



P. V. Gorskiy

P. V. Gorskiy

Institute of Thermoelectricity of the NAS and MES
Ukraine, 1 Nauky str., Chernivtsi, 58029, Ukraine

**THE CORRELATION BETWEEN THE
LATTICE THERMAL CONDUCTIVITY OF
THERMOELECTRIC MATERIALS BASED
ON Zn-Cd-Sb AND THE PHASE DIAGRAM
OF THE SYSTEM**

Based on the analysis of phase diagrams of Zn-Sb, Cd-Sb and Zn-Cd-Sb systems, a correlation between the features of these diagrams and the lattice thermal conductivity of crystalline compounds existing in these systems was established. This correlation was analyzed with considerations of symmetry. Only those compounds and their modifications were considered which exist in crystalline state in the "generation" temperature range of 300-650 K which is relevant for thermoelectric application. Formulae for the components of lattice thermal conductivity tensors of all considered phases were derived with regard to symmetry elements which enter spatial groups to which the respective crystals belong. The lattice thermal conductivity was calculated with the use of the Boltzmann equation for phonons in relaxation time approximation. The relaxation time is determined with regard to normal processes of phonon-phonon scattering, as well as umklapp processes, taking into account different correlation between the frequency of interphonon collisions determined by these processes and phonon frequency. In this case, the symmetry elements determine the number of independent components of the velocity of longitudinal and transverse sound waves, the Gruneisen tensor, and the umklapp coefficient tensor characterizing the dependence of the frequency of umklapp collisions on the phonon frequency. In this way, the number of independent components of the lattice thermal conductivity tensor is determined for each phase. In the Zn-Cd-Sb system, phases with rhombic (including orthorhombic), hexagonal and cubic symmetry are possible. This means that the lattice thermal conductivity tensor can have three, two or only one independent component, respectively. In each case, analytical expressions for these components were obtained. In addition, analysis of the influence of point structural defects on the lattice thermal conductivity of Cd-Sb system was made.

Key words: phase diagram, lattice thermal conductivity, spatial symmetry, umklapp parameters, scattering by interstitial atoms

Introduction

Phase diagrams are known to reflect the equilibrium between different phases in a particular multicomponent system and determine the temperature and concentration intervals of their existence, since, as a rule, they are constructed by experimental or calculated methods in the "temperature-composition" coordinates. In so doing, the corresponding equilibrium curves in these diagrams reflect both the equilibrium between the liquid and solid states, for instance, with the same composition, and the equilibrium between the solid phases of different composition and crystal structure. The "liquid-crystal" equilibrium is decisive when choosing a technology for the production of semiconductor thermoelectric materials, for example, by growing single crystals from melts. The "crystal-crystal"

equilibrium, in turn, determines the composition and crystalline structure of the material that exists in the solid state in a particular temperature range. And since the crystal structure determines the group of spatial symmetry to which the material belongs (at least in the form of a single crystal), then it also determines the characteristics of the tensors of the kinetic coefficients of the material, namely, the thermal conductivity, the electrical conductivity, and the thermoelectric power. And these features determine the sphere of application of the material for thermoelectric energy conversion.

In turn, phase transitions between different solid phases in a system of two or more components are associated not only with a change in composition, but also with a change in the spatial symmetry of the material, even if the composition does not change. The investigation of the effect of such transitions on the lattice thermal conductivity of the Zn-Cd-Sb system is the purpose of this article.

The correlation between the components of the lattice thermal conductivity tensor of the Zn-Cd-Sb system and the features of its phase diagram

To date, the phase diagram of the Zn-Cd-Sb system has been studied most extensively in two sections of CdSb-ZnSb and Cd₄Sb₃-Zn₄Sb₃ [1]. Both these sections are not quasibinary, since Cd₄Sb₃ is a metastable phase, and ZnSb melts with decomposition. In themselves, ZnSb and CdSb are orthorhombic crystals of D_{2h}¹⁵ group and, apparently, have the lowest symmetry. The first Brillouin zone of these crystals is a rectangular parallelepiped, so that the tensors of the kinetic coefficients of these crystals in the absence of a magnetic field are diagonal, and each of them has, generally speaking, three independent and different components, characterizing the degree of influence of deformations, and consequently anharmonicity. The same symmetry property is possessed by the velocity of sound propagation in these crystals, the Grüneisen parameter, which characterizes the degree of influence of deformations, and, consequently, the anharmonicity of the lattice vibrations, on the phonon energy spectrum, as well as the umklapp parameter characterizing the dependence of the probability of interphonon collisions with the umklapp on the phonon frequency [2]. At the same time, the Debye temperature determined by the temperature dependence of specific heat, just as the specific heat of crystal itself, is a scalar.

