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Electronic and Lattice Thermal Conductivity of $Bi_{1-x}Sb_x$ and Ag-Sb-Te Systems

**Sabir ALIEV, Agamirza MOVSUM-ZADEH,
Sadijar RAGIMOV, Eldar ZULFIGAROV**
*Institute of Physics, Academy of Sciences of Azerbaijan,
370143, Baku-AZERBAIJAN*

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Abstract

An investigation of the electronic ($\aleph_e$) and lattice ($\aleph_L$) parts of the thermal conductivity for samples of Bi_{1-x} Sb_x (0,02 < X < 0,12) and system of Ag-Sb-Te was carried out in the temperature interval 6-300K. Separation of $\aleph_L$ and $\aleph_e$ has been made by suppressing $\aleph_e$ in magnetic field. The experimental data of $\aleph_L$ and $\aleph_e$ have been compared, respectively, with Gallaway's theory and with a theory which takes into account the inelastic character of scattering. It is observed that the Lorentz number L calculated by using the experimental values of $\aleph_e$ and thermomagnetic coefficients is smaller than its Zommerfeld's value L_0 in high doping $Bi_{0.88}Sb_{0.12}$. A satisfactory agreement between the theory and the experiment allows to conclude that besides the normal and U-processes in $Bi_{1-x}Sb_x$ the defects of mass also plays a considerable role in scattering. It is observed that $\aleph_L$ of single phase samples $Ag_{0.82}Sb_{1.18}Te_{2.18}$ is smaller than $AgSbTe_2$ samples. It is shown that this is due to the presence of p-Ag_2 Te in $AgSbTe_2$. It is established that the high concentration of intrinsic defects plays a considerable role in scattering for Ag-Sb-Te as well.

Introduction

The solid solutions of $Bi_{1-x}Sb_x$ are the most favorable materials for the investigations of electron and phonon phenomena. Bismuth is a semimetal with high electron and hole mobility. When Bi is doped with Sb it undergoes semimetal-semiconductor transition. Within this process additional scattering centers of electron and holes may arise. Of special interest is investigations of thermomagnetic effects in the semiconductor phase. The high mobility of charge carriers allows to separate the electronic $\aleph_e$ and lattice $\aleph_L$ thermal conductivity. Further, investigation of $\aleph_e$ and thermomagnetic coefficients allows to determine the Lorentz number L from the Wiedemann-Franz equation $\aleph_e = L.\delta.T$, generably depending on the mechanism of electron scattering and the nature of

interelectron and electron-phonon interactions. As is known. $AgSbTe_2$ is in a two-phase state congisting of single-phase $Ag_{0.82}Sb_{1.18}Te_{2.18}$ and p-Ag_2Te. Investigations [1,2] show that the second phase strongly influence galvanomagnetic and thermoelectric properties. It is supposed that thermal conductivity of Ag_2Te is more than the basic phase at low temperatures.

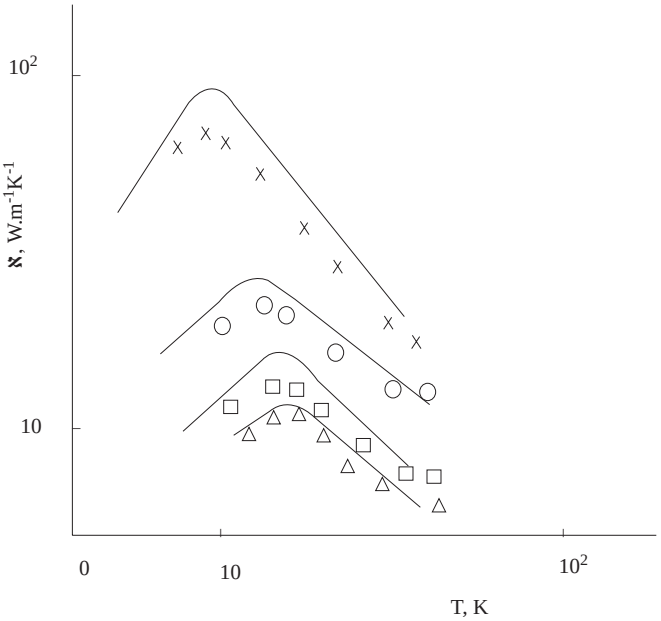

Figure 1. The temperature dependence of $\aleph_L$ in $Bi_{1-x}Sb_x$. The solid lines represent the theoretical curve from [3] (X-x=0.02; ◯-x=0.04; □-x=0.08; Δ-x=0.12)

Experimental Results and Discussion

In this work we present results of investigations of electronic $\aleph_e$ and lattic $\aleph_L$ thermal conductivity of $Bi_{1-x}Sb_x$ (0,02<X<0,12) and Ag-Sb-Te systems. Separation of $\aleph_e$ and $\aleph_L$ has been made by suppressing $\aleph_e$ in magnetic field. The $\aleph_e$ in Ag-Sb-Te was insignificant due to the low mobility of charge carriers.

