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## LATTICE THERMAL CONDUCTIVITY OF THERMOELECTRIC MATERIALS BASED ON Zn-Cd-Sb

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*Formulae for the components of the lattice thermal conductivity tensor of rhombic thermoelectric materials are derived. In the process of calculations, phonon-phonon scattering due to both normal processes and umklapp processes, is taken into account. Both the anisotropy of the sound velocity and the anisotropy of the Gruneisen tensor are taken into account. For the calculation of phonon relaxation time an approximation is used wherein this time, though anisotropic, depends on phonon frequency as a whole, rather than on the individual components of its quasi-momentum. The results of calculations are involved for the interpretation of experimental data on thermal conductivity anisotropy of cadmium and zinc antimonides. Comparison of the obtained results to the experimental data and other model theoretical approaches shows that when evaluating thermal conductivity anisotropy of rhombic crystals of cadmium and zinc antimonides one should take into account the anisotropy of the Gruneisen parameters and sound velocities, as well as frequency dependence of phonon relaxation time. However, as the Debye temperatures, their "scalar" values, determined by the calorimetric method, should be used, and not the components of the corresponding tensors determined on the basis of X-ray diffraction studies. Then it turns out that the "umklapp coefficient" which determines the frequency dependence of the intensity of the interphonon collisions due to the corresponding processes is also anisotropic. A fairly good agreement of the theoretical ratio of the components of the lattice thermal conductivity tensor of cadmium antimonide with the experimentally observed ratio of the components of the total thermal conductivity tensor indicates that the anisotropy of the lattice component of the thermal conductivity of cadmium antimonide is close to the anisotropy of the component due to free charge carriers.*

**Key words:** cadmium antimonide, zinc antimonide, symmetry, phonon spectrum, anisotropy, phonons, normal processes, umklapp processes, Gruneisen tensor, lattice thermal conductivity.

### Introduction

Despite the fact that currently bismuth telluride and alloys based on it are the dominant thermoelectric materials, there is an aspiration to replace them with other materials that do not contain tellurium. The need for a gradual abandonment of tellurium as a component of thermoelectric materials is due to a number of factors. Among them, mention should be made, in particular, of the high cost of tellurium, its limited production and reserves, its toxicity for living organisms and the environment, and the inoperability of materials based on the Bi (Sb)-Te (Se) system at high temperatures. At the same time, these shortcomings are lacking, for example, in cadmium and zinc antimonides. At room and lower temperatures, the thermoelectric figure of merit of these materials is low, so they are not used for the manufacture of thermoelectric coolers. However, they could be regarded as a worthy competitor to tellurium-containing materials as "generator" materials, since in

the range of 400 – 600 K their thermoelectric figure of merit sharply increases, and, moreover, can be substantially increased through optimization due to doping with a number of impurities in the appropriate concentrations [1].

It should be noted that thermoEMF anisotropy of cadmium and zinc antimonides, as well as possible origination in them of transverse thermoEMF, including due to thermal conductivity anisotropy, allows using these materials, especially cadmium antimonide, for the manufacture of anisotropic, including optical, thermoelements [2].

In view of the foregoing, the purpose of this paper is theoretical analysis of the mechanism of origination of lattice thermal conductivity anisotropy of rhombic crystals and application of the results to estimate of lattice thermal conductivity anisotropy of zinc and cadmium antimonides.

### **Analytical calculation of lattice thermal conductivity of rhombic crystals and discussion of the results**

Cadmium antimonide *CdSb*, as well as zinc antimonide *ZnSb*, is an orthorhombic crystal of group  $D_{2h}^{15}$ . The first Brillouin zone of these crystals is a rectangular parallelepiped, due to which the tensors of 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. The same symmetry property is typical of: sound propagation velocity in these crystals, the Gruneisen parameter characterizing the degree of influence of deformations, hence, of the anharmonism of lattice vibrations on the energy spectrum of phonons, the Debye temperature determined from X-ray diffraction studies, as well as umklapp parameter characterizing dependence of the possibility of interphonon collisions with umklapp on phonon frequency. At the same time, the Debye temperature, determined by calorimetric method, just as the specific heat of crystal, is a scalar. Based on these considerations, let us derive formulae which determine components of lattice thermal conductivity tensor of orthorhombic crystals.

