[DDI: 10.24214/jcbps.C.8.3.29606]

Journal of Chemical, Biological and Physical Sciences

An International Peer Review E-3 Journal of Sciences Available online atwww.jcbsc.org

Section C: Physical Sciences

CODEN (USA): JCBPAT

Research Article

The Existence of Anharmonic Effect due to Phonon-Phonon Interactions and its Contribution to Specific Heat of Materials.

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Received: 02 July 2018; Revised: 19 July 2018; Accepted: 26 July 2018

Abstract: Previous researchers have revealed the anharmonicity of the lattice waves to cause deviation in specific heat from Debye T^3 law. The energy spectrum of phonons can not be fully described by the Debye law. For anharmonicity of lattice waves, this contribution must be expected for metals and insulators also. Specific heat of metals, viz, Al, Cu, Ag, Ni, Fe, Pd and Pt was identified with the electronic contribution. Later experiment revealed the linear term in specific heat of almost all metals, at low temperatures. But intensive study of the transport properties namely thermoelectric properties, thermal and electrical conductivities of metals at low temperatures finds deviation of the electrons from the free electron model. The light of the recent development in this regard the earlier notion of linear temperature dependent term of specific heat appears to be doubtful, because electrons may not. Debye did not take into account the anharmonicity of the lattice wave in his theory of specific heat of solids at low temperatures.

Keywords: Thermal Conductivity, Anharmonic effect, Thermoelectric Power, Electron diffusion, Phonon drag. Wiedmann-Franz law.

²⁹⁶ J. Chem. Bio. Phy. Sci. Sec. C, May 2018 – July 2018, Vol. 8, No. 3; 296-306; [DDI: 10.24214/jcbps.C.8.3.29606]

INTRODUCTION

Thermal conductivity and phonon interaction are intimately related to the specific heat of electron. Moreover, thermoelectric properties give details of the nature of the charge carriers as well. Thus the study of electrical resistivity, thermal conductivity and thermoelectric properties would give significant information regarding the existence of anharmonic effect due to phonon-phonon interactions and its contribution to specific heat at T< θ /10. Hence we would like to study the thermal conductivity and thermoelectric properties of some metals Viz. Au, Ag, Cu, Na, Li, Zn, Fe, Al, Pt, Ni and Co at T< θ /10 where θ being the Debye-temperature for a metal.

RESULTS AND DISCUSSION

Transport of heat in a solid is due to conduction electrons and thermally excited phonons. In metals, electronic component solely dominates as the lattice component is supposed to be vanishingly small. The thermal conductivity K, for a simple metal at low temperatures (T< $\theta/10$) is written as Bloch¹, Wilson², Sondheimer³.

$$K = T/(\beta + bT^3)$$
(1)

Which may be rearranged to,

$$\frac{1}{\kappa} = \beta/T + bT^3 \tag{2}$$

Where β and b are constants for a specimen of a metal. The first term in the right hand side of equation (2) is the residual thermal resistivity caused by impurities and geometrical factors of the specimen and the second term is the ideal thermal resistivity due to lattice vibrations. On the basis of free electron gas theory, equation (1) may be understood as,

$$K = \frac{1}{3} n C_{ve} V \lambda$$
(3)

Where n = number of conduction electrons,

 $C_{ve} = specific heat of electrons,$

V = average velocity of the electrons.

 λ = mean free path of the electrons.

n and v are practically constant in noble and alkali metals.

$C_{ve} \propto T$ [Sommerfeld⁴]

and $\lambda \propto T^3$ (in pure metal) as the density of thermally excited phonons which scattered conduction electrons varies as T^3 according to Debye T^3 law at low temperatures (T< $\theta/_{10}$). In the presence of impurities in the specimen of the metal, the conduction electrons are scattered by impurities and

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In equation (1) and (2), b is expressed as,

$$b = 95.3 \text{ (n/n_a)}/K_{\infty}.\theta^2$$
and $\beta = \rho_o/L_n$
(4)

Where n/n_c = density of conduction electrons per atom.

 K_{∞} = thermal conductivity at T $\rightarrow \infty$

$$\theta$$
 = Debye- temperature

 ρ_o = residual electrical resistivity

and $L_n = normal$ Lorentz number.

