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ABSTRACT

of the dissertation for the degree of Doctor of Philosophy

KINETIC PROPERTIES OF $AgSbTe_2$ CRYSTALS WITH Ag_2Te ADDITIONAL PHASE

Speciality: 2203.01- Electronics

Field of science: Physics

Applicant: **Aygun Eyvaz Babayeva**

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The work was performed at the "Spectroscopic ellipsometry" laboratory of the Institute of Physics of Azerbaijan National Academy Sciences.

Scientific supervisor:
Doctor of physical sciences, associate professor
Sadiyar Soltan Ragimov

Official opponents:
Doctor of physical and mathematical sciences, professor
Kamal Asger Asgerov

Doctor of physical and mathematical sciences, professor
Vaqif Ali Maharramov

Doctor of physical sciences, associate professor
Tarlan Khanbaba Huseynov

Dissertation Council ED 1.14 of Supreme Attestation Commission under the President of the Republic of Azerbaijan operating at Institute of Physics of Azerbaijan National Academy Sciences under the number BED 1.14

Chairman of the Dissertation council:
Active member of ANAS,
doctor of physical and mathematical sciences, professor
Nazim Timur Mammadov

Scientific secretary of the dissertation council:
Doctor of physical sciences, associate professor
Rafiga Zabil Mehdiyeva

Chairman of the scientific seminar:
Doctor of physical and mathematical sciences,
associate professor
Ayaz Hidayat Bayramov

GENERAL DESCRIPTION OF WORK

Relevance of the research topic. One of the requirements of our time is the production and transformation of energy. A more convenient and versatile form of energy for practical use is electricity. For this reason, the search for more efficient methods of obtaining it is always relevant. Solid-state thermoelectric converters are of great interest. The advantages of converters of this type are simplicity of design, quiet operation, high reliability, the possibility of miniaturization without losing efficiency. However, the efficiency of thermoelectric converters is lower than that of conventional electric generators or refrigerators, and therefore they are not widely used in industry. They are used as a sensitive element in converters of infrared radiation, as well as in the production of compact refrigeration units in the national economy, medicine, spacecraft, etc. For widespread use of these energy converters in industry, it is necessary to significantly increase their efficiency.

$A^{I}B^{V}C^{VI}$ compounds are well known as thermoelectric materials. Preliminary studies show that this type of connection has a very low k. Although $A_2{}^{V}B_3{}^{VI}$ has low thermoelectric efficiency in relation to connections, the relation S/k in these substances is of great importance. This, in turn, allows you to produce sensory transformative elements. The overwhelming majority of recent studies on the increase in thermoelectric efficiency are focused on the nanostructuring of these materials. In this case, the increase in efficiency is due to, on the one hand, the reduction of lattice heat exchange and the total heat transfer due to the scattering of phonons on the nanomaterial's boundaries, and with another - the increase of thermoelectric power.

The thermoelectric figure of merit ZT in nanostructured AgSbTe2 compounds based on GeTe (TAGS) and PbTe (LAST) reaches 1.5 in the average temperature range (400-700K). This is mainly due to a significant decrease in heat transfer. On the other hand, an increase in efficiency can be achieved due to a decrease in thermal conductivity as a result of phonon scattering at different interphase boundaries.

$AgSbTe_2$ is **obtained** from two binary compounds such as Ag_2Te and Sb_2Te_3. The study of the Ag_2Te-Sb_2Te_3 phase diagram shows that the compounds included in the Ag-Sb-Te system are obtained in both single-phase and two-phase. In this regard, it is of particular interest to study the effect of the additional Ag_2Te phase on the kinetic, especially thermoelectric, properties of samples of the Ag-Sb-Te system.

Object and subject of research: Investigation of singlephasic and the crystals of the additional Ag_2Te phase in $AgSbTe_2$ and their kinetic properties.

Research aims and objectives:

Determination of the properties of kinetic phenomena in $AgSbTe_2$ crystals, including the additional Ag_2Te phase, and the study of their application in thermoelectric converters. To achieve this goal, it was necessary to solve the following problems.

To achieve this goal, there were determineted following tasks:

- Synthesis of the single-phase system Ag-Sb-Te crystals.
- Synthesis of $AgSbTe_2$ crystals, additional phase Ag_2Te.
- Synthesis of compounds with Ag, Te, Cd elements added to the $AgSbTe_2$ crystal in order to increase the thermoelectric figure of merit.
- X-ray diffraction and DSC analysis of the investigated compositions.
- Study of kinetic phenomena in a wide temperature range in crystals of single-phase and two-phase systems Ag-Sb-Te.
- Study of the influence of the percentage of the Ag_2Te phase on the kinetic coefficients.
- Investigation of the possibility of using the investigated components in thermoelectric converters.

Research methods:

To determine the structure and phase analysis were used X-ray diffractometry and differential scanning colorimetry methods. To determine thermoelectric and galvanomagnetic properties were used the methods of calculating the mathematical data for four-probe potentiometric and two-phase systems.

The main provisions for the defense:

1. Occurrence of an endothermic event in two-phase $AgSbTe_2$ crystals corresponding to the phase transition of Ag_2Te at 423.5K.
2. Exceeding the maximum temperature dependence of the Hall coefficient in $AgSbTe_2$ with the addition of the Ag_2Te phase.
3. Due to the presence of an additional Ag_2Te phase, the effective mass of holes in the $AgSbTe_2$ compound has a higher value than that of the singlephasic $Ag_{0.82}Sb_{1.18}Te_{2.18}$.
4. Low value of thermal conductivity of singlephasic $Ag_{0.82}Sb_{1.18}Te_{2.18}$ and two-phase $AgSbTe_2$ compounds and determination of phonon scattering mechanisms.
5. Increased thermal conductivity value of $AgSbTe_2$ compound due to the presence of additional Ag_2Te phase.
6. Interpretation of experimental results based on the theory of "effective environment" and obtaining maximum values of thermoelectric efficiency in the compound of $AgSbTe_2$ in the presence of an additional phase of 0-15% Ag_2Te.
7. $AgSbTe_2$ + 0.4at.% Te to obtain a value of $Z = 2.3 \cdot 10\text{-}3K^{-1}$ for thermoelectric efficiency at 500K and its suitability for use as a p-branch of thermoelectric converters.

