

FIG. 4. Reduced carrier density, $PD/(Iw)$, vs reduced distance, y/w , through the slab.

Curve No.	$\text{Log}_{10} W$	$\text{Log}_{10} K$	$\text{Log}_{10} S_1$	$\text{Log}_{10} S_2$
1	0	2	-3	-3
2	-1	0	-3	5
3	-1	0	5	-3
4	-1	1	5	-3
5	-1	2	5	-3
6	1	0	-3	-3
7	1	0	5	-3

absorption coefficient is plotted in Fig. 3. The ratio changes the sign at the same K as the PME response, of course, and its general characteristics become apparent by a comparison with the corresponding curves of Figs. 1 and 2. The plot shows under what conditions the effect is strongly dependent on absorption coefficient in which case extreme caution is advised when such measurements are used to determine recombination parameters (unless monochromatic light of a well-defined absorption coefficient is used). The graphs also show under what circumstances the effect is particularly sensitive to bulk lifetime or one of the surface recombination velocities leading to guidelines for the proper experimental conditions, depending on which quantity is to be measured. In certain cases it appears feasible to measure two or all three of the recombination parameters by fitting an experimental curve to the theoretical ones. Measuring the sample with the light first incident on one surface and then on the other may also prove helpful in many cases.

Figure 4 which is practically self-explanatory has been included to show the carrier density profiles across the slab for several conditions.

It is a pleasure to acknowledge the help of Mrs. H. S. Perlman in carrying out all the numerical calculations and to thank F. A. Brand and H. Mette, all of these Laboratories, for interesting discussions on the experimental aspects of the problem.

Effect of Isotopes on Low-Temperature Thermal Conductivity*

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(Received September 4, 1956)

An analysis of the experimental results of the low-temperature thermal conductivity of single crystals of Si, Ge, and KCl indicates that their maximum conductivities are limited by isotope scattering of phonons rather than by umklapp processes. The isotope scattering in solid helium and sapphire is sufficiently low so that nearly pure umklapp scattering can be observed. The measured thermal conductivities of SiO_2 , KBr, and diamond appear to still be limited by impurities. However, even in pure crystals of these three materials the isotope scattering precludes the appearance of pure umklapp scattering.

IN simple insulating crystals the thermal energy is transported by the lattice vibrations.¹ The vibrations of an infinite, perfectly periodic lattice can be described in terms of phonons.^{2,3} Any disturbance in the perfect periodicity of the lattice which produces scattering of the phonons produces a thermal resistance.

* Part of this work was done while the author was at Cornell University.

¹ P. Debye, *Vorträge Über die Kinetische Theorie der Materie und der Elektrizität* (B. G. Teubner, Berlin, 1914), pp. 17-60.

² R. E. Peierls, *Ann. Physik* **3**, 1055 (1929).

³ J. Frenkel, *Wave Mechanics—Elementary Theory* (Oxford University Press, London, 1932), p. 266.

A monatomic crystal of an element which possesses more than a single, naturally occurring isotope will not have a perfectly periodic lattice. Atoms of the several isotopes will be randomly distributed on the lattice sites. This random variation in the atomic mass will produce a thermal resistivity. This isotope effect was pointed out by Pomeranchuk⁴ in 1942, and a calculation of the magnitude of the thermal resistivity caused by isolated lattice points of differing mass was given by Klemens⁵ in 1955. The isotope resistivity will be most

⁴ I. Pomeranchuk, *J. Phys. (U.S.S.R.)* **6**, 237 (1942).

⁵ P. G. Klemens, *Proc. Phys. Soc. (London)* **A68**, 1113 (1955).

TABLE I. Thermal resistivity data for eight different crystals.

	He (solid)	Al ₂ O ₃	C (diamond)	SiO ₂	Si	Ge	KBr	KCl
No. of isotopes	2	1,3	2	3,3	3	5	2,2	2,2
$\Gamma \times 10^4$	6×10^{-5}	0.0246	0.76	0.482	2.64	5.72	0.878	1.77
V_0 in 10^{-24} cm ³	23.6	42.6	5.68	37.8	19.9	22.6	71.8	62.0
Θ_D in $^\circ\text{K}^a$	55	1010	1960	290	668	390	177	233
v in 10^6 cm/sec ^b	0.53	8.0	11.8	3.75	6.22	3.81	1.41	1.88
W_I/B in 10^{-4} cm deg/watt	0.22	13	47	290	710	2700	4400	5600
W_θ in cm deg/watt	(2700) ^e	8.7 ^d	1.05 ^e	13.9 ^f	2.2 ^g	2.13 ^h	16 ⁱ	8.4 ^j
W_M in 10^{-4} cm deg/watt	9260 ^k	72 ^d	80 ^d	830 ^f	400 ^g	910 ^l	7500 ⁱ	2180 ^{m, i}
T_M in $^\circ\text{K}$	2.25	40	80	10	25 ^l	15	7	9.1
T_M/Θ_D	0.041	0.040	0.041	0.035	0.037	0.038	0.040	0.039
W_θ/W_M	(2900)	1220	130	166	56	23.4	21	39
W_U in 10^{-4} cm deg/watt	9260	68	8.2	110	17	17	125	66
$W_M - W_U$ in 10^{-4} cm deg/watt	0	4	72	720	380	890	7400	2110
K_M watt/cm deg (observed)	1.08	140	125	12	25	11	1.3	4.4
K_M watt/cm deg (predicted)	1.08	140	400	50	40	11	6	5
K_M watt/cm deg (umklapp)	1.08	145	1200	90	600	600	80	150
Umklapp behavior?	yes	yes	?	?	no	no	no	no

^a M. Blackman, *Handbuch der Physik* (Springer-Verlag, Berlin, 1955), Vol. 7, No. 1, p. 325.