Let us start with the calculation of  $\kappa_{111}$  component. We proceed from the general formula of thermal conductivity given in [3], according to which this component is equal to:

$$\kappa_{111} = \int_0^{\omega_D} \rho v_{1g}^2 \tau_{11}(\omega) dc_V(\omega). \quad (1)$$

In this formula:  $\rho$  – crystal density,  $v_{1g}$  – group sound velocity along the corresponding axis,  $\tau_{11}(\omega)$  – depending on phonon frequency as a whole, component of relaxation time tensor,  $dc_V(\omega)$  – differential contribution to crystal heat capacity at a constant volume.

To calculate the lattice thermal conductivity of a crystal from Eq. (1), it is also necessary to specify the model phonon spectrum of an orthorhombic crystal. Since in the Debye approximation the phonon spectrum of an isotropic crystal is linear with respect to the quasimomentum modulus, it is clear that in the simplest approximation for an orthorhombic crystal it can have, for example, the form:

$$\omega(\vec{k}) = \sqrt{v_1^2 k_1^2 + v_2^2 k_2^2 + v_3^2 k_3^2}, \quad (2)$$

In this formula,  $v_1, v_2, v_3$  – phase sound velocities in the directions of principal crystallographic axes,  $k_1, k_2, k_3$  – components of quasi-momentum along the same axes. Therefore, formula (1) can be transformed as follows:

$$\kappa_{l11} = \int_0^{\omega_D} \frac{M}{V} v_1^2 \tau_{11}(\omega) d\left(\frac{dE}{MdT}\right) = \frac{\hbar^2}{(kT)^2} \int_0^{\omega_D} \Gamma_{pl1}(\omega) \tau_{11}(\omega) \frac{\omega^2 \exp(\hbar\omega/kT)}{[\exp(\hbar\omega/kT) - 1]^2} d\omega, \quad (3)$$

where

$$\begin{aligned} \Gamma_{pl1}(\omega) &= \iiint v_1^2(\vec{k}) \delta(\omega - \omega(\vec{k})) d\tau_{\vec{k}} = \iiint \frac{v_1^4 k_1^2}{(2\pi)^3 (v_1^2 k_1^2 + v_2^2 k_2^2 + v_3^2 k_3^2)} \times \\ &\times \delta\left(\omega - \sqrt{v_1^2 k_1^2 + v_2^2 k_2^2 + v_3^2 k_3^2}\right) dk_1 dk_2 dk_3 = \iint \frac{2v_1 \sqrt{\omega^2 - (v_2^2 k_2^2 + v_3^2 k_3^2)}}{(2\pi)^3 \omega} dk_2 dk_3 = \\ &= \frac{v_1}{4\pi^3} \int_0^\omega \frac{\sqrt{\omega^2 - \omega_1^2} 2\pi \omega_1 d\omega_1}{\omega v_2 v_3} = \frac{v_1 \omega^2}{6\pi^2 v_2 v_3}. \end{aligned} \quad (4)$$

In formula 3,  $M$  is crystal mass,  $V$  is crystal volume.

The relaxation time of longitudinal phonons in normal scattering in accordance with the procedure developed in [4] for an orthorhombic crystal can be represented in the following form

$$\tau_{pn11}(\omega) = \frac{3\pi v_c^5 \rho}{16\gamma_{11}^2 k_B T \omega^4} = \frac{3\pi \rho (v_1 v_2 v_3)^{5/3}}{16\gamma_{11}^2 k_B T \omega^4}. \quad (5)$$

In this formula,  $v_c = \sqrt[3]{v_1 v_2 v_3}$ ,  $\gamma_{11}$  – the Gruneisen tensor component.

