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HEAT CONDUCTIVITY OF SOME LAYERED STRUCTURES

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Abstract: Callaway model with Debye's approximation of phonon states density is used to determine heat conductivity of some layered structures of $Nb_{1-x}Sn_xSe_2$ type. In total relaxation time, a term proportional to squared frequency, typical for layered structures, is kept. Electronic heat conductivity is determined by Wiedemann–Franz law and BRT model for superconductors. In both cases, heat conductivity is determined numerically in the range of 2 – 200 K. Results correlate well with experimental data.

Keywords: phonons, Callaway model, Debye's approximation, heat conductivity.

1. INTRODUCTION

For devices to work more efficiently they should have lower operating temperature, i.e. there should be higher thermal conductivity of working elements. Therefore, the study of thermal conductivity is of great importance.

The fact is that the thermal properties of nanostructures are affected by their dimensions. Thermal conductivity of the films is a function of film thickness. In superlattices and nanowires thermal conductivity depends on the phonon properties of two distinct layers and the conditions at the interface. Optimizing the thickness of layers and number of layers in some particular device could make progress and increase thermal conductivity. Also, a question of control of phonon transport phenomena is raised in order to provide the desired thermal performance.

Thermal conductivity tensor in its most general form can be found by using the Kubo formula in which the pair correlation function figures of the heat flux operator. In the first approximation, finding the thermal conductivity tensor can be reduced to solving the Boltzmann's transport equation [4].

The model most used for the analysis of experimental data for heat conductivity of crystal lattice is Callaway's which includes different relaxation processes of phonons. In Callaway's model a series of approximations is made which limits the application of this model to the area of low temperatures [3]. Approximations are as followed:

1. Debay's approximation is used to describe the phonon spectrum
2. $$D_D(\omega)=\begin{cases}\dfrac{3V\omega^2}{2\bar{v}^3} & \omega<\omega_D\\ 0 & \omega>\omega_D\end{cases};\ \omega_D=\frac{\bar{v}}{a}\sqrt[3]{6\pi^2}$$
3. a - is the constant of crystal lattice
4. Takes into account only the average speed of the sound
5. Phonon scattering on the surface is diffused (not mirrored).
6. Normal three-phonon processes of scattering occur for low frequency longitudinal phonons
7. Relaxation time in U-process is described as similar to relaxation time for normal processes
8. Relaxation times of various phonon scattering processes are considered to be additive
9. Neither the crystal anisotropy nor polarization of phonons are taken into account (no distinction between longitudinal and transverse phonons)

Total thermal conductivity is expressed as:

$$\kappa=\kappa_e+\kappa_f \tag{1}$$

where κ_e is thermal conductivity of electrons, while κ_f is thermal conductivity of phonons.

Stationary character of heat transport process is provided through several mechanisms of relaxation. For electron heat conductivity those mechanisms are as follows:

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- Collisions of electrons with impurities κ_{ed}
- Collisions of electrons with phonons κ_{ef}
- Collisions of electrons with electrons κ_{ee}
- Relaxation mechanisms of lattice thermal conductivity are:
- Collisions of phonons with impurities κ_{fd}
- Collisions of phonons with electrons κ_{fe}
- Collisions of phonons with phonons κ_{ff}

Thermal resistance of an appropriate type of carrier can be written as the sum of thermal resistance corresponding to the different mechanisms:

$$\begin{aligned}\kappa_e^{-1} &\approx \kappa_{ed}^{-1} + \kappa_{ef}^{-1} + \kappa_{ee}^{-1} \\ \kappa_f^{-1} &\approx \kappa_{fd}^{-1} + \kappa_{fe}^{-1} + \kappa_{ff}^{-1}\end{aligned} \tag{2}$$

Depending on the conditions, different mechanisms have different effects. In metals, the main contribution to heat transfer is formed by an electronic component, while the phonon contribution makes only a few percent of the total thermal conductivity. There is an opposite situation with semiconductors and isolators in which the phonon thermal conductivity is relatively high compared to the electronic thermal conductivity.

