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High-Temperature Thermal Resistivity of Monovalent Metals and Electron-Phonon Interaction

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The electronic thermal resistivity of metals can be empirically split into two parts, one of which, the so-called "elastic" part, is directly related to the electrical resistivity through the Wiedemann–Franz–Lorenz law, and thus is of no particular interest. The remainder is the "inelastic" part which is the only part that needs investigation in conjunction with the electrical resistivity in any study of the transport properties. Such a split of the thermal resistivity is very useful at high temperatures where the inelastic part becomes independent of the lattice dynamics and, for spherical Fermi surfaces, the contributions to this resistivity of both normal and umklapp processes can be evaluated explicitly in a simple fashion. The comparison of such calculations with experimental results, which we undertake for the alkali and noble metals, forms a simple and convenient test of the electron-phonon interaction.

I. INTRODUCTION

Recent calculations of the electrical resistivity of metals,[1–3] ρ, show very satisfactory agreement with experimental values. The same cannot be said about the calculations of the thermal resistivity W which display serious deviations throughout the whole temperature range investigated.[2] No doubt part of the deviation, at low temperatures, is due to the greater complexity in the calculations of W, where, besides the usual difficulties connected with electron and lattice dynamics and electron-phonon interaction, one is faced with the further problem that the variational solution of the Boltzmann equation obtained with the conventional trial function does not approach the correct solution of that equation.[4] This is not the case at high temperatures, however, and in that region it is difficult to guess why the calculated W is in error by a much larger factor than the calculated ρ. That is the problem which we will investigate here in detail.

Unlike all previous calculations, we shall not deal with the total thermal resistivity, but only with the "inelastic" or "vertical" part W_v, that is, with that part which is not directly computable from ρ through the Wiedemann-Franz relationship with the standard Lorenz number. Obviously, that is the only part which can contain any new features not already covered in ρ. Such a separation is of no value at low temperatures, where W_v equals W. At high temperatures, W_v is only a small part of W, vanishing in the limit. In this high-temperature limit the separation proves very useful, for, as we shall show, W_v here becomes independent of the lattice dynamics. As a consequence, one can analytically reduce the calculation to a relatively simple form: For spherical Fermi surfaces, W_v can be essentially expressed as a single integral over the pseudopotential used for the electron-phonon interaction, both for normal and umklapp scattering. Since in this temperature range the variational solutions with the standard trial functions are valid,[5] one can readily calculate W_v with an accuracy limited only by the applicability of the pseudopotential; as such, our calculations form a simple and convenient test of the latter.

In the following, we will first develop very briefly the theory of the high-temperature limit of W_v. We will then compare our calculations to the experimental values for the alkali metals and, on an exploratory basis, to the noble metals; as it turns out, the problem is not one of a basic discrepancy between theory and experiment, but simply one of choice of a suitable potential to reproduce the experimental results. We will conclude the paper with a simple-minded look at the low-temperature limit of W_v, where a significant discrepancy between theory and experiment appears to exist; this area should certainly be subjected to detailed and precise calculations.

II. THEORY OF HIGH-TEMPERATURE LIMIT OF W_v

In this section we consider only the electronic part of the thermal resistivity, and limit ourselves to pure electron-phonon scattering. As mentioned above, we define the inelastic part of W, W_v, as that part which has no correspondence with ρ;

$$W_v = W - \rho/L_0 T, \tag{1}$$

where T is the absolute temperature and L_0 the standard Lorenz number $\frac{1}{3}(\pi k_B/e)^2$, k_B being the Boltzmann constant and e the electronic charge. Using the conventional trial functions for the variational solution of the Boltzmann equation, $\vec{k}\cdot\vec{u}$ for ρ and $(E-E_F)\vec{k}\cdot\vec{u}$ for W, where $\vec{k}$ is the electron wave vector and E its energy, E_F the Fermi energy, and $\vec{u}$ a unit vector in the direction of the applied field, W_v is given by[6]

$$W_v = \frac{27\hbar}{2\pi^3 MNk_B^3 k_F^2 \sigma^2 T^2} \sum_p \int\int \frac{[(\vec{K}\cdot\vec{u})^2 + 3(\vec{k}_1\cdot\vec{u})(\vec{k}_2\cdot\vec{u})]z^2(\vec{K}\cdot\vec{e}_p)^2 C^2(K)\, d\sigma_1\, d\sigma_2}{(e^z-1)(1-e^{-z})v_1 v_2}\,. \tag{2}$$