^b R. F. S. Hearmon, *Revs. Modern Phys.* 18, 409 (1946); L. Bergmann, *Der Ultraschall* (S. Hirzel, Stuttgart, 1954).

^c Reference 10.

^d Berman, Foster, and Ziman, *Proc. Roy. Soc. (London)* A231, 130 (1955).

^e Berman, Simon, and Ziman, *Proc. Roy. Soc. (London)* A220, 171 (1953).

^f R. Berman, *Proc. Roy. Soc. (London)* A208, 90 (1951).

^g G. W. Hull and T. H. Geballe, *Phys. Rev.* 96, 846 (1954).

^h K. A. McCarthy and S. S. Ballard, *Phys. Rev.* 99, 1104 (1955).

ⁱ Reference 11.

^j A. Eucken and G. Kuhn, *Z. physik. Chem.* 134, 193 (1928).

^k F. J. Webb and J. Wilks, *Phil. Mag.* 44, 663 (1953).

^l Reference 12.

^m Reference 9.

pronounced at low temperatures since it is only at these temperatures that the resistivity caused by umklapp processes^{2,6} becomes small. It is the purpose of the present paper to show how the magnitude of the isotope resistivity compares with the thermal resistivities that have been observed experimentally.

An atom with a mass differing from the average atomic mass in the crystal will act as a point scattering center. Therefore its scattering cross section will have the same frequency dependence as other point imperfections such as chemical impurities. This means that the effect of isotopes will be more pronounced in crystals of high chemical purity. The magnitude of the thermal resistivity due to atoms of differing mass is given by Klemens as

$$W_I = \frac{12\pi^2 TV_0 B \Gamma}{0.897 h v^2}, \quad \Gamma = \sum_i f_i (1 - [M_i/M])^2, \quad (1)$$

where T is the absolute temperature, V_0 is the average volume of a unit cell, v is the average phonon velocity at low temperature, M_i is the mass of a unit cell, f_i is the fraction of unit cells with mass M_i , M is the average mass of all unit cells, and h is Planck's constant. The quantity B is a constant which Klemens gives as $\frac{1}{12}$ for the case of very small concentrations of foreign atoms. Since his theory is accurate to perhaps a factor of about three, the value of B will be determined from the experimental data.

Equation (1) may also be applied to polyatomic crystals by noting that at temperatures which are low

compared with the Debye temperature the unit cells vibrate as though they were a single mass point. In crystals such as sodium fluoride all unit cells have the same mass since sodium and fluorine both possess a single naturally occurring isotope. Thus W_I for sodium fluoride will be zero, even though the atomic masses of sodium and fluorine are different.

Thermal resistivity measurements have been made at low temperatures on "pure" single crystals of several different insulators. Data collected from various sources on eight such materials are given in Table I. Pure germanium and silicon have been included since they have so very few free electrons at low temperatures that they act like insulating crystals.⁷ The experimental value of the thermal resistivity at low temperatures can be taken to be the sum of the isotope resistivity (W_I), the umklapp resistivity (W_U), the resistivity caused by chemical impurities (W_C), and the resistivity caused by crystal boundaries (W_X):

$$W_{\text{tot}} = W_I + W_U + W_C + W_X. \quad (2)$$

This additive resistance approximation leads to a value of W_{tot} which is too low by no more than 30% at the temperatures of interest. The temperature region of interest at present is that where the minimum resistivity occurs. This region lies between $0.035\theta_D$ and $0.045\theta_D$, where θ_D is the Debye temperature of the crystal. The temperature at which the minimum occurs is determined by a combination of boundary scattering and point impurity scattering (or umklapp scattering). The thermal resistivity at the minimum is, however,

⁶ Berman, Simon, and Wilks, *Nature* 168, 277 (1951).

⁷ P. G. Klemens, *Australian J. Phys.* 7, 57 (1954).

nearly equal to the resistivity that would be caused solely by the point impurities (or umklapp processes) in a crystal of infinite size.^{8,9} Thus at the resistivity minimum the W_X term in W_{tot} can be neglected for the accuracy desired.

The resistivities of a number of different crystals may be compared if the comparison is made at the same fraction of the Debye temperature. In Table I the temperature at which the minimum resistivity occurs is given under T_M . It can be seen that T_M/θ_D is nearly the same for all of the crystals. The one exception is KCl⁹ which has an observed minimum at 4.9°K. For KCl the resistivity values in Table I are all given at $0.039\theta_D = T_M$.

