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ABSTRACT

of the dissertation for the degree of Doctor of Science

HOPPING MECHANISMS OF PHASE TRANSITIONS AND CHARGE TRANSPORT OF ABX_2 (WHERE A=Cu, Tl; B=Fe, Cr, Co, Ni, Ga, In; X=S, Se,Te) TYPE LOW-DIMENSIONAL SEMICONDUCTORS

Speciality: 2222.01 – Semiconductor Physics

Field of Science: Physics

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GENERAL DESCRIPTION OF WORK

Relevance of the topic and degree of elaboration:

Phase transitions are one of the most widespread and interesting phenomena, both in fundamental scientific research and in technological applications. The theory, which describes the main aspects of the physics of phase transitions was given by L.D. Landau. The main point of Landau's theory is the concept of the order parameter- a quantity equal to zero in the disordered phase above the transition temperature and non-zero during the transition to the ordered phase. The order parameter characterizes the breaking of symmetry occurring in phase transitions. According to the standard classification, phase transitions are of two types: the first kind, when the order parameter changes abruptly, and the second kind, when the order parameter changes continuously.

In recent years, interest in the study of new magnetic states of electron spins of substances has increased. In nanoelectronics, the local magnetic field created by the rotation of an electron around itself is used. The effect of an external magnetic field or a spin-polarized electric current can change the direction of the electron spin. Such a state of materials differs from classical ferromagnets, ferrimagnets and antiferromagnets in terms of their properties.

Copper chalcopyrite compounds are one of the most promising materials for research of spintronics. The extent of magnetic diversity of magnetic structures in chalcopyrites is mainly based on the structural properties of the Cu^{2+} ion. Other magnetoactive ions prefer certain types of crystallographic positions. For example, Fe^{3+} and Mn^{2+} ions mainly occupy octahedral, sometimes tetrahedral positions. Besides the octahedral and tetrahedral chalcogen surroundings, the Cu^{2+} ion can also have square and pyramidal surroundings. Such "properties" of the Cu^{2+} ion are explained by the features of its electronic structure, which allows it to adapt to different types of chalcogens.

$TlBX_2$ (B=Fe, Cr, Co, Ni, Ga, In; X=S, Se, Te) type triple semiconductor compounds have a layer-chain structure. They are characterized by anisotropic physical properties, which ensure the free movement of their charge carriers within the layers. This limits the

movement of carriers between layers as a result of van der Waals interactions and the fact that the wave functions of adjacent layers overlap less.

The object and subject of the research:

ABX2: $CuFeTe_2$ (a. b, c),$TlFeS_2$, $TlFeSe_2$, $TlCrS_2$, $TlCrSe_2$, $TlCoS_2$, $TlCoSe_2$, $TlNiS_2$, $TlNiSe_2$ and solid solutions in $(TlFeS_2)_{1-x}(TlGaS_2)_x$, $(TlFeSe_2)_{0.5}(TlGaSe_2)_{0.5}$, $(TlGaS_2)_{0.95}(TlCoS_2)_{0.05}$, $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ type compounds, the temperature regions of phase transitions and their charge carrier transfer phenomena were revealed.

As a result, investigation of phase transitions (PT), electrical conduction mechanisms, and magnetic properties of low-dimensional semiconductors under various external conditions is a pressing issue. Researches were carried out in magnetic fields up to 50kOe and in the temperature range of 2÷400K.

The purpose and tasks of the research:

ABX_2 (A=Cu, Tl; B=Fe, Cr, Co, Ni, Ga, In; X=S, Se, Te) type of low-dimensional semiconductor chalcogenides and their solid solutions of phase transitions (magnetic, magnetoelectric and metal-dielectric type) consists of determining their compliance and determining the mechanisms of charge transfer and their prospective application as active materials in spintronics and semiconductor electronics.

For this purpose, the following tasks were set and solved in the work:

- Determination of structure symmetry and structure in $Cu_{1.15}Fe_{1.23}Te_2$ single crystal grown by gas transport method.
- Investigation of temperature dependences of conductivity below phase transition temperatures of $CuFeTe_2$, $TlFeS_2$ (Se_2) and $TlFe_{0.975}Ga_{0.025}S_2$ single crystals.
- Analysis of the transition temperature of the spin-glass state of the single crystal under the influence of the magnetic field on the chalcopyrite-type $CuFeTe_2$ single crystal.
- Analysis of the dependence of the mutual antiferromagnetic exchange between magnetic "clusters" in $CuFeTe_2$ single crystals on the arrangement of iron (Fe^{+3}) cation and tellurium (Te^{-2}) anion.
- Magnetic regularity of $TlMeX_2$ (Me=Fe, Cr, Ni and X=S, Se) compounds and their solid products with time-jump conductivity.