Here M is the mass of the ion; N the number of cells per unit volume; k_F the Fermi wave vector; v the velocity of the electron on the Fermi surface; σ the Fermi surface area; $z = \hbar\nu/k_BT$, ν being the frequency of the scattering phonon; and $\vec{K} = \vec{k}_2 - \vec{k}_1 = \vec{q} \pm \vec{g}$ is the scattering vector, $\vec{g}$ being the reciprocal-lattice vector and $\vec{q}$ the phonon wave vector. The subscripts 1 and 2 denote the original and final state of the electron. The summation over p is over the three possible polarizations $\vec{e}_p$ of the phonon. $C(K)$ is the pseudopotential used in the electron-phonon interaction.

At low temperatures, W_v dominates W; in that region it is proportional to T^2, but its magnitude is given incorrectly by Eq. (2).[4,7] As the temperature increases, W_v goes through a maximum and then decreases monotonically, approaching zero in the limit of high temperatures as T^{-2}. Our interest lies in this last region, for here not only is Eq. (2) valid to order $(\pi k_BT/E_F),^2$ but furthermore it becomes independent of lattice dynamics: This is readily seen, for $(e^z-1)(1-e^{-z}) \to z^2$ as T becomes large. Under these circumstances Eq. (2) can be relatively easily evaluated, both for normal (N) and umklapp (U) scattering processes, with only the single assumption of a spherical Fermi surface.

The reduction of Eq. (2) proceeds by standard steps.[6] For both N and U processes, the summation over p can be explicitly performed, and gives the same result: $\sum_p (\vec{e}_p\cdot\vec{K})^2 = 4k_F^2u^2$, where $u = \sin\frac{1}{2}\Omega = K/2k_F$, Ω being the scattering angle. For a spherical Fermi surface the integrals, upon transformation of coordinates, can be expressed in the following form:

$$W_v = A\left[\int_0^{u_1} V^2(u)\,F_N(u)\,du + \textstyle\sum_g \int_{u_2}^1 V^2(u)\,F_U(u)\,du\right], \tag{3a}$$

where

$$A = \frac{3(3\pi^2)^{1/3}}{2\pi}\left(\frac{\hbar}{k_B}\right)^3 \frac{1}{aMT^2}\,, \tag{3b}$$

$$V(u) = 3C(K)/2E_F\,, \tag{3c}$$

where a is the cube root of the unit cell. The first part of Eq. (3a) describes the resistance due to N processes, the second that due to U processes. u_1 depends on the Brillouin zone and $u_2 = g/4k_F$. The summation over g means a sum over the possible *types* of reciprocal-lattice vectors, and not over the total number of g's of any type, that sum being already incorporated into F_U. Thus, for both the fcc and the bcc lattices the sum extends over two terms, covering the directions [100] and [111] for the former, and [100] and [110] for the latter.

F_N and F_U can be viewed as geometrical weighting factors of the (reduced) pseudopotential V, and are given explicitly by

$$F_N(u) = (96/\pi)\,u^3(3-2u^2)\int[1-\cos z_m(\phi)]\,d\phi\,, \tag{4}$$

$$F_U(u) = (12/\pi)\,I_g u^3(3-2u^2)\int[1-\cos z_m(\phi)]\,d\phi\,. \tag{5}$$

z is the angle between K and the nearest g; z_m, its maximum value, is a function of ϕ, the angle of rotation about that g (see Fig. 1). $I_g = 1, \frac{4}{3}$, and 2 for g's in the directions [100], [111], and [110], respectively. The integration in (4) has to be carried out over the irreducible $\frac{1}{48}$th of the corresponding Brillouin zone, and in (5) over that part of the Brillouin zone which can contribute to U scattering for any particular g. It should be noted that no geometrical approximations have been made in deriving Eqs. (3)–(5), which is certainly not the case in some of the previously pub-

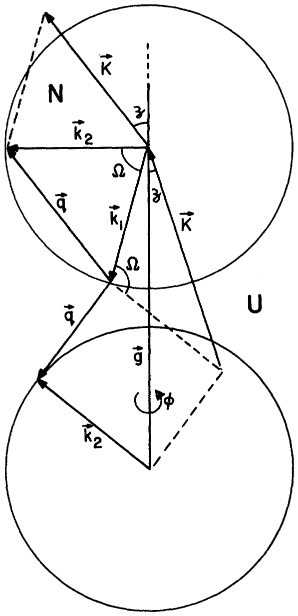

FIG. 1. Geometry for normal (N) and umklapp (U) scattering.

lished calculations.[8-10] The applicability of our equations is only limited by the assumption of a spherical Fermi surface, and of the cancellation of the terms in z. From a practical point of view, the latter assumption is valid whenever the experimental W_v displays a T^{-2} dependence.