However, normal processes occur with retention of full momentum of phonon subsystem and, therefore, they do not yield final value of lattice thermal conductivity. At the same time, in the temperature range typical for the application of "generator" thermoelectric materials (TEM), the umklapp processes play a decisive role, for which the frequency of interphonon collisions is proportional to the phonon frequency. Taking into account these processes in addition, as well as the contribution of transverse phonon legs, we find the following final expressions for the components of lattice thermal conductivity tensor:

$$\kappa_{l11} = \frac{\pi \rho \hbar}{32\gamma_{11}^2 \theta^3 k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta) - 1)^2} \left[ \frac{(v_{1l})^{8/3} (v_{2l} v_{3l})^{2/3}}{x^4 + \mu_{11} x} + \frac{2(v_{1l})^{8/3} (v_{2l} v_{3l})^{2/3}}{x(3.125\theta^3 + \mu_{11})} \right], \quad (6)$$

$$\kappa_{l22} = \frac{\pi \rho \hbar}{32\gamma_{22}^2 \theta^3 k_B T_D^2} \int_0^1 \frac{x^4 \exp(x/\theta) dx}{(\exp(x/\theta) - 1)^2} \left[ \frac{(v_{2l})^{8/3} (v_{1l} v_{3l})^{2/3}}{x^4 + \mu_{22} x} + \frac{2(v_{2l})^{8/3} (v_{1l} v_{3l})^{2/3}}{x(3.125\theta^3 + \mu_{22})} \right], \quad (7)$$

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

In these formulae, through  $\gamma_{11}, \gamma_{22}, \gamma_{33}$  we denote the Gruneisen tensor components, and through  $\mu_{11}, \mu_{22}, \mu_{33}$  – the components of umklapp coefficients. Indexes  $l, t$  refer to the longitudinal and transverse legs, the components of the Gruneisen parameter tensors and the umklapp coefficients are assumed to be independent of the phonon polarization. Besides,  $T_D$  – caloric Debye temperature,  $\theta = T/T_D$ .

At high temperatures, when the Peierls law and the Leibfried-Schlemann formula are valid, formulae (6) – (8) go over into the following:

$$\kappa_{l11} = \frac{\pi\rho\hbar}{32\gamma_{11}^2\theta k_B T_D^2} \left[ (v_{1l})^{8/3} (v_{2l}v_{3l})^{2/3} F(\mu_{11}) + \frac{(v_{1t})^{8/3} (v_{2t}v_{3t})^{2/3}}{3.125\theta^3 + \mu_{11}} \right], \quad (9)$$

$$\kappa_{l22} = \frac{\pi\rho\hbar}{32\gamma_{22}^2\theta k_B T_D^2} \left[ (v_{2l})^{8/3} (v_{1l}v_{3l})^{2/3} F(\mu_{22}) + \frac{(v_{2t})^{8/3} (v_{1t}v_{3t})^{2/3}}{3.125\theta^3 + \mu_{22}} \right], \quad (10)$$

$$\kappa_{l33} = \frac{\pi\rho\hbar}{32\gamma_{33}^2\theta k_B T_D^2} \int_0^1 \left[ (v_{3l})^{8/3} (v_{1l}v_{2l})^{2/3} F(\mu_{33}) + \frac{(v_{3t})^{8/3} (v_{1t}v_{2t})^{2/3}}{3.125\theta^3 + \mu_{33}} \right]. \quad (11)$$

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

$$F(\mu) = \int_0^1 \frac{x dx}{x^3 + \mu}. \quad (12)$$

We apply these results to estimate the anisotropy of lattice thermal conductivity of zinc and cadmium antimonides. However, first of all, we note that in [5] the following relation was used to estimate the lattice thermal conductivity of rhombic crystals and its anisotropy:

$$\kappa_{lji} \propto T_{Di}^3 / \gamma_{ii}^2, \quad (13)$$

In this formula,  $T_{Di}$  – the Debye temperatures determined from X-ray diffraction data,  $\gamma_{ii}$  – the Gruneisen tensor components.

