Journal of Thermal Analysis and Calorimetry, Vol. 55 (1999) 187-196

DEBYE TEMPERATURE DEPENDENT LATTICE THERMAL CONDUCTIVITY OF SILICON

A. H. Awad

Department of Physics, College of Education, University of Basrah, Basrah, Iraq

(Received November 20, 1997; in revised form March 18, 1998)

Abstract

The temperature dependence of the Debye temperature $\theta_D(T)$ was applied to analyze the lattice thermal conductivity of Si between 2 and 300 K. The analysis of experimental data in terms of the Dubey model of the two modes of conduction has been carried out by combining the relaxation time for phonon–phonon scattering, point defect scattering and boundary scattering. The relative importance of the contribution of each mode was examined by estimating their percentage contribution to the phonon conductivity. Agreement between theory and experiment is achieved over the whole temperature range of study.

Keywords: Debye temperature, lattice thermal conductivity, silicon

Introduction

In general, the measurement of the lattice thermal conductivity in a solid is a complex problem and has recently attracted considerable attention. Formulas extended to span temperatures from the boundary scattering regime to the Debye temperature often require adjustable parameters that do not necessarily possesses direct physical significance. Phonon–phonon scattering processes have been widely investigated by several authors [1–10] in terms of simple expressions of the three phonon scattering relaxation rates. These expressions indicate that the Debye temperature θ_D is an important factor in the estimation of the three phonon scattering relaxation rates. Using these scattering relaxation rates, several researchers [11–16] have calculated the phonon conductivity for different samples, but due to complications, none of them have considered the contribution of the temperature dependent Debye temperature to the lattice thermal conductivity integral.

Previous measurements [17, 18] on different samples having different values of Debye temperature θ_D have shown a significant effect of Debye temperature on the lattice thermal conductivity. However, in these studies the effect of the

1418–2874/99/ $ 5.00

Akadémiai Kiadó, Budapest
Kluwer Academic Publishers, Dordrecht

variation in the Debye temperature θ_D with temperature on the lattice thermal conductivity was not considered. In their attempts to analyze experimental data available for the lattice thermal conductivity of Si, Dubey and Verma [19] stated that in the low temperature range where boundary scattering of phonons dominates over other phonon scattering processes, the lattice thermal conductivity should be proportional to θ_D^{-2}. Klemens [2] has also found that the lattice thermal conductivity is proportional to $\theta_D^{-2}T^3$. Considering the contributions of three phonon normal and umklapp processes, the effects of the variation of Debye temperature with temperature on the three phonon scattering relaxation rate and on the lattice thermal conductivity of Ge, have been studied by the author [20] in the framework of the Dubey model [10]. Recently, Awad and Shargi [21] have studied the effect of the variation in the Debye temperature with temperature on the lattice thermal conductivity of Si in the framework of the SDV model [7]. They pointed out that at high temperatures, the percentage change in lattice thermal conductivity is small, while at 40 K it is as high as 75%. They have also expected that the discrepancy between the theoretical and experimental phonon conductivity of Si [19] can be modified by using the temperature dependent Debye temperature $\theta_D(T)$.

The temperature dependence of the thermal conductivity of Si has been experimentally and theoretically investigated by several workers [22–25]. It was Holland [22] who made the first attempt to interpret the lattice thermal conductivity of Si by proposing the two-mode conduction theory of phonons. As a matter of fact, he used only one process in his calculation in one conductivity integral. Joshi and Verma [8] tried to explain their measurement results by incorporating the contribution of three phonon normal processes only. They assumed an empirical relationship between the phonon frequency and the phonon wave vector to calculate the phase and group velocity, and they observed that the four phonon processes play an important role at high temperatures. Previously, Dubey and Verma [19] measured the lattice thermal conductivity of Si trying to propose their expression by considering the contribution of the three phonon umklapp processes only, but they found near the conductivity maxima ($20<T<60$ K) some discrepancies between the predicted and observed values.