In the past, many of the calculations of transport properties were done by integration not over the Brillouin zone, but over the equivalent Wigner-Seitz sphere, with a maximum vector Q_m. For such a scheme, both F_N and F_U can be put into analytical form:

$$F_N(u) = 8u^3(3 - 2u^2)\,, \tag{4a}$$

$$F_U(u) = 6I_g u^2(3 - 2u^2)[Q^2 - (G - 2u)^2]/G\,, \tag{5a}$$

where $G = g/k_F$ and $Q = Q_m/k_F$. Here, $u_1 = \frac{1}{2}Q$ and $u_2 = \frac{1}{2}(G - Q)$. Although the F's calculated for the Wigner-Seitz sphere differ considerably from those for the Brillouin zone (Fig. 2), it turns out that upon integration to yield W_v the final results differ only marginally. For the two test cases that we have calculated, one for a bcc lattice (Na) and one for a fcc lattice (Au), the W_v's differed at most by 2.4% (Table I). Such variations are negligible when compared to the uncertainties in the potentials V, or even in the experimental data for W_v. One may conclude therefore that for most cases the much simpler equations (4a) and (5a) are perfectly adequate.

Fortunately, the insensitivity of W_v to variations in the F's is not coupled with an equal insensitivity to variations in the potentials V. The difference between the two responses is probably due to the fact that the variations in the F's are of a balanced nature, decreases in one part being made up by increases in another, whereas the V's tend to differ monotonically in u, and produce quite significant changes in the calculated W_v, as will be shown in Sec. III.

III. COMPARISON WITH EXPERIMENTAL RESULTS

A. Alkali Metals

The obvious check of our calculations is through comparison with the observed resistivities of the alkali metals, as these satisfy best the require-

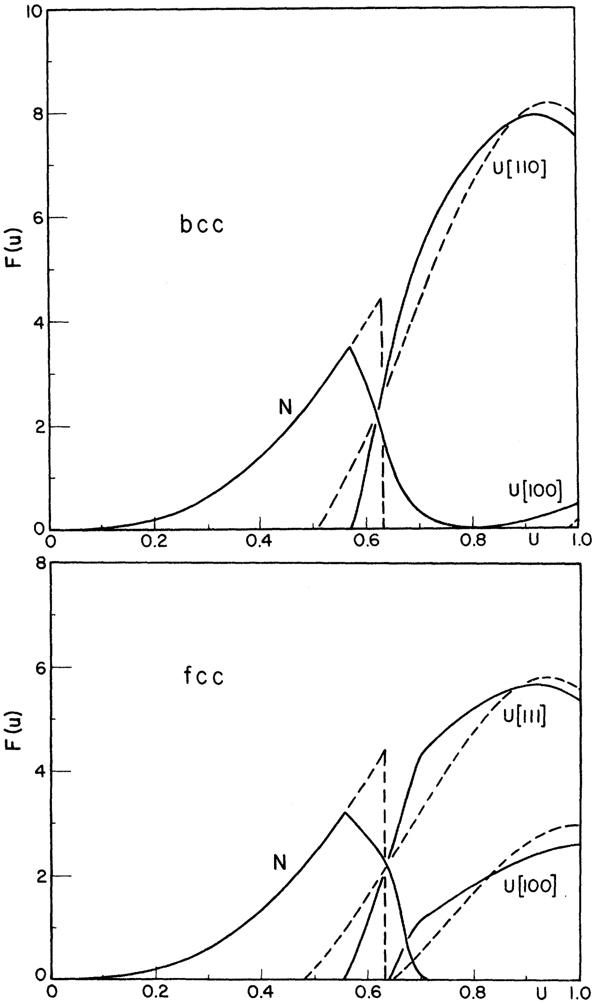

FIG. 2. Geometrical weighting factors $F(u)$ calculated for the bcc and fcc structures for the proper Brillouin zones (solid lines) and for the Wigner-Seitz spheres (broken lines). The labels N and U show the curves appropriate to normal and umklapp scattering, the latter depending upon the orientation of the reciprocal-lattice vectors. By coincidence, the U[100] curve for the bcc lattice appears as an extension of the N curve in the Brillouin-zone scheme; in the Wigner-Seitz scheme, this curve is barely visible in the lower-right-hand corner.

