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TRANSPORT PROPERTIES OF ALKALI METALS

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RESUMEN

La dependencia en temperatura de las resistividades eléctricas y térmicas de los metales alcalinos litio, sodio, potasio y rubidio se determina a partir del elemento de matrix de Bardeen para la interacción electrón-fonón, utilizando el modelo de Sharma y Joshi para su dinámica reticular. Un procedimiento más realista permite separar las contribuciones a la resistividad debido a procesos normales y umklapp. Los valores calculados para las resistividades se comparan con la información experimental disponible; resulta que las curvas de resistividad teóricas concuerdan satisfactoriamente con el experimento para sodio y potasio, pero no para litio y rubidio. Aunque el aspecto de las curvas experimental y teórica resulta ser similar, el acuerdo es pobre para temperaturas bajas y altas.

ABSTRACT

The temperature dependence of the electrical and thermal resistivities of alkali metals lithium, sodium, potassium and rubidium is determined from Bardeen's electron-phonon matrix element and using Sharma and Joshi's model for their lattice dynamics. The separation of normal and Umklapp contributions to the resistivity is effected by adopting a more realistic procedure. The calculated values of the resistivities are compared with available experimental information. It is found that the theoretical resistivity curves show satisfactory agreement with experiments for sodium and potassium, but not for lithium and rubidium. Though the nature of the theoretical and experimental thermal resistivity curves is found to be similar, the agreement is poor at low and high temperatures.

I. INTRODUCTION

One of the basic problems in solid-state physics has been to give an adequate description of the transport properties of monovalent metals. It has been a subject of investigation by many workers for the past four decades when Bloch[1] first propounded a theory to explain qualitatively the resistivity of metals at very low and at high temperatures. The transport phenomena in metals depend in a complicated way both upon the electronic band structure and the anisotropy in the lattice spectrum and this situation complicates the evaluation of the scattering probability of the conduction electrons. Since alkali metals have the most simple electronic structure, most of the theoretical work has been done on these metals. In a famous paper work Bardeen[2] first calculated the electrical conductivity of monovalent metals, assuming the electronic behaviour to be free-electron like and using simple Debye model for the phonon spectrum. These approximations were forced to be adopted, because detailed information concerning them was not available at that time. Still Bardeen's results showed good quantitative agreement in the high-temperature region.

During the last few years, there has been a good deal of awakening of interest to improve upon these approximations. Bailyn[3] first introduced the use of the Born-von Kármán model for the phonon spectrum in the study of the transport coefficients of alkali metals. Recently Darby and March[4] have directly used the experimentally determined phonon-dispersion curves of sodium to calculate its electrical resistance. They have also taken into account the exchange and correlation effects and in an approximate way the temperature dependence of the elastic constants. Their results show good agreement with the experimental data throughout the temperature range studied. Collins and Ziman[5] have used twelve-cone approximation for the Fermi surface to calculate the transport coefficients of alkali metals. They have, however, adopted a simple Debye model for the phonon spectrum. On the other hand, Hasegawa[6] and Bross and Holz[7] have computed the electrical reistivity of alkali metals by taking into account both the phonon spectrum and electronic band structure. Recently Greene and Kohn[8] have theoretically studied the electrical resistivity of solid and liquid sodium utilizing the observed inelastic neutron scattering data for the dynamics of ions. Although the calculation incorporates accurately the many-body effects, Umklapp processes, the time-dependent effects etc., yet the agreement with experiment is not satisfactory.

One of the authors (P.K.S.) and Joshi[9,10] have propounded an elastic force model for the lattice dynamics of monovalent metals taking into account explicitly the presence of electron gas. It was assumed that the forces on an ion arise from a central interaction limited to first and second nearest neighbours and from certain energies due to the compressibility of electron gas and its interaction with ions. The model has been shown to give a plausible explanation of thermal properties of alkali and noble metals[11].

In the present paper, we report the calculation of the temperature dependence of the electrical and thermal resistivities of alkali metals lithium, sodium, potassium and rubidum within the free-electron approximation using our model for the phonon spectrum. The choice of these metals for this study was dictated by the fact that they very nearly correspond to free-electron metals. This considerably simplifies the electronic band-structure aspect of the problem and allows us to use more effectively the phonon spectrum.

II. THEORY

The general method for evaluating expressions for the transport coefficients is to use the variational principle. The method has been developed by Kohler[12] and Sondheimer[13] and is clearly described by Ziman[14]. The Boltzmann equation determining the distribution function is a linear inhomogeneous integral equation with a positive definite self-adjoint kernel. It is known that such an equation can be deduced from the variation of a certain integral and the solution can be constructed formally by applying the variational principle to a general trial function. The variational principle states that the solution of the Boltzmann integral equation gives to

$$\frac{<\phi, P\phi>}{[<\phi, X>]^2} \tag{1}$$

a minimum value, where ϕ is defined as a trial function, P is the scattering operator and $X = P\phi$. The variational function (1) is calculated and the parameters are varied until it is an extremum which is the nearest approximation to the true solutions.

(a) Electrical Resistivity

The variational expression for the electrical resistivity is[14]

$$\rho_L = \frac{(1/k_B T) \iiint [\phi_k - \phi_{k'}]\, P(kq, k')\, dk\, dk'\, dq}{\left| \int e v_k \phi_k \frac{\partial f_k^0}{\partial E_k} dk \right|^2} \tag{2}$$