The present investigation is a continuation of earlier studies, its major purpose, for the first time, is to elucidate the nature of lattice thermal conductivity of Si within the temperature range 2–300 K, using the temperature dependent Debye temperature $\theta_D(T)$. The basis of this investigation the two-mode conduction proposed by Dubey [10]. This also enables us to study the contribution of both transverse and longitudinal phonons to the lattice thermal conductivity of Si.

Guthrie [5, 6] stated that phonon–phonon scattering events occur in two different ways: class I events in which the phonon is annihilated by combination and class II events in which annihilation takes place by splitting. Guthrie's classification leads to the participation of the transverse phonons in class I events

only, whereas the longitudinal phonons participate in both class I and class II events.

Considering Guthrie's classification, Dubey [10] proposed an expression for the three phonon scattering relaxation rate as

$$\tau_{3ph,T}^{-1} = (B_{TN,I} + B_{TU,I}\, e^{-\theta/\alpha T})\omega T^{m_{T,I}(T)} \tag{1}$$

for transverse phonons, and for longitudinal phonons it takes the following form:

$$\tau_{3ph,L}^{-1} = (B_{LN,I} + B_{LU,I}\, e^{-\theta/\alpha T})\omega^2 T^{m_{L,I}(T)} + (B_{LN,II} + B_{LU,II}\, e^{-\theta/\alpha T})\omega^2 T^{m_{L,II}(T)} \tag{2}$$

Here B_N and B_U are the scattering strengths of the three phonon normal and umklapp processes respectively, θ is the Debye temperature of the sample and α is a constant. The values of the temperature exponents $m(T)$ can be calculated from the following relationships [7]:

$$m_I(T) = \frac{X_{max}}{e^{X_{max}} - 1} + 0.5X_{max} + \frac{\ln(1 + \theta/\alpha T)}{\ln T} \tag{3}$$

for class I events and

$$m_{II}(T) = 0.5 + \frac{0.5X_{max}\, e^{0.5X_{max}}}{e^{X_{max}} - 1} + \frac{\ln(1 + \theta/\alpha T)}{\ln T} \tag{4}$$

for class II events, where $X_{max}=\hbar\omega_{max}/K_B T$ and ω_{max} is the phonon frequency at the boundary of the Brillouin zone. In the present study the modified dispersion relation $k=(\omega/v)(1 + r\omega^2)$ of Verma *et al.* [7] is utilized, where r is the dispersion correction constant.

If the spherical symmetry of the Brillouin zone is taken into account, the lattice thermal conductivity can be expressed as the sum of two contributions as:

$$K = K_T + K_L \tag{5}$$

where K_T and K_L are the contributions arising from the transverse and longitudinal phonons respectively, and can be given by

$$K_T = \frac{C}{v_{T1}} \int_0^{\theta_1/T} \tau_{c,T} F(X) dX + \frac{C}{v_{T2}} \int_{\theta_1/T}^{\theta_2/T} \tau_{c,T} F(X) dX \tag{6}$$

$$K_L = \frac{C}{v_{L1}} \int_0^{\theta_3/T} \tau_{c,L} F(X) dX + \frac{C}{v_{L2}} \int_{\theta_3/T}^{\theta_4/T} \tau_{c,L} F(X) dX \tag{7}$$

where $C=(K_B/3\pi^2)(K_BT/\hbar)^3$, $\theta_i=\hbar\omega/K_B$, $F(X)=X^4e^X(e^X-1)^{-2}(1+R_iX^2)^2(1+3R_iX^2)$, $R_i=r_i(K_BT/\hbar)^2$, K_B is the Boltzmann constant, $\hbar$ is the Planck constant divided by 2π, the ν's are the velocities of the corresponding modes, the θ's are the temperatures relating to the Brillouin zone boundary (for more details, see [10, 13]), $\tau_{c,T}$ and $\tau_{c,L}$ are the combined scattering relaxation time of the transverse and longitudinal phonons, respectively and can be written as

