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# Lattice Thermal Conductivity at Low Temperatures

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The theoretical models of Callaway and Klemens are compared to experimental results in (a) silicon and diamond, (b) solidified argon and neon, and (c) KCl and NaF, which are taken from the literature. Three sources of scattering are postulated: phonon-phonon processes, isotopic point defect scattering, and boundary scattering, except in NaF where isotopic scattering is absent. The Callaway model, in all cases, gives a good fit and excellent agreement is obtained for diamond and solidified argon and neon. In the vicinity of the conductivity maxima the Callaway model gives a better fit with the experimental curve. The disagreement between the results of the two models is most pronounced in solidified argon and neon where the Debye temperature is low and Umklapp processes are predominant at the conductivity maximum. In diamond, where the Debye temperature is quite high and Umklapp processes are unimportant at the temperatures of interest the results of both models are more or less similar. The Casimir model for finding the characteristic length for the boundary scattering fails to explain the observed results in solidified argon and neon. This indicates the existence of microscale fluctuations in the composition of the solid. In NaF the characteristic length has to be decreased by a factor of about 5. For diamond the value of the parameter $A$, which gives the best fit with experiment, is a hundred times the value calculated from Klemens' expression for $A$. This supports the earlier suggestion that there occur clusters of point defects in diamond. Good agreement with the experimental results beyond the conductivity maximum is obtained by taking $(B_1+B_2)$ to be temperature independent in all the cases.

## I. INTRODUCTION

KLEMENS[1–4] has worked out a detailed theory of thermal conductivity and influence of lattice defects. In perfect crystal lattices, the thermal conductivity is determined by intrinsic processes, i.e., phonon-phonon collisions which become less frequent as the temperature decreases. This explains the increase in the thermal conductivity with decrease in temperature. At low temperatures the phonons are scattered at the crystal surfaces and boundary scattering becomes dominant with a constant mean free path determined by dimensions of the crystal. In addition, any departure from perfect periodicity causes phonon scattering. In the present paper only three sources of phonon scattering are postulated; phonon-phonon processes, isotopic point defect scattering, and boundary scattering. Naturally occurring isotopes differ in mass from each other and this means departure from perfect periodicity in the lattice. Such defects, known as isotopic point defects, scatter phonons with a Rayleigh scattering law if they are randomly distributed. For the detailed discussion of mass-difference scattering, boundary effects, and other scattering processes, the reader is referred to the review article by Carruthers.[5]

Recently Callaway[6] has given a phenomenological model to explain lattice thermal conductivity at low temperatures and applied his theory to germanium with considerable success. Since then it has been used in the study of the influence of $F$ centers on the thermal conductivity of LiF,[7] conductivity of solid solutions of cadmium telluride in mercury telluride,[8] of germanium-silicon alloys,[9] of[10] LiF containing various relative concentrations of the isotopes of $\mathrm{Li}^6$ and $\mathrm{Li}^7$, and of isotopic mixtures of solid helium.[11] In the present paper, the theoretical models of Callaway and Klemens are compared to the experimental results in (a) silicon and diamond, (b) solidified argon and neon, and (c) KCl and NaF.

## II. THEORETICAL MODELS

Using the Debye expression for the specific heat, the expression for the thermal conductivity $\kappa$ is

$$\kappa=\frac{k}{2\pi^2 c}\int_0^{\omega_D}\tau_c\frac{\hbar^2\omega^2}{k^2T^2}\frac{e^{\hbar\omega/kT}\omega^2}{(e^{\hbar\omega/kT}-1)^2}d\omega, \tag{1}$$

where $k$ is the Boltzmann constant, $c$ is the sound velocity, $\omega$ is the phonon frequency, $\omega_D$ is the Debye limiting frequency, and $\tau_c$ is the combined relaxation time. According to Callaway, $\tau_c$ is given by

$$\begin{aligned}\tau_c&=(\tau_B^{-1}+\tau_N^{-1}+\tau_P^{-1}+\tau_u^{-1})^{-1}\\&=(cL^{-1}+B_1T^3\omega^2+A\omega^4+B_2T^3\omega^2)^{-1},\end{aligned} \tag{2}$$

where (1) $\tau_B=L/c$ is the relaxation time for boundary scattering and $L$ is the characteristic length of the specimen, (2) $\tau_N$ is the relaxation time for the normal three-phonon processes, and is taken to be proportional to $\omega^{-2}T^{-3}$, (3) $\tau_P$ is the relaxation time due to impurity scattering and is taken to be independent of temperature and proportional to $\omega^{-4}$, (4) $\tau_u$ is the relaxation time for the Umklapp processes and is proportional to $e^{\Theta/\alpha T}\omega^{-2}T^{-3}$, where $\Theta$ is the Debye temperature and $\alpha$ is a constant characteristic of the vibrational spectrum of the material.