An excellent agreement between the observed and calculated values of β is advocated, but not in the values of b [Berman and Mac Donald⁵, Hulm⁶]. The calculated value of θ is too much larger than the observed value [Wilson²]. These discrepancies point out the short commings in the assumptions that

- (i) the electrons are free even at very low temperatures.
- (ii) the transport of heat in metals is solely by conduction electrons.
- (iii) no phonon other than those obeying Debye T³-law is available for scattering the conduction electrons, on which the two term equation of K is based. However, a three term empirical equation,

$$\mathbf{K} = \mathbf{T} / \left(\beta + \boldsymbol{\delta} \mathbf{T}^2 + \mathbf{b} \mathbf{T}^3 \right)$$

is found to be more competent.

According to Schriempf⁷ (1968), the term δ T² is due to electron-electron scattering which leads to a T²-term in electrical resistivity also. β , δ and b are constants. Equation (5) may be obtained from the following considerations.

Thermal resistivity

$$\frac{1}{\kappa} = \rho_o / (L_n T) + \rho_i / (L_n T) + \Delta \rho / (L_n T)$$
(6)

Which has been based on the validity of Widemann-Franz law i.e.

$$K/\sigma = L_nT$$

 σ is the electrical conductivity. At low temperatures, electrical resistivity ($\rho = \frac{1}{r}$) may be expressed as,

$$\rho_T = \rho_o + \rho_i + \Delta \rho$$

(7)

(5)

Where ρ_o = residual electrical resistivity

 ρ_i = ideal electrical resistivity.

And $\Delta \rho$ = resistivity due to electron-electron scattering.

Equation (6) may be written as,

$$\omega = \omega_{0} + \omega_{i} + \Delta \omega \tag{8}$$

Where ω = thermal resistivity and $\Delta \omega$ is the thermal resistivity due to electron-electron scattering. For $\Delta \omega = 0$, from equations (2) and (8) we get,

$$\boldsymbol{\omega}_{\rm o} + \boldsymbol{\omega}_{\rm i} = \beta/T + bT^2 \tag{9}$$

Equation (6) may now be written, as

 $T/_{K} = \beta + bT^{3} + \Delta \rho / L_{n}$

Since $\Delta \rho \propto T^2$; $\Delta \omega = \Delta \rho / L_n = \delta T^2$ (say) which leads to

 $T_{K} = \beta + bT^{3} + \delta T^{2}$. This is equation (5).

If we express $\Delta \rho = AT^2$, then $AT^2 = L_n \delta T^2$ i.e. $A = L_n \delta$ (10)

Equation (10) may be used to examine the contention of electron-electron scattering in thermal conductivity.

The present study finds the following equations fit to the observed thermal conductivity data [Ho, Powell and Liley⁸] in the temperature zone T< $\theta/_{10}$, b and δ have been obtained from the slope and intercept of the curve (T/_K- β) Vs T on the basis of least square method. The correlation factor γ for the linear relationship, listed in **Table 1** is very satisfactory in each case. β is taken from the literature [Ho, Powell and Liley⁸]. Value of β , b and δ are reported in the table.

A1 : $K = T/(0.0243 - 0.26 \times 10^{-5} T^2 + 1.883 \times 10^{-5} T^3)$	
(2 < T < 20 K)	
Standard Deviation 1.3 %	
Cu : K = T/ $(0.0237 - 3.49 \times 10^{-5} \text{ T}^2 + 2.146 \times 10^{-5} \text{ T}^3)$	(12)
(2 < T < 20 K)	
Standard Deviation 0.996 %	
Ag : $K = T/(0.0254-4.309 \times 10^{-5} T^2 + 3.853 \times 10^{-5} T^3)$	(13)
(2 < T < 14 K)	
Standard Deviation 1.84 %	
Au : K = T/ $(0.188-15.703 \times 10^{-5} \text{ T}^2 + 13.483 \times 10^{-5} \text{ T}^3)$	(14)
(2 < T < 20 K)	
Standard Deviation 2.47 %	
Na : K = T/ (0.06-24.267×10 ⁻⁵ T ² + 41.482×10 ⁻⁵ T ³)	(15)
(2 < T < 30 K)	

