giving

$$\kappa = \frac{k}{2\pi^2 v (AC_U T)^{1/2}} \tan^{-1} \left[\frac{k\theta}{\hbar} \left(\frac{A}{C_U T} \right)^{1/2} \right].$$

Writing κ_0 for the thermal conductivity without mass difference scattering, we have

$$\frac{\kappa}{\kappa_0} = \frac{\omega_0}{\omega_D} \tan^{-1} \left(\frac{\omega_D}{\omega_0} \right) \quad (6)$$

† However, Klemens later (1958) gives reasons for preferring

$$\tau_U^{-1} \propto \omega$$

where

$$\left(\frac{\omega_D}{\omega_0}\right)^2 = \frac{2\pi^2 v \kappa_0 \omega_D A}{k} = \frac{\pi \Omega_0 \Gamma \kappa_0 \theta}{2\hbar v^2}.$$

ω_D is the maximum frequency in the Debye model and ω_0 is the frequency at which the mass difference and Umklapp relaxation times are equal. This is Klemens' (1960) formula.

This formula is compared with the data of Abeles *et al.* in figure 1. The value of κ_0 was obtained by interpolation between germanium and silicon using the method of Keyes (1959). It will be seen that for large values of ω_D/ω_0 Klemens' formula predicts too large a value of κ/κ_0 . Abeles *et al.* tentatively attributed this to a dependence of C_U on Γ , arising from considerations in second-order perturbation theory; there are simultaneous three- and two-phonon processes and the three-phonon processes no longer conserve crystal momentum and hence contribute directly to thermal resistance.

§ 3. THE EFFECT OF NORMAL PROCESSES ON HIGH TEMPERATURE THERMAL RESISTANCE OF ALLOYS

It is quite straightforward to allow for the N-processes in the way which Callaway did in his low temperature calculations. One puts

$$\tau_c^{-1} = A\omega^4 + (C_N + C_U)T\omega^2 \quad (7)$$

and, if one puts $C_N/C_U = k_0$, direct integration of I_1 , I_2 and I_3 gives

$$\frac{\kappa}{\kappa_0} = \frac{1}{1+5k_0/9} \left\{ \frac{1}{y} \tan^{-1}y + \frac{[1-(1/y) \tan^{-1}y]^2}{[(1+k_0)/k_0](y^4/5)-(y^2/3)+1-(1/y) \tan^{-1}y} \right\} \quad (8)$$

where

$$y^2 = \frac{\pi \kappa_0 \Omega_0 \Gamma \theta}{2(1+5k_0/9)v^2\hbar} = \frac{(\omega_D/\omega_0)^2}{1+5k_0/9}. \quad (9)$$

One can now vary k_0 so as to try to fit the experimental results. Values for $k_0 = 4$ and $k_0 = 2$ are shown in figure 1. If $k_0 = 4$ there is fit at high values of ω_D/ω_0 but not at low values; $k_0 = 2$ fits at lower values of ω_D/ω_0 but it is clear that no value of k_0 will give a fit over the whole range. What is needed is to make k_0 decrease with ω_D/ω_0 .

This suggests that a fit would be obtained if τ_N^{-1} varied less rapidly with ω than τ_U^{-1} . We will therefore try $\tau_N^{-1} \propto \omega$; later the theory of three-phonon scattering will be examined to see if this is plausible. We put

$$\tau_N^{-1} = D_N T \omega$$

specifying D_N by putting

$$D_N = k_t C_U \omega_D.$$

This means that at the surface of the Debye sphere the N-processes are k_t times as strong as the U-processes.