$$\frac{1}{\tau_{c,T}}=\frac{1}{\tau_B}+\frac{1}{\tau_{pt}}+\frac{1}{\tau_{3ph,T}}+\frac{1}{\tau_{4ph}} \tag{8}$$

$$\frac{1}{\tau_{c,L}}=\frac{1}{\tau_B}+\frac{1}{\tau_{pt}}+\frac{1}{\tau_{3ph,L}}+\frac{1}{\tau_{4ph}} \tag{9}$$

where τ_B^{-1}, τ_{pt}^{-1} and τ_{4ph}^{-1} are the scattering relaxation rates of the boundary [26], point defect [27] and four phonon [28, 29] processes respectively, and can be given by

$$\frac{1}{\tau_B}=\frac{\nu}{L},\ \frac{1}{\tau_{pt}}=A\omega^4 \text{ and } \frac{1}{\tau_{4ph}}=B_H\omega^2T^2$$

where L is the Casimir length, A and B_H are the scattering strengths of the respective processes.

Results and discussion

The experimental data of the lattice thermal conductivity of Si are taken from the report of Glassbrenner and Slack [23]. The variations in θ_D with temperature are taken from the earlier report of Flubacher *et al.* [30]. The dispersion constant of Si was calculated with the help of the dispersion curve of Brockhouse [31].

Following the earlier work of the author [13–15], the strength of the different scattering processes were adjusted. The constants and the parameters used in the present analysis are listed in Table 1. By estimating the separate contributions of K_T and K_L with the help of (6) and (7), the lattice thermal conductivities of Si were calculated between 2 and 300 K. The results obtained are shown in Fig. 1. Figure 2 illustrates the plot of the percentage contributions of transverse and longitudinal phonons to the total lattice thermal conductivity.

The variation of the temperature exponent $m(T)$ with the temperature can be studied with the help of Table 2. The percentage contributions of the three phonon N- and U-processes to τ_{3ph}^{-1} have also been studied for both modes of phonon and are illustrated in Table 3. The percentage contributions of $\tau_{3ph,T}^{-1}$, $\tau_{3ph,L}^{-1}$ (class I + class II), $\tau_{3ph,L}^{-1}$ (class I) and $\tau_{3ph,L}^{-1}$ (class II) to the combined scattering relaxation rate of the respective modes $\tau_{c,T}^{-1}$ and $\tau_{c,L}^{-1}$ have been calculated for $\omega=\omega_{max}$ and are

Table 1 Values of parameters and constants used for calculating theoretical curves

Parameter	Unit	Value
θ_1	K	180
θ_2	K	210
θ_3	K	350
θ_4	K	570
α_1	–	1.3
v_{T1}	cm s^{-1}	$5.86 \cdot 10^5$
v_{T2}	cm s^{-1}	$2.0 \cdot 10^5$
v_{L1}	cm s^{-1}	$8.48 \cdot 10^5$
v_{L2}	cm s^{-1}	$4.24 \cdot 10^5$
r_1	s^2	$6.39 \cdot 10^{-28}$
r_2	s^2	$1.934 \cdot 10^{-27}$
r_3	s^2	0
r_4	s^2	$5.588 \cdot 10^{-29}$
$\tau_{B,T}^{-1}$	s^{-1}	$2.6 \cdot 10^6$
$\tau_{B,L}^{-1}$	s^{-1}	$3.0 \cdot 10^6$
A	s^3	$2.1 \cdot 10^{-46}$
$B_{TN,I}$	deg^{-m}	$3.8 \cdot 10^{-12}$
$B_{TU,I}$	deg^{-m}	$2.0 \cdot 10^{-7}$
$B_{LN,I}$	s deg^{-m}	$3.0 \cdot 10^{-25}$
$B_{LU,I}$	s deg^{-m}	$5.0 \cdot 10^{-22}$
$B_{LN,II}$	s deg^{-m}	$1.0 \cdot 10^{-22}$
$B_{LU,II}$	s deg^{-m}	$5.0 \cdot 10^{-28}$
B_{HT}	s deg^{-2}	$1.7 \cdot 10^{-22}$
B_{HL}	s deg^{-2}	$4.0 \cdot 10^{-23}$

listed in Table 4. With the help of these Figures and Tables, one can draw the following conclusions:

1. Figure 1 reveals that an excellent fit to the experimental data, as shown by the continuous line, is also obtained near the conductivity maxima in which region Dubey and Verma [19] did not observe good agreement. This Figure indicates that the model used in the present study gives a very good response to the experimental data and in order to analyze the phonon conductivity of Si, one should take into account the use of the temperature dependent Debye temperature $\theta_D(T)$ instead of taking it as a constant.

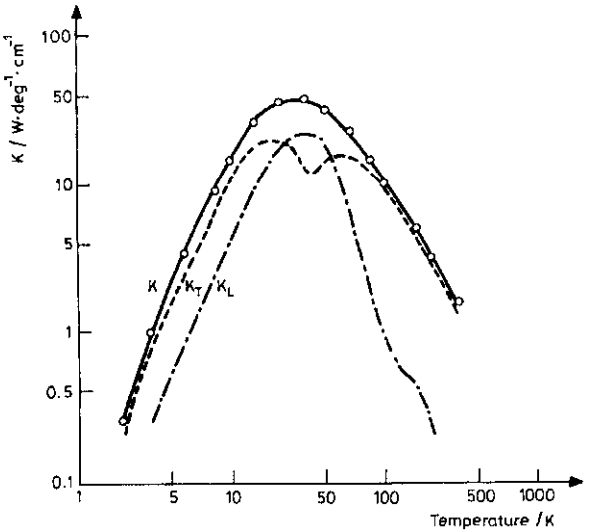

Fig. 1 Lattice thermal conductivity as a function of the temperatures

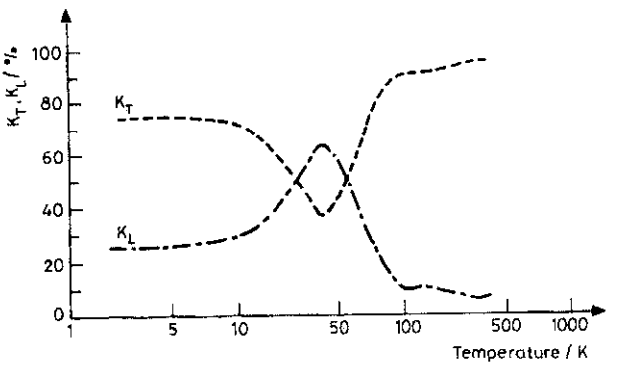

Fig. 2 Percentage contributions of K_T and K_L to the total lattice thermal conductivity as a function of the temperature

2. A better demonstration is provided by the data for Si (Fig. 2) which clearly illustrate two opposite trends. At very low temperatures, the temperature dependent lattice thermal conductivity is mainly governed by the boundary and point defect scattering. Accordingly the ratio $\%K_T/\%K_L$ depends upon the ratio of $2(\nu_L\tau_{B,L}^{-1}/\nu_T\tau_{B,T}^{-1})$ which indicates that the percentage contributions of longitudinal and transverse phonons to the total lattice thermal conductivity of Si are approximately in the ratio of 1:3. At the same time at a little higher temperature (10 K), $\%K_T$ begins to decrease while the opposite tendency is observed for $\%K_L$ and it is very clear that just above 30 K, $\%K_T$ begins to increase again. In fact, whithin

the temperature range 25–45K, the percentage contribution of longitudinal phonons exceeds the percentage contribution of transverse phonons. The basic reason for this kind of behaviour undoubtedly resides in the role of point defect scattering [7], which is in accordance with the prediction of Sherma *et al.* [7] for Ge, Dubey and Verma [19] for Si, Awad and Dubey [13] for Mg_2Ge and Mg_2Si and Awad [14] for InSb. It should also be noted that at high temperatures $\%K_T$ is much greater than $\%K_T$. This Figure also shows that most of the heat is transported by the transverse phonon alone.