Therefore, in the case of the lowest symmetry the following expressions hold true for the components of lattice thermal conductivity tensor [3]:

$$\kappa_{l11} = \frac{\pi\rho\hbar}{32\gamma_{11}^2\theta^3k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta)-1)^2} \left[\frac{(v_{1l})^{8/3} (v_{2l}v_{3l})^{2/3}}{x^4 + \mu_{11}x} + \frac{2(v_{1l})^{8/3} (v_{2l}v_{3l})^{2/3}}{x(3.125\theta^3 + \mu_{11})} \right], \quad (1)$$

$$\kappa_{l22} = \frac{\pi\rho\hbar}{32\gamma_{22}^2\theta^3k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta)-1)^2} \left[\frac{(v_{2l})^{8/3} (v_{1l}v_{3l})^{2/3}}{x^4 + \mu_{22}x} + \frac{2(v_{2l})^{8/3} (v_{1l}v_{3l})^{2/3}}{x(3.125\theta^3 + \mu_{22})} \right], \quad (2)$$

$$\kappa_{l33} = \frac{\pi\rho\hbar}{32\gamma_{33}^2\theta^3k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta)-1)^2} \left[\frac{(v_{3l})^{8/3} (v_{1l}v_{2l})^{2/3}}{x^4 + \mu_{33}x} + \frac{2(v_{3l})^{8/3} (v_{1l}v_{2l})^{2/3}}{x(3.125\theta^3 + \mu_{33})} \right]. \quad (3)$$

In these formulae, γ_{11} , γ_{22} , γ_{33} – are used to denote the components of the Gruneisen tensor, and μ_{11} , μ_{22} , μ_{33} , – the components of tensor of umkapp coefficients. Indices l, t refer to the longitudinal and

transverse legs, the components of tensors of the Gruneisen parameter and umklapp coefficients are considered to be independent of polarization of phonons. Moreover, T_D is the Debye temperature determined by the specific heat, $\theta = T/T_D$.

At high temperatures, when the Peierls law and the Leibfried-Schlemmann formula are valid, formulae (1) – (3) go over into the following:

$$\kappa_{l11} = \frac{\pi\rho\hbar}{32\gamma_{11}^2\theta k_B T_D^2} \left[(v_{1l})^{8/3} (v_{2l}v_{3l})^{2/3} F(\mu_{11}) + \frac{(v_{1l})^{8/3} (v_{2l}v_{3l})^{2/3}}{3.125\theta^3 + \mu_{11}} \right], \quad (4)$$

$$\kappa_{l22} = \frac{\pi\rho\hbar}{32\gamma_{22}^2\theta k_B T_D^2} \left[(v_{2l})^{8/3} (v_{1l}v_{3l})^{2/3} F(\mu_{22}) + \frac{(v_{2l})^{8/3} (v_{1l}v_{3l})^{2/3}}{3.125\theta^3 + \mu_{22}} \right], \quad (5)$$

$$\kappa_{l33} = \frac{\pi\rho\hbar}{32\gamma_{33}^2\theta k_B T_D^2} \int_0^1 \left[(v_{3l})^{8/3} (v_{1l}v_{2l})^{2/3} F(\mu_{33}) + \frac{(v_{3l})^{8/3} (v_{1l}v_{2l})^{2/3}}{3.125\theta^3 + \mu_{33}} \right]. \quad (6)$$

Function $F(\mu)$ is determined as follows:

$$F(\mu) = \int_0^1 \frac{x dx}{x^3 + \mu}. \quad (7)$$

In accordance with the phase diagram of the ternary system *Zn-Cd-Sb* constructed in [4], formulae of the form (1) – (6) with the corresponding parameters are valid for *ZnSb*, *CdSb*, an orthorhombic solid solution or the so-called triple ω phase $Zn_xCd_{1-x}Sb$, a monoclinic modification of β - Zn_4Sb_3 into which the γ - Zn_4Sb_3 compound transforms at 766 K, as well as for orthorhombic antimony and the high-temperature orthorhombic modification of η - Zn_3Sb_2 at a temperature below 720 K. These formulae are also formally valid for β - Cd_4Sb_3 and η - Cd_3Sb_2 though the above compounds are metastable.

