PHONON SCATTERING IN $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ SOLID SOLUTION

R.N. RAHIMOV, D.H. ARASLY, A.A. KHALILOVA

Institute of Physics, Nat. Acad. Sci. of Azerbaijan
(33, H.Javid Ave., Baku Az-1143, Azerbaijan; e-mail: rashad@physics.ab.az)

UDC 532

The thermal conductivity of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ (with $0 \le x \le 0.1$) solid solutions at temperatures between 80 and 300 K has been studied. Experimental data has been analyzed on the basis of the existing theories of solid-state thermal conductivity. The essential role of N processes in the phonon scattering in $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ solid solution has been established. The observed dip in the temperature dependence of thermal conductivity is explained by the phonon resonance scattering by various complexes.

1. Introduction

$(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ solid solutions have defect structure. In the In—Sb—Ge—Te system, Ge atoms occupy places in both the anionic and cationic sublattices owing to the varying valence. As a consequence, they turned out to be in crystallographic nonequilibrium positions. A change of the composition heterogeneity degree of In_2GeTe in $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ solid solutions and a change in the anion valence should affect the physical properties of the solutions, especially the thermal ones.

In our previous works [1, 2], the electrical and optical properties of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ solutions have been studied, and it is shown that the quasilocal level of a donor is formed by 0.21—0.19 eV above the bottom of the conductivity band. In this case, this level is displaced in the direction of the band gap with increasing temperature and the In_2GeTe-component content [2].

The present paper deals with the thermal conductivity of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$depending on the In_2GeTe content and temperature.

2. Experimental Results and Discussion

$(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ solid solutions have been obtained by the method of quick cooling of the InSb and In_2GeTe "ternary" compound and quenching from a melt. The obtained samples then were annealed for 14 days [3]. The electron concentration is varied within of $n = (3 \div 7) \times 10^{18}$ cm^{-3}.

The thermal conductivity of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ ($0 \le x \le 0.1$) solid solution has been measured by the absolute stationary method and flash light method between 80 and 300 K.

The temperature dependence of the thermal conductivity of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ solid solutions, $K(T)$, is shown in Fig. 1. As seen from the figure, the value of K reduced with increasing the In_2GeTe content and the temperature dependence of the thermal conductivity is weakened. About 100 K, the anomalous dip in $K(T)$ is observed.

The electron share of the thermal conductivity computed in accordance with the Widemann—Franz formula is 5% of $K_{\exp}$. Therefore, changes in the thermal conductivity are related to phonon processes in the investigated range of temperatures.

The thermal conductivity can be reduced by the phonon scattering by introduced lattice imperfections of the In_2GeTe component. In the case of solid solutions, such point defects are substitutional atoms, vacancies, and interstitial atoms. When larger numbers of foreign atoms are added to the host lattice by melting, the thermal conductivity decreases significantly. Consequently, the analysis of the $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ thermal conductivity dependence on the content of In_2GeTe at room temperature has been carried out by the well-known Klemens formula [4, 5] taking into account the Umklapp three-phonon processes and the phonon scattering by point defects:

$$K_p = K_0 \left(\frac{\omega_0}{\omega_{\rm D}}\right) \arctan\left(\frac{\omega_D}{\omega_0}\right), \tag{1}$$

where

$$\left(\frac{\omega_0}{\omega_{\rm D}}\right)^2 = \frac{\hbar v^2}{2\pi^4 K_0 \theta A}, \quad A = \frac{V\Gamma}{4\pi v^3}, \tag{2}$$

$$\Gamma = x(1-x)\left[\left(\frac{\Delta M}{\bar{M}}\right)^2 + \varepsilon\left(\frac{\Delta\delta}{\delta}\right)^2\right]. \tag{3}$$

Here, K_0 is the thermal conductivity of the lattice in the absence of defects; $\omega_{\rm D}$ is the maximum phonon