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	Standard Deviation 1.73 %	
$K: K = T/(0.082-33.482 \times 10^{-5} T^2 - 10^{-5} T^2)$	$+ 218.06 \times 10^{-5} \text{ T}^{3}$	(16)
	(2 < T < 10 K)	
	Standard Deviation 2.4 %	
Li : K = T/ (1.52-25.11×10 ⁻⁵ T ² + 2	$16.642 \times 10^{-5} \text{ T}^3$	(17)
	(2 < T < 20 K)	
	Standard Deviation 1.108 %	
Pt : K = T/ (0.433-43.85×10 ⁻⁵ T ² +	42.741×10 ⁻⁵ T ³)	(18)
	(2 < T < 16 K)	
	Standard Deviation 0.8 %	
Fe : K = T/ (0.585-0.2633×10 ⁻⁵ T ²	$+ 8.876 \times 10^{-5} T^{3}$)	(19)
	(2 < T < 20 K)	
	Standard Deviation 0.4 %	
Co : K = T/ $(3.71-15.173 \times 10^{-5} \text{ T}^2 - 10^{-5} \text{ T}^2)$	+ $10.873 \times 10^{-5} \text{ T}^3$)	(20)
	(6 < T < 20 K)	
	Standard Deviation 0.57 %	
Ni : K = T/ (0.46-1.985×10 ⁻⁵ T ² + 2	9.372×10 ⁻⁵ T ³)	(21)
	(2 < T < 20 K)	
	Standard Deviation 1.55 %	
Zn : K = T/ (0.0525-165.73×10 ⁻⁵ T	$T^{2} + 32.64 \times 10^{-5} \text{ T}^{3}$	(22)
	(8 < T < 20 K)	
	Standard Deviation 0.56 %	

Metals	Correlaions	Slope × 10 ⁻⁵	Intercept × 10 ⁻⁵	Temperature Zone
	Factor (γ)			in K
Li	0.9997	15.9599	1.4889	$5 \leq T \leq 25$
Na	≈ 1.000	35.1878	0.0599	$3 \le T \le 6$
К	0.9985	149.9239	0.8606	$10 \le T \le 18$
	0.9998	218.7000	0.0778	$2 \le T \le 9$
Cs	0.9964	249.9739	16.4682	$14 \le T \le 20$
Mg ₁	0.9999	8.0673	0.0967	$5 \leq T \leq 25$
Mg ₂	0.9999	8.3887	0.7700	$6 \le T \le 25$
Be	0.9993	1.0045	-0.0797	$4 \le T \le 100$
Cu	0.9996	1.8509	0.0231	3 ≤ T ≤ 13
Ag	0.9988	3.9717	0.0211	$5 \le T \le 16$
Au	0.9994	13.3883	0.1727	$5 \leq T \leq 20$

300 J. Chem. Bio. Phy. Sci. Sec. C, May 2018 – July 2018, Vol. 8, No. 3; 296-306; [DDI: 10.24214/jcbps.C.8.3.29606.]

В	0.9994	5.1380	6.0016	$35 \le T \le 50$
Ge	0.9982	8.9100	0.5934	$15 \le T \le 25$
S	0.9908	10329.3700	19.1283	$5 \le T \le 20$
	0.9998	25.8122	-0.0632	$8 \le T \le 25$
Zn	0.9946	9.4093	0.0511	$2 \le T \le 6$
Th	0.9996	137.6804	0.8801	$5 \le T \le 20$
Sb	0.9974	91.7308	1.3228	$8 \le T \le 20$

In all the above equations from (11 to 22); T^2 term exists uniformly with -ve sign. Here (n/n_a) has also been calculated from equation (4) using the values of b in **Table 2.** For each metal, the value of (n/n_a) is much smaller than the expected value (for noble and alkali metals, n/n_a \approx 1)

Table – 2: Parameters β , δ and b of three term equations of Thermal conductivity of metals at low temperatures.