TABLE I. Calculated values of W_v in the high-temperature limit. *A* or *B* denotes the use of the Ashcroft or Bardeen potentials; W-S or Br denotes integration over the Wigner-Seitz sphere or over the Brillouin zone. Note that for all cases the resistance due to normal processes exceeds that due to umklapp processes.

				$10^{-4}W_vT^2$ (cm K^3/W)						
					Umklapp				Total	
Element	Potential	$[V_0(r_s)-E_0]$ (eV)	Zone	Normal	[100]	[111]	[110]	Total	calc.	Obs.
Na	*A*	···	W-S	0.12437	0.00000	···	0.04466	0.04466	0.16903	0.30
Na	*A*	···	Br	0.12267	0.00004	···	0.04233	0.04237	0.16505	0.30
Na	*B*	1.45	W-S	0.17708	0.00002	···	0.12393	0.12395	0.30102	0.30
Na	*B*	1.45	Br	0.17494	0.00054	···	0.12337	0.12391	0.29885	0.30
Na	*B*	0.09	W-S	0.12181		···		0.05447	0.17628	0.30
K	*A*	···	W-S	0.06217	0.00000	···	0.02168	0.02168	0.08384	0.1
K	*B*	0.02	W-S	0.06231	0.00000	···	0.03006	0.03006	0.09238	0.1
Rb	*A*	···	W-S	0.03005	0.00000	···	0.01416	0.01416	0.04421	0.01 (?)
Cu	*B*	1.3	W-S	0.06386	0.00455	0.02520	···	0.02975	0.09361	0.21
Ag	*B*	1.1	W-S	0.03630	0.00286	0.01541	···	0.01827	0.05457	0.11
Ag	*B*	2.3	W-S	0.04565	0.00491	0.02423	···	0.02914	0.07479	0.11
Au	*B*	3.7	W-S	0.03187	0.00463	0.02140	···	0.02603	0.05790	0.062
Au	*B*	3.7	Br	0.03133	0.00567	0.02095	···	0.02662	0.05795	0.062

ment of a spherical Fermi surface. Unfortunately, barring Na, experimental results for these metals in their high-temperature ranges are scarce and not too reliable; for K, Rb, and Cs we have found only one source, and the scatter of the experimental points for the latter two metals precludes any useful comparisons.

Figure 4 illustrates W_v for Na and K. For the former, we have used the results of Berman and MacDonald,[11] MacDonald *et al.*,[12] and Cook *et al.*[13]; for the latter those of MacDonald *et al.*[12] In computing W_v for Na we have subtracted a phonon contribution estimated on the basis of the Leibfried and Schlomann equation from the total thermal conductivity.[14] For Na we show the results in the whole temperature range from 10 to 350 K; for K only in the upper part.

We have used two pseudopotentials for our calculation: that of Ashcroft[15] and that of Bardeen.[16] The Ashcroft potential is completely specified; the Bardeen potential contains one parameter $[V_0(r_s) - E_0]$ about which there is some lack of unanimity in the published literature. Bardeen suggests that for the alkali metals this is about zero, and using his prescription and the calculations of Ham,[17] we have evaluated it as 0.09, 0.02, and 0.08 eV for Na, K, and Rb, respectively, values which are essentially in agreement with those used recently by other authors.[2,3] Some of the potentials that we have used are shown in Fig. 3, and the results of our calculations are given in Table I and are also illustrated in Fig. 4.

For Na, the calculated values for the above potentials underestimate the observed values by a substantial amount. To get any agreement between the two for the Bardeen potential, $[V_0(r_s) - E_0]$ has to be increased to about 1.45 eV, a value very close to the 1.3 eV given for this parameter by Mott and Jones,[18] and used in the calculations of Ziman.[8] Exact comparison here is difficult because of the sudden upturn in the observed W_v for temperatures in excess of 260 K, which Cook *et al.* have tentatively attributed to a contribution from electron-electron scattering.