Here k and k' are the wave vectors of the initial and final electron states, v_k is the velocity and E_k the energy of that state, $P(kq, k')$ is the scattering probability that an electron in state k is scattered to a state k' with the absorption of a phonon of wave vector q, and ϕ_k is a trial function defining a perturbed distribution of the form

$$f_k = f_k^0 - \phi_k (\partial f_k^0 / \partial E_k) \, . \tag{3}$$

The probability of occupancy at equilibrium is given by the Fermi function

$$f_k^0 = \{ \exp [(E_k - E_F)/k_B T] + 1 \}^{-1} \, . \tag{4}$$

In the case of a lattice of cubic symmetry use of $\phi_k = k \cdot u$, where u is a unit vector in the direction of the applied electric field, as a first order approximation to trial function leads to the ideal resistivity ρ_L as the four-dimensional integral

$$\rho_L = \frac{3\pi\hbar}{2e^2 M N k_B T \, k_F^2 S^2} \sum_p \iint \frac{K^2 (\mathbf{K} \cdot \mathbf{e}_{\mathbf{q},p})^2 C^2(K)}{(1 - e^{-\beta\omega_{\mathbf{q},p}})(e^{\beta\omega_{\mathbf{q},p}} - 1) v v'} \, ds \, ds' \, , \tag{4}$$

where $\omega_{\mathbf{q},p}$ is the angular frequency of phonon of wave vector $\mathbf{q}$ and polarization vector $\mathbf{e}_{\mathbf{q},p}$, the suffix p distinguishes the three modes of lattice vibrations, v denotes the velocity of an electron on the Fermi surface S_F, k_F is the Fermi radius, $\mathbf{K} = k' - k$ is the scattering vector, N is the number of ions per unit volume, M is the mass of an ion, $C(K)$ is the matrix element corresponding to the transition from k to k' and β is $\hbar/k_B T$, k_B being the Boltzmann constant. The two surface integrals are over the Fermi surface. For a spherical Fermi surface, the matrix element $C(K)$ is given by the relation[2]

$$C(K) = \frac{W(K) \, q_s^2(K) + \{ V(r_s) - E_0 \} K^2}{K^2 + q_s^2(K)} \, g(K r_s) \, , \tag{5}$$

in which

$$W(K) = \frac{2E_F}{3}\left[\frac{1}{2} + \frac{2k_F^2 - K^2}{8Kk_F}\, ln \left| \frac{2k_F + K}{2k_F - K}\right|\right]^{-1} \tag{6a}$$

$$g(x) = 3(\sin x - x \cos x)/x^3 , \tag{6b}$$

$$q_s^2(K) = \frac{4\pi n e^2}{W(K)} , \tag{6c}$$

where $[E_0 - V(r_s)]$ is the kinetic energy of an electron in the lowest state at the boundary of the atomic polyhedra of radius r_s, E_F is the Fermi energy and n is the electron number density.

(b) Thermal Resistivity

The variational calculation of the thermal resistivity gives[14]

$$W_L = \left(\frac{1}{k_B}\right) \frac{\iiint [\phi_k - \phi_{k'}]^2 \, P(kq, k')\, dk\, dk'\, dq}{\left| \int (E_k - E_F)\, v_k\, \phi_k \, \frac{\partial f_k^0}{\partial E_k}\, dk \right|^2} \tag{7}$$

Here the trial function for the elastic scattering can be taken as

$$\phi_k = (E_k - E_F)\, k \cdot u ,$$

where u is now a unit vector parallel to the heat current. This finally leads to the expression

$$W_L = \frac{9\pi\hbar}{2e^2 L_0 MN k_B T^2 k_F^2 S_F^2}$$

$$\times \sum_p \iint \frac{(\mathbf{K}\cdot \mathbf{e}_{\mathbf{q},p})^2 C^2(K) \left\{\frac{K^2}{3} - \frac{K^2}{6\pi^2}(\beta\omega_{\mathbf{q},p})^2 + \frac{k_F^2}{\pi^2}(\beta\omega_{\mathbf{q},p})^2\right\}}{(1 - e^{\beta\omega_{\mathbf{q},p}})(e^{\beta\omega_{\mathbf{q},p}} - 1)}\, ds\,ds' \tag{8}$$

where L_0 is the Lorentz number and the other symbols have their usual significance.

III. EVALUATION OF THE DOUBLE AVERAGE

Even for a spherical geometry, the evaluation of the double average in Eqs. (4) and (8) is quite difficult. We use Bailyn's averaging procedure[3] to compute the double average and write expressions (4) and (8) in the form

$$<<F(\mathbf{K})>>_{\overline{\mathrm{F.S.}}} = \frac{1}{S^2}\iint F(\mathbf{K})\, ds\,ds' \;. \tag{9}$$

The phonon frequencies $\omega_{\mathbf{q},p}$ and the polarization factor $(\mathbf{K}\cdot \mathbf{e}_{\mathbf{q},p})^2$ vary with the direction of the scattering vector $\mathbf{K}$, and this situation makes these integrals very complicated. To effect the average (9), we first choose vectors k and k' that have the same difference vector $\mathbf{K} = k' - k$ and average over all such vectors. In the case of a spherical Fermi surface all these vectors can be obtained by visualizing a rigid rod of length K oriented in the direction of $\mathbf{K}$ to roll on the inside of a spherical shell of radius k_F. The ends of the rigid roll will each describe a circle of circumference $s(\mathbf{K}) = \pi(4k_F^2 - K^2)^{\frac{1}{2}}$. The advantage in performing a preliminary average over all such combinations with a given $\mathbf{K}$ is that the phonon parameters will not change during it. We denote this average of the integrand as $\{\{F(\mathbf{K})\}\}$. Next one averages over all $\mathbf{K}$ for a given magnitude of $\mathbf{K}$.

This average equals the integral over all directions of $\mathbf{K}$ with with weighting factor equal to the ratio of the length $s(\mathbf{K})$ to the integral of $s(\mathbf{K})$ over the whole solid angle for a given K magnitude. This average can be expressed by the relation

$$[\{\{F(\mathbf{K})\}\}] = \int_{(K)} d\Omega\, \{\{F(\mathbf{K})\}\}\, \frac{s(\mathbf{K})}{\int d\Omega\, s(\mathbf{K})} \tag{10}$$

Here $\int_{(K)} d\Omega$ indicates the integration over the solid angle of $\mathbf{K}$ on a constant K-magnitude surface. Finally we average over all K magnitudes. This is equal to the integral over all K with weighting factor

$$\frac{\int d\Omega\, s(\mathbf{K})}{\int dK \int d\Omega\, s(\mathbf{K})} \,.$$

This finally gives the required average over the Fermi surface as

$$<<F(\mathbf{K})>>_{\text{F.S.}} = \frac{\int dK \int d\Omega\, [\{\{F(\mathbf{K})\}\}]\, s(\mathbf{K})}{\int dK \int d\Omega\, s(\mathbf{K})} \tag{11}$$