Scientific novelty of the research:

1. Kinetic phenomena in a wide temperature range in the singlephasic and different percentage Ag_2Te phase components of the Ag-Sb-Te system were studied.
2. Based on the results of DSC and X-ray structure analyzes in single-phase and two-phase $AgSbTe_2$ crystals, it was shown that an endothermic event corresponding to the phase transition of Ag_2Te occurs around 423.5K in two-phase compositions.
3. The effective masses of the carriers in $Ag_{0.82}Sb_{1.18}Te_{2.18}$ and $AgSbTe_2$ were calculated and it was shown that the high value of the effective mass of $AgSbTe_2$ is explained by both the thermal activation of the carriers in the conduction zone and the presence of the second Ag_2Te phase.
4. It has been shown that the low value of lattice thermal conductivity in $AgSbTe_2$ and $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compounds

depends on several factors, such as irregularity of the crystal lattice, scattering on point defects, and structural components.

5. Despite the formation of additional thermal resistance due to point defects and interfacial scattering, the presence of an additional Ag_2Te phase at low temperatures leads to an increase in the total thermal conductivity of the $AgSbTe_2$ compound.
6. Calculations based on the theory of effective medium have shown that the maximum values of thermoelectric efficiency of two-phase $AgSbTe_2$ compounds are obtained when the second phase of 0-15% Ag_2Te is present.
7. For the purpose of application in thermoelectric heat converters, Z thermoelectric efficiency of singlephasic, two-phase, alloyed with Ag, Te and Cd elements of Ag-Sb-Te system compounds were determined at different temperatures and it was shown that the value of thermoelectric efficiency in $AgSbTe_2$ + 0.4at.% Te is higher and obtained the value $Z = 2.3 \cdot 10^{-3}\ K^{-1}$ at 500K.

Theoretical and practical significance of the research:

The results of the study of thermoelectric properties over a wide temperature range of single phase and Ag_2Te additional phase compounds belonging to the Ag-Sb-Te system can be used in the preparation of other chemical compounds and solid solutions based on them. The obtained experimental results allow to substantiate that the irregular structure in the studied compositions plays an important role in increasing the value of their thermoelectric efficiency. It has been shown that both $AgSbTe_2$ and $Ag_{0.82}Sb_{1.18}Te_{2.18}$ crystals can be used as a sensitive element in the p-branch of thermoelectric converters. In addition, the obtained results can be used in the interpretation of kinetic phenomens in two-phase systems.

Approbation and application:

The results of the dissertation were presented at the following conferences

- ✓ International scientific conference "Actual problems of physics" dedicated to the 80th anniversary of Academician B.M. Asgarov (Baku-2013);

- ✓ Ist International Scientific Conference of young scientists and specialists; «The role of multidisciplinary approach in solving actual problems of fundamental applied sciences (earth, technical and chemical)» (Baku- 2014);
- ✓ International Conference dedicated to the 80th anniversary of the corresponding member of RAN I. K. Kamilova “Phase transitions, critical and nonlinear phenomena in condensed environments” (Chelyabinsk, Russia-2015);
- ✓ XI International Seminar, "Magnetic Phase Transitions", dedicated to the 80th anniversary of the corresponding member of the Russian Academy of Sciences I. Kamilova. K., (Russia, Makhachkala- 2015);
- ✓ Republican scientific conference "Actual problems of physics" (Baku-2015);
- ✓ “KhazarNeftGasYataq-2016” scientific-practical conference (Baku-2016);
- ✓ XVIII Republican Scientific Conference of Masters and Young Researchers "Problems of Physics and Astronomy" (Baku, BSU-2016);
- ✓ VIII International Scientific and Technical Conference (Russia, Nalchik-2016) "Micro- and nanotechnology in electronics";
- ✓ X International Scientific and Technical Conference (Russia, Nalchik 2018) "Micro- and nanotechnology in electronics."

18 works on the topic of the dissertation were published. 9 of them were published in the form of articles in local and foreign magazines, and 9 in the form of materials and theses at national and international conferences.

The name of the organization where the dissertation work was perfomed: The dissertation work was carried out at the Institute of Physics of the National Academy of Sciences of Azerbaijan.

The structure and scope of the dissertation: The dissertation consists of an introduction, four chapters, results and a bibliography of 133 titles, commented on 147 pages. Excluding figures, tables and references, the introduction consists of 17506, Chapter I 44952, Chapter II 10301, Chapter III 34831, Chapter IV 52832, Results

2393 symbols. The total volume of the dissertation is 166963 symbols.

MAIN CONTENT OF THE DISSERTATION

The **Introduction** provides information about the general characteristics, relevance, purpose, scientific novelty, practical significance of the dissertation, the main provisions, approbations and publications. The chapters are then briefly explained.

Chapter First of the dissertation examines the literature on the crystal structure of $A^{I}B^{V}C_2^{VI}$ triple semiconductor compounds. It was shown that the triple compound $AgSbTe_2$ belongs to the group $A^{I}B^{V}C_2^{VI}$ and has a cubic structure centered on the surface of the NaCl type. This compound is based on two binary Ag_2Te and Sb_2Te_3 compounds and is known as a thermoelectric material operating in a wide temperature range. In addition, the kinetic phenomena in the solid solution crystals of $AgSbTe_2$ based on PbTe and GeTe (LAST, TAGS) were analyzed, the literature on the effect of alloying on the thermoelectric properties of Ag-Sb-Te system compounds was analyzed.

Chapter Second first presents the methodology for the synthesis of Ag-Sb-Te system compounds obtained by single phase, two-phase and addition. Taking into account the probability of the formation of the second Ag_2Te phase in the samples of Ag-Sb-Te system, the studied components were synthesized by various methods, such as slow cooling, sharp cooling (zone cooling) and zone melting, and this is given in detail. The studied compounds at firstly were synthesized in furnace with a constant temperature gradient of 3K/cm.

In order to change the amount of additional Ag_2Te phase, the cooling rates of $AgSbTe_2$ during synthesis were in firstly changed in the range of 1^0C/min-8^0C/min. The most satisfactory results were obtained at a less cooling rate - 1^0C/min. Although the cooling rate in the crystallization region was different, the formation of the Ag_2Te phase in $AgSbTe_2$ compounds was inevitable. To maintain the high-temperature phase, the ampoule of $AgSbTe_2$ alloy, was removed

from the furnace at 500°C and solidified by immersion in cold water. However, in such a sharply cooled sample, several more phases were observed along with $AgSbTe_2$. Some of the synthesized samples were subjected to zone melting. In this case, the stoichiometric sample was first synthesized by slow cooling, and then the ampoule was passed through a zone furnace 4 times.