- Effect of changing their ratios (x=0.025, 0.05, 0.075 and 0.01) on the Neel temperature in $(TlFeS_2)_{1-x}(TlGaS_2)_x$ solid solutions.
- Study of the activation energies of $TlCoS_2$ and $TlCoSe_2$ ferrimagnets before and after the magnetic phase transition.
- Determination of the metal-dielectric phase transition in the hexagonal $TlNiS_2$ compound at the transition temperature. Investigation of the transition of this phase transition at the temperature of 240-245K.
- Investigation of the proportionality between alternating current conductivity and current frequency in the range of 3.2÷35 kHz of $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ (x=0.4 and 0.6) solid solutions.

Research methods:

The quality and composition of the samples were controlled by following the production technology of the conducted studies, using XRDD2 PHASER x-ray device and EDX (Energy Dispersive X-ray Analysis), Analyst-800 Perkin Elmer type analyzers.

It was studied using the standard research methods of magnet (SQUID magnetometer, SQUID - Superconducting Quantum Interference Device) and kinetic properties applied for the study of semiconductor materials, as well as ПИУС-1ум-К - device for determining the type of semiconductor materials. Experimental methods tested on relevant materials were used and results studied by other methods were cited. Magnetic properties of ABX_2 type (A=Cu, Tl; B=Fe, Cr, Co, Ni, Ga In; X=S, Se, Te) low-dimensional semiconductors and their solid solutions studied in wide temperature ranges (2÷400K). electrical, thermoelectric and thermal properties were studied.

The main provisions submitted to the defense:

1. It is shown that $Cu_{1.15}Fe_{1.23}Te_2$ single crystal grown by gas transport has a tetragonal structure with structural symmetry D_{2h}^2=P4/nmm and lattice constants a=0.402nm, c=0.604nm.

2. It was determined that in the chalcogen compounds $CuFeTe_2$, $TlFeS_2$, $TlFeSe_2$ and in their solid solution, a break in the temperature dependence of conductivity is observed below the phase transition temperature of $TlFe_{0.975}Ga_{0.025}S_2$ single crystals.

3. It was determined that in single crystals of chalcopyrite type $CuFeTe_2$ (a, b, c) there is an anomaly in temperature dependences of heat capacity and magnetic susceptibility $\chi(T)$ at Neel temperature and spin-glass state.

4. It was determined that the transition temperature of the spin-glass state decreases with increasing magnetic field in $CuFeTe_2$ single crystals.

5. It is shown that in single crystals of $CuFeTe_2$, there is mutual antiferromagnetic exchange between magnetic "clusters". In this case, the iron (Fe^{+3}) cation is located between two tellurium (Te^{-2}) anions, and the magnetic moments of the spins are aligned opposite to each other parallel to the (001) plane. While $CuFeTe_2$ single crystals have Z=1 for their crystal structure, the magnetic crystal structure consists of eight formula units, i.e. Z=8.

6. It has been shown that the charge transport of $TlMeX_2$ (Me=Fe, Cr, Ni and X=S, Se) compounds and their solid products in the temperature ranges of magnetic order occurs with a jump.

7. It was shown that in $(TlFeS_2)_{1-x}(TlGaS_2)_x$ solid solutions, changing their ratios (x=0.025, 0.05, 0.075 and 0.01) leads to a decrease in Neel temperatures and an increase in activation energies.

8. It was shown that the jump length of the specific electrical conductivity along the natural layers of the single crystal of $(TlFeSe_2)_{0.5}(TlGaSe_2)_{0.5}$ solid solution was $R_{or.}$=23.3nm. At the same time, a temperature hysteresis related to the proportional and non-proportional phases of the $TlGaSe_2$ single crystal is observed in the temperature dependence of the specific resistance.

9. It was determined that the activation energy of $TlCoS_2$ and $TlCoSe_2$ ferrimagnets was ΔE_a=0.009eV, ΔE_a=0.005eV before the magnetic phase transition, and ΔE_a=0.116eV, ΔE_a=0.014eV after the transition, respectively.

10. It is shown that a metal-dielectric phase transition occurs in the hexagonal $TlNiS_2$ compound at the transition temperature (240K). At temperatures above this phase transition, the conductivity changes from p-type semiconductor to metallic conductivity.