For a spherical Fermi surface, the denominator of (11) equals $4\pi^3 k_F^2$. Hence

$$<<F(\mathbf{K})>>_{\text{F.S.}} = \frac{1}{2\pi^2 k_F} \int dK \int d\Omega\, [\{\{F(K)\}\}]\,(1 - u^2)^{\frac{1}{2}} \,, \tag{12}$$

where $u = K/2k_F$. Using this result, the expressions (4) and (8) for the electrical and thermal resistivities can be written as

$$\rho_L = \frac{3\hbar}{4\pi e^2 M N k_B T v_F^2 k_F^3} \int d\Omega \int A(K)\, dK\,, \qquad (13a)$$

and

$$W_L = \frac{9\hbar}{4\pi e^2 L_0 M N k_B T^2 v_F^2 k_F^2} \int d\Omega \int B(K)\, dK\,, \qquad (13b)$$

where

$$A(K) = \sum_p \frac{K^2 (1-u^2)^{\frac{1}{2}} (\mathbf{K}\cdot\mathbf{e}_{\mathbf{q},p})^2 C^2(K)}{(1-e^{-\beta\omega_{\mathbf{q},p}})(e^{\beta\omega_{\mathbf{q},p}}-1)} \qquad (14a)$$

and

$$B(K) = \sum_p \left\{ \frac{(1-u^2)^{\frac{1}{2}} (\mathbf{K}\cdot\mathbf{e}_{\mathbf{q},p})^2}{(1-e^{-\beta\omega_{\mathbf{q},p}})(e^{\beta\omega_{\mathbf{q},p}}-1)} \times \left[\frac{K^2}{3} - \frac{K^2}{6\pi^2}(\beta\omega_{\mathbf{q},p})^2 + \frac{k_F^2}{\pi^2}(\beta\omega_{\mathbf{q},p})^2 \right] C^2(K) \right\} \qquad (14b)$$

IV. NUMERICAL COMPUTATIONS

We have calculated the electrical and thermal resistivities from Eqs. (13) by using a modification of Houston's method. The integration over K was performed numerically and the integration over Ω was carried out using the modified Houston's spherical six-term integration procedure as elaborated by Betts et al.[15] Houston's method gives proper weight to each reciprocal lattice point and is inherently preferable to the sampling technique which relies merely on taking a very large number of points in the reciprocal space. For the evaluation of the integral

$$J = \int I(\theta, \phi)\, d\Omega\,, \tag{15}$$

where the integrand is invariant under the operations of the complete symmetry group, the value of $I(\theta, \phi)$ in the [100], [110], [111], [210], [211] and [221] directions is required. The expansion of the integrand in cubic harmonics retaining only six terms leads to the following expression for J:

$$J = \frac{4\pi}{1081080}\,[117603\, I_A + 76544\, I_B + 174961_C$$

$$+ 381250\, I_D + 311040\, I_E + 177147\, I_F]\,, \tag{16}$$

where the subscripts A, B, C, D, E and F denote the values of the integrand $I(\theta, \phi)$ along the directions [100], [110], [111], [210], [211] and [221], respectively. The direction cosines of the polarization vector $\mathbf{e}_{q,p}$ are obtained from the solutions of the secular equation for a body-centred cubic lattice.

In calculating the integrals (13) we have distinguished normal (N) processes from the Umklapp (U) processes in the conservation law of the wave vector. Early workers have completely ignored the Umklapp process, but in recent years its importance has been realised. Bailyn[3], Hasegawa[6] and others assume that the normal processes operate in the range of the variable of integration u from 0 to 0.63 in the Brillouin zone, while the range 0.63 to 1 corresponds to the Umklapp processes. This type of separation of the range of the normal and the Umklapp processes is rather artificial when one goes beyond the spherical approximation. In the present work a more realistic approach has been adopted.

(a) Normal processes

The normal processes are governed by the conservation law

$$\mathbf{K} = \mathbf{k}' - \mathbf{k} = \mathbf{q}\,, \tag{17}$$

where q is restricted to be in the first Brillouin zone. Using Ziman's formulation, we have taken account of the elastic anisotropy by integrating over K numerically, restricting the phonon wave vector to points within the first Brillouin zone. The limiting values of K vectors were obtained by finding the points of intersection of a line having the direction of K with the planes of the first Brillouin zone boundary. These limiting values for a body-centred cubic structure are given in Table I.

(b) Umklapp processes

n an Umklapp process the scattering vector is given by the selection rule

$$K = k' - k = q + G , \tag{18}$$

where G is a reciprocal lattice vector. In this case vector K goes beyond the boundary of the Brillouin zone, but q is constrained to be within the Brillouin zone. From the knowledge of the reciprocal lattice vectors of a body-centred cubic structure, the values of K along any direction at which the Umklapp processes crop up can be determined. The phonon wave vectors which are needed for the average are then obtained from the conservation law (18).

V. RESULTS AND DISCUSSION

The values of the elastic constants and the other data used in the present calculation of the electrical and thermal resistivities of the alkali metals are listed in Table II. The electrical and thermal resistivities of lithium, sodium, potassium and rubidium are calculated as a function of temperature. For the phonon spectrum we have used our electron-gas model. In the case of sodium and potassium, the computations have been performed with two sets of elastic constants. It was found that the Umklapp processes contribute appreciably to the total resistivity, being most important at higher temperatures. In some cases the contribution from U-processes exceeded that from N-processes over a wide range of temperature.

The calculated values of the electrical resistivities of the four metals along with the experimental values are plotted in Figs. 1-4. The experimental values of the electrical resistivities for lithium are taken from Dugdale and Gugan[16] and MacDonald et al.[17] and for sodium and potassium from Dugdale and Gugan[16], while those for rubidium are due to Dugdale and Phillips[18]. It is seen from Figs. 2 and 3 that the calculations using elastic-constant data at 90°K for sodium and 83°K for potassium are in better agreement with the experimental values at low temperatures, while those from the elastic constants at room temperature give a better experimental fit at higher temperatures.