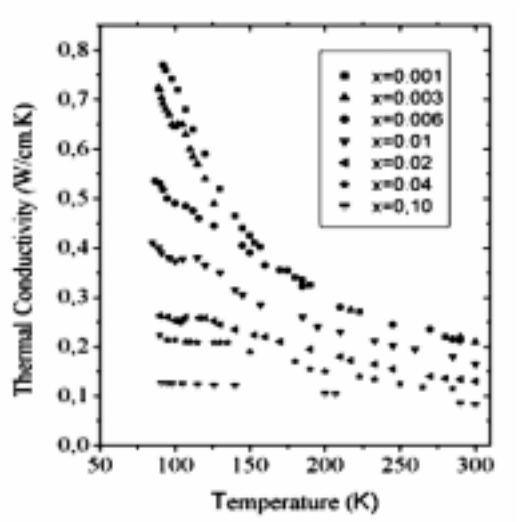

Fig. 1. Thermal conductivity of $(InSb)_{2(1-x)}(In_2GeTe)_x$ solid solutions versus temperature

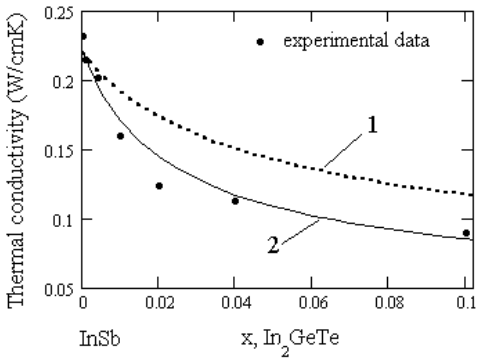

Fig. 2. Thermal conductivity of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ at room temperature versus the alloy composition (dots) and the result of computations (curves *1*, *2*) with formulas (1), (4), respectively

frequency in the Debye model; ω_0 is the frequency, at which the relaxation rates due to the point-defect scattering ($\tau_D^{-1} = A\omega^4$) and U-processes ($\tau_U^{-1} = B\omega^2 T$), in which total momentum is changed by the reciprocal lattice vector, are equal; V is the atomic volume, v is the phonon velocity; θ is Debye temperature; Γ is the disorder parameter including local changes in the density and elastic properties of the alloy, when one atom is replaced with another, and $\bar{M}$ is determined as $\bar{M} = 2(1-x)M_{InSb} + xM_{In_2GeTe}$. In general, ε should be regarded as a phenomenological adjustable parameter. In the case of A^3B^5 semiconductor compound, the parameter ε is determined from the relation between elastic constants and atomic volume. The relation $\Delta\delta/\delta$, according to Abeles [6], may be substituted for $\Delta a/\bar{a}$, where $\bar{a}$ is the lattice constant. The parameters, which are necessary for the computation, have been taken from work [7] and are linearly extrapolated for solid solutions. It should be noted that, in a solid solution, the term related to a local change of the material elastic properties makes its significant contribution to the disorder parameter, Γ.

The thermal conductivity of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ at room temperature versus the alloy composition and the result of computations are presented in Fig. 2. As seen from Fig. 2, the experimental data are below the computed curve *1*. It may be supposed that a significant quantitative divergence between the theoretical and experimental data is related to that the normal three-phonon process was not taken into consideration.

It is known that heat is generally transported by longitudinal phonons in solid solutions because of the strong scattering of high-frequency phonons by point defects. However, the interaction between longitudinal phonons, with the conservation of the quasi-momentum, may influence the lattice thermal conductivity at high temperatures.

The influence of N-processes on thermal conductivity at $T > \theta$ in alloys was considered in [6, 8]. Abeles [6] proposed a phenomenological approach based on the model presented in [9] for the lattice thermal conductivity of disordered semiconductor alloys at high temperatures. He considered three kinds of the relaxation (normal three-phonon processes, $\tau_N^{-1} = B_N\omega^2 T$, Umklapp three-phonon processes, $\tau_U^{-1} = B_U\omega^2 T$, and the phonon scattering by point defects, $\tau_D^{-1} = V\omega^4\Gamma/4\pi v^3$) and obtained the following formula for the lattice thermal conductivity:

$$K = K_0\left(\frac{1}{1+5C/9}\right)\left[\frac{\arctan y}{y} + \right.$$

$$\left. + \frac{(1-\arctan y/y)}{((1+C)/C)(y^4/5) - (y^2/3) + 1 - \arctan y/y}\right], \qquad (4)$$

where

$$y^2 = \frac{(\omega_D/\omega_0)^2}{1+5C/9}, \quad C = B_N/B_U;$$

C being the adjustable parameter indicating how many times N-processes are stronger than the U-processes.

