

Electron-phonon interaction and lattice thermal conductivity

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Phonons in a metal have a finite lifetime due to the emission of electron-hole pairs. This process leads to an electron-phonon contribution W_{ep}^p to the thermal resistance, which limits the lattice thermal conductivity κ_p . We present a summary of the available experimental data on W_{ep}^p , emphasizing uncertainties and contrasting results from different types of experiments. Methods of obtaining better data are suggested. We show that the available data are in fair agreement with a simple theoretical estimate $\lim(T \rightarrow 0) W_{ep}^p (T/\theta_D)^2 = 0.42 \Omega_a^{1/3} N \lambda$ (Kcm/W), where Ω_a is the atomic volume in \AA^3 , N is the Fermi-energy density of states (1 spin) in states/eV, and λ is the electron-phonon mass enhancement. The fact that most of the experimental points fall below this estimate is probably the result of anisotropy and mode dependence of the electron-phonon coupling.

I. INTRODUCTION

In a metal heat is transported by the phonons as well as the electrons. The phonon thermal conductivity κ_p is limited both by processes in which a phonon scatters off another phonon and by processes in which a phonon decays by emission of electron-hole pairs.¹ This latter process leads to the electron-phonon component of the lattice thermal resistance W_{ep}^p , which is the subject of this paper. It also makes a contribution $\gamma_i^{ep}(q)$ to the phonon linewidth $\gamma_i(q)$ which is the width in energy of a phonon of momentum q and mode index i .

$\gamma_i^{ep}(q)$ has been observed recently²⁻⁴ by inelastic neutron scattering. It is of great theoretical interest since $\gamma_i^{ep}(q)/\omega_i^2(q)$ is proportional to the amount of phonon-mediated electron-electron coupling which arises from the phonon with momentum q , mode index i , and frequency $\omega_i(q)$. The electron-phonon coupling parameter λ , which determines the electronic mass enhancement and the superconducting transition temperature, may be written as an average over i and q of $\gamma_i^{ep}(q)$ as⁵

$$\lambda = \sum_{q,i} \frac{\gamma_i^{ep}(q)}{\hbar^2 \omega_i^2(q) \pi N}, \quad (1.1)$$

where N is the Fermi-energy density of states (1 spin) per atom. We shall show later that W_{ep}^p may be written as an average over q and i of the phonon lifetime $\tau_i^{ep}(q)$ which is related to the linewidth through the uncertainty relation $\tau_i^{ep}(q) = \hbar/\gamma_i^{ep}(q)$. Thus λ and W_{ep}^p are intimately related, and measurements of W_{ep}^p can, in principle, yield valuable information about the basic electron-phonon interaction process.

In this paper we present a summary of the available experimental information which is, unfortunately, limited in its extent and accuracy. We

suggest some methods for improving this situation and show that the available data are in fair agreement with theoretical estimates of the average strength of the electron-phonon interaction. Section II describes the methods used to extract W_{ep}^p values from measurements of thermal and electrical conductivity. In Sec. III we review the experimental data on W_{ep}^p for 15 elements, in Sec. IV we develop a formula that relates W_{ep}^p to the density of states and λ , and, finally, in Sec. V we compare the experimental data with this formula.

II. EXPERIMENTAL IDENTIFICATION OF THE ELECTRON-PHONON THERMAL RESISTANCE

The phonon thermal conductivity κ_p of a pure metal cannot be directly determined because the parallel electronic conductivity κ_e is usually much larger. In this section we summarize the three experimental techniques that have been used to identify κ_p , emphasizing the experimental and analytical difficulties associated with each method. Klemens⁶ has reviewed this type of research, but considerable progress has been made during the last 20 years.

A. Low-temperature alloying technique

The electronic conductivity of alloys can be greatly reduced by impurity scattering, and this effect has been widely used to identify κ_p . The difficulties associated with the method can be seen by writing the measurable total conductivity in terms of its two components, and by showing explicitly the various resistances that limit them⁷:

$$\kappa = \kappa_e + \kappa_p = (W_i^e + W_{ep}^e + \Delta W^e)^{-1} + (W_{ep}^p + W_u^p + W_d^p + W_i^p)^{-1}. \quad (2.1)$$

In Eq. (2.1) the superscripts on the W 's identify

the carriers of the heat flux, while the subscripts denote the scattering mechanism. ΔW^e denotes the "deviational" thermal resistivity which arises from the failure of Mattheissen's rule. At "sufficiently" low temperatures the electronic resistances reduce to the impurity scattering contribution W_i^e , because the electron-phonon (W_{ep}^e) and deviational (ΔW^e) terms decrease with decreasing temperature. Similarly, the phonon resistances W_u^p and W_i^p , which are due to phonon-phonon scattering and to impurity scattering, respectively, also tend to zero at low temperatures, leaving the electron-phonon W_{ep}^p and dislocation W_d^p resistances. Prolonged annealing should reduce W_d^p , and the remaining terms W_i^e and W_{ep}^p can be identified both by using the theoretical temperature dependence and by calculating W_i^e from the resistivity measurements and the Sommerfeld-Lorenz number L_0 . Examination of (2.1) shows the principal deficiencies of the method: (i) The dislocation resistance must be minimized or identified because it has the same temperature dependence as W_{ep}^p . This requires additional measurements and electron microscopy. (ii) Alloying can change W_{ep}^e and, more importantly, W_{ep}^p . These possibilities must be evaluated experimentally and, if possible, theoretically. (iii) Separation based on the low-temperature limit,

$$\kappa = (W_i^e)^{-1} + (W_{ep}^p)^{-1},$$

can only be valid if the temperature is "sufficiently" low, and this condition must be verified by estimating the other resistances and making electrical resistivity measurements, which can show where Mattheissen's rule begins to break down. (iv) On the other hand, if the temperature is too low in an alloy for which the electronic mean free path is very short, one can enter a regime in which the wavelength of the important phonons exceeds the electronic mean free path. Pippard⁸ has shown that the standard quantum perturbation theory treatment of the electron-phonon interaction breaks down in this region and the expected $\kappa_p \propto T^2$ temperature variation is no longer obtained.