The samples were then cut into a parallelepiped shape with a diamond saw and polished with polishing powder. The finished samples were boiled in ethyl alcohol and distilled water. Electrical contacts were then placed on the samples and copper wires were used as a measuring probe during the measurement. This chapter also describes the construction of the cryostat and the electrical measurement scheme used to determine the kinetic coefficients.

In Chapter III of the dissertation, the results of the study of the crystal structure, thermoelectric and galvanomagnetic properties of single phasic $Ag_{0.82}Sb_{1.18}Te_{2.18}$ and two-phase $AgSbTe_2$ compounds are explained, and DSC analyzes are presented.

The results of X-ray structure analysis showed that the singlephasic compound $Ag_{0.82}Sb_{1.18}Te_{2.18}$ belongs to the space group Fm3m and crystallizes in a surface-centered cubic structure with lattice parameters a=6.08Å. According to the results obtained, no peaks corresponding to other phases were observed in all studied $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compositions, which in turn showed that these samples were singlephasic.

Based on the results of X-ray structure analysis of $AgSbTe_2$ (Figure 1), it was determined that peaks related to the Ag_2Te phase are observed in $AgSbTe_2$ compared to the singlephasic $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compounds. The analysis showed that in these samples, along with $AgSbTe_2$, there is a small amount of Ag_2Te phase that crystallizes in the orthorhombic structure with lattice parameters a = 16.27, b = 26.68, c = 7.55Å.

In contrast to the slow cooling method, the X-ray structure analysis of different parts of the $AgSbTe_2$ sample synthesized by the zone melting method was carried out in the presented dissertation work. It was shown that the percentage of additional phase Ag_2Te in

the part of the alloy that begins to crystallize is very small and corresponds to the singlephasic state.

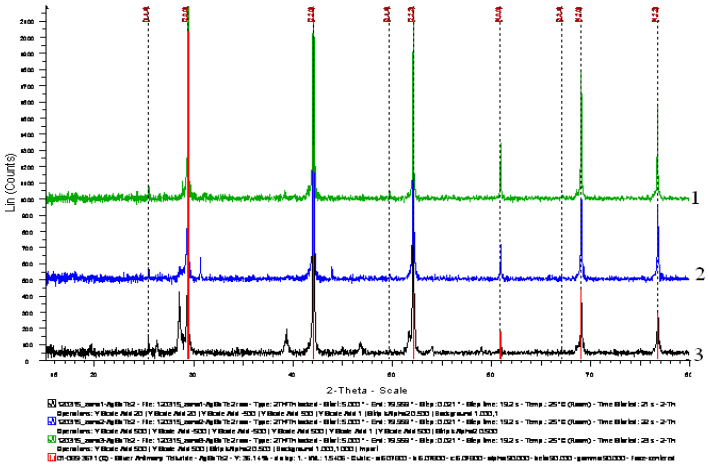
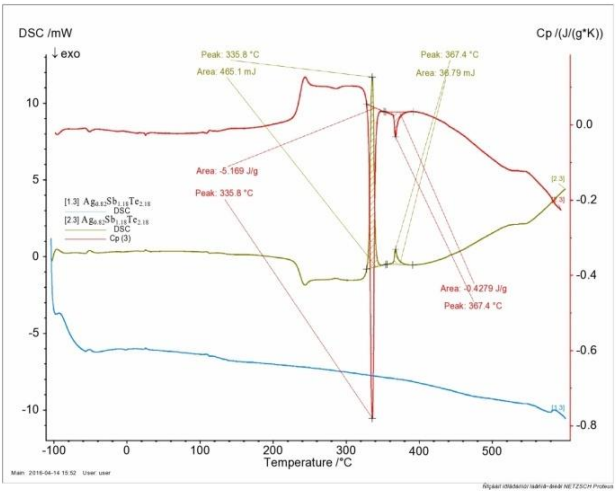


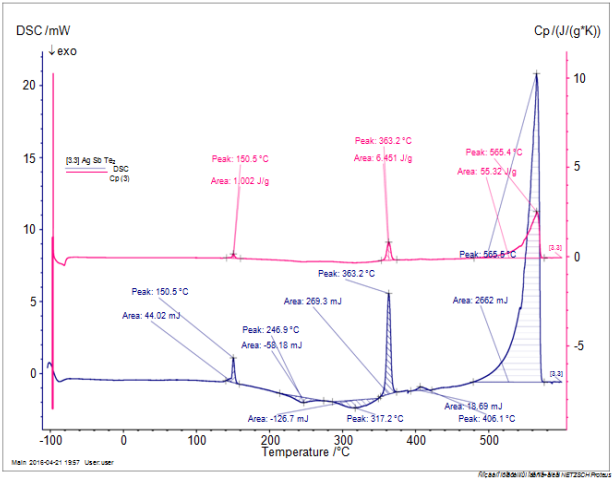
Figure 1. Diffractograms of the $AgSbTe_2$ compound passed through the zone: anterior part of sample 1, intermediate part 2 and end part of sample 3

However, the X-ray analysis of the final part of the alloy is consistent with the X-ray analysis of the additional phase $AgSbTe_2$ sample. Figure 1 shows the results of X-ray structure analysis of samples cut into three longitudinal sections. As can be seen, the results of the X-ray structure analysis for all three samples coincide, but the presence of an additional phase of Ag_2Te is observed in the middle and end parts of the sample.

Differential scanning calorimetric analyzes were performed for the singlephasic $Ag_{0.82}Sb_{1.18}Te_{2.18}$ and two-phase $AgSbTe_2$ samples studied. The experiments were carried out at DET 204F1 Phoenix's NETZSCH-Geratebau GmbH (Germany). Two thermal effects (150.50˚C, 363.20˚C) were observed in $AgSbTe_2$ compared to the singlephasic $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compound (Figure 2b).



a)



b)

Figure 2. DSC analysis of singlephasic $Ag_{0.82}Sb_{1.18}Te_{2.18}$ (a) and $AgSbTe_2$ (b) compounds in the temperature range of -100-300ºC

These thermal effects suggest that the $AgSbTe_2$ triple compound under study consists of two phases. As can be seen from the figure, the exo effect is observed at a 246.9^0C. This can be attributed to the deformation of the crystal lattice as the temperature increases. Deformation of the crystal lattice occurs during heating, and when cooled, the original regular structure is restored.