To compare our results with the results obtained within model (13) for *ZnSb*, we use the data given in Table [5].

Table

*Elastic constants (in the Voigt notation) and the Gruneisen tensor components of ZnSb single crystal*

$c_{11}$	$c_{22}$	$c_{33}$	$c_{44}$	$c_{55}$	$c_{66}$	$c_{12}$	$c_{23}$	$c_{13}$	$\gamma_{11}$	$\gamma_{22}$	$\gamma_{33}$
9.22	10.38	9.38	2.13	4.65	3.46	3.31	3.10	3.80	1.30	1.08	0.86

Elastic constants are given in units of  $10^{10}$  Pa.

Moreover, the Debye temperatures determined from X-ray diffraction data are equal to:  $T_{D1} = 223$  K,  $T_{D2} = 271$  K,  $T_{D3} = 283$  K.

Sound velocities for the longitudinal and transverse waves in orthorhombic crystal can be defined by the formulae:

$$\begin{aligned} v_{1l} = \sqrt{c_{11}/\rho}; v_{1t} = \sqrt{2c_{55}c_{66}/\rho(c_{55} + c_{66})}; v_{2l} = \sqrt{c_{22}/\rho}; v_{2t} = \sqrt{2c_{44}c_{66}/\rho(c_{44} + c_{66})}; \\ v_{3l} = \sqrt{c_{33}/\rho}; v_{3t} = \sqrt{2c_{44}c_{55}/\rho(c_{44} + c_{55})}. \end{aligned} \quad (14)$$

Taking into account that zinc antimonide density according to [6] is  $6380 \text{ kg/m}^3$ , we obtain the

following values of said velocities (in m/s):

$$\begin{aligned} v_{1l} &= 3.802 \cdot 10^3; v_{1t} = 2.494 \cdot 10^3; v_{2l} = 4.034 \cdot 10^3; \\ v_{3l} &= 2.033 \cdot 10^3; v_{3t} = 3.834 \cdot 10^3; v_{3t} = 2.140 \cdot 10^3. \end{aligned} \quad (15)$$

Using formula (13) for the estimate of thermal conductivity anisotropy we find that components of lattice thermal conductivity tensor of zinc antimonide correlate as 1.0:2.6:5.2.

However, the procedure used in [5] does not take into account either the contribution of transverse phonons or the fact that the temperature whereby said estimate is performed is not substantially higher than the Debye temperature, or sound velocity anisotropy in crystal. Therefore, we clarify the impact of these factors on lattice thermal conductivity anisotropy of *ZnSb*. Note that due to limited amount of experimental data on this matter we have to assume that “umklapp parameter”  $\mu$  is isotropic and, hence, thermal conductivity anisotropy is governed only by the anisotropy of sound velocity, the Gruneisen tensor and X-ray characteristic temperatures, if such are used in formulae (6) – (8) instead of isotropic caloric Debye temperature.

In this case, we select the value of the "umklapp parameter" from the requirement of "coincidence of theory with experiment", i.e. in such a way that thermal conductivity averaged over the directions, which, as can easily be shown, is equal to the "experimental" value given in the handbook [6]; i.e. 1.4 W/(m·K) at 293 K. Then, taking into account the above values of *ZnSb* parameters, we get  $\mu = 4.997$ , and, hence,  $\kappa_{111} = 0.968$  W/(m·K),  $\kappa_{122} = 1.256$  W/(m·K),  $\kappa_{133} = 1.977$  W/(m·K). These values correlate as 1:1.298:2.042, which is substantially lower than in conformity with formula (13).

If, however, we assume, as it is done in [5], that “experimental” value of thermal conductivity is equal to 2 W/(m·K) at 293 K, we obtain that  $\mu = 3.328$ , and, hence,  $\kappa_{111} = 1.375$  W/(m·K),  $\kappa_{122} = 1.800$  W/(m·K),  $\kappa_{133} = 2.826$  W/(m·K). These values correlate as 1:1.309:2.055, i.e. such a correction does not significantly change thermal conductivity anisotropy.