B. Intermediate-temperature alloying technique

As the temperature is increased, the analysis required to separate κ_p and κ_e becomes more difficult because W_{ep}^e , W_u^p , and W_i^p become more important. We have used a procedure⁹ that requires measurements of both thermal and electrical conductivity on at least two samples, one of which is the pure metal and the others are dilute alloys. These measurements typically extend from $T \lesssim \Theta_D/5$ to $T \gtrsim 2\Theta_D$.

By assuming that (a) the electronic Lorenz func-

tion $L_M(T)$ and the phonon conductivity κ_p are not altered by alloying, and (b) the impurity and deviational terms in the electrical and thermal resistivities are related by the Sommerfeld-Lorenz number L_0 , one can derive equations of the form

$$\kappa = [\rho_M/L_M(T)T + (\rho - \rho_M)/L_0T]^{-1} + \kappa_p \quad (2.2)$$

for each sample. Here ρ_M is the electrical resistivity of the pure metal, while κ and ρ are the measured thermal conductivity and electrical resistivity of the particular sample. Two equations of the form (2.2) can be solved for the unknowns $L_M(T)$ and κ_p in terms of the measured quantities κ , λ_M , ρ , and ρ_M . Equations for any other sample provide at least a partial check on the consistency of assumptions (a) and (b) above.

Once $W^p = 1/\kappa_p$ is known, it must be separated into phonon-phonon (W_u^p) and electron-phonon (W_{ep}^p) contributions. This can be done either by examining the temperature dependence of W^p for $T \gtrsim \Theta_D$, where W_u^p should be linear in T and W_{ep}^p constant, or by estimation of W_u^p from the Leibfried-Schlömann equation.¹⁰ We have used the latter method, performing the calculations at the lowest possible temperature so as to minimize the magnitude of the W_u^p correction.

Difficulties with this method include: (i) possible variation of W_{ep}^e or κ_p with solute concentration and (ii) uncertainty in the phonon-phonon correction. It should be noted that even if these difficulties are overcome $W_{ep}^p(T)$ at intermediate temperature samples $\tau_{ep}(q)$ at higher frequencies than $W_{ep}^p(T \rightarrow 0)$. In Sec. IV we discuss a method for comparing low- and intermediate-temperature results.

C. Magnetothermal resistance technique

The magnetothermal resistance effect can be used to obtain κ_p values for high-purity samples of compensated metals at low temperatures. Here one assumes that κ_p is not changed by the field, and applies large fields to increase the electronic thermal resistance. If sufficiently large fields were available, the electronic conductivity could be effectively eliminated, but this is not yet possible and various methods for deriving the infinite field values must be employed. Two possibilities that have been employed are to extrapolate the measured thermal resistance using the theoretical field dependence¹¹ and to employ the "modified Wiedemann-Franz law" with magnetoresistivity measurements.¹² Pernicone and Schroeder¹³ have discussed the difficulties associated with analyzing the experimental data and extrapolating to find the lattice conductivity.

The theory of the thermal-conductivity change associated with the normal-superconducting transition¹⁴ has been used to derive normal-state κ_p

values for some elements which exist in the superconducting state over appreciable temperature ranges. The separation involves using the theory to calculate the electronic thermal conductivity in the superconducting state, obtaining the phonon part by subtraction, and again using the theory to account for the increased electron-phonon scattering in the normal state. This method can be applied for a limited number of elements but, of course, is subject to uncertainties arising from the approximations used in the Bardeen-Rickayzen-Tewordt¹⁴ (BRT) theory as well as possible complications that arise when both electron and impurity scattering are significant in the superconducting state.¹⁵ Most of the experimental work on this problem has been done to establish the theory and not to obtain normal-state lattice conductivity values, but this separation method provides a good consistency test for some elements.

D. Low- versus intermediate-temperature measurements

The experimental data are mainly from low-temperature experiments ($T/\Theta_D < 0.05$), but with some results from intermediate temperature ($0.05 < T/\Theta_D < 0.5$). In order to include the intermediate-temperature data and compare them with the low-temperature results, it is necessary to make some assumption about the temperature variation of W_{ep}^p . We have derived the temperature variation of W_{ep}^p based on the assumption that $\tau_i^{ep}(q)$ is proportional to $[\omega_i(q)]^{-1}$. The details of this approximation are discussed in Sec. IV. We find it important to take account of the effect of phonon dispersion on the temperature variation of W_{ep}^p .

Since W_{ep}^p varies as T^{-2} at sufficiently low temperatures, we have chosen to discuss the quantity $W_0 \equiv \lim_{T \rightarrow 0} [W_{ep}^p(T)(T/\Theta_D)^2]$. In Sec. IV we show that W_0 gives a fairly direct measure of the average electron-phonon lifetime at long wavelengths.

III. SURVEY OF EXPERIMENTAL RESULTS

A summary of the experimental results for W_0 is shown in Table I. In this section, we give a fairly detailed account of how these W_0 values were deduced from the various experiments. The non-specialist who is not interested in such details may want to skip to Sec. IV. He should remember, however, that one important conclusion of this section is that most of the W_0 values in Table I have substantial uncertainties.

Much of the older work was located through the review article by Klemens,⁶ and where possible the discussion includes attempts to define the experimental uncertainties. These uncertainties do not include inadequacies in the formulas used to de-

rive the κ_p values. For instance, the reader should note that the low-temperature alloy results were not all calculated in exactly the same way. Data for several anisotropic elements (Be, Ga, In and Sn) are included in the survey, because there is a limited amount of information and our goal was to examine the gross features of the variation of W_{ep}^p .

A. Copper, silver, and gold

The low-temperature alloy studies have been summarized by White, Woods, and Elford¹⁶ and by Kemp and Klemens.¹⁷ W_{ep}^p values derived for concentrated alloys tend to be high, and a cusp-like W_{ep}^p minimum was found by varying the Pd and Cd concentration in Ag.¹⁷ Annealing reduced, but did not eliminate, the effect. Therefore, the most appropriate values for elements are obtained from very dilute alloys or extrapolation of values for a series of alloys. For Cu, the data are from a Cu-Fe alloy¹⁶ and from a series of Cu-Zn alloys.¹⁸ For Ag the average of values for Ag-Sn,¹⁶ Ag-Pd,¹⁷ and Ag-Cd,¹⁷ were used, and for Au data for Fe,¹⁶ Pt, and Cr solutions¹⁷ were extrapolated and averaged to yield the final values shown in Table I. Debye temperatures from low- T specific-heat measurements¹⁹ and inelastic neutron scattering²⁰ were used to calculate W_0 values. The value for Ag seems to be fairly well defined by the data, and a tentative uncertainty of $\pm 15\%$ is estimated based on experimental scatter and a ± 10 K variation in Θ_D . On a similar basis, the W_0 values for Au and Cu are uncertain by about $\pm 30\%$.