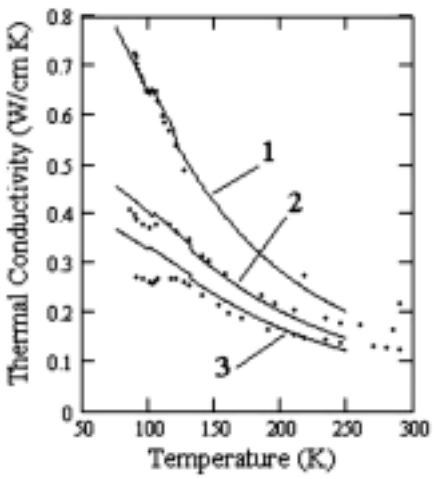

Fig. 3. Comparison of the experimental data on thermal conductivity versus temperature and the result of computations by formula (5) (curves *1*, *2*, *3*) for $(InSb)_{2(1-x)}(In_2GeTe)_x$ alloys with $x = 0.003$, 0.01, and 0.02, respectively

The results of computations have been plotted in Fig. 2. Curve *2* calculated with formula (4) with the adjustable parameter $C = 1.6$ fits well our experimental data. It shows the essential role of N-processes in the phonon scattering in $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ solid solution.

The dip observed in the temperature dependence of the thermal conductivity near $T = 100$ K (Fig. 1) is of great interest. The observed features in the narrow interval of temperatures evidently indicate a realization of the resonance phonon scattering. A similar K(T) behavior was also observed for pure A^3B^5 compounds at a temperature below 50 K [10—13], and we consider that it is related to the resonance phonon scattering. However, the nature of the resonance phonon scattering center is not clear yet.

In $In_{1-x}Ga_xAs$ [14] and GaSb—Ga_2Te_3 [15] solid solutions, a dip in $K(T)$ is also observed and is connected with the resonance phonon scattering by impurity-vacancy complexes.

The resonance phonons scattering is observed at a comparatively "high" temperature near $T = 100$ K in A^3B^5 alloys in contrast to binary compounds. The analysis of the temperature dependence of thermal conductivity is carried out in the framework of the well-known Callaway—Klemens' model by the following formula:

$$K = \frac{k}{2\pi v}\left(\frac{2\pi k}{h}\right)^3 T^3\left[I_1 + \frac{I_2^2}{I_3}\right], \qquad (5)$$

where

$$I_1 = \int_0^{\theta/T} \frac{\tau_c x^4 e^x}{(e^x-1)^2}dx, \quad I_2 = \int_0^{\theta/T} \frac{\tau_c}{\tau_N}\frac{x^4 e^x}{(e^x-1)^2}dx,$$

$$I_3 = \int_0^{\theta/T} \frac{1}{\tau_N}\left(1-\frac{\tau_c}{\tau_N}\right)\frac{x^4 e^x}{(e^x-1)^2}dx, \quad x = \frac{\hbar\omega}{kT}.$$

The combined relaxation time τ_c includes phonon-scattering processes, crystal boundary scattering, normal three-phonon process, Umklapp three-phonon process, Rayleigh scattering by point defects, and phonon resonance scattering:

$$\tau_c^{-1} = v/L + A\omega^4 + (B_N + B_U)\omega^2 T^3 + \tau_R^{-1}, \qquad (6)$$

where L is the sample length. For the relaxation rate of phonon resonance scattering, the following expression given in [16] should be used:

$$\tau_R^{-1} = \frac{R\omega^2 T^2}{(\omega^2 - \omega_R^2)^2},$$

where ω_R is the resonance frequency.

For the comparison of the calculated data with experiments, we have used the method of least squares. In Fig. 3, the experimental data and the curves computed by formula (5) for three of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ alloys (with $x = 0.003$, 0.01, and 0.02) are plotted. As seen from Fig. 3, the introduction of the relaxation time of the resonance phonon scattering in the combined relaxation time enables one to describe qualitatively the dip observed in the temperature dependence of the thermal conductivity. Values of the approximation coefficients are given in Table.

It should be noted that the adjustable parameter, A, is considerably different from the computed parameter of alloy-disorder scattering determined by formula (2). Probably, this is connected with the presence of the other defects. It is assumed that the nonequivalence of the nearest environment of the same atom in a solid solution can result in the formation of various complexes, whose presence brings about the features of phonon scattering.

