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[33]See, for example, J. M. Ziman, *Electrons and Phonons* (Oxford U. P., London, 1962), pp. 357–367.

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Electron-Phonon Umklapp-Scattering Processes in the Low-Temperature Thermal Resistivity of Potassium*

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Using the variational method, the *electronic* contribution to the low-temperature thermal conductivity of potassium is calculated for a number of different pseudopotentials and a realistic phonon spectrum. A detailed evaluation of electron-phonon umklapp-scattering effects is presented. In particular, umklapp processes have a negligible effect on the thermal resistivity and Wiedemann-Franz ratio below ~2 °K, but significantly enhance both at temperatures above this. Higher-order corrections to the variational calculation are evaluated and it is found that these are significantly larger for the umklapp component of the thermal resistivity than for the normal component. The *lattice* contribution to the low-temperature thermal conductivity is calculated and compared with the results for the electronic component.

I. INTRODUCTION

In an earlier paper[1] we reported calculations of the ultrasonic attenuation and electrical resistivity of potassium which included a detailed evaluation of the role of electron-phonon umklapp-scattering processes at low temperatures. In this paper, a similar treatment has been extended to the thermal resistivity of potassium.

In Secs. II and III, the electronic contribution to the thermal conductivity is calculated, including an explicit evaluation of umklapp-scattering effects. It is found that above ~2 °K umklapp processes significantly enhance both the thermal resistivity and the Wiedemann-Franz ratio. The results for the total thermal resistivity (umklapp plus normal components) are in reasonable agreement with experimental observation,[2] although a detailed comparison must await more precise experimental study. In Sec. IV, higher-order corrections to the variational calculation are evaluated. The umklapp component is affected by such corrections to a much greater extent than the normal component. In Sec. V, the lattice conductivity is calculated in order to quantitatively evaluate the relative importance of heat conduction by the lattice compared to that by the electronic system.

II. THEORY

Taking the phonon system to be in equilibrium, the variational expression for the thermal resistivity W of a metal due to electron-phonon scattering (i.e., in the absence of electron-impurity scattering) is given by[3]

$$W = \frac{(1/k_B)\int\int\int [\phi(\vec{k}') - \phi(\vec{k})]^2 P\, d\vec{q}\, d\vec{k}\, d\vec{k}'}{|\vec{v}(E-\mu)\phi(\vec{k})(\partial f_0/\partial E)\, d\vec{k}|^2}\,, \tag{1}$$

where P is the probability for an electron in a state of wave vector $\vec{k}$ to be scattered to a state of wave vector $\vec{k}'$ through the absorption (or creation) of a phonon of wave vector $\vec{q}$ ($\vec{k}' - \vec{k} = \vec{q} + \vec{G}$, with $\vec{G}$ a reciprocal-lattice vector). Here $\vec{v}$ is the electron group velocity, E is the electron energy, k_B is Boltzmann's constant, and μ is the chemical potential. The trial function ϕ represents the deviation of the true electron distribution f from the equilibrium Fermi distribution f_0:

$$f - f_0 \equiv \left(-\frac{\partial f_0}{\partial E}\right)\phi\,; \tag{2}$$

it is chosen to minimize the right-hand side of Eq. (1). As a first approximation, we shall use the standard trial function appropriate to the solution

of the Bloch equation in the relaxation-time approximation

$$\phi(\vec{k}) \propto \vec{k}\cdot\vec{u}(E-\mu)\ . \tag{3}$$

Here a spherical Fermi surface has been assumed such that $\vec{v}$ and $\vec{k}$ are parallel; $\vec{u}$ is a unit vector in the direction of the heat current.

In the approximation of a local pseudopotential $V(\vec{k}'-\vec{k})$, spherical Fermi surface, one-plane-wave electron states, and cubic-crystal symmetry, this trial function leads to the expression

$$W=\frac{m}{L_0 Tne^2}\,\frac{3\Omega_0 N(0)}{8k_F^4}\,\frac{\hbar}{Mk_BT}\sum_\lambda\int_{q\le 2k_F} d^3q\,\frac{|\vec{\epsilon}(\vec{q};\lambda)\cdot\vec{q}\,|^2\,|V(\vec{q})|^2}{q(1-e^{-z})(e^z-1)}\left[\frac{q^2}{3}-\left(\frac{z^2}{6\pi^2}\right)q^2+\left(\frac{z^2}{\pi^2}\right)k_F^2\right]. \tag{4}$$