Van Witzenburg and Laubitz¹² have reported magnetoresistance measurements for Cu, Ag, and Au at intermediate temperatures. Unfortunately, calculations based on these data, which assume the existence of a modified Wiedemann-Franz law in the presence of a magnetic field, produce κ_p values that are 2 or 3 times larger than the Leibfried-Schlömann (LS) estimate¹⁰ (which completely neglects electron-phonon scattering), so W_{ep}^p values cannot be obtained from these measurements.

B. Tungsten

Results on this element can be used to compare the magnetic and intermediate-temperature alloying techniques. The alloy results, obtained using W-Re and W-Ta alloys,⁹ indicate that the total lattice resistance is about 2.0 K cm/W at 90 K, and using the LS equation to estimate W_{ep}^p ($\sim 30\%$) yields a W_{ep}^p value of about 1.3 at 90 K. This base temperature was chosen because it was the lowest available and W_{ep}^p decreases in importance at higher T . W_0 was calculated using the "constant- α^2 "

TABLE I. Comparison of low-temperature lattice thermal resistivity W_0 with theoretical "constant- α " values.

Element	Θ_D (K)	$\langle \omega^{-1} \rangle^{-1}$ (K)	$\Omega_a^{1/3}$ (Å)	N (states/eV)	λ	Experimental		Experimental technique ^a	W_0 [Eq. (4.12)]
						W_0 (cmK/W)	W_0 (cmK/W)		
Cu	342	210	2.27	0.13	0.14 ± 0.04	0.0047 ± 0.0014		A	0.018 ± 0.006
Ag	228	140	2.57	0.13	0.10 ± 0.04	0.009 ± 0.0014		A	0.014 ± 0.006
Au	165	169	2.57	0.14	0.14 ± 0.04	0.012 ± 0.004		A	0.036 ± 0.01
Be	1160	(0.6 θ_D)	2.00	0.040	0.23	0.0037		A	0.0078
Be						0.0028		B	
Al	430	265	2.55	0.20	0.43	0.058 ± 0.005		A	0.096
Al						0.054		B	
Ga	320	(0.6 θ_D)	2.70	0.091	0.40	0.018		A	0.042
Ga						0.014		B	
In	112	80	2.90	0.20	0.83	0.15 ± 0.06		A	0.24
In						0.084		B	
Sn	200	110	3.00	0.22	0.72	0.26		A	0.18
Sn						0.067		B	
Nb	277	182	2.62	0.84	0.97	0.43		A	0.98
Ta	260	153	2.62	0.75	0.69	0.22 ± 0.06		A	0.56
Ta						0.54 ± 0.24		C	
Mo	460	261	2.50	0.27	0.44	0.10 ± 0.04		C	0.12
W	388	219	2.51	0.15	0.28	0.013		B	0.042
W						0.023 ± 0.009		C	
W						0.007		D	
Fe	470	283	2.27	(0.76)	(0.17)	0.12 ± 0.03		C	(0.12)
Pd	283	182	2.45	1.17	(0.29)	0.40 ± 0.08		A	0.38
Pt	234	(0.6 θ_D)	2.47	0.86	(0.27)	0.15 ± 0.08		A	0.24

^aA, low- T alloying; B, low- T magnetothermal resistance; C, intermediate- T alloying; D, intermediate- T magnetothermal resistance.

approximation discussed in Sec. IV. The inelastic neutron scattering data of Chen and Brockhouse²¹ were used to account for the effect of phonon dispersion on the temperature variation of W_{ep}^p . It is difficult to assess the uncertainties associated with W_0 obtained in this way; however, we estimate an uncertainty of about 40% based on experimental uncertainties, inadequacies of the κ_p separation model,⁹ and uncertainties associated with the phonon-phonon resistance correction.

The magnetoresistance data of van Witzenburg and Laubitz¹² which were also obtained at 90 K can be analyzed along similar lines, and yield a W_0 value of 0.007 K cm/W. This value is somewhat lower than the value of 0.013 derived from the low-temperature magnetoresistance data of Wagner.¹¹

C. Platinum and palladium

Fletcher and Greig²² have reported κ_p values derived from low- T data on Pt-Au, Pt-Ir, and Pd-Ag alloys. Buhl and Giaque²³ have also determined a κ_p value for a Pt-9-wt.-%-W alloy, but this result was not used in the analysis because there is no basis for correcting to infinite solute dilution. The Pd-Ag data are consistent with earlier results,²⁴ and indicate that W_{ep}^p is large for this element. Difficulties arise because the W_{ep}^p values do not exactly show the expected T dependence and the values shown in Table I are averaged over the 10–20 K range. This variation also gives a rough lower limit for the uncertainty in the W_0 values, and it should also be noted that the W_{ep}^p values for Pd-Ag alloys do not show the solute composition dependence noted for Pt, W, and the noble metals. The total minimum uncertainty estimate also includes a contribution from the Θ_D value.

D. Molybdenum

The W_0 value is based on unpublished data²⁵ on alloys containing 1%–2% of Nb and Zr and on the previously reported⁹ κ_p value for Mo-0.5-wt.-% Ti, which is about 35% lower than the later results. The results were analyzed using the method employed for the W alloy data, and at 90 K the W_u^p correction amounted to about 10%. The derived κ_p values do not show consistent trends with composition, and the best value was taken to be an average value determined at 90 K. The total minimum uncertainty estimate shown in Table I contains contributions from the scatter in the derived W_{ep}^p values, phonon-phonon resistance correction, and the Θ_D choice.

