THERMOELECTROMOTIVE FORCE AND THERMAL CONDUCTIVITY IN Ag_2Te

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In this work the temperature dependences of the thermoelectromotive force and the thermal conductivity in Ag_2Te have been analyzed at next approaches the elektron spectrum dispersion law, the charge carries scattering mechanizm, Debay model of fonon spectrum and Kallarvay thery of the relaxation time.

The telluride of argentum (Ag_2Te) is respected to compounds by the type $Ag_2{}^{I}X^{VI}$ (X-Te, Se, S), having the polimorphysm property [1], caused by the content of the big amount of intrinsic defects [2] - metal atoms in the interstices and vacancies in the lattice points of crystals. The number of these defects increases with the temperature increase and at the given temperature T the disorder of the cation sublattice appears with the simultaneous reconstruction of the close lattice of chalcogen, that leads to the phase transformation (PT). The excess or lack of each of the components leads to the according change of defects concentration [3]. The investigation of the temperature dependence of the thermoelectromotive force (α_0) and thermal conductivity (χ), is the one of the methods of the revealing and studying of the defects nature and interphonone processes, especially at the low temperatures. That's why their combined investigation represents the special interest.

The given paper is devoted to the investigation α_0 and χ Ag_2Te of the stoichiometric composition and compositions with the excess Te and Ag in the temperature interval 3÷300K.

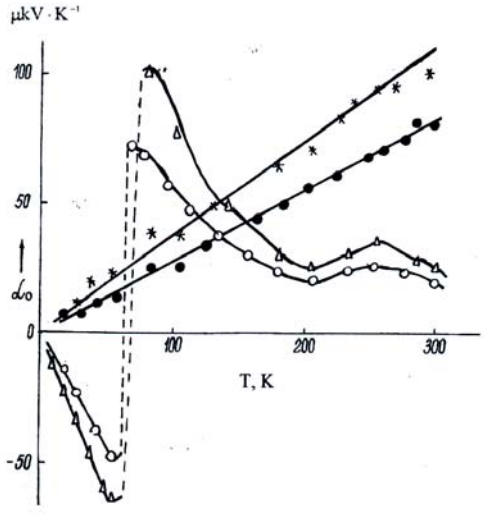

Fig.1. The temperature dependences of thermoelectromotive force α_0(1) Δ-0.25; 0-0.75 at % Te; •-0.25 at Ag. The solid lines are calculated.

The samples had been obtained by the unit technology [4, 5]. The experimental and calculated data are given on the figures 1-2. From the fig. 1 it is seen, that $\alpha_0(T)$ for p-Ag_2Te increases linearly up to T≤45K, later the sign p changes on n. At T≈80 and 270K the maximums of the dependence $\alpha_0(T)$ appear. The analysis has been carried out for the revealing of the dependence $\alpha_0(T)$ taking into consideration the dispersion law of the electronic spectrum and mechanism of scattering on the ion impurities and acoustic phonons [4].

From the fig. 1 it is seen, that $\alpha_0(T)$ for n-Ag_2Te is characteristics for the semiconductors with the one kind of the carriers of current in the degenerate state and Kein's dispersion law [5]. From the analysis $\alpha_0(T)$ in the p- and n-Ag_2Te it is established, that Ag atoms in the Ag_2Te create the small donor levels [5], and Te atoms create the acceptor levels [6], situating from the bottom of the conduction band on the distances ($7\cdot10^{-5}T\cdot K^{-1}$-0.002) and ($7\cdot10^{-5}T\cdot K^{-1}$-0.030) eV, accordingly.