11. It was determined that dielectric loss in $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ (x=0.4 and 0.6) solid solutions occurs due to direct conduction. In the

range of 3.2÷35 kHz, their alternating current conductivity is proportional to the 0.8 power of the frequency ($\sigma_{ac} \sim f^{0.8}$).

Scientific novelty of the study:

1. It was determined that a phase with symmetry D_{2h}^2 - P4/nmm is formed in the $Cu_{1.15}Fe_{1.23}Te_2$ monocrystals grown by the gas transport method in the tetragonal structure. The X-ray study found that for $Cu_{1.15}Fe_{1.23}Te_2$, the main parameters of the crystal lattice, a=0.402nm, c=0.604nm and the ratio of lattice parameters c/a=1.5, volume v=0.09745nm^3, densities ρcalc=6.764g/cm^3 and ρrentgen=6.76g/cm^3.

2. It has been shown that in single crystals of $CuFeTe_2$ type (a, b and c) below the semiconductor-metal phase transition temperature (PTT=125K) there is a deviation in the temperature dependence of the conductivity, which is explained by the change in activation energy. In that temperature range, the charge transport process occurs with a variable hopping length. The activation energy of the semiconductor-metal phase transition increases with an increase in the amount of iron in the compound.

3. In the temperature dependences of the magnetic susceptibility, $\chi^{ZFC}{}_{H=0}(T)$ and $\chi^{FC}{}_{H=1кOe}(T)$ broadening is observed, which is related to $CuFeTe_2$ (a, b and c) clustering. It was determined that in single crystals of CuFeTe2 (a, b and c) a decrease in the transition temperature to the spin-glass state is observed with increasing magnetic field.

4. It was determined that the mutual magnetic exchange between magnetic "clusters" (Cu^+-Te^{2-}-Fe^{3+}=Te^{2-}) in $CuFeTe_2$ type compounds (a, b and c) is antiferromagnetic. At this time, Fe^{3+} cations are located between two Te^{2-} anions, and the magnetic moments of the spins are parallel to the (001) plane and opposite to each other. Effective magnetic moments of paramagnetic clusters are calculated for weak (100÷1000 Oe) and strong (10÷50 kOe) magnetic fields, paramagnetic Curie temperature (Θ_P) is

negative. It was determined that the magnetic crystal structure of CuFeTe2 consists of eight formula units - 8($CuFeTe_2$) Z=8, and the crystal structure is Z=1. Anisotropy parameter η=0.309, three-dimensional Debye temperature Θ_D=176.9K and two-dimensional Debye temperature Θ_2=122.2K of $CuFeTe_2$ type compound were found.

5. It is shown that in $TlFeS_2$, $TlFeSe_2$, $TlCrS_2$ samples, $(TlFeS_2)_{0.975}(TlGaS_2)_{0,025}$ and $(TlGaSe_2)_{0.5}(TlFeSe_2)_{0.5}$ solid solutions are determined by Moπttt coordinates in the magnetically arranged temperature range lgϭ=f($T^{-1/4}$) and has a variable length hopping conduction mechanism.

6. It was found that the increase of x=0.025, 0.05, 0.075 and 0.1 in the composition $(TlFeS_2)_{1-x}(TlGaS_2)_x$ leads to a decrease in the Neel temperature (T_N) and an increase in the activation energy, gallium and iron in this magnetic structure is explained by the redistribution of cations.

7. It was determined that the Neel temperature changes from 150K to 180K when samples of $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ solid solution are compressed under pressure P=1.5x10^3kg/cm^2.

8. The temperature dependence of the resistance of $(TlGaSe_2)_{0.5}(TlFeSe_2)_{0.5}$ solid solution shows a temperature hysteresis of T_{gis}=109÷120K, which is related to the commensurate and incommensurate phases of $TlGaSe_2$ single crystal.

9. It is shown that the activation energy values are calculated before and after the magnetic phase transition temperature in Tl(Fe, Cr, Co, Ni)S_2(Se_2) crystals and their solid solutions. The difference between these values (before and after PT) indicates the energy of the magnetic phase transition collapse.