At the same time, during heating, two endo effects are observed on the thermogram: first at a small size of 150.5°C and then at 363.2°C. According to the results of DSC analyzes, it can be said that in contrast to singlephasic $Ag_{0.82}Sb_{1.18}Te_{2.18}$, the weak endo effect observed in 150.5°C (423.5K) $AgSbTe_2$ at belongs to Ag_2Te. This endo effect is due to the structural phase transition that occurs in Ag_2Te. The endo effect observed at 363.2°C is due to the formation of the Sb_2Te_3 phase in $AgSbTe_2$ in this temperature range, which is described in detail in the literature.

The temperature dependences of kinetic coefficients of singlephasic and two-phase compositions were studied to comprehensively study the effect of Ag_2Te addition phase on the kinetic properties of $AgSbTe_2$. Studies have shown that the temperature dependence of the electrical conductivity of the compound $Ag_{0.82}Sb_{1.18}Te_{2.18}$ has a metallic course (Figure 3).

One of the parameters determining the thermoelectric efficiency is S. Semiconductor thermoelements, which have a high value of S, are used in thermoelectric generators and converters that convert heat energy directly into electrical energy. Thermoelectric power is one of the main quantities characterizing electronic transport properties. Thermoelectric power of semiconductors is determined by physical characteristics such as carrier concentration, energy spectrum, scattering mechanism of carge carriers and temperature range. In addition, S is also one of the parameters that allows to determine the type of conductivity.

The temperature dependence of the S of the $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compound was also observed to be metallic and showed p-type conductivity over the entire temperature range.

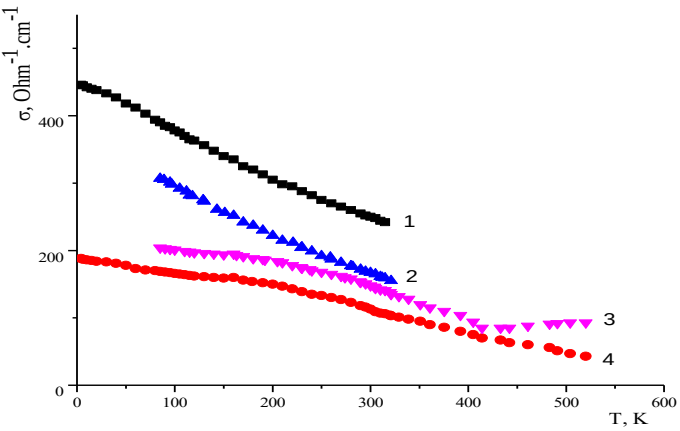


Figure 3.Temperature dependences of specific electrical conductivity of $Ag_{0.82}Sb_{1.18}Te_{2.18}$(1-[1], s.332], 4) and $AgSbTe_2$ (2, 3) compounds

It should be noted that the thermoelectric driving force of the studied singlephasic compound $Ag_{0.82}Sb_{1.18}Te_{2.18}$ is quite large at room temperature (250μkV·K^{-1}) and increases with temperature.

Chapter III of the dissertation presents the results obtained from the temperature dependence of the Hall coefficient of the compound $Ag_{0.82}Sb_{1.18}Te_{2.18}$. In the whole studied temperature range (4.2-550K), the sign of the carge carriers according to the Hall coefficient of $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compound was always p-type, in contrast with Ag_2Te additional phase $AgSbTe_2$ one.

The Hall coefficient is independent of temperature due to the constant concentration of carriers. It should be noted that, unlike the single-phase $Ag_{0.82}Sb_{1.18}Te_{2.18}$, the temperature dependence of the

[1] Алиев, С.А., Рагимов, С.С. Термоэлектрические свойства образцов системы Ag-Sb-Te // Журнал “Неорганические Материалы”, – 1992. 28(2), – с. 329-334.

Hall coefficient of the additional phase $AgSbTe_2$ Ag_2Te exceeds the maximum in the range of 180-200K. In $AgSbTe_2$, the (+) and Hall (-) signs of S also differ over a wide temperature range, which is described in detail in Chapter IV of the dissertation.

The dependence of the Hall coefficient (R) on the magnetic field of the compound $Ag_{0.82}Sb_{1.18}Te_{2.18}$ was also considered, and it was observed that R is independent of the magnetic field. This suggests that there is a kind of load-carrying holes in the conduction. Based on Hall measurements, the carrier concentration and Hall mobility values were determined in $Ag_{0.82}Sb_{1.18}Te_{2.18}$ ($p = 10^{19}cm^{-3}$, μ=15-20$cm^2 \cdot V^{-1} \cdot sec^{-1}$). The value of S is associated with an increase in the effective mass of the carriers and a decrease in the concentration. Therefore, based on the experimental values of thermoelectric motive force and concentration, the effective masses of the holes in the temperature range 250-300K were calculated for the $Ag_{0.82}Sb_{1.18}Te_{2.18}$ ($m_p^*=0.61m_0$)[2].

According to Hall effect measurements, the $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compound has holes in the entire studied temperature range, and the $AgSbTe_2$ compound has an electron-type conductivity in the 60-415K range. In addition, it should be noted that the Hall coefficient of the $AgSbTe_2$ compound, S and sharp changes are observed in the 420K region of the temperature dependence of electrical conductivity. However, for the $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compound, no change was observed in the kinetic coefficients studied in that temperature range. The changes observed in the kinetic coefficients of the $AgSbTe_2$ compound at a temperature around 420K are due to the presence of an additional Ag_2Te phase, which is described in detail in Chapter IV.

The low value of Hall's mobility is due to the high concentration of specific defects. Such a high concentration of

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[2] Рагимов, С.С. Эффективная масса дырок в $Ag_{0,82}Sb_{1,18}Te_{2,18}$ и $AgSbTe_2$ / С.С. Рагимов, Р.И. Селим-Заде, А.Э. Бабаева [и др.] // Известия высших учебных заведений-Физика, – 2020. т.63, №4, – с. 150-153.

defects affects the reduction of the value of k, which is one of the main requirements for thermoelectric substances.