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[1] P. G. Klemens, Proc. Roy. Soc. (London) **A208**, 108 (1951).

[2] P. G. Klemens, Proc. Phys. Soc. (London) **A68**, 1113 (1955).

[3] P. G. Klemens, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1956), 2nd ed., Vol. 14, p. 198.

[4] P. G. Klemens, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1958), Vol. 7, p. 1.

[5] P. Carruthers, Revs. Modern Phys. **33**, 92 (1961).

[6] J. Callaway, Phys. Rev. **113**, 1046 (1959).

[7] R. O. Pohl, Phys. Rev. **118**, 1499 (1960).

[8] J. Blair, Bull. Am. Phys. Soc. **5**, 164 (1960).

[9] A. M. Toxen, Phys. Rev. **122**, 450 (1961).

[10] J. Callaway and H. C. von Baeyer, Phys. Rev. **120**, 1149 (1960).

[11] J. Callaway, Phys. Rev. **122**, 787 (1961).

Callaway's expression for thermal conductivity is

$$\kappa=\frac{k}{2\pi^2 c}(I_1+\beta I_2), \tag{3}$$

where

$$I_1=\int_0^{\omega_D}\tau_c\frac{\hbar^2\omega^2}{k^2T^2}\frac{e^{\hbar\omega/kT}\omega^2}{(e^{\hbar\omega/kT}-1)^2}d\omega, \tag{4}$$

$$I_2=\int_0^{\omega_D}\left(\frac{\tau_c}{\tau_N}\right)\frac{\hbar^2\omega^2}{k^2T^2}\frac{e^{\hbar\omega/kT}\omega^2}{(e^{\hbar\omega/kT}-1)^2}d\omega, \tag{5}$$

and

$$\beta=\int_0^{\Theta/T}\left(\frac{\tau_c}{\tau_N}\right)\frac{x^4e^x}{(e^x-1)^2}dx \Bigg/ \int_0^{\Theta/T}\frac{1}{\tau_N}\left(1-\frac{\tau_c}{\tau_N}\right)\frac{x^4e^x}{(e^x-1)^2}dx. \tag{6}$$

According to Klemens, the relaxation time due to scattering by point defects is given by

$$\frac{1}{\tau_P}=\left[\frac{V_0}{4\pi c^3}\sum f_i\left(1-\frac{m_i}{\bar{m}}\right)^2\right]\omega^4=\left[\frac{V_0}{4\pi c^3}\Gamma\right]\omega^4=A\omega^4, \tag{7}$$

where $V_0$ is the atomic volume, $m_i$ is the mass of the $i$th species of the atom and $\bar{m}$ is the average atomic mass. If this expression for relaxation time is substituted in Eq. (1), the resultant integral diverges for the low-frequency phonons. In order to remove this divergence Klemens assumed that the long-wavelength phonons which cause the divergence are scattered by normal three-phonon processes, which by themselves conserve wave vector and, therefore, do not cause thermal resistance. This scattering annihilates long-wavelength phonons and creates phonons with a typical frequency such that $1/\tau_I=A(kT/\hbar)^4$ for $\omega<kT/\hbar$ and $1/\tau_I=A\omega^4$ for $\omega>kT/\hbar$. Combining the boundary scattering with Klemens relation for point defect scattering, the resultant expression for thermal conductivity is

$$\kappa=\frac{\hbar}{2\pi^2 cAT}\left[\frac{1}{1+R}\int_0^1\frac{x^4e^x}{(e^x-1)^2}dx+\frac{1}{R}I_4(R)\right], \tag{8}$$

where

$$R=\frac{c}{LA}\left(\frac{\hbar}{kT}\right)^4 \text{ and } I_4(R)=\int_1^\infty\frac{x^4e^x}{[1+(x^4/R)](e^x-1)^2}dx.$$

The additive resistance approximation is used to compare the theory with experiment when Umklapp processes are also present.

## III. COMPARISON OF THE THEORETICAL MODELS WITH THE EXPERIMENTAL RESULTS

The thermal conductivity at different temperatures has been computed numerically on the basis of the Callaway model and the Klemens model and compared with the experimental results. The substances which we have chosen for comparison are (a) silicon and diamond, (b) solidified gases like argon and neon, and (c) alkali halides such as KCl and NaF. In all the cases, data are available at temperatures below the thermal conductivity maximum, where boundary scattering makes important contribution to thermal resistivities. In NaF there will be no isotopic scattering as Na and F both occur as single isotopes.