For K, both the Ashcroft and Bardeen potentials fit reasonably well, in agreement with the detailed calculations of ρ at low temperatures by Trofimenkoff and Ekin,[3] which, for these two potentials, bracketed the experimental results. The agreement may be somewhat spurious, for we suspect that the results of MacDonald *et al.* yield too low

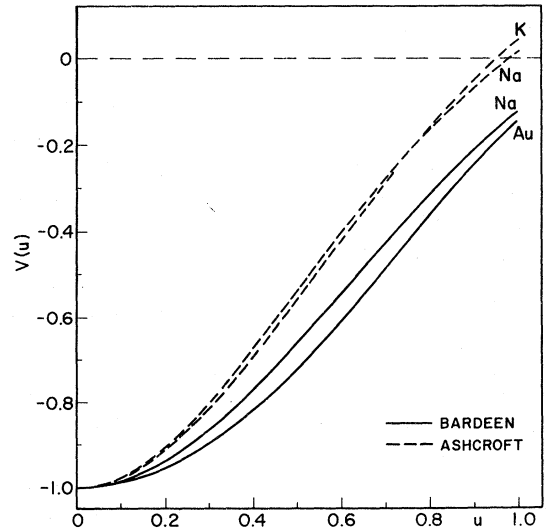

FIG. 3. Reduced pseudopotentials $V(u)$ used in the calculations.

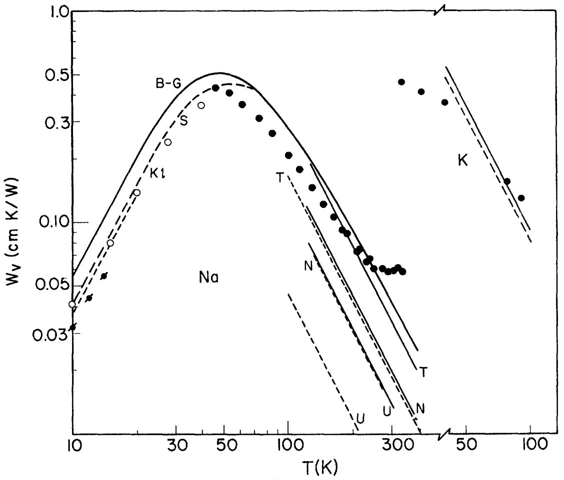

FIG. 4. Comparison of the experimental and calculated W_v for the alkali metals. The experimental results are taken from Refs. 11 (crossed dots), 12 (circles), and 13 (dots) for Na, and from Ref. 12 for K. The straight lines are our calculated values, and solid and broken lines for the Bardeen and Ashcroft pseudopotentials; for Na, we show the total resistivities (T) and their normal (N) and umklapp (U) components. For comparison, we also show the curves obtained from the Bloch-Grüneisen equation (B-G), and from the work of Sondheimer (S) and Klemens (Kl), for a characteristic temperature of 260 K.

a W_v at high temperatures, and to check this out we are currently undertaking high-temperature measurements on K similar to those that we have done for Na.

For Rb, both potentials overestimate the resistivity by a factor of about 4 (Table I); however, here we have absolutely no confidence in the high-temperature W_v data.

For purposes of comparison, we show in Fig. 4 W_v calculated for Na by the formulas of Klemens,[4] Bloch-Grüneisen,[6] and Sondheimer[7] (third-variational solution), on the assumption of a Debye temperature $\Theta \sim 260$ K, a rough estimate given for longitudinal phonons by Blackman.[19] The apparently good agreement between Klemens's values and the low-temperature results will be discussed briefly in Sec. IV.

B. Noble Metals

The extension of our calculations to the noble metals is by way of an exploration, for two factors affect the applicability of our equations to these metals: (i) the nonsphericity of their Fermi surfaces, which at best make our formulas only approximate; and (ii) the intersection of these surfaces with the Brillouin zones, which may increase the contribution of U processes to the resistivity: This certainly has a large effect on the low-temperature ρ,[20] but whether it is important at high temperatures, where only large-angle scattering occurs, is a moot point—our guess is that it is not significant.

The experimental results for the noble metals are of good precision. We have used those of Moore *et al.*[21] and of Laubitz[22] for Cu, those of Matsumura and Laubitz[23] and of Laubitz[24] for Ag, and those of White and Woods,[25] Cook and van der Meer,[25(a)] and Laubitz[24] for Au. As before, we have subtracted the phonon conductivities from the total thermal conductivities before calculating W_v; these were taken from White[26] and from White *et al.*[27] From the high-temperature results of Laubitz we have further subtracted the estimated electron-electron scattering contribution.[28] The results are shown in Fig. 5.