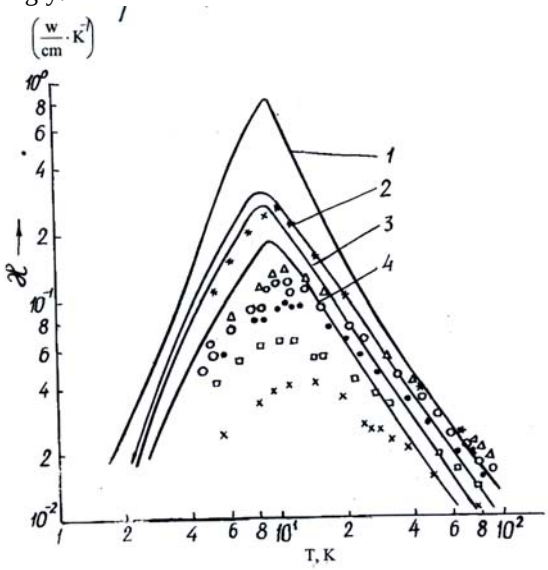

Fig.2. The temperature dependences of the thermoconductivity $\chi(T)$: •-0.25 at % Ag. The solid lines are calculated by the formulae (1), where 1 - stoichiometric composition, which $A=6.64\cdot10^{2}c^{-1}K^{-4}$
2 - with the excess Te 0.1 at % $A=6.64\cdot10^{2}c^{-1}K^{-4}$
3 -with the excess Te 0.1 at % $A=9.96\cdot10^{2}c^{-1}K^{-4}$
4 - with the excess Te 0.15 at % $A=6.64\cdot10^{2}c^{-1}K^{-4}$

The experimental results $\chi(T)$ for the five samples Ag_2Te are given in the fig. 2. As it is shown for all samples χ_{ph} has maximum at ~10K; in the interval 20-80K the dependence $\chi_{ph}(T)$ for the stoichiometric composition has the form $\chi\sim T^{1,2}$; the excess of Te and Ag decreases the value of χ_{ph} on the

law $\chi_{ph} \sim T^{\alpha}$. The going throw the maximum of $\chi_{ph}(T)$ is caused by the phonons scattering on the crystal boundary and defects, and relatively weak temperature dependence in the interval 20-80K and decrease of the exponent α in the dependence on the deflection of Ag_2Te from the stoichiometric is connected with the presence in Ag_2Te of the intrinsic defects and increase of their concentrations at the presence of excess of Ag and Te atoms.

For the approving of the such quality conclusion and quantity estimation of defects concentrations, it is need the obtained results combine with the existing theoretic models of thermal conductivity in the solid bodies. In the given case the excess atoms of Te and Ag can be considered as point defects of Raileigh type. According to the Kallarvay theory [7], takes into consideration the phonon scattering on the boundaries, point defects and phonons (processes of transfer and normal processes), the temperature dependence of thermal conductivity is calculated by the Debay model of phonon spectrum in the approximate relaxation time by the following formulae:

$$\chi_{ph} = GT^3 \left\{ \int_0^{\theta/T} \frac{\tau_c x^4 dx}{Sh^2\left(\frac{x}{2}\right)} + \frac{\left[\int_0^{\theta/T} \frac{\tau_c}{\tau_N} \frac{x^4 dx}{Sh^2\left(\frac{x}{2}\right)}\right]^2}{\int_0^{\theta/T} \frac{\tau_c}{\tau_N \tau_R} \frac{x^4 dx}{Sh^2\left(\frac{x}{2}\right)}} \right\} = GT^3 \left(I + \frac{I_2^2}{I_3} \right) \quad (1)$$

where $G=\hbar/2(2\pi)^2 \upsilon (k/\hbar)^{-4}$, υ - mean velocity, $X=\hbar\omega/KT$, ω - phonon frequency, $\tau^{-1}=\tau^{-1}_R+\tau^{-1}_N$, τ_R τ_N - relaxation times of the resistive and normal processes.

$\tau_R^{-1}=\tau^{-1}_{pp}+\tau^{-1}_b+\tau^{-1}_{pd}$ where τ^{-1}_{pp}, τ^{-1}_b, τ^{-1}_{pd} - relaxation times of phonon processes of the transfer, boundary and phonon-defect scattering accordingly. The following dependences of the outside relaxation times from X and T are as follows [8],

$$\tau^{-1}_{pp}=AX^2T^4e^{-\theta/aT};\ \tau^{-1}_B=\upsilon/L.$$

$$\tau^{-1}_{pp}=BX^4T^4;\ \tau^{-1}_N=cx^nT^4.$$

Here L=0,2 cm, the effective size of the crystal. The parameters A, c, a, n and θ had been defined by the method of the shortest squares numerically by the way of the comparison of the expression (1) with the experimental curves $\chi(T)$. As at the breath of the stoichiometric in Ag_2Te the changes of the fluctuating density $\Delta\delta/\delta$ aren't mentionable, so the scattering on the point defects appears only at the mass change $\frac{\Delta M}{M}$. At this approximation we write according with [8].