2. $NBSE_2$ THERMAL CONDUCTIVITY

The analyzed compound $NbSe_2$ has a hexagonal crystal structure (layer structure-layer along which the transfer is made of heat and charge, will be marked as a layer ab) and for the interval $T = 4.5 - 7K$ temperature exceeds in the superconductive state [2]. If it is doped with Sn the compound $Nb_{1-x}Sn_xSe_2$ for $x < 0.5$ has the properties of metal and for $x \geq 0.5$ it has semiconductor properties. This compound is interesting from the aspect of thermal analysis because: it has a layered structure (the influence of dimensions), it transits from normal to superconducting state and depending on stoichiometry rate of Nb and Sn it has metal or semiconductor properties.

In the case of $x < 0.5$, the compound has metal conductivity and the main share in thermal conductivity is electronic thermal conductivity which is calculated by Wiedeman-Franz law as:

$$\kappa_e^n = \frac{L_0 T}{\rho} = \frac{L_0 T}{\rho_0 + A\left(\frac{\theta}{T}\right)^3 J_3\left(\frac{\theta}{T}\right)} \tag{3}$$

where $\rho_0 = 5.56 \cdot 10^{-8}\ \Omega\text{m}$, $A = 5.85 \cdot 10^{-6}\ \Omega\text{m}$, $\theta = 222\,\text{K}$ is Debay's temperature, and

$J_3\left(\frac{\theta}{T}\right) = \int_0^{\frac{\theta}{T}} \frac{z^3 e^z}{\left(e^z - 1\right)^2} dz$ is Debay's integral.

Graphical representation of dependence of equation (3) is given in Figure 1.

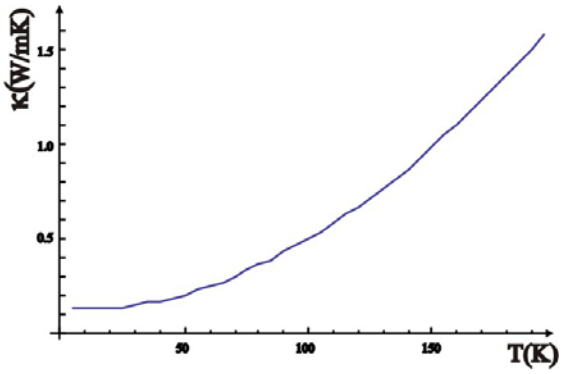

Figure 1. Dependence of κ_e^n on temperature

The phonon component of heat conductivity is determined from the expression for heat conductivity of bulk while the layered structure is taken into account through additional term $B_3\,\omega^2$ in total relaxation time [6]:

$$\begin{aligned}\kappa_f^n &= \frac{1}{3}\int_0^{\frac{\theta}{T}} v^2 \tau(x,T) C_v(x) dx = \\ &= \frac{k_B}{2\pi^2 v}\left(\frac{k_B}{\hbar}\right)^3 T^3 \int \frac{x^4 e^x}{\left(e^x - 1\right)^2 \tau^{-1}(x,T)} dx\end{aligned} \tag{4}$$

where k_B is Boltzman's constant, $\hbar$ - Dirac's constant, $x = \frac{\hbar\omega}{k_B T}$, $v = 1.75 \cdot 10^3$ m/s is the average speed of sound in compound, $\tau(x,T)$ - is total relaxation time of phonons. Relaxation time of phonons is determined from

$$\tau^{-1}(x,T) = \frac{v}{d} + B_1\omega^4 + B_3\omega^2 \tag{5}$$

where the first term corresponds to the scattering of phonons on borders (d is the corresponding length of the average free path), the second term corresponds to scattering of phonons on dot impurities and the third term is typical for scattering of phonons on dot impurities for layered compounds.

For the case where $x > 0.5$ the compound is acting as a semiconductor and phonon component of thermal conductivity dominates. In this case, thermal conductivity is determined by expression (5) with corrected relaxation time

$$\tau^{-1}(x,T) = \frac{v}{d} + B_1\omega^4 + B_3\omega^2 + B_2 T\,\omega^3 e^{-\frac{\theta}{bT}} \qquad (6)$$

where the last term describes scattering of phonons on phonons (the so-called U-processes).