However, if in formulae (6) – (8) instead of X-ray characteristic temperatures we use isotropic caloric Debye temperature 225 K and assume “experimental” value of thermal conductivity equal to 1.4 W/(m·K) at 293 K, we get that  $\mu = 5.973$ , and, hence,  $\kappa_{111} = 0.821$  W/(m·K),  $\kappa_{122} = 1.301$  W/(m·K),  $\kappa_{133} = 2.078$  W/(m·K). These values correlate as 1:1.585:2.531. Strange as it may seem, within the framework of the above approach, the use of the isotropic Debye temperature slightly increases the expected anisotropy of the lattice thermal conductivity. The latter estimate of thermal conductivity anisotropy seems to be “most shrewd”, but a deviation from it may indicate, for instance, the anisotropy of umklapp coefficient  $\mu$ .

We now proceed to estimate of cadmium antimonide thermal conductivity anisotropy. Moduli of elasticity of cadmium antimonide at temperature 293 K in units  $10^{10}$  Pa are equal to [7]:  $c_{11[100]} = 7.97$ ;  $c_{22[010]} = 9.50$ ;  $c_{33[001]} = 8.40$ ;  $c_{44[001]} = 1.257$ ;  $c_{44[010]} = 1.259$ ;  $c_{55[001]} = 2.982$ ;  $c_{55[100]} = 2.997$ ;  $c_{66[010]} = 1.883$ ;  $c_{66[001]} = 1.867$ .

By virtue of this, with regard to cadmium antimonide density equal to 6900 kg/m<sup>3</sup> [7], the values of sound velocities in this single crystal along the principal directions are, respectively, equal to:  $v_{1l} = 3.399 \cdot 10^3$  m/s;  $v_{1t} = 1.828 \cdot 10^3$  m/s;  $v_{2l} = 3.711 \cdot 10^3$  m/s;  $v_{2t} = 1.477 \cdot 10^3$  m/s;  $v_{3l} = 3.489 \cdot 10^3$  m/s;  $v_{3t} = 1.602 \cdot 10^3$  m/s.

Components of the Gruneisen tensor of *CdSb* have the following values [7]:  $\gamma_{11} = 1.28$ ;  $\gamma_{22} = 0.48$ ;  $\gamma_{33} = 0.64$ . In so doing, the characteristic Debye temperatures are equal to:  $T_{D1} = 180$  K,  $T_{D2} = 215$  K,  $T_{D3} = 204$  K [7].

Guided by thermal conductivity value of *CdSb* given in [6] and equal to 1 W/(m·K), on the assumption of umklapp parameter isotropy, but with regard to the anisotropy of X-ray characteristic Debye temperatures, we get  $\mu = 7.256$ , and, hence,  $\kappa_{11l} = 0.696$  W/(m·K),  $\kappa_{22l} = 0.894$  W/(m·K),  $\kappa_{33l} = 1.41$  W/(m·K). These values of components of lattice thermal conductivity tensor correlate as 1:1.284:2.026. Using caloric isotropic Debye temperature equal to 180 K, we get  $\mu = 8.651$ , and, hence,  $\kappa_{11l} = 0.59$  W/(m·K),  $\kappa_{22l} = 0.925$  W/(m·K),  $\kappa_{33l} = 1.485$  W/(m·K). These values correlate as 1:1.568:2.517. At the same time, the estimate of lattice thermal conductivity anisotropy by formula (13) yields the ratio 1:12.118:5.823, which, as we shall see later, is obviously exaggerated in comparison with the true one.