Callaway's integrals must now be solved numerically and it is convenient to do a certain amount of reformulation at this point leading to

$$\kappa = \frac{k^2 \theta}{2\pi^2 v \hbar C_U T} \left[\mathcal{J}_1(\beta; k_t) + \frac{k_t \mathcal{J}_2^2(\beta; k_t)}{\mathcal{J}_4(\beta; k_t) + \beta^2 \mathcal{J}_6(\beta; k_t)} \right] \quad (10)$$

where

$$\mathcal{J}_j(\beta; k_t) = \int_0^1 \frac{z^j dz}{\beta^2 z^3 + z + k_t}.$$

Then

$$\beta^2 = \left(\frac{\omega_D}{\omega_0} \right)^2 \left/ \left[\mathcal{J}_1(0; k_t) + \frac{k_t \mathcal{J}_2^2(0; k_t)}{\mathcal{J}_4(0; k_t)} \right] \right. \quad (11)$$

and the ratio is then

$$\frac{\kappa}{\kappa_0} = \frac{\mathcal{J}_1(\beta; k_t) + k_t \mathcal{J}_2^2(\beta; k_t) / [\mathcal{J}_4(\beta; k_t) + \beta^2 \mathcal{J}_6(\beta; k_t)]}{\mathcal{J}_1(0; k_t) + k_t \mathcal{J}_2^2(0; k_t) / \mathcal{J}_4(0; k_t)}. \quad (12)$$

The results for $k_t = 0.1, 1$ and 10 are compared with Abeles' data in figure 2 and it is seen that the curve for $k_t = 0.1$ is in good agreement with experiment, providing

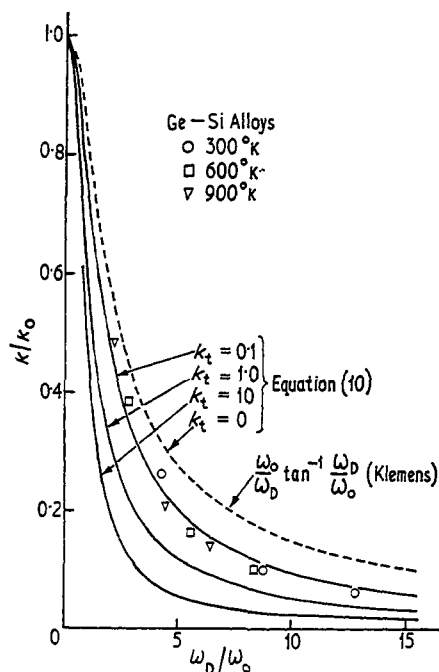


Figure 2. Comparison of experimental data with equation (10).

empirical justification for the chosen form of τ_N^{-1} . There remains a slight tendency for the predicted value of κ/κ_0 to be low at small values of ω_D/ω_0 , which might disappear if a slightly slower dependence of τ_N^{-1} on ω were used. This possibility will not be pursued further. The important question is whether the form of τ_N^{-1} and τ_U^{-1} can be made plausible by examining the scattering processes involved and whether $k_t = 0.1$ is a reasonable value.

§ 4. THREE-PHONON SCATTERING PROCESSES

The theory of the scattering of phonons by other phonons through anharmonic interatomic forces is discussed by Drabble and Goldsmid (1961) and in more detail by Ziman (1960). It is not possible to determine a true relaxation time, only a 'single-mode relaxation time', meaning that the phonons with which the scattered phonon interacts are assumed to be in thermal equilibrium. For long wavelength phonons the use of this relaxation time in conductivity calculations is justified by saying that the short wavelength phonons with which they mainly interact are themselves very strongly scattered and are therefore very close to equilibrium.

For long wavelength phonons the most important processes are those where the phonon of interest (wave vector \mathbf{q}) combines with a phonon \mathbf{q}' to produce a third phonon \mathbf{q}'' .

Conservation of energy and of wave vector (modulo a reciprocal lattice vector in the case of U-processes) means that the relaxation time can be written in the form of an integral over a surface ('energy conservation surface') in \mathbf{q}' space. This can be written as

$$\tau^{-1} = \frac{\hbar}{8\pi^2\rho^3} \int (N' - N'') \frac{qq'q''}{vv'v''} A^2 \frac{dS'}{|v'|}. \quad (13)$$

Here N' and N'' are the Planck distribution functions for q' and q'' , v, v', v'' are the phase velocities for q, q', q'' , which will differ if different polarizations are involved, and

$$|v'| = \nabla_{\mathbf{q}'}(\omega + \omega' - \omega'')$$

on the energy conservation surface. The last expression gives the density of states. A is a quantity approximately independent of the magnitudes of q, q' and q'' , but dependent on the angles between them and on the strength of the anharmonic coupling constants. The evaluation of τ is made much simpler by the assumption of a Debye model, particularly if q is a low frequency phonon. The shape of the energy conservation surface S' depends on whether N- or U-processes are being considered.