10. It was determined that the $TlNiS_2$ compound has a jump conduction mechanism with a variable length (R_L=3.0nm)

localized near the Fermi level in a constant electric field. It was determined that the metal-dielectric (MD) phase transition in $TlNiS_2$ with hexagonal structure occurs at the Curie temperature TC=240K and at temperatures below T_C and is accompanied by a sharp jump of electrical resistance (ρ/ρ_{room}) 10^3. Thermo-EMF is greater than S(T) by more than 10^2 (S/S_{Room}). The temperature dependence ρ(T) for TlNiSe2 crystals during the MD phase transition is observed at TN=120K, and at this temperature the inversion of the thermo-EMF S(T) sign occurs.

11. It has been shown that the dielectric losses in samples of solid solutions $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ (x=0.4 and 0.6) are related to conductivity. In the f=3.2÷35kHz frequency range, their variable conductivity ($б_{ac}$) obeyed the regularity $б_{ac}$~$f^{0.8}$, which is characteristic of the hopping mechanism of charge transfer through states localized near the Fermi level.

Theoretical and practical significance of the study:

The importance of the research conducted is determined by the fact that its results can be used to improve production and quality control methods. The results of the work can be used in the selection of electrical conductivity and magnetic properties, in the development of magnetic energy-independent memory. In addition, the research results facilitate the use of the studied materials in the devices of spin-dependent carriers of these or other charge carriers, including in magnetic tunnel structures. The optimal conditions for the synthesis of $CuFeTe_2$ single crystals were determined. A photo-accumulator can be created with the help of $CuFeTe_2$ bonding layer. These layers of $CuFeTe_2$ single crystals are used to measure the pressure of oxygen gas (O_2). Based on copper chalcogenides such as Al-Al_2O_3-$CuFeTe_2$ ($Cu_{2-x}Te$), it is possible to create multi-position converters in tunnel structures with Al_2O_3 thickness from 0.3 nm to 10 nm. $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ (where x=0.1, 0.2, 0.4 and 0.6) solid solutions can be a base material for spintronics. The activation energies calculated before and after the magnetic phase transition of Tl(Fe, Cr, Co, Ni)S_2 (Se_2) low-size single crystals can be used in theoretical calculations.

The obtained results have a prospective application as an active material in both spintronics and semiconductor electronics.

Approbation of work:

The main materials of the dissertation were presented and discussed at the following international conferences, meetings and seminars:

- Inorganic Materials Conference, Konstanz, Germany.7-10 September 2002, Abs. Ref. Number 38.
- Abstracts of 13-th International Conference on ternary and multinary Compounds - ICTMC-13, Paris, France. Code Number 20 (SO3 A020), October 14 – 18, 2002, P.P. 2-1.
- International conference on Technical and Physical Problems in Power Engineering, Baku – Azerbaijan, № 93, 2002, p. 366-369.
- TPE – 06 3-rd International Conference on technical and Physical Problems in Power Engineering Turkey, Ankara,, May 29-31, 2006, p. 607- 609.
- Сб. док.IV-ой Международной научной конференции «Актуальные проблемы физики твердого тела ФТТ-2009». Минск. Беларусь. 20 – 23 октиября. 2009, т. 2, с.127-129.
- Bakı Dovlət unvtrsiteti. Fizikasının müasir problemləri V-ci respublika konfransı "Opto-, nanoelektronika və kondensə olunmuş mühit fizikası" material. Bakı 16-17 dekabr, 2011, s. 85-86.
- Fizikanın Müasir Problemləri. VI Respublika Konfransının Materialları "Opto, nanoelektronika, kondensə olunmuş mühit və yüksək enerjilər fizikası". Bakı. Azərbaycan. 14 – 15 dekabr, 2012. s.163 – 165.
- Труды XVII-ой Международной конференции «Опто-наноэлектроника, нано технологии и микросхемы». Ульяновск, Россия. 15-19 сентября 2014, с. 79-80.
- Тезисы докладов Шестой Международной конференции «Кристаллофизика и деформационное поведение перспективных материалов», посвященной 90-летию со дня рождения проф. Ю.А. Скакова. Москва. 26-28 мая 2015, с. 223.
- Nineteenth Symposium on Thermophysical Properties, University of Colorado at Boulder, CO, USA. June 21-26, 2015, pp. 272.

- Труды международной конференции: «Фундаментальные и прикладные вопросы физики», 13-14 июня Ташкент, т. 2, 2017, с. 44-46.
- Восьмая Международная конференция «Кристаллофизика и деформационное поведение перспективных материалов» 5–8 ноября 2019 Москва, с. 162.

Publications:

36 scientific articles(12 of them in journals with international citation indexes - ICI) have been published on the topic of the dissertation: 25 articles in publications recommended by the SAC under the President of the Republic of Azerbaijan and 11 conference materials.