Table 2 Values of the temperature exponent $m(T)$ for transverse phonons class I events ($m_{T,I}(T)$), longitudinal phonons class I events ($m_{L,I}(T)$) and longitudinal phonons class II events ($m_{L,II}(T)$)

T/K	$m_{T,I}(T)$	$m_{L,I}(T)$	$m_{L,II}(T)$
2	4.0	3.0	1.0
4	4.0	3.0	1.0
6	4.0	3.0	1.0
8	4.0	3.0	1.0
10	4.0	3.0	1.0
20	4.0	3.0	1.0
30	4.0	3.0	1.0
40	3.275	3.0	1.0
50	2.707	3.0	1.0
60	2.346	3.0	1.0
70	2.105	3.0	1.0
80	1.931	3.0	1.0
90	1.805	3.0	1.0
100	1.708	3.0	1.0
150	1.445	2.274	1.0
200	1.325	1.834	1.0
300	1.256	1.485	1.0

3. From the data in Table 2, one can see that at low temperatures are the temperature exponents for transverse and longitudinal phonons $m_I(T)$ reduced to 4 and 3, respectively, moreover, at low temperatures, the factor $e^{-\theta/\alpha T}$ is negligibly small. As a result, τ_{3ph}^{-1} shows ωT^4 dependence for transverse phonons and $\omega^2 T^3$ dependence for longitudinal ones. At high temperatures, both $m(T)$ and $e^{-\theta/\alpha T}$ reduce to unity which yield a $\omega T e^{-\theta/\alpha T}$ dependence of τ_{3ph}^{-1} for transverse phonons and $\omega^2 T e^{-\theta/\alpha T}$ dependence for longitudinal phonons. However, it would be instructive to note that at high temperatures, the phonon conductivity is propor-

tional to $1/T$ for both polarizations. It is worth noting that the same proportionality of τ_{3ph}^{-1} has been observed by Herring [1].

Table 3 The percentage contributions of $\tau_{3ph,N}^{-1}$ and $\tau_{3ph,U}^{-1}$ to τ_{3ph}^{-1} for transverse and longitudinal phonons

T/K	Transverse phonons		Long. phonons class (I)		Long. phonons class (II)	
	$\%\tau_{3ph,N}^{-1}$	$\%\tau_{3ph,U}^{-1}$	$\%\tau_{3ph,N}^{-1}$	$\%\tau_{3ph,U}^{-1}$	$\%\tau_{3ph,N}^{-1}$	$\%\tau_{3ph,U}^{-1}$
2	100	0	100	0	100	0
4	100	0	100	0	100	0
6	100	0	100	0	100	0
8	100	0	100	0	100	0
10	100	0	100	0	100	0
20	99.99	$1.0\cdot10^{-2}$	99.99	$1.0\cdot10^{-2}$	99.99	$1.0\cdot10^{-2}$
30	81.49	18.51	99.34	$6.6\cdot10^{-1}$	83.42	16.58
40	11.80	88.20	82.10	17.90	13.34	86.66
50	2.74	97.26	49.15	50.85	3.22	96.78
60	1.05	98.95	25.49	74.51	1.12	98.88
70	$5.30\cdot10^{-1}$	99.47	15.39	84.61	$6.0\cdot10^{-1}$	99.40
80	$3.20\cdot10^{-1}$	99.68	9.75	90.25	$3.6\cdot10^{-1}$	99.64
90	$2.00\cdot10^{-1}$	99.80	6.44	93.56	$2.3\cdot10^{-1}$	99.77
100	$1.40\cdot10^{-1}$	99.86	4.60	95.40	$1.6\cdot10^{-1}$	99.84
200	$2.00\cdot10^{-2}$	99.98	$7.2\cdot10^{-1}$	99.28	$2.0\cdot10^{-2}$	99.98
300	$1.00\cdot10^{-2}$	99.99	$3.2\cdot10^{-1}$	99.68	$1.0\cdot100^{-2}$	99.99