The same notation has been used as in Ref. 1: $\vec{q}\equiv\vec{k}'-\vec{k}$; m is the electron mass; Ω_0 is the volume per ion; k_F is the Fermi wave number; $N(0)$ is the single-spin electron density of states per unit volume at the Fermi surface, $N(0)=mk_F/2\pi^2\hbar^2$; m is the electron mass; M is the ion mass; T is the absolute temperature; n is the valence electron density; e is the electronic charge. The phonons are described in a repeated-zone scheme; they are specified by their frequencies $\omega(\vec{q};\lambda)$ and polarization vectors $\vec{\epsilon}(\vec{q};\lambda)$ for wave vector $\vec{q}$ and polarization branch λ (one longitudinal, two transverse). In Eq. (4), we have introduced the dimensionless parameter z to represent the ratio of $\hbar\omega(\vec{q};\lambda)$ to k_BT. The integral extends over the volume of a sphere of radius $2k_F$.

III. RESULTS AND DISCUSSION

In evaluating expression (4), the same phonon spectrum[4] and pseudopotentials were used as in previous papers[1,5,6] (for a listing of the pseudopotentials, see Table IV of Ref. 6). The results for the total thermal resistivity have been explicitly separated into normal and umklapp components and are tabulated in Table I and plotted in Fig. 1. As in the case for the electrical resistivity and ultrasonic attenuation, the normal component at low temperatures is nearly identical for all three pseudopotentials. It is the umklapp component which is entirely responsible for the variation between the total (umklapp plus normal) results for the various pseudopotentials. Umklapp processes freeze out at low temperatures, contributing only a negligible amount to the thermal resistivity at temperatures below ~2 °K. At temperatures above this, however, the umklapp component significantly enhances the thermal resistivity over that due to the normal component alone. This results in a temperature dependence in the range ~2–6 °K that is slightly steeper (depending on the pseudopotential) than the usual T^2 dependence predicted by the Bloch theory, which neglects umklapp processes. The effect, however, is much less pronounced than for the electrical resistivity.[1]

In Fig. 2 is presented the temperature dependence of the Wiedemann-Franz ratio (divided by the square of the temperature and expressed in units of the Lorenz number L_0). The ratio has been calculated individually for both the normal $[\rho_N/(L_0T^3W_N)]$ and umklapp $[\rho_U/(L_0T^3W_U)]$ resistivity components as well as the total $[\rho_T/(L_0T^3W_T)]$. Values of the electrical resistivity ρ used are those of Table III in Ref. 6. Large-angle umklapp-scattering events are more effective in degrading the electrical current than the thermal

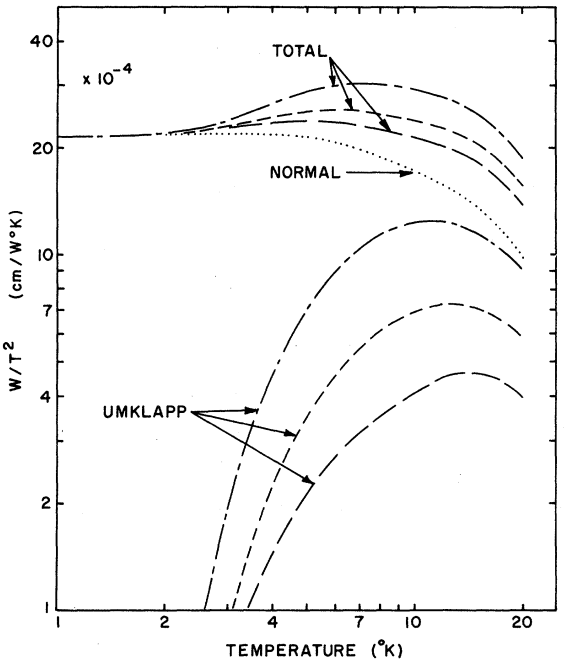

FIG. 1. Temperature dependence of the thermal resistivity of potassium W as calculated using several different pseudopotentials: long and short dashes, Bardeen pseudopotential; short dashes, lower Lee-Falicov psuedopotential; long dashes, Ashcroft pseudopotential. In each case the total thermal resistivity has been explicitly separated into normal and umklapp components. The normal components from the three pseudopotentials were so nearly identical they could not be resolved on this scale.