For transverse phonons of long wavelength the most important process is where $\mathbf{q}', \mathbf{q}''$ are longitudinal phonons. Then S' is an hyperboloid cut off by the Debye sphere and

$$\begin{aligned} \frac{dS'}{|v'|} &\simeq \frac{\pi}{2^{1/2}v_1} \frac{q'^2 dq'}{q} \\ N' - N'' &\simeq \frac{kT}{\hbar v_1} \left(\frac{v_t}{v_l} \right) \frac{q}{q'^2} = f \frac{kT}{\hbar v_1} \frac{q}{q'^2} \\ A^2 &\simeq 8\beta^2 \sin^2\theta' \cos^2\theta' \end{aligned}$$

θ' being the angle between \mathbf{q} and \mathbf{q}' , and β a third-order elastic coefficient, $A_{112}^{112} = A_{111}^{122}$ for an isotropic substance.

Then one finds

$$\frac{1}{\tau_N^t} = \frac{2^{1/2} \beta^2 f^2 (1 - f^2) q q_D^3 kT}{3\pi \rho^3 v_1^5}. \quad (14)$$

where q_D is the radius of the Debye sphere. This expression is probably valid for $q < \frac{1}{2}q_D$. Thus $(\tau_N^t)^{-1}$ has the form assumed for τ_N^{-1} in the calculations of § 3. Fortunately in all cases, except those where the alloys are very dilute, the strong point defect scattering will dominate at high frequencies so the form of τ_N^{-1} at these frequencies is not too important.

For the U-processes one has to introduce into the Debye model something equivalent to making \mathbf{q}'' differ from $\mathbf{q} + \mathbf{q}'$ by a reciprocal lattice vector. It seems in keeping with the spirit of the approximation to do this by adding to \mathbf{q}'' a parallel vector of length $2q_D$ and calculating the corresponding energy conservation surface. For small q this is a sphere of radius $q_D/2$, $|v'|$ is $2v_1$ so

$$\frac{1}{\tau_U^t} = \frac{4}{15\pi} \frac{\beta^2 q_D^2 kT}{\rho^3 v_1^5}. \quad (15)$$

This has the same dependence on q as that assumed in § 3, and comparing (15) with (14) suggests that $k_t \sim 0.4$, which is somewhat larger than the value found empirically. The very approximate nature of the calculations leading to (14) and (15) probably removes any significance from this discrepancy.

The calculation of the relaxation times for longitudinal phonons presents greater difficulties since for an isotropic medium one finds $(\tau_N^l)^{-1} \propto \omega^4$. This appears to lead to a divergence at the lower limit of the thermal conductivity integral, but Herring (1954) showed that, if elastic anisotropy was allowed for, $(\tau_N^l)^{-1} \propto \omega^2$ in cubic crystals. Elastic anisotropy lifts the degeneracy of the transverse modes and makes possible a transition

longitudinal + transverse (lower branch) \rightarrow transverse (higher branch).

Simons (1957) made an explicit calculation of the relaxation time for this process in the case of very long wavelength phonons. Extending his results to the high temperature case one has

$$\frac{1}{\tau_N^l} = \frac{1}{40\pi^2} \frac{q^2 q_D^2 kT}{\rho^3 v_t^5} \frac{(\kappa + 1)\Gamma^2}{t(\kappa - 1)(2\kappa + 1)} \quad (16)$$

where

$$\kappa = \frac{c_{12} + c_{44}}{c_{11} - c_{44}} \quad t = \frac{c_{11}}{c_{44}} - 1$$

$$\Gamma^2 = (\beta - \gamma)^2 [E(h) - (1 - h^2)K(h)] + 4\gamma^2 [K(h) - E(h)]$$