For lithium and rubidium the calculated values of the electrical resistivity are lower than the experimental ones throughout the temperature range studied and the discrepancy increases with the rise of temperature. The disagreement is more pronounced in the case of lithium. For example at 100°K, the experimental value of the electrical resistivity is larger than the theoretical one by a factor of 3.77. This large variation in the values can be attributed to the distorted Fermi surface in lithium.

The calculated values of the thermal resistivities are compared with the experimental observations in Figs. 5-8. The experimental data plotted in these figures are due to MacDonald et al.[17] for lithium, potassium and rubidium, while those for sodium are from Berman and MacDonald[19]. It is seen that the theoretical and experimental resistivity curves are somewhat inconsistent. Though the two curves are of similar nature, the agreement is poor at low and high temperatures. At low temperatures the calculated resistivity is substantially lower than the experimental values, while at higher temperatures the theoretical curve lies below the experimental measurements. However, the experimental data at higher temperatures are not reliable.

The disagreement between the theory and experiment can be attributed to the following reasons: (1) We have used here Bardeen's expression for the electron-phonon matrix element which overlooks the exchange and correlation effects. (2) The expressions (4) and (8) are based on a first-order variational solution of the Boltzmann equation. This somewhat over-estimates the resistance[13]. (3) In the calculation we have not incorporated the temperature dependence of the elastic constants. It is known that the elastic constants vary with temperature.

(4) Lithium[20] and sodium[21] undergo a martensitic type of phase transformation into a mixture of body-centred cubic and hexagonal close-packed structures at low temperatures below about 78°K and 37°K, respectively. Because of the occurrence of this transformation, the significance of the theoretical values below these temperatures is somewhat doubtful. Thus a rigorous comparison with the experimental values below these temperatures is not feasible.

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TABLE I

Limiting values of K for normal processes in body-centred cubic structure.

Direction	Limiting value
[100]	$\frac{\pi}{a}$
[110]	$\frac{1}{\sqrt{2}} \frac{\pi}{a}$
[111]	$\frac{\sqrt{3}}{2} \frac{\pi}{a}$
[210]	$\frac{\sqrt{5}}{3} \frac{\pi}{a}$
[211]	$\frac{\sqrt{6}}{3} \frac{\pi}{a}$
[221]	$\frac{3}{4} \frac{\pi}{a}$

TABLE II

Constants for alkali metals

Metal	Notes	Elastic constants (10^{11} dyn/cm^2) C_{11}	C_{12}	C_{44}	Temperature* (°K)	Density (g/cm^3)	Lattice parameter (Å)	$[V(r_s) - E_0]$ g) (eV)	E_F (eV)
Lithium	a)	1.481	1.248	1.077	78	0.534	3.491	-0.8	4.72
Sodium	b)	0.741	0.624	0.419	300	0.9660	4.291	0.08	3.16
	c)	0.808	0.664	0.586	90	1.0014	4.240	0.08	3.16
Potassium	d)	0.3715	0.3153	0.188	295	0.851	5.344	-0.02	2.06
	e)	0.457	0.374	0.263	83	0.870	5.344	-0.02	2.06
Rubidium	f)	0.296	0.244	0.160	80	1.532	5.699	0.03	1.86

* Temperature at which the elastic constants are measured.

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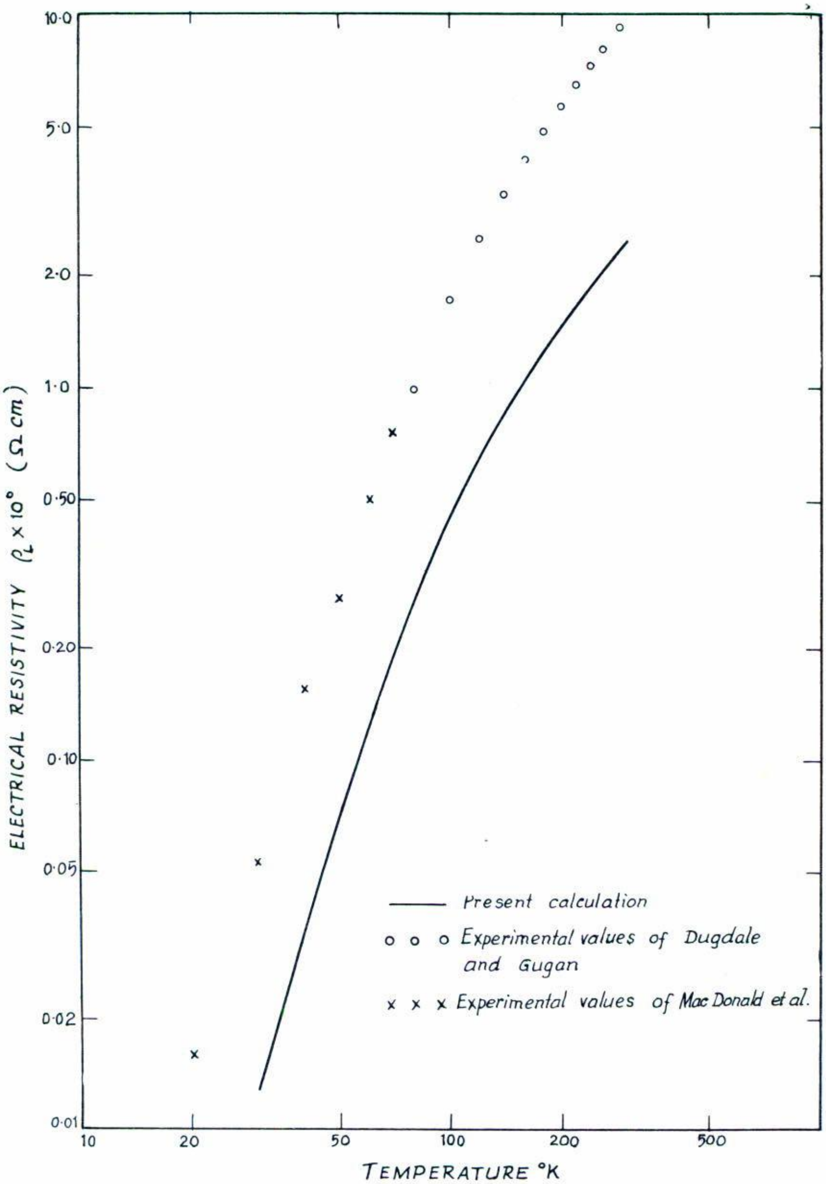

Fig. 1. The electrical resistivity of lithium as a function of temperature.