However, in the *Zn-Cd-Sb* system, even higher-symmetry compounds are possible, for example, hexagonal or cubic. In the case of hexagonal symmetry, two axes lying in a plane perpendicular to the hexagonal axis are equivalent. Therefore, for the velocities of longitudinal and transverse phonons, as well as the components of the tensors that appear in formulas (1) – (6), the following relations hold:

$$\begin{aligned} v_{1l} = v_{2l} \equiv v_{l\perp}, v_{3l} \equiv v_{l\parallel}, v_{1t} = v_{2t} \equiv v_{t\perp}, v_{3t} \equiv v_{t\parallel}, \\ \gamma_{11} = \gamma_{22} \equiv \gamma_{\perp}, \gamma_{33} \equiv \gamma_{\parallel}, \mu_{11} = \mu_{22} \equiv \mu_{\perp}, \mu_{33} \equiv \mu_{\parallel}. \end{aligned} \quad (8)$$

Taking them into account, we obtain the following expressions for independent components of lattice thermal conductivity tensor of the hexagonal phases of *Zn-Cd-Sb* system:

$$\begin{aligned} \kappa_{l11} = \kappa_{l22} \equiv \kappa_{l\perp} = \frac{\pi\rho\hbar}{32\gamma_{\perp}^2\theta^3 k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta) - 1)^2} \times \\ \times \left[\frac{(v_{l\perp})^{10/3} (v_{l\parallel})^{2/3}}{x^4 + \mu_{\perp} x} + \frac{2(v_{t\perp})^{10/3} (v_{t\parallel})^{2/3}}{x(3.125\theta^3 + \mu_{\perp})} \right], \end{aligned} \quad (9)$$

$$\kappa_{l33} \equiv \kappa_{ll} = \frac{\pi\rho\hbar}{32\gamma_{\parallel}^2\theta^3k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta)-1)^2} \times \left[\frac{(v_{\parallel})^{10/3} (v_{\perp})^{2/3}}{x^4 + \mu_{\parallel}x} + \frac{2(v_{\parallel})^{10/3} (v_{\perp})^{2/3}}{x(3.125\theta^3 + \mu_{\perp})} \right]. \quad (10)$$

When the Peierls law and the Leibfried-Schlemmann formula are valid, formulae (9) and (10) go over into the following:

$$\kappa_{l\perp} = \frac{\pi\rho\hbar}{32\gamma_{\perp}^2\theta k_B T_D^2} \left[(v_{\perp})^{10/3} (v_{\parallel})^{2/3} F(\mu_{\perp}) + \frac{(v_{\perp})^{10/3} (v_{\parallel})^{2/3}}{3.125\theta^3 + \mu_{\perp}} \right], \quad (11)$$

$$\kappa_{ll} = \frac{\pi\rho\hbar}{32\gamma_{\parallel}^2\theta k_B T_D^2} \left[(v_{\parallel})^{10/3} (v_{\perp})^{2/3} F(\mu_{\parallel}) + \frac{(v_{\parallel})^{10/3} (v_{\perp})^{2/3}}{3.125\theta^3 + \mu_{\parallel}} \right]. \quad (12)$$

For single-crystal materials with a hexagonal symmetry, and, consequently, layered, and therefore superlattice, as a rule, inequality $\kappa_l > \kappa_{ll}$ is valid. This inequality holds mainly because sound propagation speed in the direction perpendicular to hexagonal axis is higher than along the hexagonal axis, i.e. perpendicular to layers.

Formulae (9) – (12) are valid for hexagonal limited solid solutions of cadmium and zinc, as well as for hexagonal modifications $\beta\text{-Zn}_4\text{Sb}_3$ and $\beta\text{-Cd}_4\text{Sb}_3$, though the latter of said compounds is metastable.

For phases with cubic symmetry the following relations are valid:

$$\begin{aligned} v_{1l} = v_{2l} = v_{3l} &\equiv v_l, v_{1t} = v_{2t} = v_{3t} \equiv v_t, \\ \gamma_{11} = \gamma_{22} = \gamma_{33} &\equiv \gamma, \mu_{11} = \mu_{22} = \mu_{33} \equiv \mu \end{aligned} \quad (13)$$

Consequently, for these phases the components of the lattice thermal conductivity tensor are compared to each other, and it turns into a scalar:

$$\kappa_{l11} = \kappa_{l22} = \kappa_{l33} \equiv \kappa_l = \frac{\pi\rho\hbar}{32\gamma^2\theta^3k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta)-1)^2} \left[\frac{v_l^4}{x^4 + \mu x} + \frac{2v_t^4}{x(3.125\theta^3 + \mu)} \right]. \quad (14)$$

When the Peierls law and the Leibfried-Schlemmann formula are valid, formula (14) goes over into the following:

$$\kappa_l = \frac{\pi\rho\hbar}{32\gamma^2\theta k_B T_D^2} \left[v_l^4 F(\mu) + \frac{v_t^4}{3.125\theta^3 + \mu} \right]. \quad (15)$$

Temperature dependences of the components of the lattice thermal conductivity tensors of cadmium and zinc antimonides obtained with the use of the derived relationships are given in Fig. 1a –b.