Metals	β cm. deg. w ⁻¹	$\delta \times 10^4$ (cm.w ⁻¹)	$b \times 10^4 (cm.w^{-1} deg^{-1})$
Na	0.06	-2.4267	4.1482
Cu	0.0237	-0.3490	0.2146
Pt	0.433	-4.3849	4.2741
Ni	0.46	-0.1985	0.9372
Pb	0.0353	-128.81	59.102
Zn	0.0525	-16.573	3.2636

These results stand against the contention of electron-electron scattering leading to resistivity and quasi free conduction electron model at very low temperatures.

The thermo-electric power of metals (viz Au, Ag, Cu, Pt, Zn) have been widely studied by several workers, [Blatt ⁹Bernard^{10,} Ziman¹¹; Lawrence ¹², Rumbo¹³; Mac Donald ¹⁴; Guenaultand Hawksworth¹⁵; Poddar and Ranju¹⁶] at low temperatures. The main findings of these studies may be summarised as-

- (i) a peak in thermoelectric power,
- (ii) the variation of thermoelectric power as T^3
- (iii) negative in character for noble metals viz. Ag, Au and Cu,
- (iv) straining of the specimen suppresses the T^3 component of thermo-electric power and
- (v) on alloying the metal, the peak is attenuated.

The peak in thermopower and T^3 – variation are interpreted as due to the phonon drag effect. This phenomenon may be understood as follows-

(26)

The phonons at low temperatures, if they find nothing else to collide with (including themselves) must always collide ultimately with conduction electrons, thus imparting their momentum to the electrons and giving rise to thermoelectric force. The presence of impurity in the sample reduces the probability of phonon-electron collision and hence reduces the thermoelectric force. The phonon-drag thermo-electric power S_g is written as S_g = 1 / (3 N_e S_g) C_g where C_g = lattice specific heat per unit volume. N = number of conduction electrons per unit volume. Since C_g \circ CT³ (According to Debye)

Hence $S_g \propto T^3$

Alloying attenuates the lattice thermal conductivity which causes attenuation in the peak of phonon drag. The –vethermopower has been interpreted as due to electron-electron and electron-phonon scattering. These explanations however have been criticized by several workers [Guenaultand Hawksworth¹⁵; Lawrence¹²; Bernard ¹⁰; Poddar and Ranju¹⁶]. Guenault and Hawksworth¹⁵ suggested a crossover from dirty regime at low temperatures towards a pure regime at higher temperatures for the change of sign of the thermopower. A change of scattering mechanism is expected to give a change in thermopower, whether S arises from electron diffusion or phonon drag. One should, therefore expect a change in the form of thermopower between the lowest temperature (where electron-impurity scattering dominates) and the region above about 10K (where electron-phonon scattering dominates). The temperature T* around which the cross-over occurs may be estimated as follows. For a single band approximation, the thermopower of a metal at temperature T is given by,

$$\mathbf{S} = (\mathbf{S}_{o}\boldsymbol{\omega}_{o} + \mathbf{S}_{T}\boldsymbol{\omega}_{T})/\boldsymbol{\omega}_{o} + \boldsymbol{\omega}_{T})$$
(23)

Where ω_{o} and ω_{T} are the thermal resistances due to residual and phonon scattering respectively; S_{o} and S_{T} are the characteristic thermopowers for the two scattering mechanisms. Clearly the cross-over from S_{o} to S_{T} occurs at temperature T* at which $\omega_{o} \approx \omega_{T}$. Using the method of Guenaultand Howksworth¹⁵. T* is obtained as

$$T^{*3} = 300 \theta^2 / CRR$$
 (24)

Where θ is the Debye temperature; R R is the resistance ratio i.e. $\rho_{300k}/\rho_{4.2K}$ and C is defined by the equation

$$\boldsymbol{\omega}_{\mathrm{T}} = \mathrm{C}\mathrm{T}^{2}\boldsymbol{\omega}_{\infty}/\theta^{2} \tag{25}$$

Where ω_{∞} is thermal resistivity at T $\rightarrow \infty$.

From equations (24) and (25), we get.

$$T^* = (300T^2 \omega_{\infty} \rho_{4,2}) / \omega_{T,} \rho_{300}$$

At 300 K, we may take $\omega_{300} \approx \omega_{\infty}$ for simple metals as thermal conductivity remains constant.