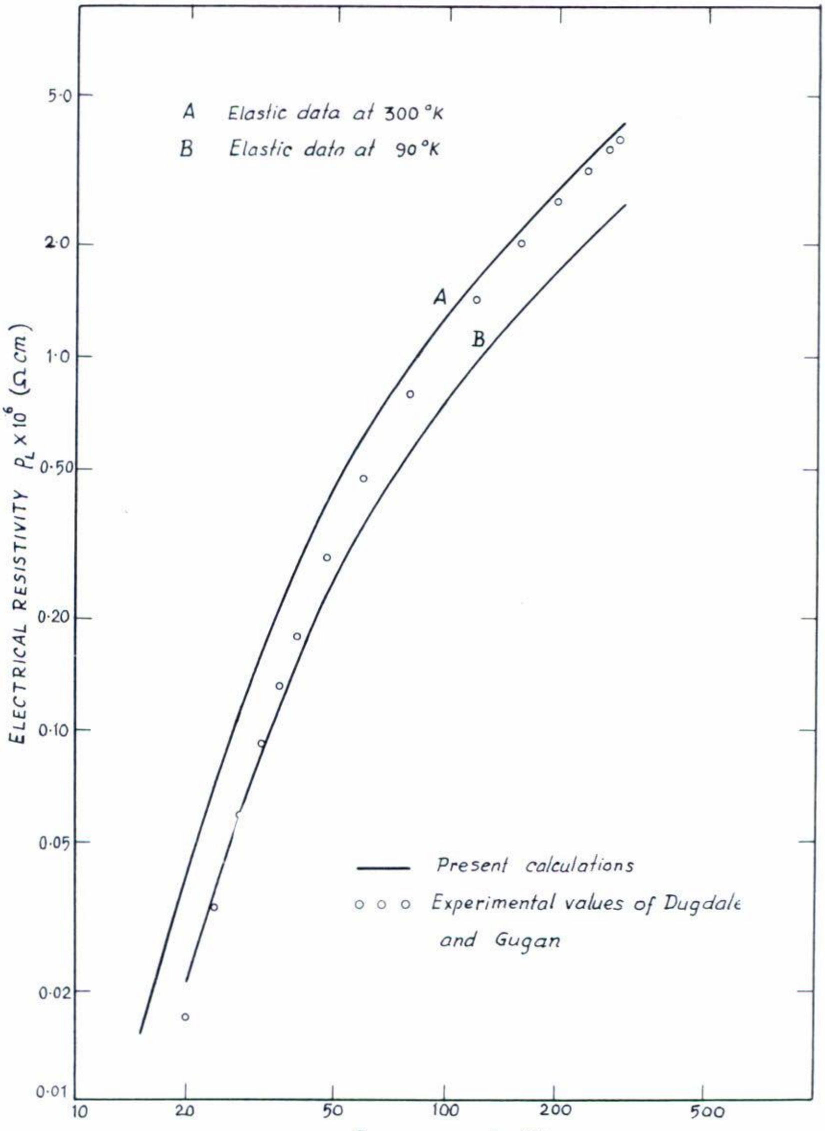

Fig. 2. The electrical resistivity of sodium as a function of temperature.

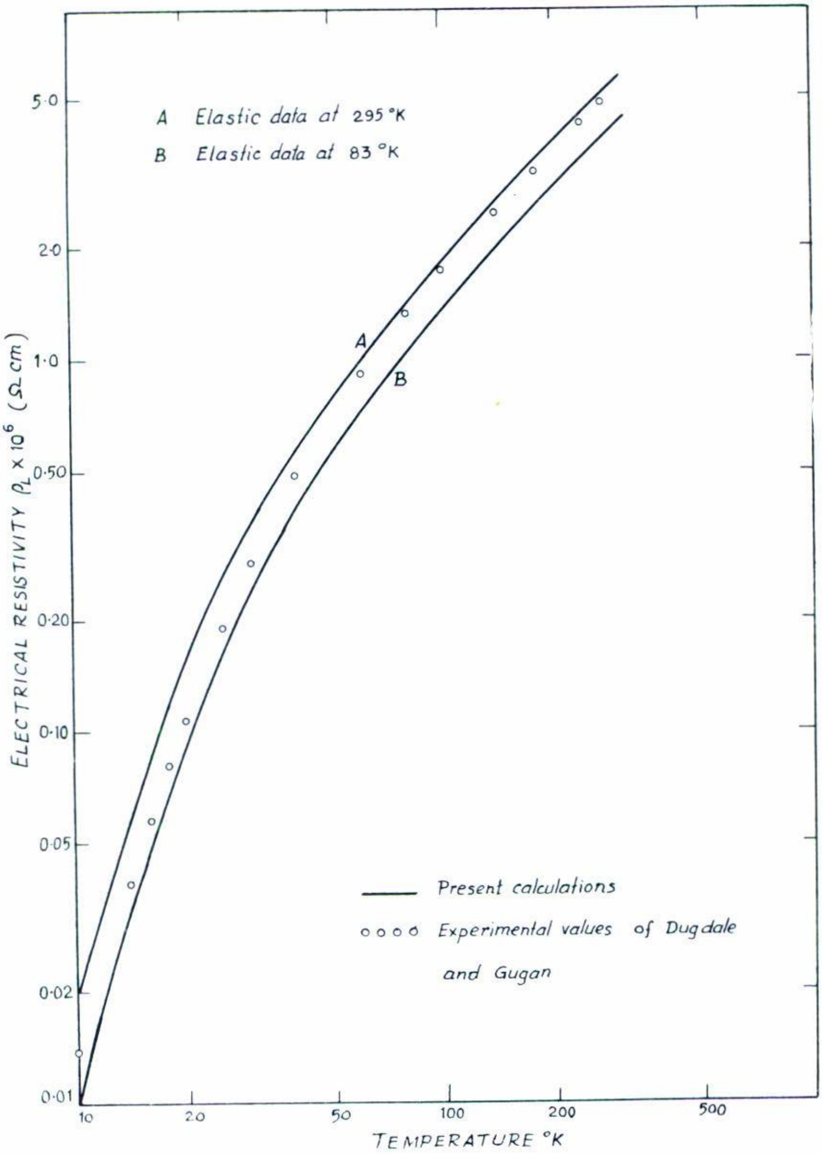

Fig. 3. The electrical resistivity of potassium as a function of temperature.