4. By examining the values in Table 3 it becomes quite clear that at low temperatures $\tau_{3ph,N}^{-1}$ predominates over $\tau_{3ph,U}^{-1}$ below certain temperature (about 35 K), and the opposite is true above that temperature. In other words $\tau_{3ph,N}^{-1}$ decreases with increasing temperature, an opposite trend is shown for $\tau_{3ph,U}^{-1}$ with temperature. Meanwhile, one can conclude that at low temperatures the most of the heat is transported by phonons which conserve momentum, while at high temperatures the role of those phonon processes which do not conserve momentum in the lattice thermal resistivity becomes predominant. It is of interest that the results are similar to those obtained by previous workers [10, 13–16]. Table 3 shows zero percentage contribution due to U-process at very low temperatures. In fact, it is not zero, but the value is so small that it can be considered zero.

5. The data in Table 4 clearly indicate that at temperatures just above the conductivity maxima τ_{3ph}^{-1} predominates over τ_B^{-1} and τ_{pt}^{-1} for both the polarization branches of phonons, which reflects the effectiveness of the phonon–phonon scattering in the thermal resistance at high temperatures. At the same time, at

Table 4 The percentage contributions of $\tau^{-1}_{3ph,T}$, $\tau^{-1}_{3ph,L}$ (class I + class II), $\tau^{-1}_{3ph,L,I}$ and $\tau^{-1}_{3ph,L,II}$ to the combined scattering relaxation rate $\tau^{-1}_{c,T}$ and $\tau^{-1}_{c,L}$ for $\omega=\omega_{max}$

T/ K	$\tau^{-1}_{3ph,T}/\tau^{-1}_{c,T}$	$\tau^{-1}_{3ph,L}/\tau^{-1}_{c,L}$	$\tau^{-1}_{3ph,L,I}/\tau^{-1}_{c,L}$	$\tau^{-1}_{3ph,L,II}/\tau^{-1}_{c,L}$
		%		
2	$1.25\cdot10^{-3}$	$1.72\cdot10^{-2}$	$2.05\cdot10^{-4}$	$1.70\cdot10^{-2}$
4	$2.00\cdot10^{-2}$	$3.58\cdot10^{-2}$	$1.64\cdot10^{-3}$	$3.41\cdot10^{-2}$
6	$1.01\cdot10^{-1}$	$5.67\cdot10^{-2}$	$5.53\cdot10^{-3}$	$5.12\cdot10^{-2}$
8	$3.47\cdot10^{-1}$	$8.14\cdot10^{-2}$	$1.31\cdot10^{-2}$	$6.83\cdot10^{-2}$
10	$8.45\cdot10^{-1}$	$1.10\cdot10^{-1}$	$2.56\cdot10^{-2}$	$8.53\cdot10^{-2}$
20	12.00	$3.74\cdot10^{-1}$	$2.04\cdot10^{-1}$	$1.70\cdot10^{-1}$
30	45.49	$9.94\cdot10^{-1}$	$6.90\cdot10^{-1}$	$3.04\cdot10^{-1}$
40	54.27	4.37	1.91	2.46
50	53.32	16.81	5.42	11.38
60	54.13	38.80	12.68	26.12
70	53.29	61.00	22.28	38.71
80	55.48	76.49	31.67	44.82
90	56.94	86.21	39.96	46.25
100	59.41	91.61	46.84	44.76
200	80.87	98.63	0.823	97.80
300	89.59	99.59	0.133	99.46