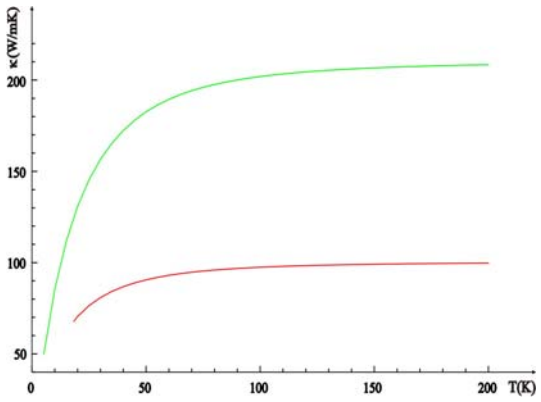

Figure 2. Dependence of κ_f^n on temperature (green line corresponds $x = 0$, while red line corresponds $x = 0.30$)

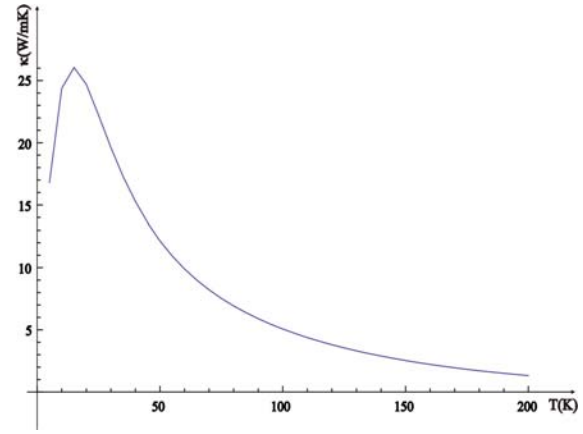

Figure 3. Dependence of κ_f^n on temperature ($x = 0.60$)

When the compound has entered the superconducting state electronic thermal conductivity is given as [1]:

$$\kappa_e^s = \frac{L_0 T}{\rho_0 + A\left(\frac{\theta}{T}\right)^3 J_3\left(\frac{\theta}{T}\right)} \times \times \frac{2\int_0^\infty \frac{x\,dx}{e^{x+y}+1} + 2y\ln\left(1+e^{-y}\right) + \frac{y^2}{1+e^y}}{2\int_0^\infty \frac{x\,dx}{e^x+1}} \qquad (7)$$

where $y = 1.1\frac{k_B T_C}{k_B T} = \frac{7.7}{T}$. Just fitted value $\frac{\Delta(0)}{k_B T_C} = 1.1$ is better for presentation of experimental data for thermal conductivity of specified layered compound [2] than the ratio given by *BCS* theory the value of which is $\frac{\Delta(0)}{k_B T_C} = 1.76$.

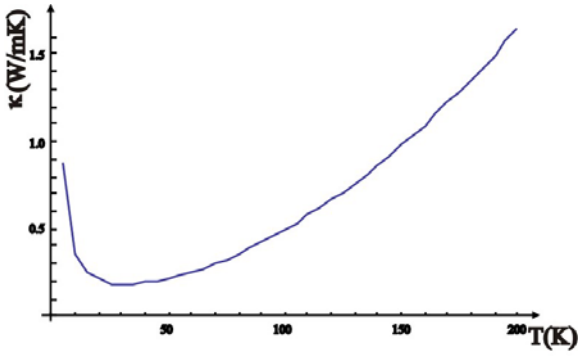

Figure 4. Dependence of κ_e^s on temperature

In superconducting state, phonon thermal conductivity is determined from (4) where the total relaxation time is corrected by adding one more term [1,2] which describes scattering of phonons on electrons:

$$B_4\,\omega\left(1-e^x\right)\sqrt{\frac{2\frac{7.7}{T}}{x+2\frac{7.7}{T}}}\,e^{\frac{x}{2}+\frac{7.7}{T}} \times \times \left\{K_1\left(\frac{x}{2}\right) + \frac{\left(3x+4\frac{7.7}{T}\right)x}{8\frac{7.7}{T}\left(x+2\frac{7.7}{T}\right)}\left[K_1\left(\frac{x}{2}\right) - K_0\left(\frac{x}{2}\right)\right]\right\} \qquad (8)$$

where $K_1\left(\frac{x}{2}\right)$ and $K_0\left(\frac{x}{2}\right)$ are Besels functions of the first and zeroth order of an imaginary argument.