$h = (1 + 2\kappa)^{1/2}/(1 + \kappa)$ and γ is the third-order elastic constant, $A_{123}^{123} = A_{231}^{312} = A_{122}^{133}$, if third-order elastic isotropy is assumed. $K(h)$ and $E(h)$ are complete elliptic integrals of the first and second kind respectively. Experimental data for germanium (Bateman, Mason and McSkimin 1961) averaged to third-order isotropy suggest that

$$\frac{\tau_N^t}{\tau_N^l} \simeq 0.5 \frac{q}{q_D} \quad (17)$$

which means that at low energies the longitudinal phonons are scattered much less than the transverse. It should be noted that the frequency dependence of τ_N^l is the same as that assumed by Callaway (Callaway 1959, 1961, Callaway and von Baeyer 1960).

It is difficult to calculate τ_U^l even in the approximate manner used for τ_U^t . Ziman (1960), on the basis of a consideration of the geometry of the scattering, states that U-processes are impossible for long wavelength longitudinal phonons. It may be that this prohibition may be relaxed for some types of frequency surface near the zone boundary but in any case it is to be expected that τ_U^l will be greater than τ_N^l at long wavelengths and that it increases more rapidly than q^2 . (Incidentally this means that Callaway's use of $\tau_U \propto \omega^2$ for longitudinal phonons must be incorrect.)

The result of all these considerations is that the dependence of thermal conductivity on alloy concentration is that to be expected if the heat transport were almost entirely by transverse phonons. However, one must first eliminate the possibility that the use of relaxation times appropriate to longitudinal phonons gives equally good agreement. To do this the calculation of thermal conductivity has been carried out for the case where

$$\tau_N^{-1} = C_N T \omega^2$$

and

$$\tau_U^{-1} = B_U T \omega^3$$

and

$$C_N = k_1 B_U \omega_D.$$

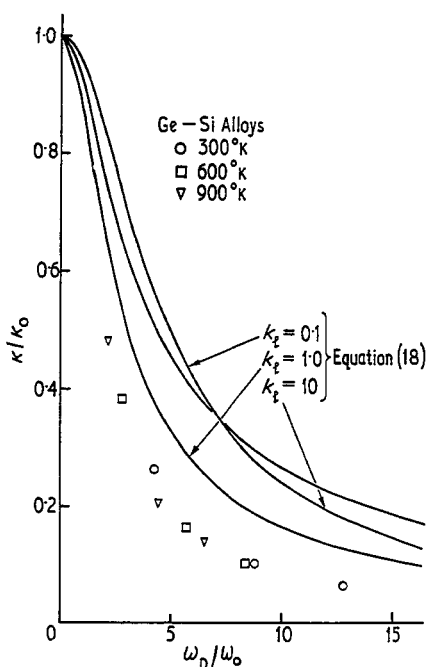


Figure 3. Comparison of experimental data with equation (18).

The result is that

$$\frac{\kappa}{\kappa_0} = \frac{\mathcal{G}_0(\beta; k_1) + k_1 \mathcal{G}_2^2(\beta; k_1) / [\mathcal{G}_5(\beta; k_1) + \beta^2 \mathcal{G}_6(\beta; k_1)]}{\mathcal{G}_0(0; k_1) + k_1 \mathcal{G}_2^2(0; k_1) / \mathcal{G}_5(0; k_1)} \quad (18)$$

where

$$\mathcal{G}_j(\beta; k_1) = \int_0^1 \frac{z^j dz}{\beta^2 z^2 + z + k_1}$$

and

$$\beta^2 = \left(\frac{\omega_D}{\omega_0} \right)^2 \left/ \left[\mathcal{G}_0(0; k_1) + \frac{k_1 \mathcal{G}_2^2(0; k_1)}{\mathcal{G}_5(0; k_1)} \right] \right.$$

The results for $k_1 = 0.1, 1, 10$ are shown in figure 3, showing that this form of dependence for κ/κ_0 is incapable of accounting for the experimental results. (It should be

noted that without any N-processes at all ($k_1 = 0$) the integral for the thermal conductivity would diverge at the lower limit.)