temperatures below the conductivity maxima, one can see an opposite behaviour of the percentage contribution and it can be said that the lattice thermal resistivity is mainly due to scattering of phonons by boundary and point defects. The same behaviour was also observed by Awad and Dubey [13] in the lattice thermal conductivity of Mg_2Ge and Mg_2Si based on the scattering relaxation approach. From Table 4, it is obvious that $\tau^{-1}_{3ph,L}$ for class II events, dominates over $\tau^{-1}_{3ph,L}$ for class I events, but in the meantime at the conductivity maxima ($10<T<40$ K) an opposite behaviour is shown.

6. In conclusion, the present investigation provides an account of the effect of temperature dependent Debye temperature $\theta_D(T)$ on the lattice thermal conductivity of Si.

References

1 C. Herring, Phys. Rev., 95 (1964) 954.
2 P. G. Klemens, Solid State Physics, Vol. 7 (Ed. F. Seitz and D. Turnbull) Academic Press Inc., New York 1958, p.1.
3 J. Callaway, Phys. Rev., 113 (1959) 1046.

4 M. G. Holland, Phys. Rev., 132 (1963) 2461.
5 G. L. Guthrie, Phys. Rev., 152 (1966) 801.
6 G. L. Guthrie, Phys. Rev., B3 (1971) 3373.
7 P. C. Sharma, K. S. Dubey and G. S. Verma, Phys. Rev., B3 (1971) 1985.
8 Y. P. Joshi and G. S. Verma, Phys. Rev., B1 (1970) 750.
9 K. S. Dubey and R. H. Misho, J. Thermal Anal., 12 (1977) 223.
10 K. S. Dubey, J. Thermal Anal., 19 (1980) 263.
11 M. C. Al-Edani and K. S. Dubey, Phys. Status Solidi, (b) 86 (1978) 741.
12 K. S. Dubey, Indian J. of Pure and Applied Physics, 15 (1977) 455.
13 A. H. Awad and K. S. Dubey, J. Thermal Anal., 24 (1982) 233.
14 A. H. Awad, Acta Physica Hungarica, 63 (1988) 331.
15 A. H. Awad, J. Thermal Anal., 39 (1993) 559.
16 A. H. Awad and Q. M. A. Hussan, Engineering & Technology, 13 (1994) 1.
17 K. R. Wilkinson and J. Wilks, J. Proc. Phys. Soc., London A64 (1951) 89.
18 F. J. Weeb and J. Wilks, J. Phil. Mag., 44 (1953) 664.
19 K. S. Dubey and G. S. Verma, Phys. Rev., B7 (1973) 2879.
20 A. H. Awad, M. Sc. Thesis, Basrah University, Basrah (1982).
21 A. H. Awad and S. N. Shargi, J. Thermal Anal., 37 (1991) 277.
22 M. G. Holland and L. J. Neuringer, Proc. Int. Conf. Semiconductor, Exeter (1962).
23 C. J. Glassbrenner and G. A. Slack, Phys. Rev., 134 (1964) A1058.
24 W. Fulkerson, J.P. Moore, R. K. Willians, R. S. Graves and D. L. Mcelroy, Phys. Rev., 167 (1968) 765.
25 H. R. Shanks, P. D. Maycock, P. H. Sidles and G. C. Danielson, Phys. Rev., 130 (1963) 1743.
26 H. B. G. Casimir, Physica, 5 (1938) 945.
27 P. G. Klemens, Proc. Roy. Soc. London, 68 (1955) 1113.
28 I. Pomeranchuk, Phys. Rev., 60 (1941) 82.
29 I. Pomeranchuk, J. Phys. U.S.S.R., 4 (1941) 259.
30 P. Flubacher, A. J. Leadbetter and J. A. Morrison, Phil. Mag., 4 (1959) 273.
31 B. N. Brockhouse, Phys. Rev. Lett., 2 (1959) 256.