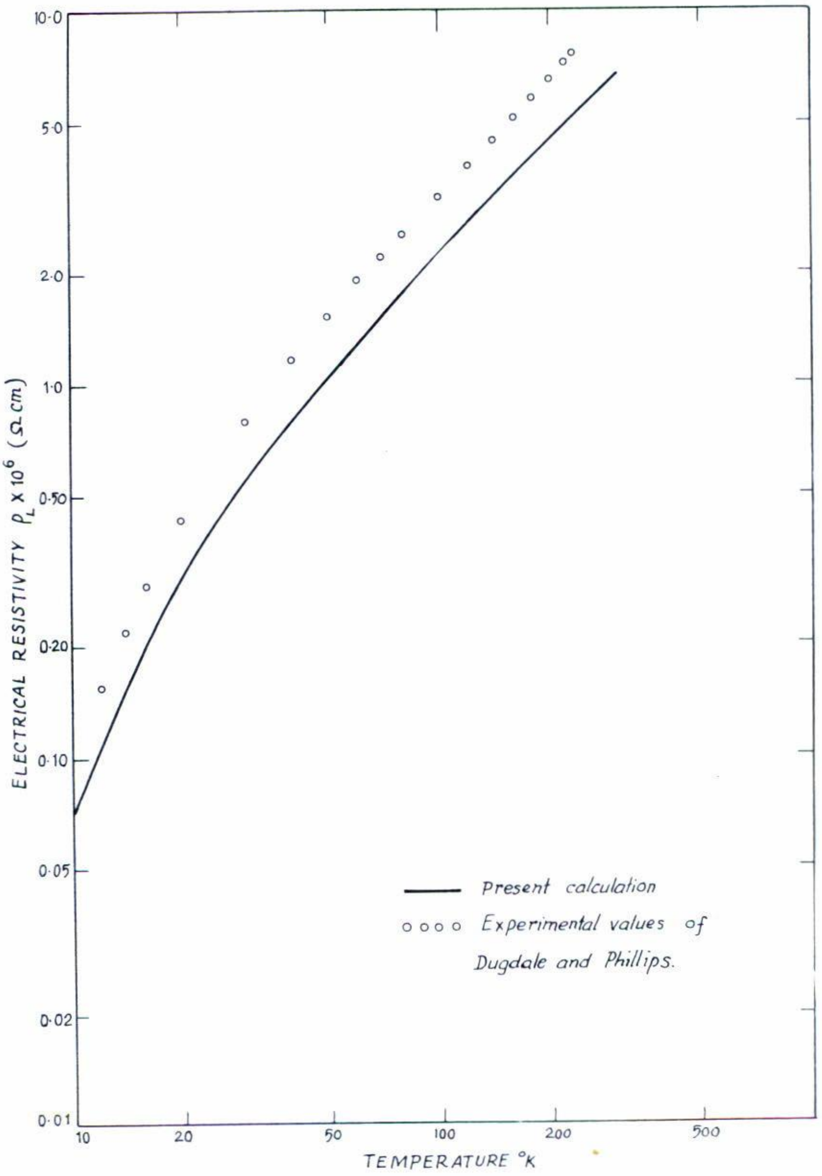

Fig. 4. The electrical resistivity of rubidium as a function of temperature.