It can be seen from the plots that the approach used, in contrast to traditional approaches, makes it possible to obtain estimates of the components of the lattice thermal conductivity tensors of ZnSb and CdSb , consistent with the experimental data, and the degree of its anisotropy, which was mentioned in [3] (without citing specific temperature dependences). However, it should be borne in mind that total thermal conductivity of a single crystal, consisting of lattice and electron or hole components, is always determined experimentally.

These components in a variety of thermoelectric materials, in particular in cadmium and zinc antimonides, can be comparable. Therefore, in order to estimate the microscopic characteristics of phonon-phonon scattering, in particular, the umklapp coefficient, a preliminary separation of these components from each other is required. This separation is carried out exceptionally by calculation on the basis of experimental data on electrical conductivity, the Hall effect, and thermoEMF using the Wiedemann-Franz relation.

And temperature dependences of its anisotropy – in Fig.2a – b.

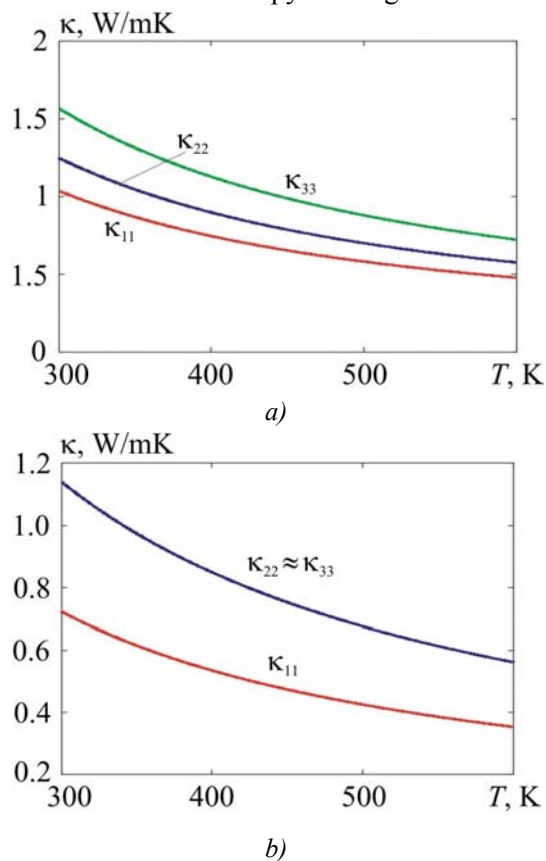
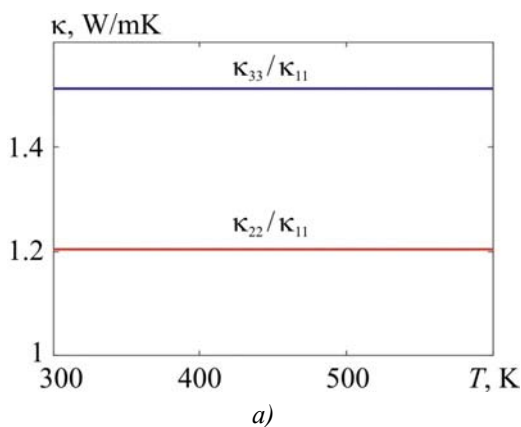


Fig. 1. Temperature dependences of the components of the lattice thermal conductivity tensors of zinc antimonide ZnSb (a) and cadmium antimonide CdSb



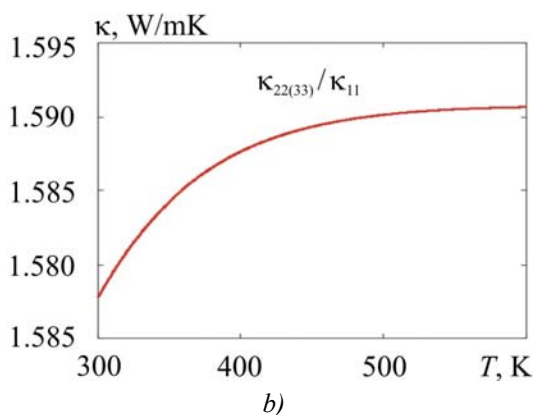


Fig. 2. Temperature dependences of thermal conductivity anisotropy of zinc antimonide ZnSb (a) and cadmium antimonide CdSb