This chapter also presents the results of the temperature dependence of the thermal conductivity (k) of the compound $Ag_{0.82}Sb_{1.18}Te_{2.18}$ over a wide temperature range (4.2-550K).

It should be noted that the study of the temperature dependence of thermal conductivity, in addition to its use in thermoelectric converters, also provides insight into the mechanisms of scattering of phonons.

The thermal conductivity of $Ag_{0.82}Sb_{1.18}Te_{2.18}$ exceeds the maximum in the temperature range of 18-20K, decreases depending on the temperature and obtained a fairly low value at room temperature. The maximum dependence of k(T) is explained by the scattering of phonons across the crystal boundaries.

The electronic part of the thermal conductivity is calculated by Videmann-France law and is shown to have a very small value ($k_e=0.2\%k_{total}$). The experimental and calculated results of heat transfer were analyzed comparatively. Calculations were made on the basis of Kalavey's theoretical model, which took into account the scattering of phonons from point defects, boundaries and phonons. The k(T) dependence for the $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compound is mainly explained by scattering from acoustic phonons.

The lack of a silver atom in the $Ag_{0.82}Sb_{1.18}Te_{2.18}$ compound causes the atomic mass to change throughout the crystal and a large number of point defects to form. Such a change in atomic mass and irregularity ensures that short-wave phonons are scattered. As is well known, acoustic vibrations are the main heat carriers in a crystal lattice. Because optical phonons have a small group velocity, they carry heat poorly. They can affect the thermal conductivity by interacting with acoustic phonons. In the literature, calculations have been made for electronic, optical, and lattice oscillations, and it has been shown that optical modes in the $AgSbTe_2$ compound have low frequencies and are covered by acoustic modes. This can lead to strong interactions between low-frequency optics and high-frequency acoustic modes. The scattering of acoustic phonons from optical modes, as well as the irregularity of silver and lead atoms, lead to an

increase in thermal resistance. Thus, the irregularity of the crystal lattice and scattering from point defects increase the scattering speed of the phonons and consequently decrease the thermal conductivity of the lattice.

In the **fourth chapter**, the effect of different volume percentages of the additional phase of Ag_2Te on the kinetic properties of $AgSbTe_2$ was studied. To comprehensively study the effect of the Ag_2Te addition phase on the kinetic properties of $AgSbTe_2$, the temperature dependences of the kinetic coefficients in both singlephasic and two-phase compositions were studied. According to the phase diagram, the compounds included in the Ag-Sb-Te system are obtained in both single and two-phase.

It should be noted that there are materials in the literature on the effect of the additional phase on the $AgSbTe_2$ on thermal and cargo transport events[1]. The analysis of kinetic coefficients in the Ag-Sb-Te system was carried out, the effect of the additional phase on these kinetic coefficients was partially analyzed, the R coefficient and thermoelectric power calculations were made for only two samples. At the same time, no calculations were made on the effect of different volumes of the second phase on the electrical conductivity, the Hall coefficient, and the thermal conductivity.

Therefore, in the presented dissertation, the effect of the additional phase of the $AgSbTe_2$ compound on thermal and load-carrying events was studied and calculations were made within the framework of the theory of percolation (flow) and "effective environment" in different percentages of Ag_2Te. During the calculations, the experimental results were obtained in a lower and wider temperature range than in the existing literature.

It should be noted that the analysis of multiphase systems is based on percolation theory and "effective environment" methods. As is well known, the "effective environment" method is applied outside the percolation boundaries. In practice, this is done by constructing curves that depend on the percentage of the additional phase of the normalized conductivity in the unit.

The linear part of the dependence corresponds to the scope of effective environmental methods. At the same time, as we approach

the flow boundary, this dependence goes beyond the linearity, becoming superficial at the intersection with the percentage axis in the part corresponding to the critical value. Given that most studies are conducted outside the boundaries of percolation or in the field of "effective environment", it is necessary to emphasize this or that approach to the experimental data.

Figure 4 shows the dependence of the normalized conductance on the amount of the second phase for the samples of the Ag-Sb-Te system. As can be seen, the curve, built on the appropriate coordinates, goes beyond the line in the 40% region and has a superficial character. This indicates an approach to the percolation boundary. According to the experimental results, the critical value of the percolation limit was calculated (p_c = 0.4) and this value coincides with the theoretical basis.

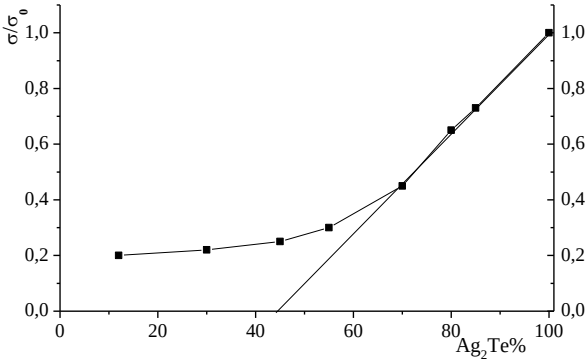

Figure 4. Volume percentage dependence of conductivity on Ag_2Te phase for $AgSbTe_2$ compound

Below this area, the theory of effective environment is applied and the electrical conductivity, thermoelectrical power, Hall coefficients are calculated.

Calculations were made for the electrical conductivity of different volumes of Ag_2Te as a percentage, using the results of the

compound $Ag_{0.82}Sb_{1.18}Te_{2.18}$ as the main phase and the p-Ag_2Te compound as the second phase. The specific resistance of the samples was calculated using the following expression for matrix systems with inputs of approximately the same distance:

$$\sigma = \sigma_0\left(1+\frac{X_1}{\frac{1-X_1}{3}+\frac{\sigma_0}{\sigma_1-\sigma_0}}\right), \qquad \rho = \rho_0\left(1+\frac{x}{\frac{1-x}{2}+\frac{\rho_0-\rho_1}{\rho_1}}\right) \qquad (1)^3$$

Where X is the unit volume concentration of the second phase; ρ_0 and ρ_1 are the specific resistances of the main matrix and the auxiliary phase, respectively. The calculation results are shown in solid lines in Figure 5.