For the evaluation of the basic parameters of the Callaway model, the following procedure is adopted. The term $\beta I_2$ in Eq. (3) is in general small and is not considered in evaluating the conductivity at different temperatures. However, its values has been estimated in each case near the conductivity maximum where it is expected to be important. At a temperature far beyond the maximum where the phonon-phonon scattering predominates and contributions due to isotopic and boundary scatterings may be negligible, the parameter $(B_1+B_2)$ is roughly determined from

$$\kappa=\frac{k^2}{6\hbar(B_1+B_2)cT^2}\left[1-\frac{3}{\pi^2}e^{-\Theta/T}\left(\frac{\Theta^2}{T^2}+2\frac{\Theta}{T}+2\right)\right], \tag{9}$$

where $\Theta$ is the Debye temperature. The above expression is the same as Eq. (36) of Callaway[6] after neglecting the terms proportional to $B_2/B_1$ and containing $c/L$. Using this value of $(B_1+B_2)$, the value of $A$ is roughly determined by numerically integrating Eq. (4) after neglecting the $\beta I_2$ term. Lastly, different values of $A$ and $(B_1+B_2)$ are tried and Eq. (4) is numerically integrated for each set of the above parameters till the agreement between theory and experiment is the best for the entire temperature range.

In order to compare the Klemens model with the experimental results, values of the parameters $c/L$ and $A$ have been taken to be the same as obtained from the analysis based on Callaway's theory. Values of the integral $I_4(R)$ are given in Slack's[12] paper. For calculating conductivity due to Umklapp processes, the expression $\kappa_u=BT^3e^{\Theta/\alpha T}$ is used, where $B$ and $\alpha$ are constants for the substance concerned. $B$ is calculated from the experimental value of conductivity at a temperature far beyond the conductivity maximum where the boundary and isotope scatterings are negligible. $\alpha$ is chosen to fit the Umklapp curve in the high-temperature region. For solid argon and neon the conductivity due to umkalpp processes is plotted separately. For diamond the scattering due to Umklapp processes is unimportant in the region of present interest due to the high value of $\Theta$ and has not been considered. The conductivity due to boundary and isotopic scattering is calculated from Eq. (8). For Si and KCl, the total resistivity is obtained by adding the resistivity due to Umklapp processes to the resistivity obtained from Eq. (8).

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[12] G. A. Slack, Phys. Rev. **105**, 832 (1957).

### (a) Silicon and Diamond

The thermal conductivity in silicon at low temperatures has been studied by Carruthers *et al.*[13] The area of the cross section of the specimens is $2.15\times1.98$ mm$^2$. According to Casimir's model the characteristic length $L$ of the specimen is given by $L=2R$ in the case of a circular cross section of radius $R$. For a rectangular cross section of area $S$, $L=1.12S^{\frac{1}{2}}$. The value of $c$, the average phonon velocity, was taken to be $5\times10^5$ cm/sec. Using this value of $c$, $c/L$ was calculated to be $2.14\times10^6$ sec$^{-1}$. Values of $A$ and $(B_1+B_2)$ which give the best fit, are $A=0.5\times10^{-44}$ sec$^3$ and $(B_1+B_2)=5\times10^{-24}$ sec deg$^{-3}$. This value of $A$ can be compared with the $A$ calculated from Klemens' expression. Using $\Gamma=2.64\times10^{-4}$, $V_0=19.9\times10^{-24}$ cm$^3$, $c=5\times10^5$ cm/sec' $A$ comes out to be $0.33\times10^{-44}$ sec$^3$. The agreement seems to be quite good.

It may be seen from Fig. 1 that the Callaway model gives better agreement with experiment. However, at temperatures lower than 10°K, both models give almost the same values for the conductivity. In the temperature region of maximum conductivity the Klemens model gives a poor fit to the data. It may be seen further that calculated results on the basis of the Callaway model beyond 100°K are lower than the experimental results. If we modify the value of $(B_1+B_2)$ so that agreement is better in the temperature range 100°K to 200°K, the fit becomes poorer in the region of the maximum. The maximum contribution due to the $\beta I_2$ term, in the limiting case when $B_1=0$, at the conductivity maximum is 0.36 w/cm-deg.