Participation of author:

The topic, idea and main direction of research of the dissertation work are defined by the author. The main goal and the issues that need to be solved during the research were evaluated by the author. ABX_2 (A=Cu, Tl; B=Fe, Cr, Co, Ni, Ga, In; X=S, Se, Te) and their basic solutions, processing of results and their analysis, general provisions for defence, conclusions and recommendations compiled.

Name of the organization where the work is done:

The dissertation work was carried out in the "Resonance Phenomena in Solids" laboratory of the Physics Institute of the Azerbaijan National Academy of Sciences.

The scope, structure and main content of the dissertation:

The submitted dissertation consists of an introduction, six chapters, main conclusions and a bibliography of 203 names. The work of the dissertation consists of a total of 381,754 signs, 82 pictures and 13 tables.

The content of the work

The introduction substantiates the relevance of the chosen research topic, formulates the purpose and main tasks of the dissertation work, scientific novelty, practical significance of the work, presents the main scientific provisions submitted for defense,

and briefly outlines the main content of individual chapters of the dissertation work.

The first chapter is devoted to the review of research studies of phase transition and jump conductance of low-dimensional crystals.

This chapter discusses the fluctuation magnetic structures that arise in magnetic systems of low-dimensional crystals with antiferromagnetic exchange and interactions. Due to strong quantum fluctuations in the competition of exchange interactions between the nearest ions and second neighbors in the chain, such systems exhibit a wide range of unusual magnetic structures.

One of the most promising classes for spintronics research and study is copper chalcogen compounds or chalcopyrites.

In a number of semiconductor compounds with the general formula ABX_2, the metal atoms A and B tend to exhibit different valences, which allows obtaining new materials for spintronics and semiconductor optoelectronics.

Compared to other groups of substances, 3d compounds have a surprisingly wide variety of electrical properties. Triple $TlMeX_2$ (Me=Fe, Cr, Co, Ni and X=S and Se) compounds belong to the class of anisotropic magnetic semiconductors.

Electronic structures, electrical and magnetic properties in chalcogenides of transition metals are complicated by the presence of 3d-electrons. These properties can best be analyzed by extending the molecular-orbitals and band model to include d-electrons.

The metal-dielectric Transition (MDT) belongs to the class of electronic phase transitions, because its main characteristic is the reconstruction of the electronic energy spectrum - the generation of a spin density wave (SDW) and a charge density wave (CDW) energy band that separates the under-occupied states and the over-unoccupied states. In most cases, the temperature dependence of the electrical resistivity $\rho(T)$ changes abruptly at the Curie temperature T_C point, which indicates a phase transition of the first type. Sometimes this case is small or non-existent, so it is possible to have a first-type phase transition or a second-type phase transition close to a second-type phase transition.

In the last few years, structural phase transitions in layered thallium compounds of type $TlBX_2$ (where B=In^{3+}, Ga^{3+} and X=S, Se, Te) have been discovered and are intensively studied. In all these layered crystals, below a certain temperature T_0, displacement of atoms occurs from the equilibrium state of the extreme structure - the basic lattice. The period of this extreme structure is, as a rule, not proportional to the period of the main lattice (lattice above T_0), and with the subsequent decrease in temperature, the extreme structure approaches a structure proportional to the initial structure, either continuously or abruptly.

Studying the lattice dynamics of such materials provides information that reflects and characterizes the nature of changes in intra- and interlayer interactions, on the basis of which various microscopic models can be constructed.

The second chapter is devoted to structural-crystalline chemical properties of chalcopyrite and $CuFeTe_2$ kind compounds of its various types. In this chapter, all the known facts are summarized and a large group of tetrahedral coordination semiconductor compounds is considered based on the principle of close approach. After looking at the crystal-chemical properties of this class of compounds, the reasons for the formation of phases in chalcopyrite and its different types of structures and their compliance with the law are explained. The main physical-chemical properties of triple and more complex compounds are analyzed and appropriate conclusions are drawn about the differences and contradictions obtained from the experimental results of various authors.

X-ray structure studies showed that plate-like crystals are tetragonal: a=0.402 nm, c=0.604 nm, space group P4/nmm, Z=1, ρ=6.5g/cm^3. X-ray structure studies also showed that chalcopyrite-type $CuFeTe_2$ (a, b, c) single crystals are the most perfect.