E. Tantalum

The lattice conductivity of this element has been determined at low^{26,27} and intermediate²⁸ temperatures using fairly concentrated alloys, and the property changes associated with the normal-superconducting transition have also been used to obtain the normal-state lattice conductivity.²⁸ The low-temperature data of Sousa on 10- and 20-at.-%-Nb alloys can be linearly extrapolated to yield a W_0 value of about 0.26 K cm/W, but it should be noted that Lowell's²⁷ κ_p result for Ta-20-at.-% Nb is about 60% higher than the Sousa value. Treatment of the data for more dilute Ta-Nb alloys²⁶ in the superconducting range yields a W_0 value of about 0.19 K cm/W. The low-temperature data thus indicate that W_0 is about 0.22 K cm/W.

The estimate of W_0 that was obtained from intermediate-temperature data is somewhat higher. Lattice resistance estimates for 6- and 12-at.-%-W alloys at 80 K were extrapolated to yield a W^p value of 18.7 K cm/W. The W additions probably decrease W_{ep}^p somewhat, while in this temperature range impurity scattering should be appreciable and thus the W^p values for alloys could be either greater or smaller than the true value for Ta. The data indicate that the W_u^p of the Ta-6-at.-%-W alloy is about 15% larger than the W_u^p of pure Ta. The LS equation was used to estimate W_u^p , and a correction of 9% was applied yielding $W_{ep}^p \approx 17$ K cm/W. The W_0 value 0.54 K cm/W was calculated by using the same "constant- α^2 " assumption as was used for Mo and W. We estimate an uncertainty in this value of W_0 of about 45%.

F. Niobium

Sousa^{26,29} has reported data on Nb-Mo alloys at low temperatures and shown that the results are consistent with the BRT theory and that they indicate a dependence on the density of states. The W_0 value shown in Table I was computed with a Debye temperature derived from low-temperature specific-heat results, and there is no basis for estimating a probable uncertainty range.

G. Iron

Results for three Fe-Cr alloys and one Fe-Cr-Ni alloy were used to determine W_L at 100 K.^{30,31} The data for all four alloys yield W^p values with a range of only 11%, so this seems to be a case in which W^p is fairly well defined. A W_u^p correction amounting to 26% was applied and W_0 was obtained in the same way as for W. The uncertainty estimate for this element was obtained with the procedure used for Mo and W. Unfortunately, we cannot compare the experimental W_0 with theory

because we do not have a good estimate of λ for Fe.

H. Beryllium

Kohler³² analyzed magnetoconductivity data obtained near 20 K to obtain an estimate of the lattice conductivity, 0.12–0.17 W/cm K. Assuming that this is within the $T^2-W_{ep}^p$ range and using the low-temperature specific-heat Θ_D yields an average W_0 value of 0.0028 K cm/W. This value is only about 30% less than the W_0 value derived from the White and Woods³³ data on a sintered Be specimen.

I. Indium

Lindenfeld and Rohrer³⁴ published κ_p values for two In-Bi and two In-Sn alloys in the normal state and showed that these results were consistent with the data for the superconducting state and the BRT¹⁴ theory. Averaging the values for the four alloys yields a W_0 value of 0.15 ± 0.06 K cm/W, but the trends with composition would favor a result on the low end of this range. The data of Hulm³⁵ on In-10-at.% Tl were reanalyzed by the authors using the Sommerfeld-Lorenz number with the residual resistivity, and also indicate a W_0 value of 0.15 K cm/W. Van Kempen³⁶ *et al.* performed magnetothermal measurements, and their data yield a W_0 of 0.084 K cm/W.

J. Aluminum

Klaffky *et al.*⁷ obtained data on a series of annealed Al-Mg alloys and derived values for the lattice conductivity of pure, defect-free Al in the appropriate (<20 K) temperature range, while Amundsen *et al.*³⁷ obtained similar data on Al-Cu alloys. W_0 values calculated from these two sets of data are 0.063 and 0.052 K cm/W, respectively. DeLang *et al.*³⁸ obtained κ_p via the magnetothermal technique, obtaining a W_0 value of 0.054, in excellent agreement with the alloying measurements.

K. Gallium

Gorter and Noordermeer³⁹ have presented alloy and magnetoconductivity results on this anisotropic element, and the magnetoconductivity data were used to derive κ_p values along the three principal axes. These results show a maximum anisotropy ratio of 1.6, and for the *a* axis the alloy method gave a lattice resistance that was about 15% higher than the magnetoconductivity value.

L. Tin

Pernicone and Schroeder¹³ have published magnetoconductivity data for this element and com-

pared the κ_p results with the alloy data of Karargin *et al.*⁴⁰ The magnetoconductivity data show an anisotropy ratio of about 2.2, and the basal plane κ_p values are about 5 times higher than the results from the alloy work. Pernicone and Schroeder also noted a departure from the usual T^2 dependence and suggested that this was due to the rapid variation of Θ_D with T . The Θ_D variation was included in the W_0 calculation by using the tabulated $\kappa_p - T$ values with the appropriate Θ_D to calculate individual W_0 values at a series of temperatures for both principal axes and averaging these results to get mean values for both principal axes. The anisotropy ratio from magnetoconductivity was used to estimate an $\langle 001 \rangle$ alloy κ_p from the $\langle 100 \rangle$ alloy measurement, and mean W_{ep}^p values for both experimental methods were then obtained from the averaging formula⁴¹ for polycrystalline conversions. A low temperature Θ_D of 200°K was used to calculate W_0 from the alloy data; however, a lower value of Θ_D might be more appropriate and would yield a correspondingly higher W_0 .

Inspection of the W_0 values shown in Table I reveals an interesting point. The W_0 values vary from a low of 0.003 K cm/W for Be to a high of 0.5 for Ta and Nb, and this range is more than 10 times larger than the high-temperature electrical and thermal-conductivity variations for these elements. Another interesting point is that there appear to be consistent, significant differences between the results obtained from the three experimental techniques.

IV. THEORETICAL ESTIMATE FOR W_{ep}^p

It is clear from Table I that there is substantial uncertainty, which averages about $\pm 40\%$, in most of the derived values of W_0 . Only the fact that there is a very large variation from the lowest to the highest of the tabulated values gives us some hope of achieving a meaningful correlation with theory. Generally, W_0 should be high for those metals that show strong electron-phonon coupling, and in this section we shall derive an approximate relation between W_{ep}^p and λ , the electron-phonon coupling parameter, which accounts in a semi-quantitative way for the interelement variation of W_0 .