The influence of point defects on the lattice thermal conductivity of Zn-Cd-Sb system

As long as [5] deals with the possibility of increasing the thermoelectric figure of merit of zinc antimonide due to introduction into its composition of a certain atomic fraction of cadmium isovalent impurity, it is of certain interest to study the influence of such impurities on the lattice thermal conductivity of zinc antimonide. The authors of [5] note that cadmium isovalent impurity sufficiently strongly reduces the lattice thermal conductivity of zinc antimonide. Presumably, this decrease may be due to the fact that the presence of such an impurity leads to the appearance of an additional mechanism for scattering of phonons due to the difference in the masses of cadmium and zinc atoms. We consider this effect in the isotropic approximation, assuming that the impurity atoms are randomly distributed in the bulk of a single crystal. In this case, in accordance with the approach described in [6], the relaxation time due to scattering by impurity atoms can be represented as:

$$\tau_i = \frac{4\pi\rho^2 v_1 v_2 v_3}{N_i (\Delta M)^2 \omega^4}. \quad (16)$$

In this formula, N_i – volumetric concentration of impurity atoms, $\Delta M = M_{Cd} - M_{Zn}$ – mass difference of cadmium and zinc atoms. The density of crystal ρ in this case should be understood to mean its density in the presence of isovalent impurity. In turn, for $Cd_x Zn_{1-x} Sb$ composition N_i can be determined as follows:

$$N_i = \frac{\rho N_A x}{2(xA_{Cd} + (1-x)A_{Zn} + A_{Sb})}. \quad (17)$$

In this formula, N_A – Avogadro number, A_{Cd} , A_{Zn} , A_{Sb} – tabular atomic masses of cadmium, zinc and antimony, respectively, other notations are explained above.

Therefore, (16) can be finally rewritten as:

$$\tau_i = \frac{8\pi\rho v_1 v_2 v_3 (xA_{Cd} + (1-x)A_{Zn} + A_{Sb})}{N_A x (M_{Cd} - M_{Zn})^2 \omega^4}. \quad (18)$$

Thus, in the most general form with regard to the contribution of transverse phonon legs, the lattice thermal conductivity can be represented as:

$$\kappa_{l11} = \frac{\pi\rho\hbar}{32\gamma_{11}^2\theta^3k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta)-1)^2} \times \left[\frac{(v_{1l})^{8/3} (v_{2l}v_{3l})^{2/3}}{x^4 (1 + \mu_{il11}) + \mu_{11}x} + \frac{2(v_{1l})^{8/3} (v_{2l}v_{3l})^{2/3}}{x(3.125\theta^3 + \mu_{11}) + \mu_{il11}x^4} \right], \quad (19)$$

$$\kappa_{l22} = \frac{\pi\rho\hbar}{32\gamma_{22}^2\theta^3k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta)-1)^2} \times \left[\frac{(v_{2l})^{8/3} (v_{1l}v_{3l})^{2/3}}{x^4 (1 + \mu_{il22}) + \mu_{22}x} + \frac{2(v_{2l})^{8/3} (v_{1l}v_{3l})^{2/3}}{x(3.125\theta^3 + \mu_{22}) + \mu_{il22}x^4} \right], \quad (20)$$

$$\kappa_{l33} = \frac{\pi\rho\hbar}{32\gamma_{33}^2\theta^3k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta)-1)^2} \times \left[\frac{(v_{3l})^{8/3} (v_{1l}v_{2l})^{2/3}}{x^4 (1 + \mu_{il33}) + \mu_{33}x} + \frac{2(v_{3l})^{8/3} (v_{1l}v_{2l})^{2/3}}{x(3.125\theta^3 + \mu_{33}) + \mu_{il33}x^4} \right]. \quad (21)$$