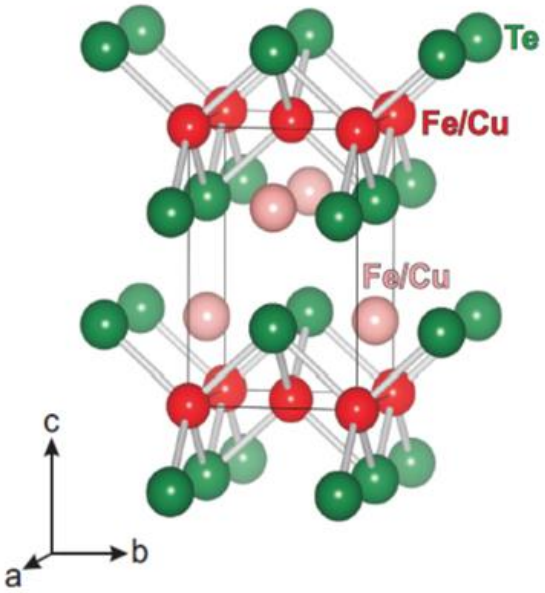

Image. 1. Crystal structure of $CuFeTe_2$

Figure 1. shows the crystal structure of $CuFeTe_2$, where the Cu and Fe atoms (marked in red) are located in 2a (0 0 0) crystallographic positions with about 50% for each of these atoms. Te atoms (marked in green) are located in 2c(0½z) crystallographic positions. Additional sites in [2c′ -(01/2z′)] crystallographic positions (marked in pink) are partially filled with Cu and Fe atoms, less than 15% capacity. Therefore, in order to determine the nominal composition of single crystals and clarify their crystal structures, they were analyzed in an EDX-type analyzer (Energy Dispersive X-ray Analysis) in an x-ray device and repeated by the analysis method in a Perkin Elmer Analyst-800 device.

The automatically calculated atomic percentages correspond to the following values:

- for single crystals of $CuFeTe_2$ grown from stoichiometric composition: Cu=24.53 ± 1.85; Fe=28.68 ± 1.86; Te=46.79 ± 2.27;

- for $Cu_{1.15}Fe_{1.23}Te_2$ single crystals grown from polycrystalline composition: Cu=27.95 ± 1.98; Fe=25.47 ± 1.61; Te=46.59 ± 2.18 (in

atomic percent for composition). The chemical composition of single crystals calculated on the basis of the above analysis values corresponds to the formula $Cu_{1.15}Fe_{1.23}Te_2$.

The structural formula of chalcopyrite type single crystals is as follows:

$Cu^{1+}Fe^{3+}Te_2$ ($Cu_{0.22}$ and $Fe_{0.1}$), $Cu^{1+}Fe^{3+}Te_2$ ($Cu_{0.13}$ and $Fe_{0.22}$) and $Cu^{1+}Fe^{3+}Te_2$ ($Cu_{0.15}$ and $Fe_{0.23}$) and "excess" Cu (metal atoms (one- or two-valent) and Fe (two- or three-valent) are chaotically located in the cavities of the crystal lattice and behave as additives (nanoparticles).

In normal group metals, the exchange forces in the system of collective electrons are broken by the Fermi energy and cannot lead to the formation of magnetic order. The situation may be more favorable for exchange forces in compounds of transition 3d-metals. Considering that the crystal structure of a solid body has 230 spatial symmetry groups, and the atomically ordered magnetic structure has 1651 spatial symmetry groups, therefore, in some cases, ferromagnetism or antiferromagnetism is observed in them as a result of the dominance of exchange relationships. By maintaining a significant localization of charge and spin density in the crystal nodes, the existence of narrow energy bands of the former internal 3d-electrons makes possible the spin regularization of the collected s- and d-electrons in the 3d-system of metals and compounds. Thus, the origin of the exchange bond of electronic magnetic moments in semimetals and semiconductors is due to the active influence of conduction electrons on the system of uncompensated magnetic moments of former 3d-electrons.

In crystals which have antiferromagnetic order, when conduction electrons move from one magnetic sublattice node to another sublattice node, electron spin moves from one node with a certain value and direction to another node with a different spin value and direction. Therefore, the state of conduction electrons in a crystal with several magnetic sublattices must differ significantly from the state of conduction electrons in a crystal with one sublattice or in a paramagnetic crystal. The difference is that in an antiferromagnetic crystal, spatial degeneration of conducting electrons is partially removed, because the presence of several magnetic sublattices causes a change in the

periodicity of the potential field in which the electrons move (that is, a decrease in its symmetry). In this case, for conduction electrons with different projected spins, the shift of energy subbands does not occur, but nevertheless the energy spectrum can change significantly. For example, in the middle part of the 4s-energy band of conduction electrons, it is possible to create a forbidden part ΔE_{af}, similar to what happens in alloys during atomic ordering.