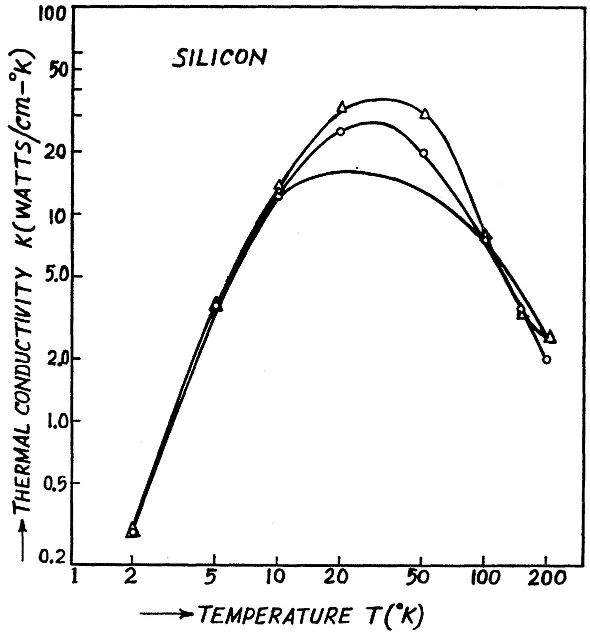

FIG. 1. Thermal conductivity of silicon. Solid curve represents experimental values by Carruthers *et al.* Circles denote the values calculated on the basis of Callaway's model with the parameters $A=0.5\times10^{-44}$ sec$^3$, $(B_1+B_2)=5\times10^{-24}$ sec deg$^{-3}$, and $c/L=2.14\times10^6$ sec$^{-1}$. Triangles denote the values calculated with Klemen's expression with the same values of $A$ and $c/L$. $B=8.25\times10^{-8}$ w/cm deg$^4$ and $\alpha=1$.

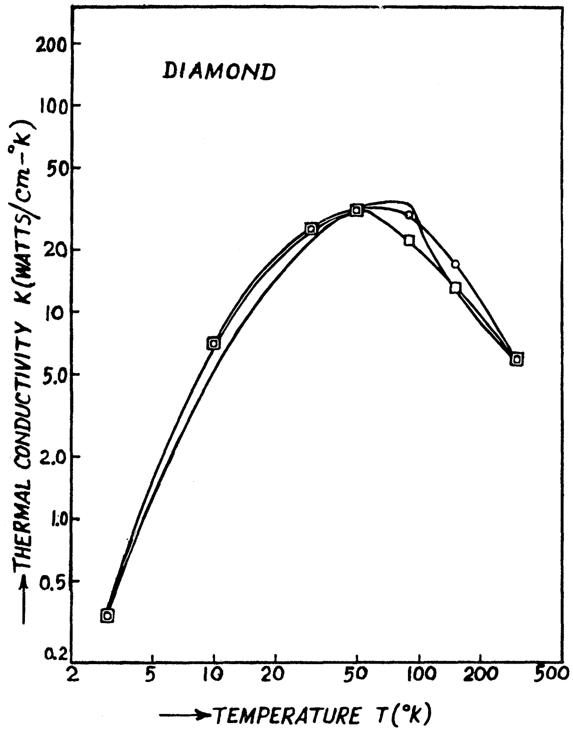

FIG. 2. Thermal conductivity of diamond. Solid curve represents the experimental values by Berman *et al.* Circles denote the values calculated with Callaway's model with the parameters $A=2.09\times10^{-45}$ sec$^3$, $(B_1+B_2)=2.0\times10^{-25}$ sec deg$^{-3}$, and $c/L=2.68\times10^6$ sec$^{-1}$. Squares denote the values obtained with Klemens' model taking the same values of $A$ and $c/L$.

For diamond the experimental results, as shown in Fig. 2, are from Berman *et al.*[14] The area of the cross section is $(0.39)^2$ cm$^2$. Using[15] $c=1.18\times10^6$ cm/sec, the ratio $c/L$ is $2.68\times10^6$ sec$^{-1}$. The values of $A$ and $(B_1+B_2)$ which give the best fit are $A=2.09\times10^{-45}$ sec$^3$ and $(B_1+B_2)=2\times10^{-25}$ sec deg$^{-3}$. It is interesting to note that the value of $A$ which gives the best fit with the experiment is 100 times the value calculated from Klemens' expression. The above result suggests that in diamond there occur groups or clusters as suggested by Berman *et al.*, and previously by Klemens.[16] The over-all dimensions of such defects may be as large as 30 to 100 A. For calculating $A$ from Klemens' expression we have used $\Gamma=0.76\times10^{-4}$ and $V_0=5.68\times10^{-24}$ cm$^3$ (from Slack's[15] paper), and[17] $\Theta=1960$°K for the calculation of the conductivity. If for the

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[13] J. A. Carruthers, T. H. Geballe, H. M. Rosenberg, and J. M. Ziman, Proc. Roy. Soc. (London) **A238**, 502 (1957).