Only the Bardeen pseudopotential was tried for these metals. The parameter $[V_0(r_s) - E_0]$ has been taken directly from Bardeen: Values of 1.3, 1.1, and 3.7 eV for Cu, Ag, and Au, with the potential for Au illustrated in Fig. 3. The calculated results (Table I, Fig. 5) show good agreement for Au, and poor agreement for both Cu and Ag. To obtain a good fit for the latter two, the potentials would have to be broadened considerably; for Cu, the parameter $[V_0(r_s) - E_0]$ must assume a value of 4.8 eV, and surprisingly, the resultant potential is virtually identical with that of Au, illustrated in Fig. 3. The case of Ag is more complicated: The experimental results do not display a T^{-2} dependence until much higher temperatures are reached than is the case with the other metals. In the intermediate range ($200 < T < 400$ K) a reasonable fit is obtained with $[V_0(r_s) - E_0] = 2.3$ eV, the value given in Mott and Jones. If, however, we accept that the correct high-temperature limit is not reached until $T > 400$ K, then the potential required to match this is again identical with that of Au.

It appears then that one and the same potential will satisfactorily describe all the noble metals. This potential appears somewhat broader than that required for the alkali metals and, for all we know, may not have the shape predicted by the Bardeen equation, shown in Fig. 3, but one such as calculated by Collins[29] for deformed Fermi surfaces. This, of course, is the main drawback in all the calculations of the transport properties: They can serve as a test of the pseudopotentials, but cannot be used to derive them.

From the above comparisons we may conclude that the calculations of the high-temperature thermal resistivity are in a much better state than could be surmised from Ref. 2. We certainly get the right temperature dependence of W_v, and reasonable agreement in magnitude for some of the pseudopotentials examined. In fact, our approach seems useful as a good and simple first test of the pseudopotential. Although it is not nearly as sensi-