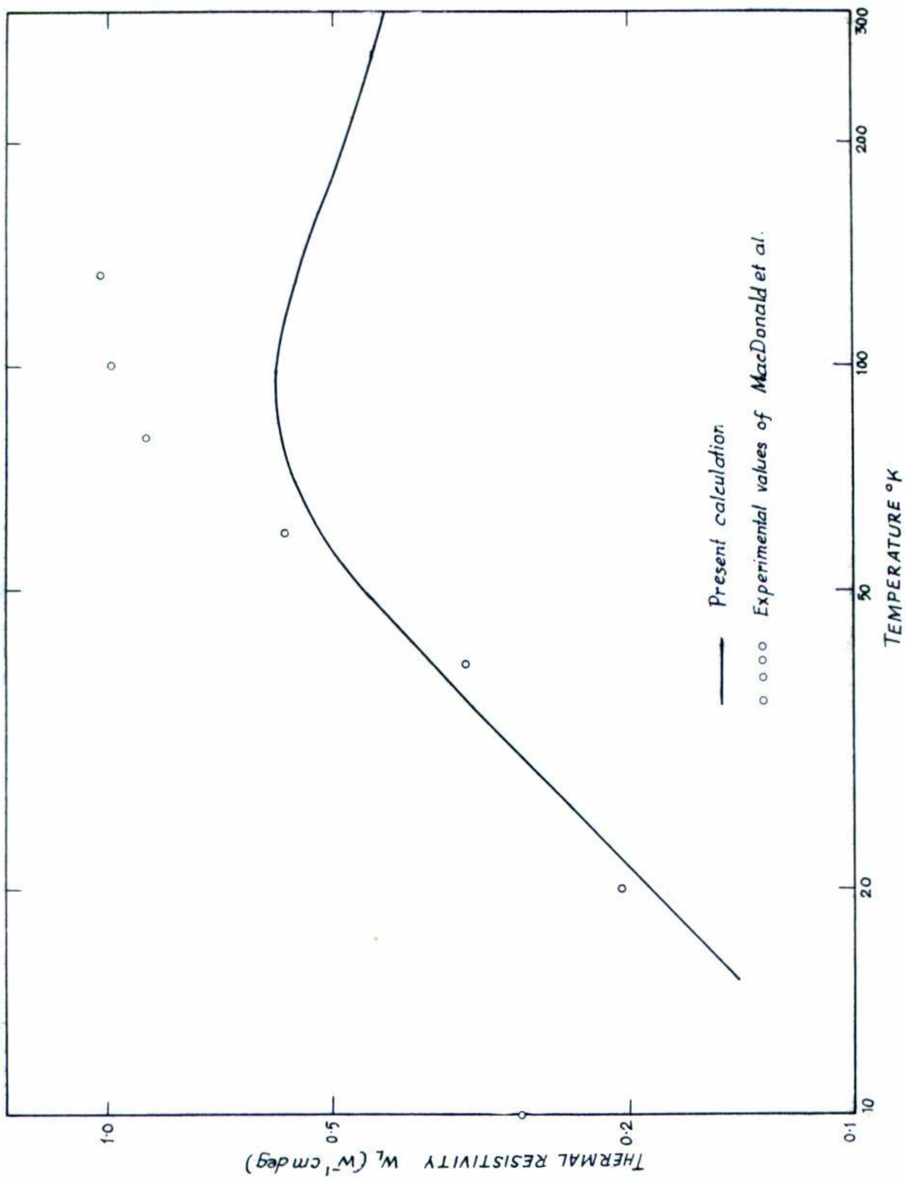

Fig. 5. The thermal resistivity of lithium as a function of temperature.

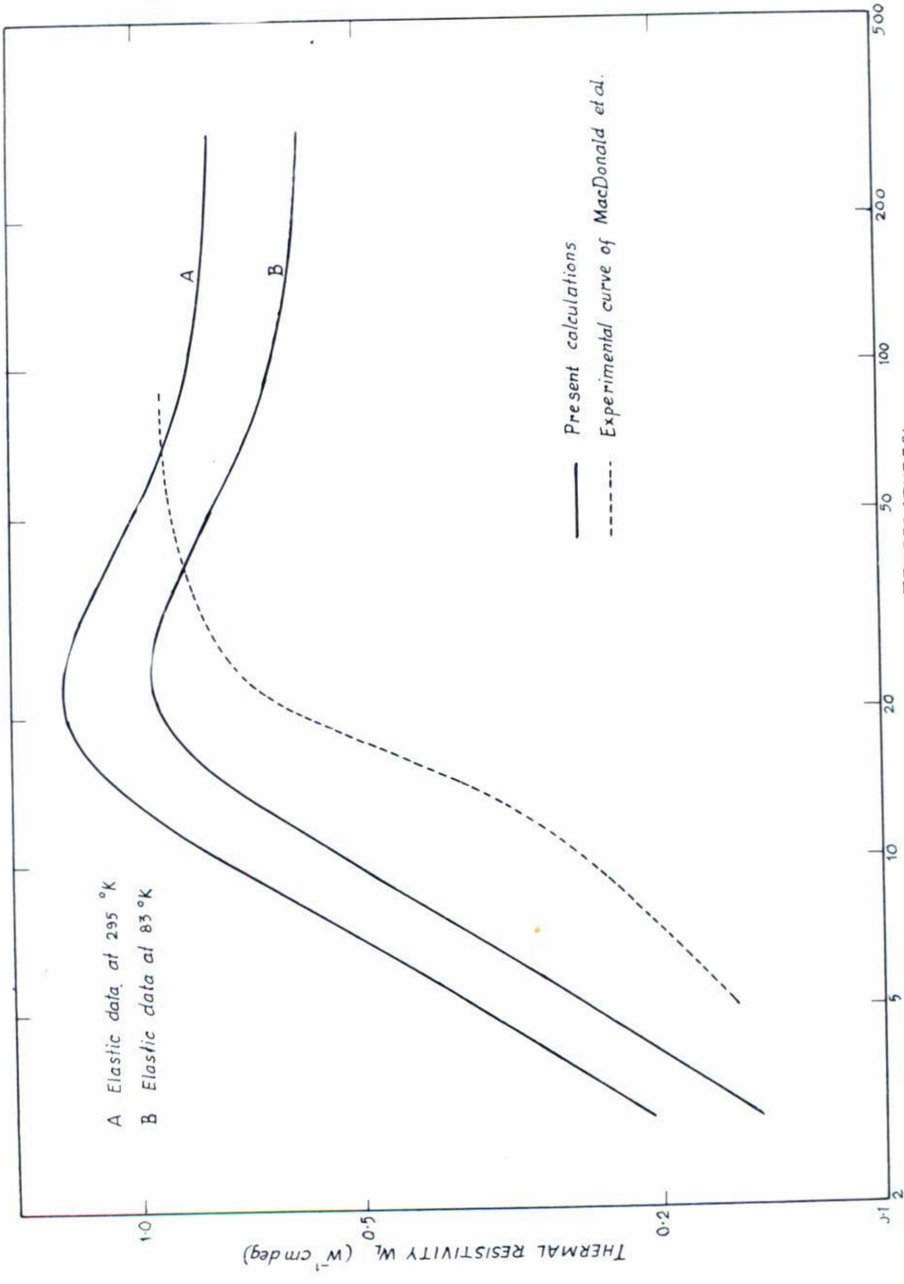

Fig. 6. The thermal resistivity of sodium as a function of temperature.