[14] R. Berman, F. E. Simon, and J. M. Ziman, Proc. Roy. Soc. (London) **A220**, 171 (1953).

[15] G. A. Slack, Phys. Rev. **105**, 829 (1957).

[16] P. G. Klemens, Phys. Rev. **86**, 1055 (1952).

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

Klemens model we use the value of $A$ which gives the best fit with experiment for the Callaway model, we see from Fig. 2 that for diamond the Klemens model also gives approximately a good fit with experiment. In diamond too the maximum contribution due to the $\beta I_2$ term in the limiting case, i.e., $B_1=0$, is 0.07 w/cm-deg.

### (b) Solidified Argon and Neon

The thermal conductivity in solidified argon and neon at low temperatures has been studied by White and Woods.[18] The radius of the circular cross section is the same in both cases and is equal to $(0.25\times 2.54)$ cm and hence the characteristic length is 1.27 cm for each case. The compressibility of solidified argon and neon has been studied by Stewart[19] and is $1.334\times 10^{-4}$ cm$^2$/kg for argon and $1.01\times 10^{-4}$ cm$^2$/kg for neon. Using the density $\rho=1.77$ g/cm$^3$ for argon and $\rho=1.61$ g/cm$^3$ for neon, the phonon velocity comes out to be $6.42\times 10^4$ cm/sec and $7.75\times 10^4$ cm/sec, respectively. Argon has three isotopes and its mean atomic weight is 39.944. Neon has also three isotopes and its average atomic weight is 20.183. Values of $\Gamma$ for argon and neon are $3.56\times 10^{-5}$ and $8.25\times 10^{-4}$, respectively. Values of $\Theta$ for argon and neon are[20] 80°K and 64°K, respectively. The parameter $A$, as calculated from Klemens' expression, is $4.01\times 10^{-43}$ sec$^3$ for argon and $2.93\times 10^{-42}$ sec$^3$ for neon. The values of the parameter $(B_1+B_2)$ which give the best fit with experiment for argon and neon are $8.0\times 10^{-19}$ sec deg$^{-3}$ and $3.5\times 10^{-18}$ sec deg$^{-3}$, respectively. For solid argon and neon the Debye temperature $\Theta$ is low and scattering due to Umklapp processes is important at the conductivity maximum. The contributions due to the $\beta I_2$ term for argon and neon at the conductivity maximum for $[B_1/(B_1+B_2)]=0.9$ are $2.2\times 10^{-3}$ and $1.4\times 10^{-3}$ w/cm-deg, respectively.

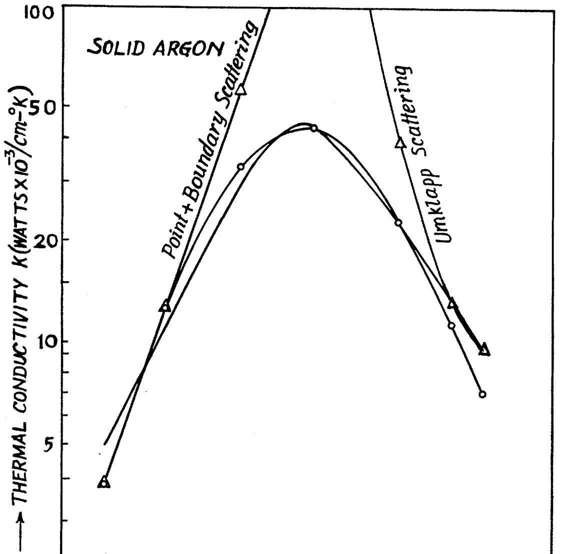

FIG. 3. Thermal conductivity of solidified argon. Solid curve denotes the experimental values given by White and Woods. Circles denote the values calculated with Callaway's model with the parameters $A=4.01\times 10^{-43}$ sec$^3$, $(B_1+B_2)=8\times 10^{-19}$ sec deg$^{-3}$, and $c/L=1.3\times 10^9$ sec$^{-1}$. Triangles denote the values on the basis of Klemens' model with the same values of $A$ and $c/L$. $B=1.11\times 10^{-8}$ w/cm-deg$^4$ and $\alpha=0.8$.