Thus, the presence of magnetic order in the transition metal crystal leads to significant changes in the energy spectrum of conduction electrons. Moreover, two main effects are observed here:

1) elimination of spin degeneration (by direction) and

2) elimination of space degeneration.

The results of the conducted experimental studies answer all the questions of the existing disagreements in these materials and allow us to draw new conclusions about the process of phase formation and the manifestation of these or other properties. In this chapter, enough space is also given to the chemical analysis of single crystals grown from different compositions of polycrystals.

Then, the results of studies of electrical conductivity (σ), thermo-EMF(S), magnetic susceptibility(χ), magnetization(M) of $CuFeTe_2$(a, b and c) type chalcopyrite single crystals in a wide temperature range are explained.

The investigated low-dimensional ABX_2 (A=Cu, Tl; B=Fe, Cr, Co, Ni, Ga, In; X=S, Se, Te) type semiconductors and their solid solutions were pressed under a pressure of P=1.5 x 10^3kg/cm^2 and have the shape of a rectangular parallelepiped designed for electrical measurements. Before striking the contacts, they are heated to a temperature of ~450K. Ohmic contacts are created by electrolytic deposition of copper. The electrical conductivity σ(T) and thermo-EMF(S) coefficients of the obtained samples were measured with four probe methods up to 3% accuracy in the temperature range of 77÷400K.

Two main directions can be distinguished in the problem of phase transitions. First, it is related to the study of the behavior of the system properties around the critical temperature. Another direction on which the microscopic theory of phase transition is based is to analyze the

properties of the ground state of the system within the framework of one or another model concepts and explain the possible causes of its instability, under the influence of which the system changes to a newer, more favorable energy state. The process of merging these two directions into a single phase transition theory is currently being investigated.

The temperature dependences of the magnetic susceptibility $\chi(T)$ and the thermal capacity $C_p(T)$ repeat the dependence of $\chi(T)$, the presence of anomalies in the Neel temperature region, the generation and dissipation of spin-glass waves (SGW), and the locking temperature, repeating the dependence of $\chi(T)$ (characterized by T_L). For $CuFeTe_2$(a, b, c) chalcopyrite-type single crystals, the Neel temperature T_N=65K and the locking characteristic of supermagnets TB>360K were determined. Fe^{3+} cations are located between two Te^{2-} anions, and the antiferromagnetic magnetic moments of the spins are directed parallel to the opposite (001) plane. The magnetic crystal structure of $CuFeTe_2$ consists of eight formula units-8($CuFeTe_2$) Z=8, whereas in the crystal structure it is Z=1. Effective magnetic moments of paramagnetic clusters were measured in weak (100÷1000Oe) and strong (10÷50kOe) magnetic fields and it was shown that weak $\mu_{eff.} \geq 0.7\mu_B$ and strong $\mu_{eff.} \sim 2.4\mu_B$ (μ_B is Boron magneton). Below T_g=55K, the spin-glass state is observed in $CuFeTe_2$(a, b, c) chalcopyrite type single crystals. The different behavior of the dependences of the $\chi^{ZFC}(T)$ magnetic field at zero and $\chi^{FC}(T)$ magnetic field at 1kOe during cooling below the locking temperature is one of the main signs indicating the clustering of the chalcopyrite-type $CuFeTe_2$ single crystal (Fig. 2).