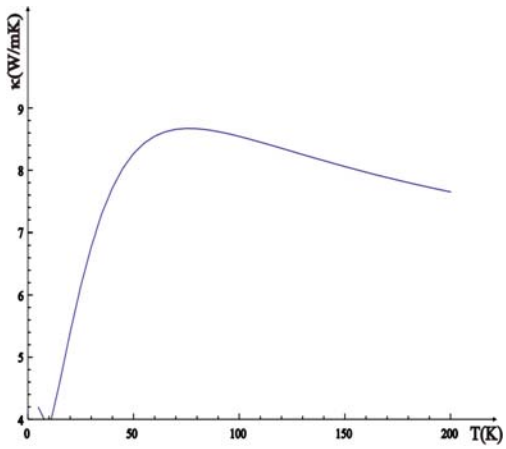

Figure 5. Dependence of κ_f^s on temperature

Parameters of heat conductivity are taken from [2] and are given in table 1.

Table 1. Heat conductivity parameters

Parameter	*x*		
	0.00	0.30	0.60
$B_1\ 10^{-42}\ s^3$	1.25	4.05	14.3
$B_2\ 10^{-30}\ K^{-1}s^2$			1.34
$B_3\ 10^{-17}\ s$	5.15	7.7	12.8
d mm	110	75	190
b			6
$B_4\ 10^{-4}$	0.3	200	

All calculations are performed numerically in the temperature interval from 2 – 200 K with the help of *Mathematica 7.0*.

3. CONCLUSION

As it can bee seen from Figure 1, electronic thermal conductivity of a compound with metal conductivity $x < 0.5$ increases with temperature, especially for the temperature interval above 100 K. As phonon heat conductivity for materials with semiconductor properties, e.g. when $x > 0.5$, does not increase in this temperature interval, Figure 2 and 3, it is clear that electronic subsystem is responsible for this increase. Estimation of the share of electronic thermal conductivity in the overall thermal conductivity for the case of normal state and $x < 0.5$ are around 10-30%. In the area of temperatures below 10 K, phonon-phonon interaction is negligible. In the layered materials acoustic phonons polarized normally to layer ab do not participate in the processes of scattering on electrons [5]. Therefore, the coefficient B_4 for them is zero. In the area of superconductive transition, all phonons are with equal probability scattered on electrons. Electron-phonon interaction is the highest for the compounds with $x = 0.3$. Coefficients B_1 and B_3 are of higher values when x is higher, indicating larger defects in the crystal structure of the compounds in which Nb replaces Sn, than in the case of a pure compound.

4. ACKNOWLEDGMENT

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ТОПЛОТНА ПРОВОДНОСТ НЕКИХ СЛОЈЕВИТИХ СТРУКТУРА

Сажетак: За израчунавање топлотне проводности код неких слојевитих структура типа $Nb_{1-x}\,Sn_x\,Se_2$ користи се Калавејев модел са Дебајевом апроксимацијом густине фононских стања. У укупном времену релаксације узет је и члан пропорционалан квадрату фреквенције који је типичан за слојевите структуре. Електронска топлотна проводност одређује се из Видеман−Францовог закона и БРТ модела за суперпроводнике. У оба случаја топлотна проводност се одређује нумерички у интервалу од 2 до 200 K. Резултати су у доброј сагласности са експерименталним подацима.

Кључне речи: фонони, Калајев модел, Дебајева апроксимација, топлотна проводност.