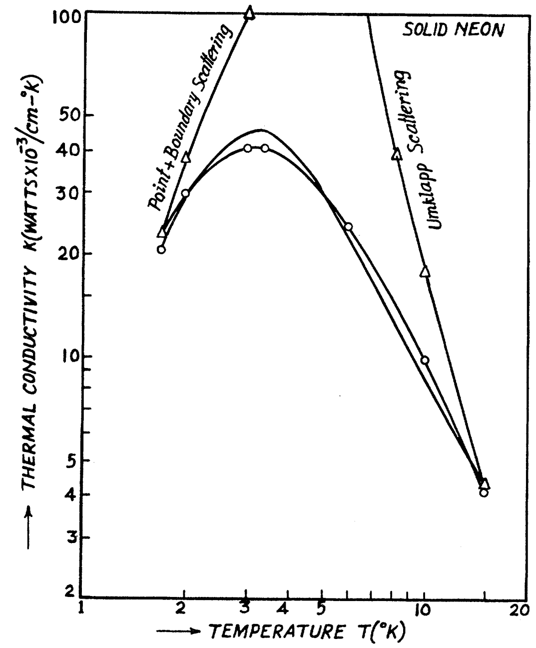

FIG. 4. Thermal conductivity of solidified neon. Solid curve represents the experimental values observed by White and Woods. Circles denote the values on the basis of Callaway's model with the parameters $A=2.93\times 10^{-42}$ sec$^3$, $(B_1+B_2)=3.5\times 10^{-18}$ sec deg$^{-3}$, and $c/L=1\times 10^8$ sec$^{-1}$. Triangles denote the values on the basis of Klemens' model using the same values of $A$ and $c/L$. $B=5.2\times 10^{-8}$ w/cm deg$^4$ and $\alpha=1.1$.

The interesting thing to be noted is that Casimir's model for finding out the characteristic length of the specimen for boundary scattering fails completely for solidified argon and neon. Values of $c/L$ have been determined from the experimental results at the lowest temperature where the conductivity is mainly dependent upon the boundary scattering and are $1.3\times 10^9$ sec$^{-1}$ for argon and $1.0\times 10^8$ sec$^{-1}$ for neon. This may be compared with $c/L=5.05\times 10^4$ sec$^{-1}$ for argon and $c/L=6.1\times 10^4$ sec$^{-1}$ for neon on the basis of Casimir's model. It was realized by White and Woods that in solidified argon and neon, severe physical disorders are produced due to strains imposed during cooling and it is natural that for such highly disordered state Casimir's model should not hold good. Low values of characteristic length as obtained from the conductivity

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[18] G. K. White and S. B. Woods, Phil. Mag. **3**, 785 (1958).

[19] J. W. Stewart, J. Phys. Chem. Solids **1**, 146 (1956).

[20] E. R. Dobbs and G. O. Jones, *Reports on Progress in Physics* (The Physical Society, London, 1957), Vol. 20, p. 516.

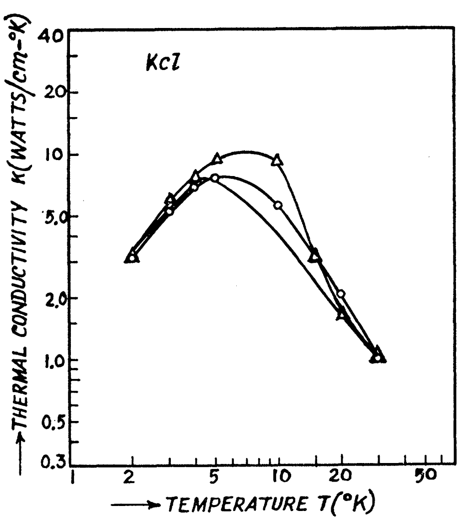

FIG. 5. Thermal conductivity of KCl. Solid curve represents the experimental values given by Slack. Curve marked by circles is drawn on the basis of Callaway's model by taking $A=1.97\times10^{-43}$ sec$^3$, $(B_1+B_2)=1.2\times10^{-21}$ sec deg$^{-3}$, and $c/L=2.48\times10^5$ sec$^{-1}$. Curve marked by triangles is based on Klemens' model with the same values of $A$ and $c/L$. $B=1.64\times10^{-6}$ w/cm deg$^4$ and $\alpha=2.3$.

measurements at low temperatures might be due to microscale fluctuations in the composition of the solid.