TABLE I. Electronic component of the thermal resistivity of potassium as calculated using several different pseudopotentials. The results have been separated into normal and umklapp contributions. In each case the resistivity has been divided by the square of the temperature, the units being 10^{-4} cm/W °K.

T	Bardeen pseudopotential			Lower Lee-Falicov pseudopotential			Ashcroft pseudopotential		
(°K)	normal	umklapp	total	normal	umklapp	total	normal	umklapp	total
1.00	21.5	...	21.5	21.5	...	21.5	21.5	...	21.5
1.25	21.6	...	21.6	21.6	...	21.6	21.6	...	21.6
1.50	21.6	0.02	21.6	21.6	0.01	21.6	21.6	0.01	21.6
1.75	21.6	0.09	21.7	21.7	0.04	21.7	21.7	0.04	21.7
2.00	21.7	0.24	21.9	21.7	0.10	21.8	21.8	0.11	21.9
2.25	21.8	0.49	22.3	21.8	0.22	22.0	21.8	0.21	22.0
2.50	21.8	0.86	22.7	21.9	0.39	22.3	21.9	0.35	22.3
2.75	21.9	1.3	23.2	22.0	0.62	22.6	22.0	0.51	22.5
3.00	22.0	1.9	23.9	22.0	0.89	22.9	22.1	0.69	22.8
3.25	22.0	2.6	24.6	22.1	1.2	23.3	22.1	0.88	23.0
3.50	22.0	3.3	25.3	22.1	1.5	23.6	22.2	1.1	23.3
3.75	22.0	4.0	26.0	22.1	1.9	24.0	22.2	1.3	23.5
4.00	22.0	4.6	26.6	22.1	2.2	24.3	22.2	1.4	23.6
4.50	21.8	6.0	27.8	21.9	3.0	24.9	22.0	1.8	23.8
5.00	21.5	7.3	28.8	21.6	3.6	25.2	21.7	2.1	23.8
6.00	20.7	9.2	29.9	20.8	4.7	25.5	20.9	2.7	23.6
8.00	18.7	11.4	30.1	18.8	6.2	25.0	18.9	3.5	22.4
10.00	17.1	12.2	29.3	17.2	6.9	24.1	17.3	4.1	21.4
12.00	15.7	12.3	28.0	15.8	7.2	23.0	15.9	4.5	20.4
14.00	14.3	11.8	26.1	14.3	7.2	21.5	14.5	4.6	19.1
16.00	12.8	11.0	23.8	12.8	6.9	19.7	12.9	4.6	17.5
18.00	11.3	10.0	21.3	11.2	6.4	17.6	11.4	4.3	15.7
20.00	9.8	9.1	18.9	9.8	5.8	15.6	9.9	4.0	13.9

current at low temperatures.[7] This leads to a considerable enhancement of the (total) Wiedemann-Franz ratio in the temperature regime where the umklapp contribution comprises a significant fraction of the thermal resistivity. At lower temperatures where umklapp processes are frozen out, the ratio is determined entirely by normal processes.

The experimental data of MacDonald, White, and Woods[2] are in reasonable agreement with these results both in absolute magnitude and temperature dependence. After subtracting the impurity-scattering contribution,[8] they obtained an approximate T^2 temperature dependence for the thermal resistivity in the temperature regime below about 15 °K. For their three relatively pure potassium samples [$\rho_0 = (13.5,\ 14.0,$ and $22.2) \times 10^{-9}\ \Omega$ cm] the magnitude of this T^2 term was determined to be between 16.5 and 17×10^{-4} cm/W °K. This is somewhat lower (by 20–30%) than what would be expected on the basis of these calculational results.

IV. HIGHER-ORDER VARIATIONAL CORRECTION TO THERMAL RESISTIVITY

The results thus far have been obtained using the first-order "variational" trial function given by Eq. (3) (which actually allows for no variation at all). Calculations by Sondheimer[9] using higher-order variational trial functions have shown that corrections to the first-order result are relatively large in the case of the thermal resistivity. At very low temperatures and in the absence of impurity scattering, Sondheimer found an 18.8% change in the thermal resistivity between results using a trial function linear and cubic in E. Sondheimer's calculations, which neglect umklapp processes, however, show that this percentage change will *decrease* when one is not in the limit of low temperatures or when substantial impurity scattering is present.