At this temperature Wiedmann-Franz law holds good i.e.

 $K/\tau = L_nT$

J. Chem. Bio. Phy. Sci. Sec. C, May 2018 – July 2018, Vol. 8, No. 3; 296-306;
 [D0l: 10.24214/jcbps.C.8.3.29606.]

This equation gives $\omega_{\infty}/\rho_{300} = 1/(300L_n)$

And so, $T^{*3} = (300T^2) / \omega T) \rho_{4,2} 1/300L_n = (T^2 \rho_{4,2}) / (\omega_T L_n)$ (28)

In a normal pure metal, at T= 4.2K the residual thermal resistivity is much greater than that due to lattice vibrations and hence we may take $\omega_{4,2} \approx \omega_0$, the residual resistivity. Also at very low temperatures, for the residual resistivity, the Wiedmann-Franz law holds well [Bermann and Mac Donald⁵; Hulm⁶].

i.e.
$$\rho_{4,2} = \omega_{4,2} L_n (4.2) \approx \omega_0 L_n (4.2).$$

At low temperatures (T < $\theta/_{10}$), the two term equation for thermal- conductivity is written as,

$$K = 1/\left(\beta + bT^3\right) \tag{29}$$

for $\beta >> bT^3$, this equation reduces to

$$1/K = \beta/T = \omega_o$$

At T = 4.2 K, we have

 $\beta/4.2 = \omega_{4.2} \approx \omega_{o}$

i.e. $\beta = 4.2 \omega_{o}$

So, equation (28) may be written as

 $\mathbf{T}^{*3} = (\mathbf{T}^2 \boldsymbol{\omega}_{\mathbf{o}} \times 4.2) / \boldsymbol{\omega}_{\mathbf{T}} = \beta \mathbf{T}^2 / \boldsymbol{\omega}_{\mathbf{T}}$ (30)

At $T^*, \omega_0 \approx \omega_T$ which is discussed above. Hence we may write from equation (29)

 $1/K^{*} = \boldsymbol{\omega}_{o} + bT^{*2} = 2bT^{*2}$ [Since $\boldsymbol{\omega}_{o} = \boldsymbol{\omega}_{T^{*}} = bT^{*2}$]
or, $\boldsymbol{\omega}_{T^{*}} = 1/K^{*} = 2bT^{*2}$ Here $\boldsymbol{\omega}T^{*}$ and K^{*} are the values of $\boldsymbol{\omega}_{T}$ and K at T^{*} . With this value of $\boldsymbol{\omega}_{T}^{*}$, equation (30) gives T^{*3} $=T^{*2}/(2b^{T*2}) \beta = \beta/2b$ or, $T^{*} = \sqrt[5]{(\beta/2b)}$ (31)
From equation (30); T_{opt} at which K_{nax} (the maximum thermal-conductivity) occurs, may be obtained as

 $T_{opt} = \sqrt[8]{(\beta/2b)}$ (32)

[from the condition, dK/dT = 0. $d^2K/dT^2 < 0$ at $T = T_{opt}$] From equations (31) and (32), it is clear that $T^* = T_{opt}$ (33) Equations (31), (32) and (33) imply that T_{opt} and T^* increase with β and decrease with b. As the metal becomes more and more impure, β increases more and the regime of impurity extends towards higher value of T^* . This is in confirmity with the contention of Guenault and Hawksworth¹⁵. On alloying the metal the amplitude of lattice vibrations lowers down and hence an attenuation in the value of b is expected and it would lead to an increase in T^* . This conclusion supports the results reported by Guenault and Hawksworth¹⁵;Poddar and Ranju¹⁶.