### (c) KCl and NaF

The thermal conductivity in KCl at low temperatures has been studied and compared with Klemens' model by Slack.[12] We have chosen KCl for the study of Callaway's model as it has not been done so far. It may be seen from Fig. 5 that Callaway's model gives a good fit with the experimental data. The parameters which give the best fit are $A=1.97\times10^{-43}$ sec$^3$, $(B_1+B_2)=1.2\times10^{-21}$ sec deg$^{-3}$, and $c/L=2.48\times10^5$ sec$^{-1}$. The values of other constants are $\Gamma=1.77\times10^{-4}$, $V_0=6.2\times10^{-23}$ cm$^3$, $\Theta=233$°K, and $c=1.88\times10^5$ cm/sec, which have been taken by Slack.[12] The value of $A$ from Klemens' expression comes out to be $1.31\times10^{-43}$ sec$^3$. The maximum value of $(k/2\pi^2c)\beta I_2$ at the conductivity maximum for the limiting case $B_1=0$ is 0.2 w/cm deg.

As Na and F are both single isotopes, there does not arise the question of isotopic scattering and only phonon-phonon and boundary scatterings are considered. The experimental results for the thermal conductivity in sodium fluoride have been taken from Cohen's[21] paper. The area of the cross section of the specimen is 0.0738 cm$^2$. The phonon velocity $c$ has been calculated from $\Theta/T=\hbar c(6\pi^2N)^{\frac{1}{3}}/kT$, where $\Theta$ is the Debye temperature and is equal to 469°K.[22] The phonon velocity comes out to be equal to $4.61\times10^5$ cm/sec and $c/L=1.5\times10^6$ sec$^{-1}$, if $L$ is the length calculated on the basis of Casimir's model. The parameters which give the best fit are $(B_1+B_2)=1\times10^{-22}$ sec deg$^{-3}$ and $c/L=7.0\times10^6$ sec$^{-1}$. It may be noticed that in order to fit the Callaway model with the experimental data $\tau_B=L/c$ had to be decreased by a factor of about 5 from the value calculated from the specimen dimensions (see Fig. 6). This indicates the existence of internal boundaries associated with the fluctuations in the composition of the solid. The contribution due to the $\beta I_2$ term for $[B_1/(B_1+B_2)]=0.9$ is 0.5 w/deg cm at the conductivity maximum where Umklapp processes are important in the absence of isotopic scattering.

## IV. SUMMARY

For all the substances we have studied on the basis of Callaway's and Klemens' models, the former gives better fit and especially in solidified argon and neon, Callaway's model gives an excellent fit. This shows that normal processes are approximated in a better way in Callaway's theory. The values of the various parameters used in the calculation together with their theoretical values are given in Table I. The following conclusions are drawn from the present calculations.

(1) In the vicinity of the conductivity maximum Callaway's model gives a better fit with the experimental curve. The disagreement between the results of the two models is most pronounced in solidified argon and neon where the Debye temperature is low and

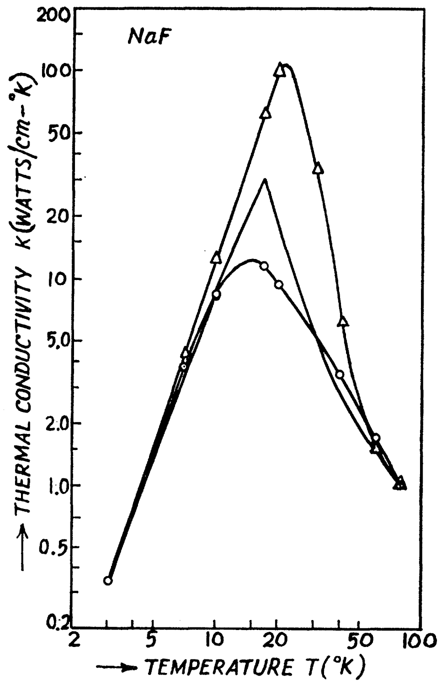

FIG. 6. Thermal conductivity of NaF. Solid curve represents the experimental values quoted from Cohen. Circles denote the values on the basis of Callaway's model with parameters $(B_1+B_2)=1\times10^{-22}$ sec deg$^{-3}$ and $c/L=7\times10^6$ sec$^{-1}$. Triangles denote values obtained from Klemens' model with the same values of $A$ and $c/L$. $B=3.92\times10^{-8}$ w/cm deg$^4$ and $\alpha=1.5$.

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[21] A. F. Cohen, J. Appl. Phys. 29, 870 (1958).

[22] S. S. Mitra and S. K. Joshi, Physica 26, 284 (1960).