Figure 1 Shows the $\aleph_L$ (T) dependence for $Bi_{1-x}Sb_x$ solid solutions. It is seen that $\aleph_L$ (T) maximum is displaced to the high temperature side. Figure 2 shows the $\aleph_L$ (T) dependences for samples of the Ag-Sb-Te system. As is seen, the $\aleph$ (T) curve of a two-phase sample pass above the basic phase curve. In Fig. 3 are presented the characteric curves of $\Delta\aleph(UH/C)$ (U-mobility of electrons) dependence for $Bi_{0.88}Sb_{0.12}$ doped by Te up to 0,01%. Since the bipolar thermal conductivity $\aleph_{Bip}$ is very small for samples with high electron concentration the limit value of $\Delta\aleph_\infty$ corresponds to the electron part of thermal conductivity $\aleph_e$. The experimental value of $\aleph_e$ made it possible to calculate the Lorentz number L from the W-F equation. The obtained data are plotted in Fig. 4

as L(T). As is seen, the L value is smaller than its Zommer feld value in the 40-220 K temperature interval. The results of $\aleph_L(\mathrm{T})$ were interpreated by Gallaway theory [3], according to which

$$\aleph_L = G.T^3 \left\{ \int_0^{\theta/T} \frac{\tau_c.x^4.dx}{Sh^2(x/2)} + \left[\int_0^{\theta/T} \frac{\tau_c}{\tau_N} \frac{x^4 dx}{Sh^2(x/2)} \right]^2 \left[\int_0^{\theta/T} \frac{\tau_c}{\tau_N.\tau_R} \frac{x^4 dx}{Sh^2(x/2)} \right]^{-1} \right\},$$

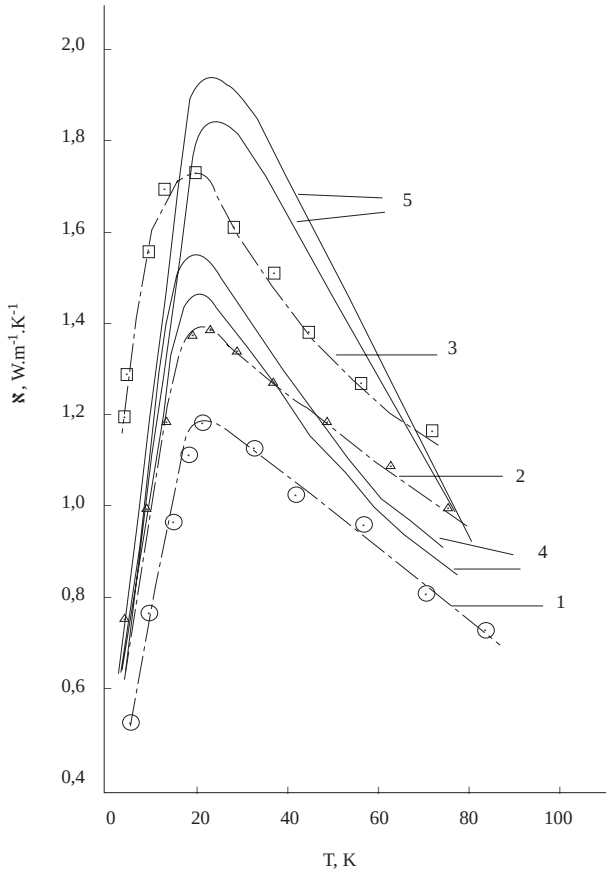

Figure 2. The temperature dependences of thermal conductivity: 1. $Ag_{0.82}Sb_{1.18}Te_{2.18}$ 2. $AgSbTe_2$, 3. AgSbTe. The solid lines are for calculations according to [4] (4), and according to [3] (5).

where $G = (\hbar/2(2\pi)^2 v(k/\hbar)^4$, v is average velocity; $X = \hbar w/kT; w$ is the frequency of phonons: $\tau_c^{-1} = \tau_R^{-1} + \tau_N^{-1}; \tau_R$ and τ_N are the relaxsation times for the, resistivity and normal processes, $\tau_R^{-1} = \tau_{pp}^{-1} + \tau_B^{-1} + \tau_{pd}^{-1}; \tau_{pp}; \tau_B$ and τ_{pd} is the relaxation times for unclup

phonon phonon processes, boundary and phonon-phonons scattering, resprectively. The following was found as a function of relaxation time T:

$$\tau_{pp}^{-1} = Ax^2T^4e^{\theta/\alpha T}; \tau_B^{-1} = BX^4T^4; \tau_N^{-1} = CX^4T^4.$$