The next step must be to compare the actual magnitudes of the thermal conductivity due to transverse and longitudinal phonons. It should suffice in fact to compare κ_0 for the two polarizations. The most reliably known relaxation times are for the N-processes compared in (17). Using this result one finds

$$\frac{\kappa_0^t}{\kappa_0^l} = 0.5 \frac{k_t \{ \mathcal{J}_1(0; k_t) + k_t \mathcal{J}_2^2(0; k_t) / \mathcal{J}_4(0; k_t) \}}{k_l \{ \mathcal{G}_0(0; k_l) + k_l \mathcal{G}_2^2(0; k_l) / \mathcal{G}_5(0; k_l) \}}. \quad (19)$$

Assuming that $k_t = 0.5$ and using the computed integrals, one finds that if $k_1 = 0.1$, $\kappa_0^t/\kappa_0^l \sim 0.7$, whilst if $k_1 = 10$, $\kappa_0^t/\kappa_0^l \sim 0.1$. Thus in the more favourable case κ_0^t and κ_0^l are comparable, but owing to the more rapid decrease for κ^t with ω_D/ω_0 there is no doubt that for $\omega_D/\omega_0 \sim 10$ the longitudinal thermal conductivity would be much the greater. In any case, from what has been said earlier, $k_1 = 0.1$ must be regarded as implausibly low.

§ 5. DISCUSSION

The position seems to have been reached where theoretical estimates of thermal conductivity based on consideration of scattering processes suggest that longitudinal phonons predominate in heat transport in germanium-silicon alloys, whilst the experimental data are best fitted by a phenomenological theory using relaxation times appropriate to transverse phonons. Since this contradiction has arisen as a result of an attempt to refine the Callaway technique for calculating thermal conductivity it seems desirable to look again at the basic assumptions of the method.

Before doing this it would be well to consider whether the neglect of scattering due to atomic misfit could be the cause of the trouble. The effect of this would be to alter Γ and therefore ω_D/ω_0 . At present Γ is quite asymmetric in terms of alloy composition because of the low mass of the silicon atom, and atomic misfit would increase it most near the germanium end of the alloy range. In fact such a change would not improve the situation and the relatively smooth experimental curve is evidence that atomic misfit is not an important cause of phonon scattering.

The assumptions of the Callaway method are likely to be particularly inadequate for higher energy phonons. The use of a Debye spectrum is quite unrealistic for transverse modes in germanium and silicon, and phonons whose wave vectors belong to points, say, more than half-way out to the zone boundary are capable of undergoing a large number of both normal and Umklapp scattering processes, which can be quite properly neglected for long wavelength phonons. Since these processes commonly involve phonons of similar energy this means that the use of single-mode relaxation times would be rather inappropriate. Such considerations might lead one to suppose that the errors arise largely in the estimate of κ_0 rather than in the actual values of κ for large ω_D/ω_0 . In fact a very rough estimate suggests that the absolute magnitude of κ (using the longitudinal phonon assumptions) for $\omega_D/\omega_0 \sim 13$ would agree with experiment for $5 < k_1 < 10$, and that the transverse phonon contribution would be relatively small. However, it is unfortunate that both the use of a more realistic dispersion law and consideration of extra scattering possibilities would tend to reduce κ_0 , making the disagreement with experiment worse. If it is the use of relaxation times as such which is the main cause of error, then correction should increase κ_0 since the scattering would probably have been overestimated.

Alternatively one might use the variational method to determine the thermal conductivity, though some improvement on the form described by Ziman (1960) would probably be needed. In particular, the use of different distribution function perturbations for longitudinal and transverse phonons is desirable. A detailed calculation, using the known dispersion curves for germanium and silicon, might yield interesting results.

The Callaway technique has proved an extremely successful phenomenological scheme for the calculation of thermal conductivity and it is not surprising that it has shown certain weaknesses on closer examination. The complexity of the processes involved in lattice thermal conductivity make it unlikely that any better method of comparable simplicity will be available in the near future.

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