To estimate the magnitude of such corrections for the present calculation of umklapp effects, the variational results of Sec. III have been similarly carried to higher order using a second trial function cubic in E:

$$\phi(\vec{k}) \propto \vec{k} \cdot \vec{u}(E-\mu)\left[1 + a(T)\left(\frac{E-\mu}{k_B T}\right)^2\right], \qquad (5)$$

where $a(T)$ is a parameter which is chosen to minimize the resultant values of the thermal resistivity.[10] Although expression (5) does not have the exact form of the true ϕ at low temperatures (see Ref. 11), it does permit a much better approximation of ϕ in the important region near the Fermi surface [$(E-\mu) \approx 0$]. Only the scattering of electrons by phonons is considered here; in particular, the variational calculation is carried out in the absence of impurity scattering.[12]

The results using this second variational trial

TABLE II. Variational corrections to the thermal resistivity of potassium. W is the electronic component of the thermal resistivity obtained using the standard trial function given by Eq. (3); W' is the thermal resistivity which results when using a higher-order variational trial function given by Eq. (5). In both cases the thermal resistivity has been divided by the second power of the temperature, the units being 10^{-4} cm/W °K.

	Bardeen pseudopotential			Lower Lee-Falicov pseudopotential			Ashcroft pseudopotential		
T (°K)	W/T^2	W'/T^2	% difference	W/T^2	W'/T^2	% difference	W/T^2	W'/T^2	% difference
1.00	21.5	17.5	18.8	21.5	17.5	18.9	21.5	17.5	18.9
1.25	21.6	17.5	18.8	21.6	17.5	18.9	21.6	17.5	18.9
1.50	21.6	17.6	18.8	21.6	17.6	18.9	21.6	17.6	18.9
1.75	21.7	17.6	18.9	21.7	17.6	18.9	21.7	17.6	19.0
2.00	21.9	17.7	19.3	21.8	17.7	19.2	21.9	17.6	19.3
2.25	22.3	17.8	20.2	22.0	17.7	19.6	22.0	17.7	19.7
2.50	22.7	17.9	21.3	22.3	17.8	20.2	22.3	17.8	20.3
2.75	23.2	18.0	22.6	22.6	17.9	20.9	22.5	17.8	20.7
3.00	23.9	18.2	23.9	22.9	18.0	21.6	22.8	17.9	21.2
3.25	24.6	18.4	25.1	23.3	18.1	22.2	23.0	18.1	21.5
3.50	25.3	18.7	26.0	23.6	18.3	22.8	23.3	18.2	21.8
3.75	26.0	19.0	26.8	24.0	18.4	23.2	23.5	18.3	21.9
4.00	26.6	19.4	27.4	24.3	18.6	23.6	23.6	18.4	22.0
4.50	27.8	20.0	28.0	24.9	18.9	23.9	23.8	18.6	21.9
5.00	28.8	20.7	28.0	25.2	19.2	23.3	23.8	18.7	21.5
6.00	29.9	21.8	27.0	25.5	19.5	23.3	23.6	18.7	20.7
8.00	30.1	22.5	25.1	25.0	19.4	22.4	22.4	17.9	20.2
10.00	29.3	22.1	24.5	24.1	18.6	22.8	21.4	16.8	21.3
12.00	28.0	21.4	23.5	23.0	17.8	22.5	20.4	16.0	21.5
14.00	26.1	20.5	21.4	21.5	17.0	20.8	19.1	15.2	20.1
16.00	23.8	19.4	18.6	19.7	16.1	18.1	17.5	14.4	17.7
18.00	21.3	18.0	15.5	17.6	15.0	15.2	15.7	13.3	14.9
20.00	18.9	16.5	12.6	15.6	13.7	12.4	13.9	12.2	12.1

function are presented in Table II. The agreement in absolute magnitude with the experimental data of MacDonald *et al.*[2] (see end of Sec. III) is considerably improved.[13] As seen in Table II, corrections to the first-order result (in the absence of impurity scattering) are slightly less than 20% in the low-temperature regime where umklapp processes are frozen out. This change agrees well with Sondheimer's very-low-temperature results. However, as umklapp processes become important, the corrections *rise* to slightly less than 30% near 5 °K for the Bardeen pseudopotential (where umklapp effects are the greatest) before diminishing at still higher temperatures. Such a rise in the variational correction is not observed when normal processes alone are considered. Higher-order variational corrections are significantly greater for the umklapp component than for the normal component.