At the temperature T_M , Eq. (2) may be rewritten as

$$W_I + W_C = W_M - W_U, \quad (3)$$

where W_M is the value of W_{tot} measured at T_M . The value of W_I/B has been computed at $T = 0.039\theta_D$ from a table of naturally occurring isotopes and the values of V_0 , and ν given in Table I. Leibfried and Schlömann¹⁰ have shown that W_θ/W_U is a function only of T/θ_D , where W_θ is the thermal resistivity measured at θ_D . Thus at $T = 0.039\theta_D$ it may be assumed that $W_\theta/W_U = A$, where A is a constant to be determined. Both W_I and W_C are small for solid helium, so that $W_M = W_U$. If the theoretical value of W_θ for helium as calculated by Leibfried and Schlömann is used, then $A = 2900$. The values of A and B can also be calculated from experimental data. As a reasonable approximation it may be assumed that W_C is small for both germanium and artificial sapphire. Then A and B can be estimated from Eq. (3)

$$\begin{aligned} 0.270B &= 0.091 - (2.13/A), & \text{Ge} \\ 0.00013B &= 0.0072 - (8.7/A), & \text{Al}_2\text{O}_3. \end{aligned} \quad (4)$$

This gives $B = 0.33$, $A = 1280$. The value of B is determined mainly by the Ge data, and that of A by the data for Al_2O_3 . If the assumption that $W_C \ll W_I$ is incorrect, then, sometime in the future, measurements on better crystals may show lower values of W_M for Ge and Al_2O_3 than given in Table I. It does not seem, however, that the above values of A and B are in error by more than a factor of four. Within this accuracy they agree with Klemens, and with Leibfried and Schlömann.

The values of W_I and $W_M - W_U$ as computed using the above values of A and B are shown in Fig. 1. It can be seen that the data for artificially produced Si, Ge, KCl, and Al_2O_3 crystals all lie on the curve $W_I = W_M - W_U$ to better than a factor of two. The C and SiO_2 crystals were natural crystals, and hence the

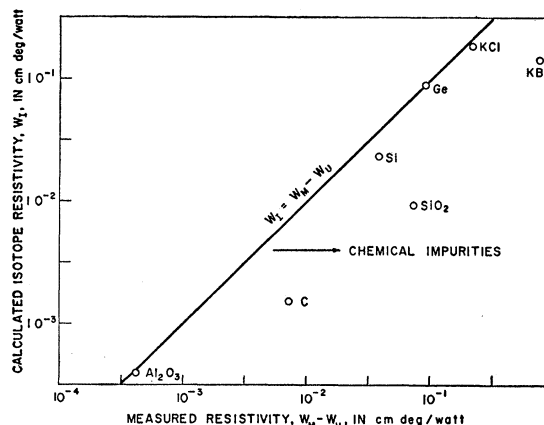


FIG. 1. Comparison of the residual measured thermal resistivity due both to isotopes and to chemical impurities with the calculated isotope resistivity. This residual resistivity is the measured resistivity minus the computed umklapp resistivity.

assumption that $W_C = 0$ is probably not valid. An increase in purity by a factor of five would bring their resistivities close to the minimum value. The data for KBr also indicates a high chemical impurity content, which may well have been present.¹¹ The point for He does not appear in Fig. 1 since W_I is so low.

Only for He and Al_2O_3 has the exponential decrease in resistivity characteristic of umklapp processes been experimentally observed. For these two crystals the isotope resistivity is very low compared to the umklapp resistivity, and thus, when the chemical impurity concentration is low, the resistivity is primarily due to umklapp processes. For both C and SiO_2 the isotope and umklapp resistivities are comparable. Thus, in sufficiently pure crystals of these materials, a behavior approaching an exponential decrease in resistivity with temperature dependence will probably not be observed for Ge, Si, KCl, or KBr because of the large isotope resistivity. This explains the question raised by White and Woods¹² of why umklapp behavior was not observed in germanium and silicon.

A comparison at $T = 0.039\theta_D$ is shown in Table I between the maximum observed thermal conductivity, the predicted maximum conductivity, and the maximum conductivity that would be observed if only umklapp processes were present. The predicted maximum combines both isotope and umklapp resistivities. The measured thermal conductivities of He, Al_2O_3 , Si, Ge, and KCl have just about reached their predicted maximum values. Diamonds free of chemical impurities should exhibit the highest thermal conductivity of any insulating crystal at a value of about 400 watt/cm-deg.

⁸ P. G. Klemens, Proc. Roy. Soc. (London) **A208**, 108 (1951).

⁹ G. A. Slack, Phys. Rev. **105**, 832 (1957).

¹⁰ G. Leibfried and E. Schlömann, Nachr. Akad. Wiss. Göttingen, Math. physik. Kl. **4**, 71 (1954).

¹¹ W. J. de Haas and T. Biermasz, Physica **4**, 752 (1937).

¹² G. K. White and S. B. Woods, Phys. Rev. **103**, 569 (1956).