Following Makinson⁴² and others, it is not difficult to obtain an expression for W_{ep}^p that takes into account the phonon lifetime due to the electron-phonon interaction:

$$(W_{ep}^p)^{-1} = \frac{1}{2} \sum_{qj} c_j(q, T) u_{jx}^2(q) \tau_j^{ep}(q), \quad (4.1)$$

where

$$c_j(q, T) = \frac{(\hbar^2 \omega_j^2 / kT^2) e^{\hbar \omega_j / kT}}{(1 - e^{\hbar \omega_j / kT})^2}$$

is the specific heat of the phonon mode having crystal momentum q and mode index j . $\omega_j(q)$ is the phonon frequency, $u_{jx}(q)$ is the x component of the phonon velocity, $\tau_j^{ep}(q)$ is the phonon lifetime as limited by the electron-phonon interaction, and Ω_a is the volume per atom. The factor of $\frac{1}{2}$ in Eq. (4.1) arises because $\tau_j^{ep}(q)$ as we have defined it describes the decay of the phonon *amplitude*, whereas the lifetime that enters the semiclassical formulas for the transport coefficients describes the decay or relaxation of the phonon *density*, i.e., the square of the amplitude.

At high temperatures ($kT \gg \hbar \omega_j$) (4.1) becomes

$$W_{\infty}^{-1} = \frac{\hbar k}{2\Omega_a} \sum_{aj} u_{jx}^2(q) / \gamma_j^{ep}(q), \quad (4.2)$$

which may be compared with Eq. (1.1). This equation emphasizes the fact that in this approximation W_{∞}^{-1} is an average over phonon modes of the phonon lifetime as limited by the electron-phonon interaction.

Unfortunately little is known in general about $\gamma_j^{ep}(q)$ at present. Calculations in the one orthogonalized plane wave (1-OPW) approximation⁴³⁻⁴⁵ which are meaningful for the alkali metals yield $\gamma_j^{ep}(q)$ proportional (at low q) to $[\hat{q} \cdot \hat{\epsilon}_j(q)]^2 / |q|$, where $\hat{\epsilon}_j(q)$ is the phonon polarization vector. Thus for these systems purely transverse phonon modes (for sufficiently low q that umklapp scattering is not allowed) have a lifetime that is not limited by the electron-phonon interaction, and W_{ep}^p , as defined by (4.1), vanishes. If the Fermi surface touches the Brillouin-zone boundary as in the polyvalent, noble, and transition metals, the 1-OPW approximation does not give the correct behavior of $\gamma_j^{ep}(q)$ at low q and a more careful treatment⁴⁶ gives $\gamma_j^{ep}(q)$ proportional to q for long wavelengths for all modes. Recent calculations by Butler *et al.*^{4,47} for Nb indicate that in the transition metals $\gamma_j^{ep}(q)$ is proportional to q for small q , but that the proportionality constant varies considerably with direction and mode index and that the proportionality does not extend far from $q=0$.

Let us now try to find a simple relation between W_{ep}^p and the electron-phonon coupling parameter. The similarities and differences between these two quantities are most evident if we write

$$\gamma_j^{ep}(q) = \frac{2}{3} \pi \hbar^2 N \omega_j(q) \alpha_j^2(q), \quad (4.3)$$

where $\alpha_j^2(q)$ is a quantity with the same dimensions as ω which measures the amount of electron-phonon coupling arising from the phonon having momentum q and mode index j . Expressing (1.1) and (4.1) in terms of integrals over constant-frequency

surfaces, we obtain

$$\lambda = \frac{\frac{2}{3} \Omega_a}{(2\pi)^3} \int \frac{d\omega}{\omega} \sum_i \int_{\omega=\omega_j(q)} \frac{dS_a}{|u_j|} \alpha_j^2(q), \quad (4.4)$$

and

$$(W_{ep}^p)^{-1} = \frac{3k}{2(2\pi)^4 \hbar N} \int \frac{d\omega}{\omega} \left(\frac{\hbar \omega}{kT} \right)^2 \frac{e^{\hbar \omega / kT}}{(e^{\hbar \omega / kT} - 1)^2} \times \sum_j \int_{\omega=\omega_j(q)} \frac{dS_a}{|u_j|} \frac{u_{jx}^2}{\alpha_j^2(q)}. \quad (4.5)$$

A. Constant α^2

In order to relate (4.4) and (4.5), we need to make an assumption about $\alpha_j^2(q)$. From superconducting tunneling experiments⁴⁸ one can extract a function

$$\alpha^2(\omega) F(\omega) \equiv \frac{1}{3} \sum_{jq} \alpha_j^2(q) \delta(\omega - \omega_j(q)),$$

which turns out to be approximately proportional to $F(\omega)$. This indicates that $\alpha_j^2(q)$, when averaged over phonon modes with a given frequency ω , is approximately a constant. In the following we will make the more restrictive assumption that $\alpha_j^2(q)$ is constant. Using this assumption in (4.4) and (4.5) yields

$$\lambda = \frac{2}{3} \alpha^2 \sum_j \int [F_j(\omega) / \omega] d\omega = 2\alpha^2 \langle \omega^{-1} \rangle \quad (4.6)$$

and

$$(W_{ep}^p)^{-1} = \frac{9}{\alpha^2} \frac{k}{4\pi \hbar N \Omega_a} \frac{1}{3} \int \frac{d\omega}{\omega} \left(\frac{\hbar \omega}{kT} \right)^2 \times \frac{e^{\hbar \omega / kT}}{(e^{\hbar \omega / kT} - 1)^2} \langle u^2 \rangle_{\omega} F(\omega), \quad (4.7)$$

where $\langle u^2 \rangle_{\omega}$ is defined by

$$\langle u^2 \rangle_{\omega} = \left(\sum_j \int_{\omega=\omega_j} \frac{dS_a}{|u_j|} u_{jx}^2(q) \right) / \left(\sum_j \int_{\omega=\omega_j} \frac{dS_a}{|u_j|} \right). \quad (4.8)$$

The factor $\frac{1}{3}$ in (4.7) arises from setting $\langle u^2 \rangle_{\omega} = \frac{1}{3} \langle u^2 \rangle_{\omega}$, and is strictly valid only for cubic systems. For low T (4.7) becomes

$$(W_{ep}^p)^{-1} = 9k \langle u^2 \rangle_0 (T / \Theta_D)^2 J_3(\infty) / (4\pi \hbar N \Omega_a \omega_D \alpha^2), \quad (4.9)$$

where ω_D is defined in terms of the low-frequency behavior of $F(\omega)$ by

$$F(\omega) = \frac{1}{3} \sum_j F_j(\omega) = \lim_{\omega \rightarrow 0} (\omega \rightarrow 0) \frac{3\omega^2}{\omega_D^3}$$

and $\Theta_D = \hbar\omega_D/k$.