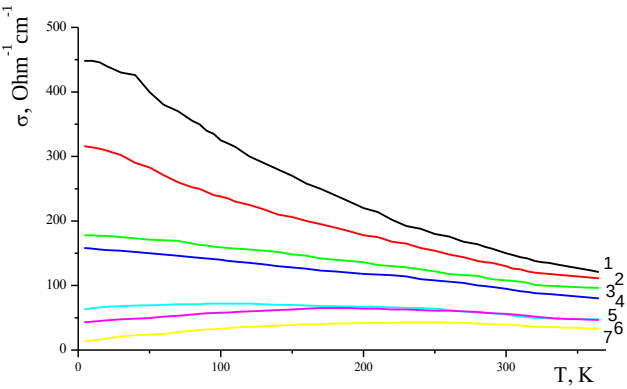

Figure 5. Theoretical calculations of temperature dependence of specific electrical conductivity for different amounts of the second phase in the Ag-Sb-Te compound (Ag_2Te): 1-0%; 2-5%; 3-8%; 4-13%; 5-15%; 6-20%; 7-30%.

[3]Herring, C. Effect of random inhomogeneities on electrical and galvanomagnetic measurements // Journal of Applied Physics, – 1960. 31(11), – p. 1939-1953.

For some examples, there are discrepancies between calculations and experimental results. Certain discrepancies in the experimental and computational results can be explained by the change in the motion of the charge carriers at the boundaries of the matrix and the auxiliary phase and the increase in the amount of the heterogeneous second phase as the crystallization rate increases.

Improper averaging of the additional phase can give an increased and at the same time reduced value of the specific resistance. This change will remain constant throughout the temperature range. It should be noted that at high crystallization rates, the linear dimensions of the main phase $Ag_{0.82}Sb_{1.18}Te_{2.18}$ decrease and the total surface area of the phase boundaries increases. It is on these surfaces that diffusion scattering or mirror reflection of carriers can occur.

If the dimensions of the phase cores coincide with the length of the free runway, the length of the free runway of the carriers may be limited by the phase boundaries.

Figure 6 shows the calculation curves of the Hall coefficient at different percentages of the additional phase (p-Ag_2Te as the second phase).

Theoretical calculations were made based on the results obtained from the experiment. These calculations are based on the theory of kinetic conditions for two-phase systems. According to this theory, the Hall coefficient is defined by the following expression:

$$R = \frac{<\sigma^2 R>}{<\sigma>^2}\left\{1+\frac{2}{3}\sum_{i=1,2} X_i\left[\frac{(\sigma_i - <\sigma>)^2}{<\sigma>^2} - \frac{(\sigma_i^2 R_i - <\sigma^2 R>)(\sigma_i - <\sigma>)}{<\sigma^2 R><\sigma>}\right]\right\} \qquad (2)$$

If we denote the average value of the quantities in the expression by <P> (P = σ, ρ, σ^2R, etc.) is determined as follows:

$$P_i \geq X_1 P_1 + X_2 P_2 \qquad (3)$$

Here P_1 and P_2- properties of separate phases; X_1 and X_2 are the volumes of the phases. The calculations used the results of

$Ag_{0.82}Sb_{1.18}Te_{2.18}$ as the main phase and p-Ag_2Te as the second phase.

The correspondence of the calculated values with the experimental values, first of all, the correct choice of the average composition of the matrix and the auxiliary phase, and the effect of the factor limiting the free path of the charge carriers in these crystals are insignificant. The average length of the free runway determines the value of moving the carriers. For this reason, the mobility of lighter carriers is limited in the first place. As can be seen, as the percentage of the additional phase increases, the value of the Hall coefficient increases and the maximum shifts towards the lower temperature.

Studies show that for singlephasic samples, the S(T) and R(T) dependencies have p-type conductivity and R(T) = const in the entire temperature range studied. When a certain amount of the second phase is introduced, the R dependence is similar to the dependence of the samples obtained in a stoichiometric ratio. As mentioned above, when T>40K, the Hall coefficient changes from positive to negative and then exceeds the maximum.

At the same time, as the phase percentage increases, a maximum temperature shift is observed in the R(T) dependence curve. It should be noted that according to the results of theoretical calculations, in large samples of the second phase, the R at low temperatures does not exceed zero and remains inversely proportional to the temperatures of liquid helium. The R(T) calculations of the Hall coefficient for the different percentages of the additional phase are shown in solid lines in Figure 6. Calculations have shown that when 5% Ag_2Te second phase is added to the $AgSbTe_2$ compound, the maximum Hall coefficient in the range of ~180-200K and the inversion of the sign in the range of ~60K are observed. This is due to the fact that the temperature dependence of p-Ag_2Te in the range of 60K changes the sign of the Hall coefficient from positive to negative. The high value of the conductivity of the second phase of Ag_2Te and the mobility of electrons in a given temperature range leads to a difference in the type of conductivity according to the Hall effect and thermoelectric power.

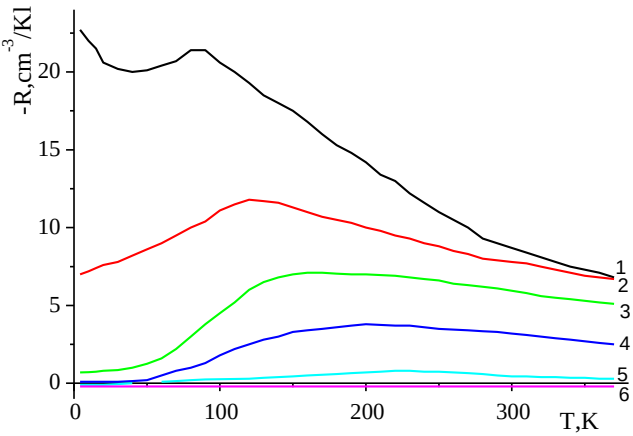

Figure 6. Calculation values of temperature dependences of the Hall coefficient in $AgSbTe_2$ at different percentages of the additional phase of Ag_2Te: 1-30%; 2-20%; 3-13%; 4-8%; 5-5%; 6-0%

Note that with the increase of the second phase, the value of conductivity decreases monotonously. Calculations have shown that the presence of a second phase of 5% Ag_2Te is sufficient for a sharp change in the coefficients studied in the phase transition.