3. Summary

The investigation of thermal conductivity of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ solid solution indicates that

Parameters used in the computation of the phonon thermal conductivity of $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ alloys with $x = 0.003$, 0.01, and 0.02

x	$A_{exp} \times 10^{42}$ (s^3)	$A_{calc} \times 10^{44}$ (s^3)	$B_U \times 10^{24}$ ($s \cdot K^{-3}$)	B_N/B_U	$R \times 10^{-26}$ ($s \cdot K^{-2}$)	$\omega_0 \times 10^{-12}$ (s^{-1})	$v/L \times 10^{-6}$ (s^{-1})
0.003	4.7	0.41	2.03	1.6	4.28	5.8	1.23
0.01	9	1.37	2.03	1.6	4.28	5.8	2.05
0.02	11.4	2.72	2.32	1.6	4.28	5.8	2.51

the normal processes play the essential role in the phonon scattering by point defects together with Umklapp processes. In the temperature dependence of the thermal conductivity at about 100 K, we have observed the dip. This dip is qualitatively described by the Klemens—Callaway formula, when the relaxation time of the resonance phonon scattering is included in the combined relaxation time. It is assumed that complexes can be centers of resonance phonon scattering in the solid solutions.

1. *Aliyev M.I., Arasly D.H., Rahimov R.N.*// Dokl. AN Azerb. SSR.— 1982.— **38**, N 3.— P.21.
2. *Rahimov R.N.*// Ukr. J. Phys. — 2004.— **49**, N 2.— P.118—120.
3. *Aliyev M.I., Suleymanov Z.I., Arasly D.H., Rahimov R.N.*// Izv. AN SSSR, Neorgan. Mater.— 1981.— **17**, N 10.— P.1763.
4. *Drabl Dj., Goldsmid G.*// Heat Conductivity of Semiconductors.— Moscow, 1963 (in Russian).
5. *Wu J., Padture N.P., Klemens P.G. et al.*// J. Mater. Res.— 2002.— **17**, N 12.— P.3193—3200.
6. *Abeles B.*// Phys. Rev.— 1963.— **131**.— P.1906—1911.
7. *Gultyaev P.V., Petrov A.V.*// Fiz. Tverd. Tela.— 1959.— **1**.— P.368.
8. *Parrot J.E.*// Proc. Phys. Soc.— 1963.— **81**.— P.726—735.
9. *Callaway J.*// Phys. Rev.— 1959.— **112**.— P.1046—1051.
10. *Gaur N.K.S., Bhandari C.M., Verma G.S.*// Physica.— 1966.— **32**.— P.1048—1049.
11. *Ivanov A.I., Lukyanov A.N.*// Fiz. Nizk. Temp.— 2002.— **28**.— P.648—652.
12. *Guckelsbergert K., Briggst A.*// J. Phys. C: Solid State Phys.— 1975.— **8**.— P.L195—L198.
13. *Pal S.K., Kumar A.*// Phys. status solidi (b).— 1985.— **128**.— P.K15—K19.
14. *Arasly D.H., Rahimov R.N, Aliyev M.I.*// Fiz. Tekhn. Polupr.— 1990.— **24**, N 2.— P.365.
15. *Aliyev M.I, Khalilova A.A., Arasly D.H., Rahimov R.N.*// Izv. RAN. Neorgan. Mater.— 1997.— **33**, N 5.— P.539.
16. *Walker C.T., Pohl R.O.* //Phys. Rev.— 1963.— **131**.— P.1433.

Received 26.07.04

РОЗСІЯННЯ ФОНОНІВ У ТВЕРДОМУ РОЗЧИНІ $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$

Р.Н. Рагімов, Д.Г. Арасли, А.А. Халілова

Р е з ю м е

Досліджено теплопровідність твердих розчинів $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$ в інтервалі температур 80—300 К. Експериментальні результати проаналізовано на основі сучасної теорії термічної провідності твердих тіл. Встановлено суттєву роль N-процесів у розсіяння фононів у сплаві $(InSb)_{2(1-x)}$—$(In_2GeTe)_x$. Виявлене заглиблення на кривій температурної залежності теплопровідності пояснено резонансним розсіянням фононів на різних комплексах.