The –vethermopower revealed in noble metals may not be limited to these metals only but it may be in other metals also as ($T^* = T_{opt}$) appears in the thermal conductivity temperature curve (i.e. K Vs T) for almost all metals [Ho, Powell and Liley⁸]. The phonon-drag peak in thermopower should occur at the sane temperature at which thermal conductivity peak occurs at $T^* = T_{opt}$; and T^* and T_{opt} are the functions of the same variables β and b. The peak in thermopower is interpreted as due to phonon-drag and we may, ultimately, reach the conclusion that the thermal conductivity peak is also due to phonon drag. The phonon-drag contribution dominates over the electron-diffusion contribution below T^* and hence the dominance of phonon-drag contribution to thermal- conductivity may clearly be expected. It means that the phonons are in thermal equilibrium in metals even at very low temperatures. This is in conformitywith the claims of Klemens¹⁷ that the anharmonicity of lattice vibrations is responsible for maintaining the thermal equilibrium in phonon system. In this regards, it also supports the reports by Tosi and Fumi¹⁸.

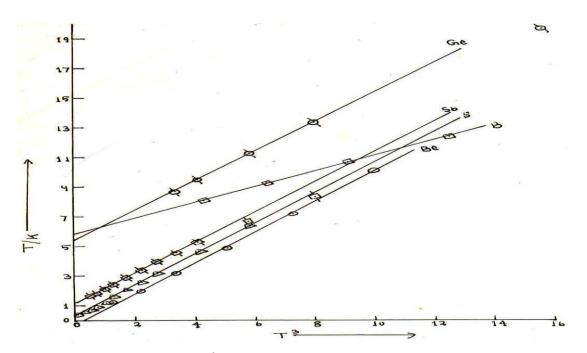


Fig. 1: Plots between $T/_{K}$ and T^{3} of insulators (Be, Ge, S, B and Sb) [The scales and origin have been suitably altered to portray the desired shape of the curves].

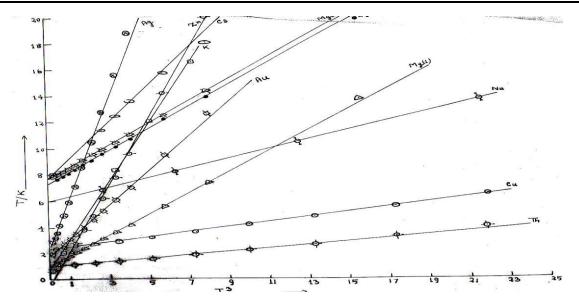


Fig. 2: Plots between T_{K} and T^{3} of metals [The scales and origin have been suitably altered to portray the desired shape of the curves].

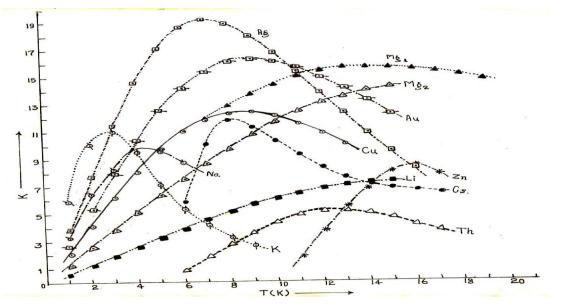


Fig. 3 Dependence of thermal conductivity (K) on temperature (T) of metals at very low temperatures [The origin and the magnification scales along the axis have been suitably altered to present just the desired shape of the curves].

CONCLUSION

The energy spectrum of phonons cannot be fully described by the Debye law. Martin suggests that the contribution to specific heat due to anharmonic effects caused by volume expansion is positive for alkali metals at temperature T. The anharmonic contribution if it exists may not be decribed completely by a few terms of function of temperature to fit in the data of thermal conductivity of Al, Cu, Ag, Au, Na, K,

Li, Pt, Fe, Co, Ni, & Zn in the temperature zone 1-30K. The first two terms dominate over, but the results of the present analysis demands the introduction of some term to the space between first and the second term of some metals in the light of equation. A unique value of θ for a metal cannot be taken because different sources. Positive contribution of either electrons or anharmonic lattice waves to specific heat whereas equation does not do. It is not unjustified because at very low temperatures anharmonicity due to volume expansion is negligible and that due to phonon-phonon interaction dominates. These results confirm the contention of Tosi and Fumi, Barron and Klemense. Since in an insulators the possibility of electronic contribution can be totally ruled out. This equation also emphasizes on the contribution of anharmonicity of lattice waves. Heat of metals at low temperatures is due to the anharmonicity of lattice waves rather than the electrons.

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