At the same time, there is a slight deviation of the calculation results from the experimental results. Such a difference between the experimental and computational results can be explained by the increase in the strong non-homogeneous second phase as a result of the scattering of charge carriers at the boundaries of the matrix and auxiliary phase and the increase in crystallization rate.

As is known, the efficiency of thermoelectric materials is determined by the following expression.

$$Z = \frac{\sigma S^2}{k} \qquad (4)$$

S-thermoelectric power;

σ-electrical conductivity;

k is the cofficient of thermal conductivity.

To increase the thermoelectric efficiency Z, it is necessary to buy materials with low k or large strength factor (σS^2). That is why the study of S for thermoelectric materials is one of the key issues.

In recent years, in the field of materials science, thermoelectric materials have been actively studied, showing an increase in the value of the Z factor, which determines the efficiency of thermoelectric devices.

It was determined from the temperature dependence of the S of the $AgSbTe_2$ with stoichiometric composition that the amount of the second phase in the studied samples depends on the rate of cooling (crystallization) and leads to a slight change in S. However, the appearance of the S(T) dependence does not change. In the temperature range of 100-220K there is a characteristic slowing down of the S-thermoelectric force due to the S(T) dependence, and above 220K there is a linear increase. For these samples, the value of the thermoelectric force at low temperatures increases with increasing temperature. All samples studied have a sufficiently large thermoelectric power at room temperature and show p-type conductivity over the entire temperature range studied.

The temperature dependences of thermoelectric power of samples with different phase quantities are of great interest. For singlephasic samples, the temperature dependence of thermoelectric power increases linearly. As the amount of the second phase increases, a slight bending around 90K is observed in the S(T) dependence. In relatively large quantities of the second phase, the stability of the coefficient of thermoelectric momentum is observed in the range of 50-240K.

In the framework of the above model, the percentage and temperature dependences of the corresponding phases used in the calculations of the electrical conductivity and the Hall coefficient were taken during the theoretical calculations of the value of thermoelectric power Within this model, the thermoelectric driving force is defined as follows:

$$S = < S > - \sum_{i=1,2} S_i X_i \left(\frac{1}{3} \frac{k_i - < k >}{< k >} + \frac{2}{3} \frac{\rho_i - < \rho >}{< \rho >} \right) \qquad (5)$$

Where <S> = $S_1X_1 + S_2X_2$ - simple volume average;

X_1 and X_2 - volume of phases;

k_1 and k_2-thermal conductivity of phases

It is the specific electrical conductivity of σ_1 and σ_2-phases.

It should be noted that this model is designed for two-phase systems with slightly different values of conductivity and conductivity.

According to the results of thermoelectric force, $AgSbTe_2$ has a hole-type conductivity in almost the entire temperature range studied. In the phase transition region of the Hall coefficient, thermoelectric power and electrical conductivity (T=420K) there are sharp changes in temperature dependence, which is explained by the presence of a structural phase transition in the separated Ag_2Te compound. It was found that the amount of the second phase in the $AgSbTe_2$ depends on the rate of cooling (crystallization). This leads to a slight change in thermoelectric power in the studied samples. However, the change in the amount of phase is sharply reflected in the temperature dependence of the Hall coefficient.

The significantly lower thermal conductivity of these components is of particular interest and is explained by several factors, such as irregular structure, point defects, and structural components. It is known that $AgSbTe_2$ crystallizes in the cubic structure of NaCl. Here, Ag and Sb ions are distributed irregularly between dense layers of Te ions. These chemically different atoms are in the same crystallographic position, but differ in the configuration of the valence electrons. This leads to distortions in the crystal lattice and, consequently, a decrease in thermal conductivity. However, given that the atomic masses of silver (107.868) and stibium (121.75) are close to each other and the differences are insignificant, they cannot cause strong scattering.

Thus, the irregularity of the crystal lattice and scattering from point defects increase the scattering rate of phonons and,

consequently, decrease the thermal conductivity of the lattice in these substances. If the phases are evenly distributed in the structure, the thermal conductivity of the two-phase system should be lower than the thermal conductivity of the main matrix. However, the opposite is observed in practice.

Then the results of the temperature dependence of the thermal conductivity of $AgSbTe_2$ with a stoichiometric composition in a wide temperature range were presented and it was shown that the value of heat transfer in the studied samples increases with increasing temperature and exceeds the maximum in the range of 180-200K. This is explained by the scattering of phonons across the crystal boundaries. These k (T) dependencies can be attributed to the phonon part of the heat transfer, since it has been shown that the electronic part of the heat transfer calculated according to Wiedemann-France law is less than 1% in a given temperature range.

At the same time, the calculation results of the dependence of the thermal conductivity on the second phase of p-Ag_2Te in the two-phase samples of the Ag-Sb-Te system are given. It has been shown that as the amount of the second phase increases at lower temperatures, the value of k also increases.[4] However, this difference decreases due to the closeness of the values of thermal conductivity of the main ($Ag_{0.82}Sb_{1.18}Te_{2.18}$) and auxiliary phase ($Ag_2Te$) around 300K. The experimental results were explained in the framework of the "effective environment" theory.

During the calculations, the experimental results were obtained in a lower and wider temperature range than in the existing literature.

In all cases, the calculations were based on a two-phase model with a matrix $Ag_{0.82}Sb_{1.18}Te_{2.18}$ and a second phase p-Ag_2Te. The correspondence between the experimental results and the calculated values is explained by the correct choice of the average composition of the matrix and the auxiliary phase.

In order to increase the thermoelectric efficiency of $AgSbTe_2$, which is used as a sensitive element in thermoelectric converters in

[4] Ragimov, S.S., Babayeva, A.E. and Aliyeva, A.İ. On the thermal conductivity of $AgSbTe_2$ and $Ag_{0.82}Sb_{1.18}Te_{2.18}$ // Low Temperature Physics, – 2018. 44(11), – p.1195-1197.

the medium temperature range, several compounds were synthesized and their thermoelectric properties were studied in the temperature range of 80-550K. Thermoelectric beam converters are characterized by high reliability, stability of parameters over time, non-selectivity in a wide wavelength range, etc. has advantages such as. The main advantage of these converters is their non-selectivity compared to other converters. The working principle of thermoelectric converters (beam receivers) is based on the thermoelectric (Seebeck) effect.