TABLE I. Parameters of Callaway's model giving the best fit to the experimental data.

| | $A_{\text{exp}}$ ($10^{-43}$ sec$^3$) | $(B_1+B_2)_{\text{exp}}$ (sec deg$^{-3}$) | $(c/L)_{\text{exp}}$ ($10^7$ sec$^{-1}$) | $A_{\text{cal}}$ ($10^{-43}$ sec$^3$) | $A_{\text{exp}}/A_{\text{cal}}$ | $(c/L)_{\text{exp}}/(c/L)_{\text{cal}}$ |
|---|---|---|---|---|---|---|
| 1. Silicon | 0.05 | $5\times10^{-24}$ | 0.214 | 0.033 | 1.5 | 1.0 |
| 2. Diamond | 0.021 | $2\times10^{-25}$ | 0.268 | $2.1\times10^{-4}$ | 100 | 1.0 |
| 3. Solid argon | 4.01 | $8\times10^{-19}$ | 130 | 4.01 | 1.0 | $2.6\times10^4$ |
| 4. Solid neon | 29.32 | $3.5\times10^{-18}$ | 10 | 29.32 | 1.0 | $1.6\times10^3$ |
| 5. KCl | 1.97 | $1.2\times10^{-21}$ | 0.025 | 1.31 | 1.5 | 1.0 |
| 6. NaF | ... | $1\times10^{-22}$ | 0.7 | ... | ... | 4.7 |

Umklapp processes are predominant at the conductivity maximum. In diamond, where the Debye temperature is quite high and Umklapp processes are unimportant at the temperatures of interest, the results of both the models are more or less similar.

(2) Casimir's model for the determination of the characteristic length for boundary relaxation time $\tau_B = L/c$ fails to explain the observed results in solidified argon and neon. This indicates the existence of microscale fluctuations in the composition of the solid. In NaF too, the calculations indicate that there exist internal boundaries, associated with fluctuations.

(3) The value of the parameter $A$ for the isotopic scattering time $1/\tau_P = A\omega^4$, which explains the experimental results in diamond, is a hundred times the value calculated from Klemens' formula. This supports the earlier suggestion that in diamond there occur groups or clusters which result in the increased value of $A$.

(4) The assumption that the parameter $(B_1+B_2)$ is temperature independent, gives approximately a good fit to the experimental curve in all the cases.

In Casimir's theory the scattering of phonons is completely diffuse, the incident phonons being absorbed and then re-emitted with the equilibrium distribution corresponding to local temperature. The temperature is assumed to be so low that the only collisions made by the phonons are with the boundary. Boundary scattering does not occur uniformly throughout the crystal, which makes the distribution function depend upon position. The phonons are also scattered by other defects and by each other. Assuming that there is specular reflection of the phonons and that there exists an equivalent relaxation time which includes the effect of normal processes, and proceeding directly from the Boltzmann equation, attempts[23,5] have been made to find an expression for the effective boundary relaxation time. Specular reflection leads to an increased value of the boundary relaxation time $\tau_B$. However, for solid argon and neon and NaF, in order to explain the experimental results we require increased values of $c/L$, which means the existence of internal boundaries.

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[23] C. Herring, Phys. Rev. **96**, 1163 (1954).

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# Scattering of Slow Neutrons by Solid and Liquid Terphenyls*

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The partial differential cross sections for scattering of slow neutrons by samples of solid and liquid terphenyls were measured and are presented in scattering law form. These data show that the difference between the scattering from the liquid and the solid is principally of magnitude and not shape. No effects due to the ortho-, meta-, or para-chemical binding is evident. Resonances attributed to transitions between vibrational states in the molecule are seen in the liquid data but not in the solid data. Fourier inversion of the scattering-law data show that the space-time self-correlation function $G_s(\bar{r},t)$ for these materials is not Gaussian. A comparison of these data with other available scattering-law curves indicate that the curves may be grouped into classes of (1) polycrystalline solids, (2) liquid–amorphous solids, and (3) gases. The total cross sections for the terphenyls for 0.0251 ev neutrons were also measured.

## A. INTRODUCTION

SLOW neutron inelastic scattering data are rapidly becoming available that reveal the extensive properties and details of the solid, liquid, and gaseous states. The inelastic neutron scattering from methane gas[1] and from propane gas[2] have been measured using the MTR

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[1] P. D. Randolph, R. M. Brugger, K. A. Strong and R. E. Schmunk, Phys. Rev. **124**, 460 (1961).

[2] K. A. Strong, G. D. Marshall, R. M. Brugger and P. D. Randolph, Phys. Rev. **125**, 933 (1962).