In these formulae, additional dimensionless parameters related to phonon scattering by impurities have the following values:

$$\mu_{il11} = \frac{3x(A_{Cd} - A_{Zn})^2 (v_{1l}v_{2l}v_{3l})^{2/3}}{128\gamma_{11}^2 [xA_{Cd} + (1-x)A_{Zn} + A_{Sb}]RT}, \quad (22)$$

$$\mu_{il22} = \frac{3x(A_{Cd} - A_{Zn})^2 (v_{1l}v_{2l}v_{3l})^{2/3}}{128\gamma_{22}^2 [xA_{Cd} + (1-x)A_{Zn} + A_{Sb}]RT}, \quad (23)$$

$$\mu_{il33} = \frac{3x(A_{Cd} - A_{Zn})^2 (v_{1l}v_{2l}v_{3l})^{2/3}}{128\gamma_{33}^2 [xA_{Cd} + (1-x)A_{Zn} + A_{Sb}]RT}, \quad (24)$$

$$\mu_{i11} = \frac{3x(A_{Cd} - A_{Zn})^2 (v_{1l}v_{2l}v_{3l})^{2/3}}{128\gamma_{11}^2 [xA_{Cd} + (1-x)A_{Zn} + A_{Sb}]RT}, \quad (25)$$

$$\mu_{i22} = \frac{3x(A_{Cd} - A_{Zn})^2 (v_{1l}v_{2l}v_{3l})^{2/3}}{128\gamma_{22}^2 [xA_{Cd} + (1-x)A_{Zn} + A_{Sb}]RT}, \quad (26)$$

$$\mu_{i33} = \frac{3x(A_{Cd} - A_{Zn})^2 (v_{1l}v_{2l}v_{3l})^{2/3}}{128\gamma_{33}^2 [xA_{Cd} + (1-x)A_{Zn} + A_{Sb}]RT}. \quad (27)$$

In formulae (22) – (27), R is a universal gas constant, other notations are explained above.

Let us estimate, for instance, μ_{il11} . At $v_{1l} = 3.803 \cdot 10^3$ m/s, $v_{2l} = 4.034 \cdot 10^3$ m/s, $v_{3l} = 3.834 \cdot 10^3$ m/s, $A_{Cd} = 112.41$, $A_{Zn} = 65.39$, $A_{Sb} = 121.75$, $\gamma_{11} = 1.5$, $x = 10^{-3}$, $T = 300$ K we get $\mu_{il11} = 7.463 \cdot 10^{-4}$, which is almost four orders of magnitude less than umklapp parameter [3]. Even if we assume $x = 0.5$, i.e. consider the composition $Cd_{0.5}Zn_{0.5}Sb$, still we will have $\mu_{il11} = 0.332$, which is more than an order of magnitude less than umklapp parameter. Moreover, scattering by point defects alone, without the action of other scattering mechanisms, in particular scattering with umklapp, in itself does not provide a finite

value of the lattice thermal conductivity, since the frequency dependence of the phonon scattering intensity on these defects is the same as for normal phonon-phonon scattering. Thus, the decrease in the lattice thermal conductivity during the transition from *ZnSb* to the ternary system $Cd_xZn_{1-x}Sb$ [5] cannot be explained by the presence of the "foreign" cadmium atoms themselves in the initial zinc antimonide lattice. It should be associated, on the one hand, with the rearrangement of this lattice, which leads to an increase in umklapp parameter, and on the other, to the appearance or increase in the concentration of such structural defects the phonon scattering intensity at which varies with a frequency more slowly than ω^4 . These can include, for instance, phase inclusions (inhomogeneities), dislocations and packing defects. Formulae for the relaxation time of phonons during scattering by these defects are given in [6] for the case of a crystal with a simple cubic lattice with one atom in the unit cell. The generalization of these formulae for the case of lattices of cadmium and zinc antimonides is far from being trivial and is not the purpose of this article.

Conclusions

1. The correlation between the lattice thermal conductivity of materials of the *Zn-Cd-Sb* system and its phase diagram is due to the existence in this system of solid phases with different spatial symmetry.
2. Since phases with orthorhombic (including orthorhombic), hexagonal and cubic symmetries can exist in the *Zn-Cd-Sb* system, the lattice thermal conductivity tensor for these phases can have three, two and one independent components, respectively.
3. In itself, the presence of "foreign" cadmium atoms in the ternary compound $Cd_xZn_{1-x}Sb$ at small x cannot explain the observed decrease in the lattice thermal conductivity of this compound in comparison with the lattice thermal conductivity of the initial *ZnSb*. Such a reduction is possible only if the addition of an isovalent cadmium impurity leads to reconstruction of the lattice, and, hence, to an increase in umklapp parameter, and also to an increase in the concentration of structural defects, the phonon scattering intensity at which increases with frequency more slowly than ω^4 , namely phase inclusions (inhomogeneities), dislocations and packing defects.

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