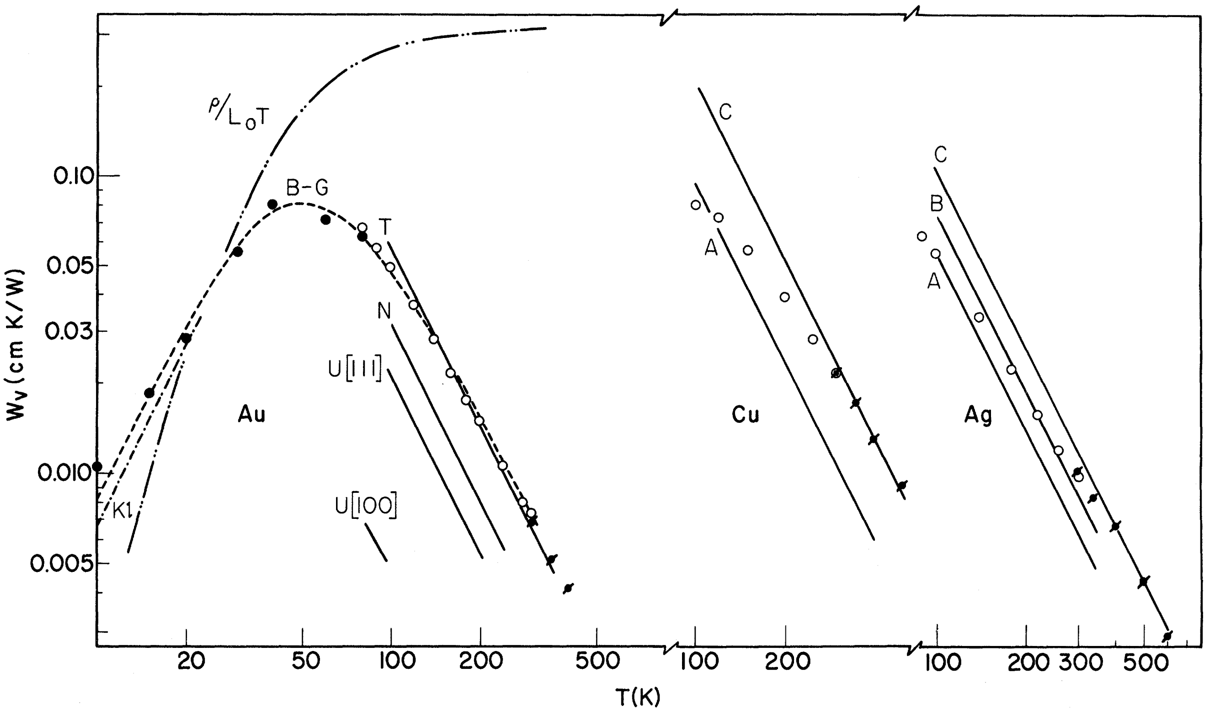

FIG. 5. Comparison of the experimental and calculated W_v for the noble metals. Dots, circles, and crossed dots are the experimental results taken from Ref. 24, 25, and 25(a) for Au, 21 and 22 for Cu, and 23 and 24 for Ag. The solid lines are the results of our calculations: For Au we show the total resistivity (*T*) and its normal (*N*) and two umklapp (*U*) components; for Cu and Ag we illustrate the total resistivity calculated with a Bardeen potential containing parameters as given by Bardeen (*A*), Mott and Jones (*B*), and as used here for Au (*C*) and shown in Fig. 3. The B-G curve is a one-parameter fit of the standard Bloch-Grüneisen equation to Au, with Kl the corresponding resistivity given by the formula of Klemens. For comparison, we also show the experimental "elastic" part of the thermal resistivity ρ/L_0T.

tive to small variations in the potential as the detailed calculations of the low-temperature resistivity,[3] it is much simpler to perform, and intrinsically more accurate, for our calculations do not depend on the lattice dynamics, and are based on valid solutions of the Boltzmann equation.

IV. LOW-TEMPERATURE LIMIT

In closing, we should like to take a simple-minded look at the low-temperature limit of W_v, where, we feel, there appears to be a genuine discrepancy between theory and experiment. Klemens[4] has numerically obtained the solution of the Boltzmann equation for W in the limit of low temperatures, which can be put into the following form:

$$W_v = 316A(T/\Theta)^4, \tag{6}$$

where A is the same as in Eq. (3b). In (6), the pertinent Θ must be that for the longitudinal phonons, Θ_L, for the following reason: At very low temperatures, the ratio of the resistances due to U and N processes is roughly $[2k_F\,C(1)/KC(0)]^2$ for ρ, while for W_v it is $\frac{1}{3}[C(1)/C(0)]^2$. Thus, the resistance due to U processes should be smaller in W than in ρ by about a factor $3(2k_F/K)^2$ which, at 10 K, is of the order of 4×10^2. On the basis of the low-temperature calculations of ρ^3, the contribution of U processes to W_v becomes completely insignificant, and W_v is dominated by $\underline{N}$ processes which, because of the factor $(\vec{e}_p\cdot\vec{K})$ in the electron-phonon interaction, are limited to the longitudinal phonons only. However, comparison of W_v calculated on the basis of realistic estimates[30] of Θ_L fall far short of those observed experimentally, as can be seen in Table II. The only possible explanation of this that we can see lies in the mixing of longitudinal and transverse modes off principal directions, and we feel that it would be useful to have this checked out by detailed calculations.

TABLE II. Comparison of the calculated and observed thermal contributions at low temperatures.

	Na	K	Cu	Ag	Au
Θ_L (K)	310	187	650	449	395
$10^4W_vT^{-2}$ (cm/W K), calc.	1.7	6.2	0.05	0.11	0.10
$10^4W_vT^{-2}$ (cm/W K), obs.	3.5	13	0.2	0.3	0.8

Finally, we should like to note with some wry amusement how well the simple Bloch-Grüneisen equation fits the experimental W_v. This is illustrated in Fig. 5 for Au, where we have plotted that equation arbitrarily adjusting only one parameter, Θ, which here turns out to be 265 K; the magnitude of the calculated W_v is essentially that predicted by that equation for that Θ. The Bloch-Grüneisen formulation suffers from many shortcomings,[8] and the good fit observed through the whole temperature range can only be viewed as fortuitous cancellation of the large and temperature-dependent effects of three unconnected simplifications employed in its development: (a) the use of the first-variational solution; (b) neglect of umklapp scattering; and (c) assumption that the electron-phonon interaction is independent of the scattering angle. Nevertheless, one has to admit that the Bloch-Grüneisen equation forms a remarkably good interpolation formula.

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[30] The Θ_L values were kindly supplied to us by Dr. Y. P. Varshni of the University of Ottawa, and were obtained by suitable numerical averaging from the largest of the three velocities in any direction. Note that for Na Θ_L differs considerably from the value of Ref. 19, which gave a reasonable fit to W (Fig. 3).