V. LATTICE CONDUCTIVITY

If we take the deviation of the phonon distribution from equilibrium, $\phi(\vec{q})$, to have the form

$$\phi(\vec{q}) \propto \vec{q}\cdot\vec{u}\,, \tag{6}$$

then the reciprocal of the lattice component of the thermal conductivity W_l is given by[14]

$$W_l = \frac{m}{Tne^2}\,\frac{\Omega_0 N(0)}{8k_F^4}\,\frac{\hbar}{Mk_BT}\left(\sum_\lambda \int_{q\le 2k_F} d^3q\,\frac{q^2\,|\vec{\epsilon}(\vec{q};\lambda)\cdot\vec{q}\,|^2\,|V(\vec{q})|^2}{q(1-e^{-z})(e^z-1)}\right)\Bigg/\left(\frac{\pi}{ek_F^3}\right)^2\left(\frac{k_B}{8\pi^3}\sum_\lambda \int d\vec{q}\,\frac{z^2e^z}{(e^z-1)^2}\right)^2. \tag{7}$$

Here it is assumed that the electron distribution is in equilibrium and only the interaction of phonons with electrons is considered (i.e., the interaction of phonons with impurities, boundaries, etc., is neglected). These assumptions are reasonable insofar as there is no net electric current and provided that $\Lambda_{\text{el-ph}} \ll \Lambda_{\text{imp}}$ (where $\Lambda_{\text{el-ph}}$ and Λ_{imp} are the phonon mean free paths determined by phonon scattering from electrons and impurities, respectively). The latter condition should be satisfied at low temperatures in relatively pure potassium samples.

The lattice contribution has been evaluated in this approximation in order to determine its importance

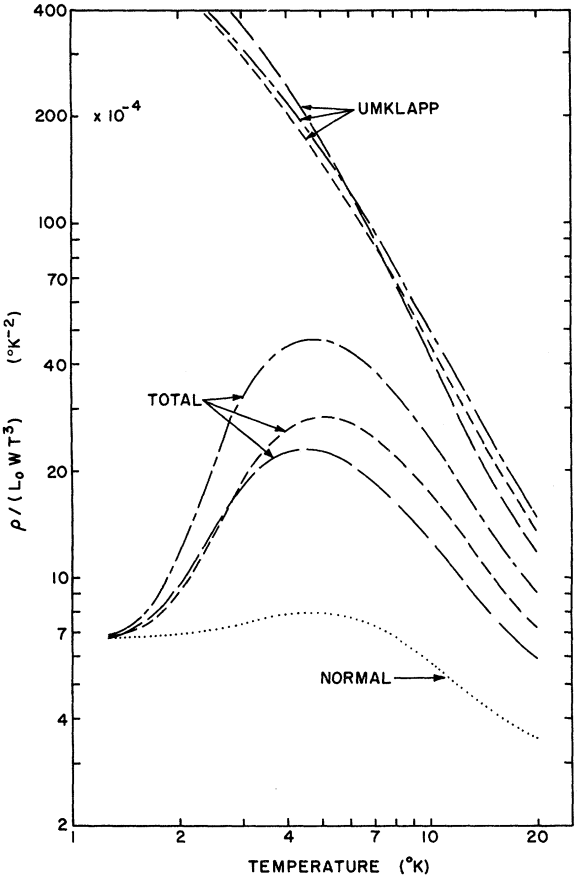

FIG. 2. Temperature dependence of the ratio $\rho/(L_0T^3W)$ as calculated individually for both the normal $[\rho_N/(L_0T^3W_N)]$ and umklapp $[\rho_U/(L_0T^3W_U)]$ resistivity components as well as the total $[\rho_T/(L_0T^3W_T)]$ (same curve codes as Fig. 1). The ratio of the normal resistivity components from the three pseudopotentials were so nearly identical at these temperatures they could not be resolved on this scale. The low-temperature rise in the ratio of the umklapp resistivity components is due principally to dividing ρ/WT by T^2 in plotting the results.

relative to the electronic heat conduction in the low-temperature regime. As before, the phonon frequencies and polarization vectors were generated from a five-nearest-neighbor Born-von Kármán fit to inelastic-neutron-scattering data.[4] The results for various pseudopotentials are presented in Fig. 3.