The main requirement for the material used in these converters is the high value of thermoelectric power.

On the other hand, since the power of the received radiation is measured mainly by increasing the temperature of the thermocouple in the temperature of the thermocouple, the value of thermal conductivity of the thermoelectric substance must be low.

The studied samples were obtained mainly as a result of the addition of the original crystal. Addition of additives leads to changes in the concentration of carriers in the substance and, accordingly, to changes in electrophysical properties.

Small changes in the metal-chalcogenide ratio in thermoelectric chalcogenide systems can lead to structural transformations. In NaCl structural tellurides (eg $AgSbTe_2$ and PbTe), the amounts of cations (M) and anions (Te) are approximately the same. The extraction of metal planes from equal distances leads to the production of compounds with an M / Te ratio less than one.

$AgSbTe_2$, which has a crystalline structure of NaCl, has a very complex lattice structure in which the Ag and Sb atoms occupy the positions of the Na atom in an irregular manner. Therefore, the thermal conductivity of $AgSbTe_2$ is very low, and the thermoelectric power is relatively high. As a result of such a low thermal conductivity and a high value of thermoelectric driving force, the thermoelectric efficiency of this substance has a high value in the medium temperature range Z. It is known that the inclusion of various additives in substances affects their zonal parameters, thermoelectric properties and dispersion mechanisms of carriers. Addition of additives leads to changes in the concentration of carriers in the substance and, accordingly, to changes in electrophysical

properties. In order to increase the thermoelectric efficiency, several substances were synthesized with the addition of Ag, Te, Cd elements to $AgSbTe_2$ and their thermoelectric properties were studied in the temperature range of 80-550K. As a result of the research, it was shown that among the components included in the Ag-Sb-Te system, the value of thermoelectric efficiency of $AgSbTe_2$ + 0.4at.% Te is higher and obtained the value $Z = 2.3 \cdot 10^{-3} K^{-1}$ at 500K and can be used as a p-branch (Table).

Table. Estimates of thermoelectric efficiency at different temperatures

$Z(T) \cdot 10^{-3}\ K^{-1}$					
Nümunə	300K	350K	400K	450K	500K
$Ag_{0.82}Sb_{1.18}Te_{2.18}$	0.96	0.98	0.9	0.78	0.7
$AgSbTe_2$	1	1	0.98	1	1.2
$AgSbTe_2$+0.2at.%Ag	0.34	0.3	0.3	0.13	0.1
$AgSbTe_2$+0.4at.%Te	1.2	1.2	1	1.5	2.3
$AgSbTe_2$+5at.%Cd	0.1	0.12	0.1	0.04	0.04

In recent years, in the field of materials science, thermoelectric materials with a high coefficient of quality ZT, which determines the efficiency of thermoelectric devices, are widely studied. The quality coefficient of thermoelectric materials currently used in practice is around ZT~1. One of the main issues is to increase the value of efficiency of thermoelectric devices and bring them to the price of traditional electromechanical systems. It should be noted that each thermoelectric material receives the highest value of

efficiency in a certain temperature range. In this sense, due to the maximum value of ZT, thermoelectric materials are more suitable for operation in the a) low temperature (<523K), b) medium temperature (523–923K) and c) high temperature (> 923K) ranges. ZT~1.15 was obtained for the quality coefficient based on the value of thermoelectric efficiency $Z=2.3\cdot10^{-3}K^{-1}$ obtained for $AgSbTe_2$+0.4at.% Te at 500K. This is the main advantage of the low temperature area, which allows it to be used in practice in thermoelectric refrigerators, generators and heat receivers.

CONCLUSION

1. The methods for obtaining of monophasic and by different percentage volumes Ag_2Te phase inclution components of the Ag-Sb-Te system with parameters suitable for use in thermoelectric heat converters have been developed. Based on DSC and X-ray structure analyzes, the endothermic effect observed at 150.5^0C (423.5K) in two-phase $AgSbTe_2$ crystals was shown to correspond to the phase transition of Ag_2Te.
2. On the basic of the experimental results of thermoelectric power and Hall coefficients were determined the concentrations and effective masses of the holes of singlephasic $Ag_{0.82}Sb_{1.18}Te_{2.18}$ ($m_p^* = 0.61m_0$) and two-phase $AgSbTe_2$ ($m_p^*=1.76m_0$) It has been shown that the presence of an additional Ag_2Te phase leads to an increase in the effective mass of $AgSbTe_2$, and the small value of the Hall mobility (15-20$cm^2/V\cdot sec$) is caused by the high carriers concentration and specific defects.
3. Irregular distribution of Ag and Sb ions between dense layers of Te ions in the crystal structure results in irregularity of Ag and Sb cation centers and a large number of point defects. In addition, structural components $AgSbTe_2$ and $Ag_{0.82}Sb_{1.18}Te_{2.18}$, such as the formation of additional phases that cause scattering from the phase boundaries, reduce the thermal conductivity coefficient and have a very small value ($(6\text{-}8)\cdot10^{-3}W/cm\cdot K$)
4. It is shown that despite the formation of additional thermal resistance as a result of interphase scattering, the presence of an

additional Ag_2Te phase leads to increase of the total thermal conductivity of the $AgSbTe_2$ due to the high thermal conductivity of that phase at low temperatures.

5. Calculations based on the effective medium theory have shown that the maximum values of thermoelectric efficiency for a two-phase $AgSbTe_2$ are obtained at the presence of a second phase of 0-15% Ag_2Te in the volume. When more than 5% of the Ag2Te addition phase is present in the $AgSbTe_2$, the inversion of the sign from p to n around ~ 60K is observed due to the high electron conductivity, and in was observed that, the Hall coefficient pass through maximum in the 150-200K temperature range
6. For application in thermoelectric heat converters, on the basis of temperature dependences of electrical conductivity, thermoelectric power and thermal conductivity were determined the thermoelectric efficiency of monophasic, twophase and alloyed with Ag, Te and Cd elements included in Ag-Sb-Te system in the 300-500K temperature range. It was shown that the value of Z of $AgSbTe_2$ + at.0.4% Te is higher and takes the value of $Z=2.3\cdot10^{-3}K^{-1}$ at 500K and may be used as a p-branch of thermoelectric converters.

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