Here, L=1,5 mm is the effective size of sample. Parameters A, B, C, d and n were determined numerically via least-squares method by comparing expression (2) with experimental curves of ℵ (T). In this case, in solid solution systems of $Bi_{1-x}Sb_x$ and Ag-Sb-Te, the change of fluctation density $\Delta\delta/\delta$ was insignificant and therefore, scattering was effectively via point defects due to change of mass $\Delta m/m$, in principal. Thus,

$$\tau_{pd}^{-1} = (V_0/n).N(\Delta m/m)(k/\hbar)^4(4\pi v^3)^{-1}x^4T^4,$$

where V_0 is the volume of a unit cell, n is the number of atoms in a unit cell, m is the average mass of atoms in crystal, and N is the number concentration of impurity atoms:

$$\beta = (V_o/n)(k/\hbar)^4(N/4\pi V^3)(\Delta m/m)^2.$$

This theory was repeatedly confirmed experimentally in [5, 6, 7, 8]. The results of calculations is plotted in Fig. 2. (solid lines). Satisfactory agreement between theory and experiment make it possible to consider that the mass defects can play a considerable role along with N and U-prosesses in $Bi_{1-x}Sb_x$ in scattering.

The experimental $\aleph_L$ data obtained for the Ag-Sb-Te system were compared with calculations of generalized thermal conductivity of inhomogeneous systems [4], according to which the generalized thermal conductivity in all concentration regions (x=01) can be expressed by

$$\aleph = \aleph_0 \left\{1 + x\left[\frac{1}{3}(1-x) + \frac{\aleph_0}{\aleph_i - \aleph_0}\right]\right\}$$

where ℵ is the resulting thermal conductivity. $\aleph_0$ is the thermal conductivity of the basic phase, $\aleph_i$ is the thermal conductivity of the second phase and X is the volume fraction of Sb.

The results of calculations for 11 and 13 vol.% p-Ag_2Te is plotted in Fig.2 (solid lines), where the calculated curves are arranged above the basic phase curves. The calculations showed that this results form Ag_2Te having a higher value of thermal conductivity of Ag_2Te than $Ag_{0.82}Sb_{1.18}Te_{2.18}$ in the indicated temperature range. In order to established the main phonon scattering mechanisms in this system the results of $\aleph_L(T)$ were compared with Gallaway's theory. The calculation results are plotted in Fig 2. As is seen, the calculated curves pass above then experimental values, from which may be concluded that, the mass defects and the intrinsic defects play an essential role along with the N and U-processes in Ag-Sb-Te systems.

In [9, 10, 11, 12, 13] it is shown that the low value of L is due to the inelastic character of electron scattering. The theoretical problem of inelastic scattering has been most fully considered in work [10]. According to this theory, at high degeneracy ($\mu^\star >> 1$) the

energy of transverse optical phonons is effectively less than the reduced energy of the charge carriers, on the order of kT ($\mu >> \hbar w, \mu \sim KT$).

Then the Lorentz number is expressed, generally, in the form:

$$\frac{L}{L_0} = \left[1 + \frac{Wee}{Wo} + \frac{U}{Uop}\left(\frac{Lo}{L} - 1\right)_{op}\right]^{-1}. \tag{1}$$

The term (U/U_{op})[(L_o/L)-1] involves inelasticity caused by the polar scattering of electrons on optic phonons, and the term Wee/Wo takes into account electron-electron interaction.