In the limit where $\Lambda_{el\text{-}ph} \ll \Lambda_{imp}$, the absolute magnitude of these results is in generally good agreement with the lattice-conductivity data of Archibald, Dunick, and Jericho obtained in several potassium-cesium alloys.[15] However, an accurate experimental determination of the temperature dependence in this limit has not yet been made. The experimental agreement in this case is as good as that of the theory of Pippard,[16] where a T^{-3} temperature dependence is predicted for W_i in the pure limit.

An analysis of lattice heat transport by individual phonon modes has also been performed. At temperatures below about 2 °K it is found that if only the fast (slow) shear phonon mode were present, the lattice conductivity would be about 250 (50) times greater than if only the longitudinal mode existed. Note that the heat conduction by the transverse modes is not infinite, even though a spherical Fermi surface was assumed in the calculation. This is a result of the phonon modes being neither purely longitudinal nor transverse in a realistic phonon model. At higher temperatures where umklapp processes become important, the difference in conductivity by the various phonon modes is considerably reduced.

In the low-temperature regime $\Lambda_{el\text{-}ph} \ll \Lambda_{imp}$, these results should serve to evaluate the relative importance of heat transport by the lattice in comparison to that by the electronic system. For example, the lattice conductivity will be less than 2% of the electronic conductivity at temperatures below ~ 6 °K in samples with $\rho_0 \lesssim 15 \times 10^{-9}$ Ω cm.[8] The calculational results for the electronic component alone should describe quite accurately the low-temperature thermal resistivity of potassium samples with purities in this regime.

VI. SUMMARY

A detailed evaluation of electron-phonon umklapp-scattering effects shows that umklapp processes have a negligible effect on the thermal resistivity and Wiedemann-Franz ratio of potassium at temperatures less than ~ 2 °K. Both are considerably enhanced by umklapp processes, however, at temperatures above this. Higher-order variational corrections are significantly larger for the umklapp component of the thermal resistivity than for the normal component.

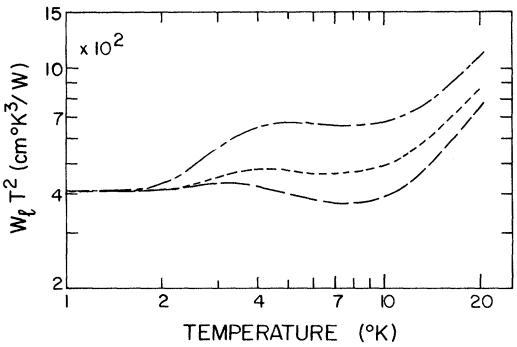

FIG. 3. Temperature dependence of the lattice component of the thermal resistivity W_l due to phonon-electron interaction (same curve codes as Fig. 1).

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[8]Here it has been assumed that electron-impurity scattering contributed an additive term $\rho_0(L_0T)^{-1}$ to the thermal resistivity (ρ_0 being the residual electrical resistivity).

[9]E. H. Sondheimer, Proc. Roy. Soc. (London) A203, 75 (1950).

[10]See J. W. Ekin, thesis (Cornell University, 1971) (unpublished), Appendix B for the modifications to Eq. (4) produced by this second trial function.

[11]In the special case of very low temperatures and in the absence of impurity and umklapp scattering, Klemens has numerically solved the Bloch integral equation and has found ϕ to be quite different from that given by Eq. (3), approaching a constant value asymptotically for large values of $(E-\mu)$. In this limit the thermal resistivity was found to be $\sim 33\%$ lower than the first-order variational result. The higher-order variational correction using the trial function cubic in E (see Table II) underestimates Klemens's exact correction in this low-temperature limit by only 14%. P. G. Klemens, Aust. J. Phys. 7, 64 (1954).

[12]This point should be emphasized, for the results using this second trial function are most appropriate to pure samples. In low-purity samples, on the other hand, the thermal resistivity is severely dominated by impurity scattering and the first-variational trial function given by Eq. (3) (obtained in the relaxation-time approximation) becomes a more accurate representation of the true ϕ. In this latter case, the results of Table I would better describe the electronic component of the thermal resistivity.

[13]A detailed test of these results, however, must await a more precise experimental study of the temperature dependence of the thermal resistivity in the low-temperature regime.

[14]See, for example, J. M. Ziman, in Ref. 3, Chap. 8, p. 319ff.

[15]M. A. Archibald, J. E. Dunick, and M. H. Jericho, Phys. Rev. 153, 786 (1967).

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