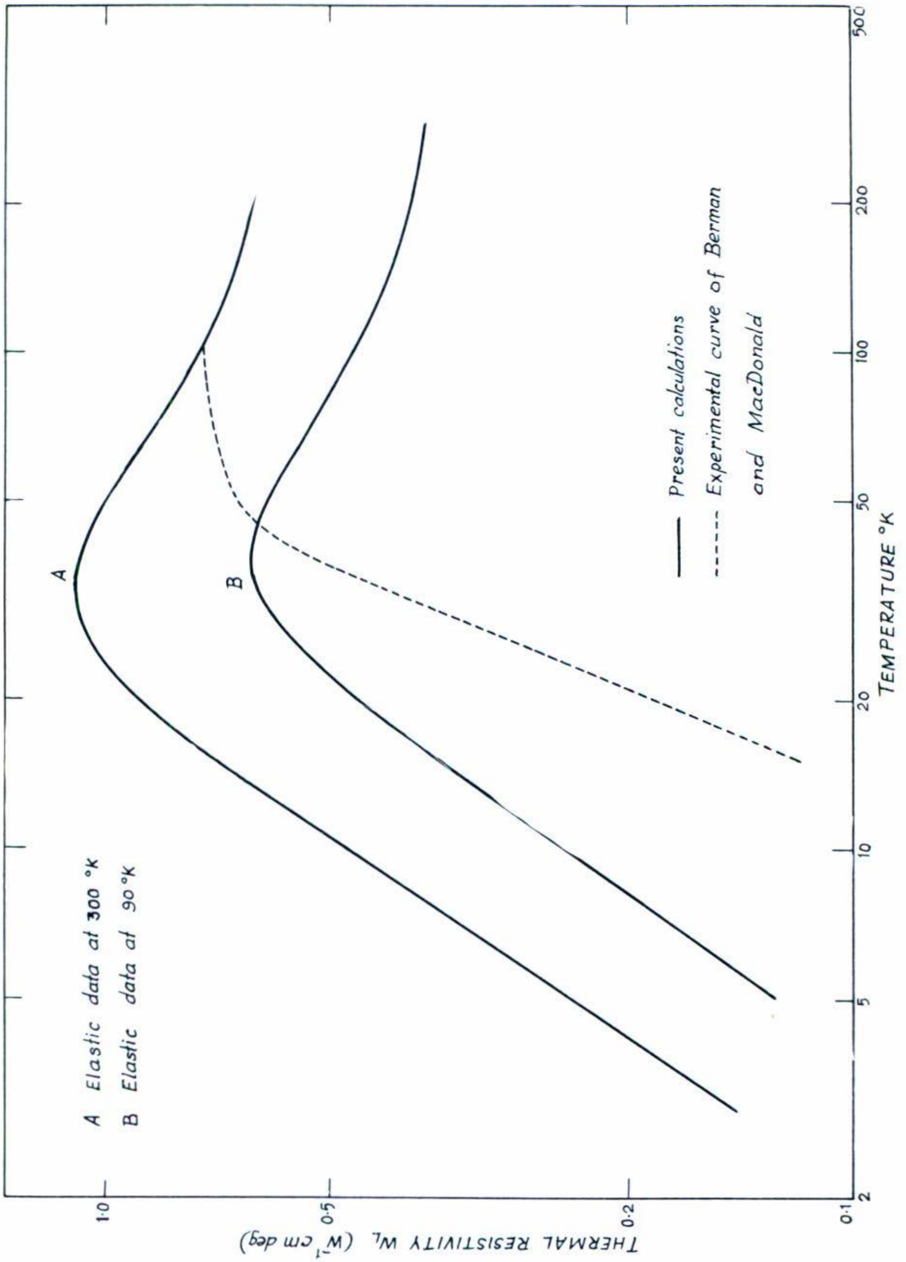

Fig. 7. The thermal resistivity of potassium as a function of temperature.

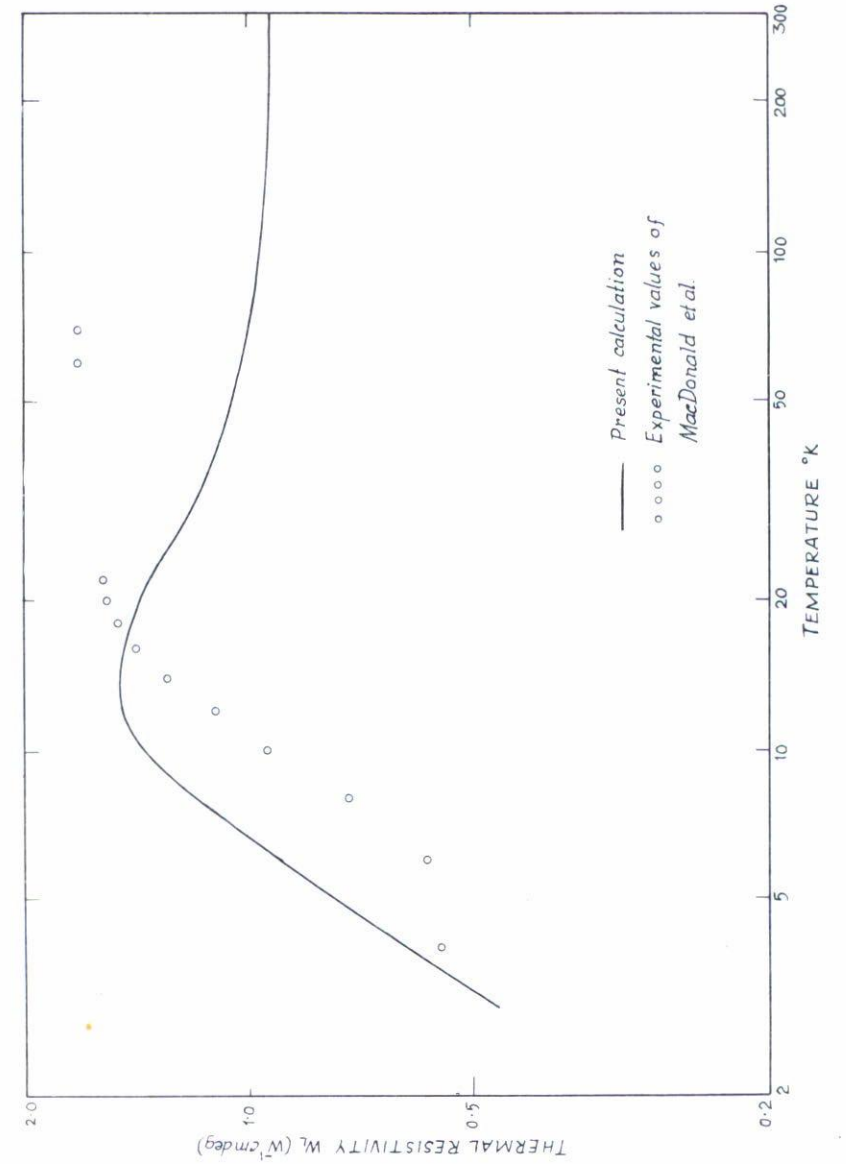

Fig. 8. The thermal resistivity of rubidium as a function of temperature.