$J_3(x)$ is defined by $J_3(x) = \int_0^x dy y^3 e^y / (e^y - 1)^2$ and $J_3(\infty) = 7.21$, while $J_3(x \rightarrow 0) = \frac{1}{2}x^2$. Thus $W_0 [\equiv \lim_{T \rightarrow 0} (W_{ep}^p T^2 / \Theta_D^2)]$ can be written

$$W_0 = [2\pi\hbar N \Omega_a \omega_D \langle \omega^{-1} \rangle^{-1} / 9J_3(\infty) k \langle u^2 \rangle_0] \lambda. \quad (4.10)$$

Using the Debye value for $\langle u^2 \rangle_0$,

$$\langle u^2 \rangle_0 = \omega_D^2 / q_D^2 = \omega_D^2 (\Omega_a / 6\pi^2)^{2/3} \quad (4.11)$$

in (4.10) yields a relation between W_0 and λ in the constant- α^2 approximation:

$$W_0 = 0.7 \frac{\text{K cm}}{\text{W}} \left(\frac{\langle \omega^{-1} \rangle^{-1}}{\omega_D} \lambda \right) \Omega_a^{1/3} (\text{\AA}) N (\text{states/eV spin}) \quad (4.12)$$

The predictions of (4.12) are compared with experiment in Table I and Fig. 1. Note that $\Omega_a^{1/3}$ is measured in \AA and N in states/eV spin.

For high temperature (4.7) becomes

$$W_\infty^{-1} = (3k \langle u^2 \rangle \langle \omega^{-1} \rangle) / (4\pi\hbar N \Omega_a \alpha^2), \quad (4.13)$$

where

$$\langle u^2 \rangle = \int \frac{d\omega}{\omega} \langle u^2 \rangle_\omega F(\omega) / \int \frac{d\omega}{\omega} F(\omega). \quad (4.14)$$

In this approximation W_0 and W_∞ are related by

$$W_\infty = W_0 21.63 \langle u^2 \rangle_0 / \langle u^2 \rangle (\langle \omega^{-1} \rangle^{-1} / \omega_D). \quad (4.15)$$

For W, Mo, Ta, and Fe data are available on $W_{ep}^p(T)$ for intermediate temperatures. In order to use this data to estimate W_0 , we employ (4.7) and assume $(W_{ep}^p)^{-1}$ to vary with temperature as $X(T) = \int d\omega F(\omega) \langle u^2 \rangle_\omega G(\hbar\omega/kT) / \omega$, where $G(x) = x^2 e^x / (e^x - 1)^2$. $X(T)$ was evaluated using force constants fit to the experimental phonon dispersion curves.

B. Other measures of $\gamma_j^{ep}(q)$

It is clear from Eq. (4.1) that W_0 is determined by an average over the low-frequency phonon modes of the phonon lifetime $[\hbar/\gamma_j^{ep}(q)]$, while λ is an average over all modes of $\gamma_j^{ep}(q)/\omega_j^2(q)$. There are experiments that yield more detailed measures of $\gamma_j^{ep}(q)$ than λ . The most detailed information can be obtained from inelastic neutron scattering. In principle, $\gamma_j^{ep}(q)$ can be determined for any phonon mode by measuring at low temperature the energy width of the phonon in question. In practice, however, this type of experiment is difficult and cost-

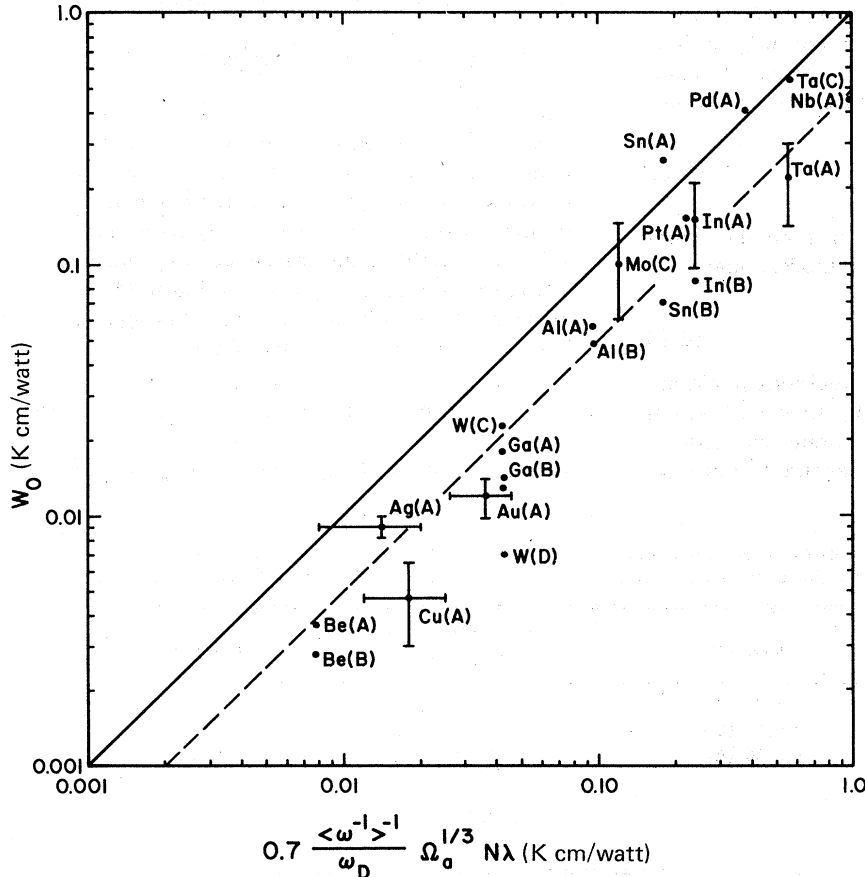


FIG. 1. Experimental values of W_0 compared with theoretical values calculated in the $\alpha^2 = \text{const.}$ approximation. The absence of error bars for some points indicates the authors' inability to reliably estimate the bounds of the probable errors. See Table I for meanings of (A, B, C, D). The solid line indicates the result of Eq. (4.12). The dashed line is $\frac{1}{2}$ of this value.