From the experimental data on thermal conductivity anisotropy of hole-type cadmium antimonide presented in [8] it follows that in given material this anisotropy is fundamentally different from electric conductivity anisotropy. Namely, for electric conductivity components the ratio  $\sigma_{22} < \sigma_{11} < \sigma_{33}$  is valid, and for thermal conductivity components – the ratio  $\kappa_{11} < \kappa_{22} \approx \kappa_{33}$ . If scattering of free charge carriers is assumed to be isotropic, then, with regard to the Wiedemann-Franz relation, such anisotropy is attributable to essential contribution of lattice thermal conductivity. If, however, scattering of free charge carriers is anisotropic, it can also contribute to increase or reduction of thermal conductivity anisotropy. Moreover, it can testify to the anisotropy of umklapp coefficient in *CdSb*.

We will estimate thermal conductivity anisotropy of *CdSb* with regard to the above facts. For this purpose we will make a model assumption that two of the three components umklapp coefficient tensor coincide. Under this assumption the following variants are possible: 1) on the assumption that  $\mu_{11} = \mu_{22} \neq \mu_{33}$ , taking into account caloric (scalar) Debye temperature we obtain that  $\mu_{11} = \mu_{22} = 6.907$ ,  $\mu_{33} = 11.533$ ,  $\kappa_{11} = 0.722$  W/(m·K),  $\kappa_{22} = \kappa_{33} = 1.139$  W/(m·K), and then the ratio  $\kappa_{22}/\kappa_{11} = 1.578$ ; 2) on the assumption that  $\mu_{11} = \mu_{33} \neq \mu_{22}$ , taking into account caloric (scalar) Debye temperature, we obtain that  $\mu_{11} = \mu_{33} = 10.423$ ,  $\mu_{22} = 6.235$ ,  $\kappa_{11} = 0.498$  W/(m·K),  $\kappa_{22} = \kappa_{33} = 1.251$  W/(m·K), and then the ratio  $\kappa_{22}/\kappa_{11} = 2.512$ ; 3) on the assumption that  $\mu_{11} = \mu_{22} \neq \mu_{33}$ , taking into account anisotropic X-ray characteristic Debye temperature, we obtain  $\mu_{11} = \mu_{22} = 5.895$ ,  $\mu_{33} = 9.662$ ,  $\kappa_{11} = 0.837$  W/(m·K),  $\kappa_{22} = \kappa_{33} = 1.082$  W/(m·K), and then the ratio  $\kappa_{22}/\kappa_{11} = 1.293$ ; 4) on the assumption that  $\mu_{11} = \mu_{33} \neq \mu_{22}$ , taking into account anisotropic X-ray characteristic Debye temperature, we obtain  $\mu_{11} = \mu_{33} = 8.627$ ,  $\mu_{22} = 5.246$ ,  $\kappa_{11} = 0.596$  W/(m·K),  $\kappa_{22} = \kappa_{33} = 1.202$  W/(m·K), and then the ratio  $\kappa_{22}/\kappa_{11} = 2.017$ . Note that the estimate of thermal conductivity anisotropy by the variant “1”, i.e. 1.578, is closest to the experimentally observed, as long as according to the evidence from [2] the above relation is 1.68, and, thus, the error is 6.2 % from the larger value. However, according to the evidence from [2], the averaged over directions thermal conductivity value at 300 K is 1.6 W/(m·K), and the value from handbook [6] – 1 W/(m·K). Both values can be simultaneously true only in the case when the higher of them characterizes full thermal conductivity of *CdSb* single crystal, and the lower – its lattice component. Therefore, it can be concluded that lattice thermal conductivity anisotropy of cadmium antimonide is close to component anisotropy due to free charge carriers.

## Conclusions

1. Lattice thermal conductivity anisotropy of cadmium and zinc antimonides is due to anisotropy of sound velocity, the Gruneisen tensor and phonon umklapp parameter.
2. The use of anisotropic X-ray characteristic Debye temperature instead of its isotropic caloric value reduces the expected estimate of lattice thermal conductivity anisotropy.
3. Estimate of lattice thermal conductivity anisotropy of cadmium and zinc antimonides without regard to sound velocity anisotropy and the frequency dependence of phonon relaxation time in said materials results in drastic overrating of this anisotropy value as compared to the experimental data.

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