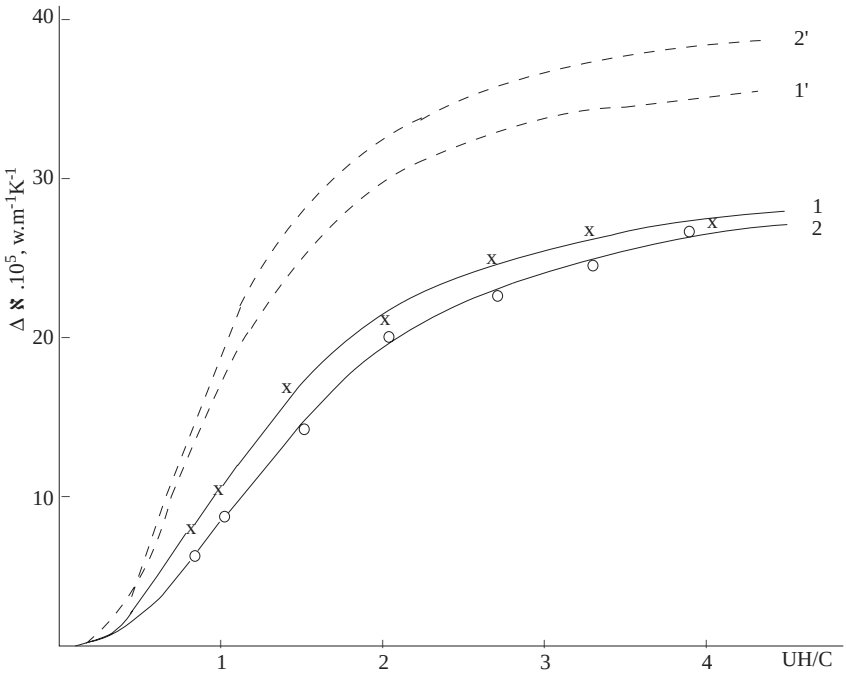

Figure 3. The magnetic field dependence of $\Delta\aleph$:(1-93K at L/L_0 = 0.75; 2-205K at L/L_0=0.65; 1′ and 2′ at L/L_0=1).

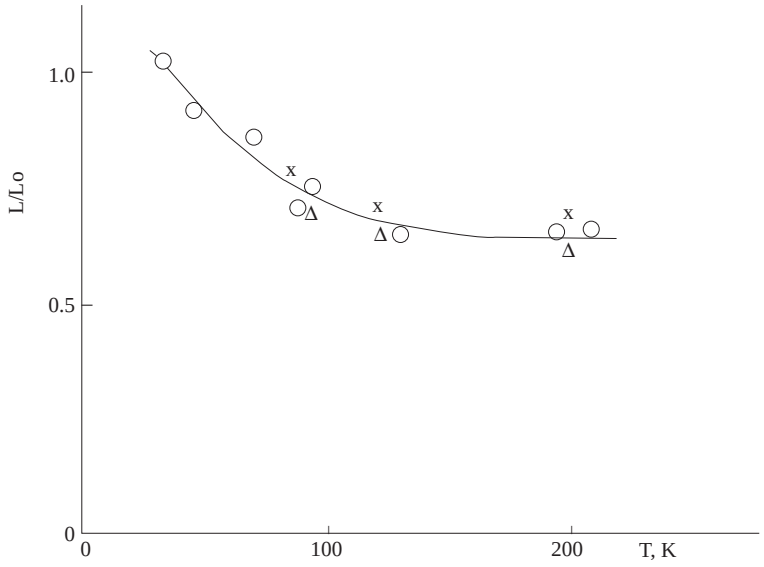

Figure 4. The temperature dependence of L/L_0 in $Bi_{0.88}Sb_{0.12}$. The solid lines represent the theoretical curve, calculated from Eq. (1).

Considering the results presented on Fig. 4, it can be concluded that, in the solid solution $Bi_{0.88}Sb_{0.12}$ with strong degeneration, the unelastic scattering of charge carriers is mainly connected with electron-electron interaction. The inelastic part connected with polar scattering on optical phonons doesn't exceed 5-7%.

References

[1] E. Wolfe, J. Wernick, S. K. Hasko, J. Appl. Phys. 31 (1960) 1959-1964.

[2] S. A. Aliev, S. S. Ragimov, Neorganicheskie materiali, 28, 1992, 329-334.

[3] J. Gallaway, J. Phys. Rev. 113, v4, 1959, 1046-1051.

[4] V. J. Odolevski Jour. Tech. Phys. 21, v6, 1951, 667-677.

[5] L. M. Vasilev, J. Gabbarov, V. S. Oskolski, L. S. Parfeneva, V. V. Popov, J. A. Smirnov, J. Phys. tv. tela v.26, N 9 1984, 2710-2715.

[6] S. A. Aliev, T. G. Gadgiev, R. J. Selimzade, J. Phys. tv. tela, v.31, N 2, 1989, 293-294.

[7] S. A. Aliev, F. F. Aliev, S. G. Abdinova, Z. S. Gasanov, D. M. Ragimova, J. Izvestija VUZ-ov, N 6, 1990, 41-45.

[8] S. A. Aliev, S. S. Ragimov, J. Tagiev, J. Izvestija VUZ-ov N 1, 1995, 123-125.

[9] L. L. Korenblit, V. E. Sherstobitov, J. Phys. Tech. polup. 2, 1968, 688.

[10] Yu. I. Ravich, B. A. Efimova, V. I. Tamarchenko, Phys St. Sol. b 43, 11, 1971.

[11] S. A. Aliev, U. K. Sujunov, D. G. Arasli, M. J. Aliev, J. Phys. tech. poluprovodnikov, v.7, 1973, 1086-1089.

[12] S. A. Aliev, D. A. Bagirov, E. R. Iskenderov, E. J. Zulfigarov, S. J. Safarova, Neorganicheskie materiali, v.29, N 4, 1993, 499-502.

[13] S. A. Aliev, A. A. Movsum-zadeh, S. S. Ragimov, J. Phys. tech. polup. v.31, N 5, 1997, 559-562.