$$\tau_{pd}^{-1}=\frac{V_\theta}{n} N\left(\frac{\Delta M}{\overline{M}}\right)^2 \frac{\omega^4}{2\pi\upsilon^3}=\frac{V_\theta}{n} N\left(\frac{\Delta M}{\overline{M}}\right)^2\left(\frac{K}{\hbar}\right)^4 \frac{1}{4\pi\upsilon^3} X^4T^4$$

where V_0 - volume of the elementary cell, n - number of atoms in the elementary cell, $\overline{M}$ - middle mass of crystal atoms, ΔM - mass variation at the exchange of the main atom on the impurity atom, N - atom concentration of impurities (in the respect of the volume of atoms number of crystal). We obtain parameter B from the following expression.

$$B = \frac{V_\theta}{n}\left(\frac{K}{\hbar}\right)^4 \frac{N}{4\pi\upsilon^3}\left(\frac{\Delta M}{M}\right)^2$$

Trimming parameters (A, C) were taken from the work [8], calculated for $L_i \cdot F$ and a=11.53, N=1.23 from the work [9], calculated by the methods of the shortest squares.

The results of the calculation $\chi(T)$ are given in the fig.2 in the form of the solid lines, in comparison with the experimental data. As it is seen, the curve $\chi(T)$, which has more stoichiometric composition Ag_2Te goes lower than calculated curve. This shows that the argentum telluride has the big amount of the intrinsic defects. From the fig.2 it is seen, that these data are suitable for the curve, calculated for Ag_2Te with the impurities concentration $7\cdot10^{19}cm^{-3}$. Later it is seen, that calculated curves $\chi(T)$ always go throw higher than experimental ones with the increase of Ag and Te content, as the divergence increases with the Te and Ag excess.

The experimental curves aren't parallel to the calculated ones, i.e. with the temperature increase the scattering probability increases, as such deflection in the samples with Te excess, is more big than with the Ag excess.

This is connected with the phonons' scattering on the carriers of the current. That's why this type of the scattering is more effective on the p-type samples.

Thus, the analysis of the experimental and calculated data on $\alpha_0(T)$ and $\chi_{ph}(T)$ approves that argentum telluride is the defect material, and Te and Ag excesses play roles of acceptor and donor centers.

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[1] *S.A. Aliyev, F.F. Aliyev, Z.S. Gasanov*. FTT, 1998, 40, 9, 1693 (in Russian).
[2] *F.F. Aliyev.* Dokl. Azerbaijana TLVI, 2000, № 1-3, 85 (in Russian)..
[3] *F.F. Aliyev, V. Atashirov, T.F. Yusifova, M.A. Kerimova, F.M. Gashimzade*. Fizika VII, №3.2001, 22.
[4] *F.F. Aliyev, E.M. Kerimova, S.A. Aliyev*. FTP, 2002, 36, 8, 932 (in Russian).
[5] *F.F. Aliyev*. Izv. RAN, Neorg. Mater., 2002, 38, 10, 1184 (in Russian).
[6] *F.F.Aliyev/* Sekond International Simpozium on Matical and computational Applications. Baku, Azerbaycan, September 1-3/80 ,1999.
[7] *J. Callarvay*. Phys.Rev., 1959, 113.4.1046.
[8] *L.N. Vasilyev, I. Dzhabbarov, V.S. Oskotskiy, L.S. Parfenyeva, V.V. Popov, I.A. Smirnov*. FTT, 1984, 26, 9, 2710.
[9] *R. Berman, I.C.Brock.* F.Proc.Roy.oc. 1965, 289, 1416, 46.

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Ag_2Te KRİSTALINDA TERMOELEKTRİK VƏ İSTİLİK XASSƏLƏRİ

Məqalədə Ag_2Te kristalında termoelektrik və istilikkeçirmə əmsallarının temperatur asılılıqları uyğun olaraq: elektron spektri üçün dispersiya qanunu və yükdaşıyıcılar səpilmə mexanizmləri, eləcədə Katavey nəzəriyyəsi əsasında fonon spektrinin debay modelində relaksasiya müddəti nəzərə alınmaqla izah olunmuşdur.

Ф.Ф. Алиев, М.А. Керимов, Ф.М. Гашимзаде

ТЕРМОЭДС И ТЕПЛОПРОВОДНОСТЬ В Ag_2Te

В работе анализированы температурные зависимости термоэдс $\alpha_0(T)$ и теплопроводности $\chi(T)$ Ag_2Te в рамках теории: с учетом закона дисперсии электронного спектра и механизмов рассеяния носителей тока, также дебаевской модели фононного спектра в приближенном времени релаксации согласно моделью Каллавея.

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