ly. A reasonably reliable value of $\gamma_j^{ep}(q)$ is known for one low- q mode in Nb.³ $\alpha_j^2(q)$ for this mode is 8.4 meV, compared with a low- q average of $1/\alpha_j^2(q)$ obtained from W_0 using (4.7) of $\langle 1/\alpha_j^2(q) \rangle^{-1}$

$$= \alpha^2 = 3.3 \text{ meV.}$$

Somewhat less-detailed information can be extracted from superconducting tunneling experiments, which yield a function $\langle \alpha^2 \rangle_\omega F(\omega)$, where

$$\langle \alpha^2 \rangle_\omega = \left(\sum_j \int_{\omega=\omega_j(q)} \frac{dS_q}{|u_j(q)|} \alpha_j^2(q) \right) / \int_{\omega=\omega_j(q)} \frac{dS_q}{|u_j(q)|}. \quad (4.16)$$

Unfortunately these experiments do not measure the electron-phonon coupling to frequencies below 1 or 2 meV. Nevertheless, we compare in Table II values of $\langle \alpha^2 \rangle_\omega$ for the lowest measured frequencies available from tunneling⁴⁸ with values of $\langle 1/\alpha_j^2(q) \rangle_{\omega \rightarrow 0}^{-1}$ derived from (4.9) for Sn, In, and Ta. In these three cases, α^2 from tunneling at low ω is in good agreement with α^2 calculated from λ using (4.6).

C. Klemens' formula

Klemens⁴⁹ derived a simple formula that relates W_{ep}^e to the electron-phonon component of the electronic thermal resistivity W_{ep}^e . Since W_{ep}^e varies as T^2 at low temperatures, let us define a temperature-independent quantity which gives a convenient measure of W_{ep}^e :

$$W_0^e \equiv \lim_{T \rightarrow 0} W_{ep}^e (\Theta_D / T)^2. \quad (4.17)$$

In terms of this quantity Klemens' formula, which was derived in the free-electron model, may be written

$$W_0 / W_0^e = Z^{4/3} / 313, \quad (4.18)$$

where Z is the number of free electrons per atom. We shall conclude this section by showing that our expression for W_0 (4.12) is consistent with Klemens' formula (4.18) and by generalizing his re-

sult to the case of general band structures.

One can obtain^{50,51} an approximate variational solution to the Boltzmann equation by assuming a trial solution for the deviation function of the form $c \epsilon_k v_k$, where ϵ_k is the electronic energy measured from the Fermi energy, v_k is the Fermi velocity, and c is a variational parameter.

The electronic thermal resistivity is given in this approximation by^{52,53}

$$W_{ep}^e = 9 \lambda_w(T) / \hbar N \Omega_a^{-1} \langle v^2 \rangle \pi k, \quad (4.19)$$

where $\langle v^2 \rangle$ is the mean-square Fermi velocity and $\lambda_w(T)$ is given (at low temperature) by

$$\lambda_w(T) \simeq \frac{12}{\pi^2} \int_0^\infty \frac{d\omega}{\omega} \alpha^2 F(\omega) \left(\frac{x}{\sinh x} \right)^2 x^2, \quad (4.20)$$

with $x = \hbar \omega / 2kT$.

It should be noted that Klemens⁵⁴ has shown by numerical solution of the Boltzmann equation that this lowest-order variational approximation overestimates the thermal resistivity by a factor of 1.5, thus (4.19) should be reduced by this factor. Using the $\alpha^2 = \text{const.}$ approximation in (4.20), substituting the result in (4.19), and taking account of Klemens' factor of 1.5, one obtains

$$W_0^e = 217 \alpha^2 / k \hbar N \Omega_a^{-1} \langle v^2 \rangle \omega_D. \quad (4.21)$$

One can also use (4.4) to relate W_{ep}^e to λ :

TABLE II. Values of the electron-phonon coupling parameter α^2 derived from lattice thermal conductivity, low ω tunneling, and from λ . See Table I for meaning of (A,B,C).

Element	From W_0	α^2 (meV) (per mode)	
		from tunneling	From λ
Sn	1.6 (A)	1.31	1.14
	0.42 (B)		
In	0.60 (A)	0.94	0.96
	0.34 (B)		
Ta	0.60 (A)	2.1	1.52
	1.5 (C)		

$$W_0^e = 108\lambda/k\hbar N\Omega_a^{-1}\langle v^2 \rangle \omega_D \langle 1/\omega \rangle. \quad (4.22)$$

Finally, we obtain our generalization of Klemens' formula by dividing (4.10) by (4.22):

$$W_0/W_0^e = N^2 \langle v^2 \rangle \hbar^2 \omega_0^2 / 1119 \langle u^2 \rangle_0. \quad (4.23)$$

The equivalence of (4.23) and (4.18) for the special case of free electrons can be seen by substituting from (4.11) for $\langle u^2 \rangle_0$ and by using the free-electron result

$$N^2 \hbar^2 \langle v^2 \rangle = 4\Omega_a^{2/3} Z^{4/3} (3\pi^2)^{4/3} (4\pi^2)^{-2}.$$

The validity of (4.18) has been verified for some simple and noble metals.^{17,38} It should be remembered, however, that the ratio W_0/W_0^e is very sensitive to one's choice of Θ_D . A systematic study of the validity of (4.22) or (4.23) is hampered at present by our lack of knowledge of mean-square Fermi velocities, but this situation should improve in the near future as more band calculations are performed. We urge band theorists to calculate $\langle v^2 \rangle$ as a matter of course, since it is just as important for transport as $N(E_F)$.

V. DISCUSSION

The experimental estimates of $W_0 \equiv \lim_{T \rightarrow 0} [W_{ep}^b(T/\Theta_D)^2]$ are compared with Eq. (4.12) in Fig. 1. Values for the bare electronic density of states were derived from electronic specific-heat coefficients^{19,55} in the usual way by dividing the enhanced density of states by $1 + \lambda$. For Pt and Pd it is necessary to allow for spin-fluctuation and Coulomb mass enhancements which do not contribute to W_0 . We assume that the total mass enhancement for Pd is 0.7 and for Pt 0.6. These values bring band calculations into agreement with electronic specific-heat measurements.^{56,57} The value of 0.7 for Pd is also in agreement with an estimate based on high-temperature specific-heat measurements.⁵⁸

Values of the electron-phonon coupling parameter were derived from the Allen-Dynes modification⁵⁹ of McMillian's formula⁶⁰ which relates λ and T_c ,

$$T_c = \frac{\langle \omega \rangle_{\log}}{1.2} \exp[-1.04(1 + \lambda)/\lambda - \mu^*(1 + 0.62\lambda)], \quad (5.1)$$

for the superconducting elements and from calculations based on resistivity measurements for the noble metals⁶¹ and Pd and Pt.^{47,62} Values of $\langle \omega^{-1} \rangle$ were calculated from force-constant fits to inelastic neutron scattering data where this data was available, and in cases where it was not available $\langle \omega^{-1} \rangle$ was estimated from superconducting tunneling data (Sn, In) or was estimated to be $(0.6\omega_D)^{-1}$ (Be, Ga).

Figure 1 shows that Eq. (4.12) provides a reasonable description of the interelement variation in W_0 . This is not a trivial accomplishment, since the data span nearly three decades. It appears, moreover, that Eq. (4.12) overestimates W_0 by a factor that varies from element to element and is somewhat obscured by the experimental scatter, but which is of the order of 2. The constant- α^2 approximation upon which (4.12) is based can fail in either or both of two ways: (i) $\langle \alpha^2 \rangle_\omega$ could depend strongly on ω with electron-phonon coupling being stronger or weaker than average at low frequencies. This effect could lead to either an overestimate or an underestimate of W_0 . This effect may be important in some materials; however, superconducting tunneling experiments indicate that $\langle \alpha^2 \rangle_\omega$ for many materials has rather weak frequency dependence. (ii) A more likely explanation for the factor of 2 overestimate of W_0 in the constant- α^2 approximation is the \hat{q} and mode dependence of $\alpha_j^2(q)$. W_0 is proportional to $(\langle u^2/\alpha^2 \rangle_{\omega \rightarrow 0})^{-1}$, where the angular brackets denote an average over q and modes for a given value of ω ; however, this quantity is approximated in the constant- α^2 approximation by $(\langle u^2 \rangle)^{-1} \langle \alpha^2 \rangle$. This "anisotropy" effect will almost always lead to an overestimate of W_0 .⁶³

There seem to be significant consistent differences in the results of the different experimental techniques for measuring W_0 . The magnetothermal resistance techniques (B, D) seem to yield consistently lower results for W_0 than the alloying techniques (A, C). This may indicate fundamental difficulties with the magnetothermal technique, or alternatively, significant phonon scattering from dislocations in the alloying technique.

Comparing alloying techniques, the intermediate temperature results (C) seem to be higher than those obtained at low temperature. We have tried to take the effects of phonon dispersion into account in obtaining W_0 from the intermediate temperature results. The discrepancies between the intermediate-temperature and low-temperature derived values for W_0 (if they are not due to experimental inaccuracies) may arise from $\langle \alpha^2 \rangle_\omega$ being greater at the higher frequencies sampled by the intermediate-temperature techniques. The intermediate-temperature lattice thermal resistance may also be enhanced by the presence of normal phonon-phonon scattering. This type of scattering does not contribute directly to the lattice thermal resistance, but it can contribute indirectly by scattering phonons from modes with low $\alpha_j^2(q)$ into modes with higher $\alpha_j^2(q)$. In this way normal phonon-phonon scattering can eliminate the effect of anisotropy in $\alpha_j^2(q)$ and make the constant- α^2 approximation more nearly exact. It is interesting

that the intermediate-temperature derived values of W_0 lie quite near the constant- α^2 prediction. The analogous effect of normal phonon-phonon processes on the umklapp lifetime has been discussed by Callaway⁶⁴ and Guyer and Krumhansl.⁶⁵

For Fe we were unable to find an independent estimate of λ . The observed value of W_0 implies a λ of 0.17 if one assumes α^2 is constant. This is a surprisingly small value.

Turning to individual elements, the discrepancies between the different techniques for determining W_0 seem especially large for W and Sn. The intermediate-temperature magnetothermal value for W seems rather low to us. For Sn the discrepancies are somewhat worse. In this case the alloy data may also need scrutiny. Points that fall below the theoretical curve can be understood in terms of anisotropy in α^2 ; however, it is more difficult to understand points that lie substantially above the α^2 =constant prediction.

An obvious way to improve the experimental values would be to concentrate on a few metals, applying all of the applicable experimental methods to systematically define and resolve the differences for these cases. Tungsten is a good candidate because magnetoconductivity values are already available at low and intermediate temperatures. Low-temperature alloy studies are needed, and the intermediate-temperature values should also be supplemented because theory suggests that the

$N\lambda$ product may be altered by alloying and only binary alloys were used. An obvious remedy for this would be to use isoelectronic ternary alloys for cases in which the rigid band theory is believed to be valid, and to supplement the conductivity measurements with specific-heat data and a determination of λ . Ternary, isoelectronic Nb, and Ta base alloys could also provide useful information, since the alloy and superconductivity techniques could be further compared in these two cases. This, of course, presumes that the normal state κ_p of fairly dilute ternary alloys could be directly determined, and it should be noted that this could not be done in the earlier work (Refs. 26 and 27). An advantage here is that both λ and N can be more easily determined in these high T_c alloys than in W base materials.

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