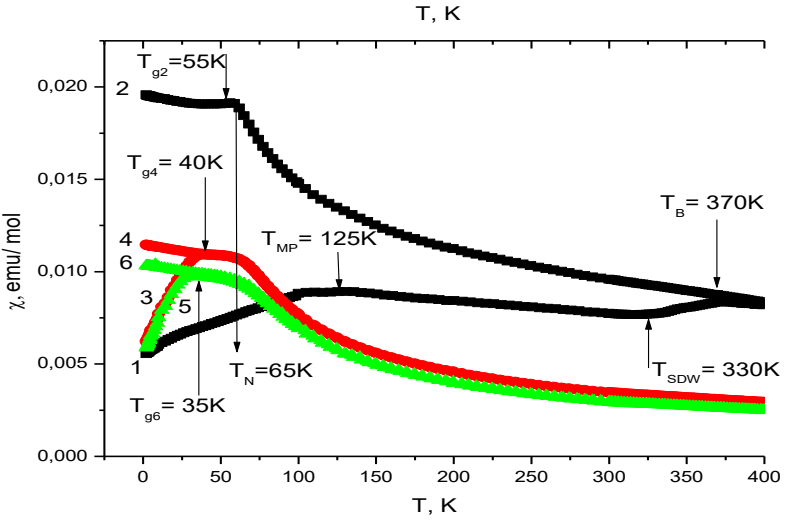

Fig. 2. Temperature dependence of magnetic susceptibility of $Cu_{1.13}Fe_{1.22}Te_2$ single crystal:
1-1.0, 3-10 and 5-20kOe in ZFC and FC – 2 – 1.0, 4 - 10 and 6 - 20kOe. Temperatures: T_{MP}=125K, spin-glass case in magnetic fields: 1.0 кОе – T_{g2}=55K, 10kOe – T_{g4}=40K and 20kЭ – T_{g6}=35K. Spin density wave T_{SDW}=330K and locking temperature T_L=370K Effective magnetic moment $\mu_{eff}=2.0\div2.4\mu_B$.

Fig. 2 shows the field dependence of magnetization (hysteresis) of chalcopyrite-type $CuFeTe_2$ single crystal at different temperatures. It is clear that at high temperatures (above 100 K), the magnetization (with field) increases almost linearly, as is characteristic of the paramagnetic region. At low temperatures (2K and 4K), a hysteresis loop is observed, which changes its position in the field dependence of magnetization (measurements were made in FC – 50÷0÷50kOe and the opposite mode). The hysteresis loop is wider and ferrimagnetic type. In all chalcopyrite-type $CuFeTe_2$(a, b and c) single crystals, iron (Fe^{3+}) is also in the trivalent state, which agrees with Messbauer effect. It has been shown in the literature that the Messbauer spectrum of $CuFeTe_2$ consists of a single peak that splits into six components

due to the extremely weak magnetic interaction.This is due to the magnetically ordered atoms of iron (Fe^{3+}) occupying a position in the structure. The isomer shift is small, and the quadrupole splitting is absent. The observed parameters are consistent with the idea that magnetically bonded Fe^{3+} atoms are located in tetrahedral positions in high spin states. The residual magnetization was determined from the field dependence of the M(H) magnetization (hysteresis) at different temperatures of $CuFeTe_2$ single crystal. It can be seen that at high temperature (above 100 K) the magnetization (with field) increases almost linearly, as is characteristic of the paramagnetic region. At low temperatures (2K and 4K), a shifted hysteresis loop is observed in the dependence of the magnetization on the field (measurements were made in the mode of the magnetic field –50–0 + 50 kOe). The hysteresis loop is wider and ferrimagnetic type. In all single crystals of $CuFeTe_2$(a, b and c) type chalcopyrite, iron (Fe^{3+}) is in the trivalent state according to Mössbauer effect. It is shown in the literature that the Mössbauer spectra of $CuFeTe_2$ consist of a single peak divided into six components due to the extremely weak magnetic interaction. It is due to the magnetically arranged iron atoms (Fe^{3+}) occupying one position in the structure. The isomeric shift is small and there is no quadrupolar splitting. The observed parameters are consistent with the concept of magnetically bound Fe^{3+} atoms in a high spin state in tetrahedral positions.

Interatomic interaction in layered crystals leads to unusual temperature dependence of heat capacity with special laws at low temperatures. The same has been observed in anisotropic layered magnets where the phase transitions are variable in nature.

The heat capacity of $Cu_{1.04}Fe_{1.12}Te_{1.84}$ was measured on a commercial Quantum Design PPMS (Physical Property Measurement System) in the range 2÷306K. Fig.3 shows the temperature dependence of the heat capacity (C_p), showing the experimental results, and the difference between the experimental heat capacity and the lattice heat capacity calculated according to the Debye model. C_{pexp} - C_{pD}=$\Delta C_p(T)$, where C_{pexp} is the difference between experimental results and C_{pD} values of the lattice heat capacity calculated according to the Debye model. (The Debye temperature is

the temperature at which all frequencies of the given $CuFeTe_2$ compound are excited. Further increase in temperature does not lead to the creation of new oscillations, but causes an increase in the amplitudes of the existing ones, that is, the average energy of the oscillations increases with the increase in temperature.

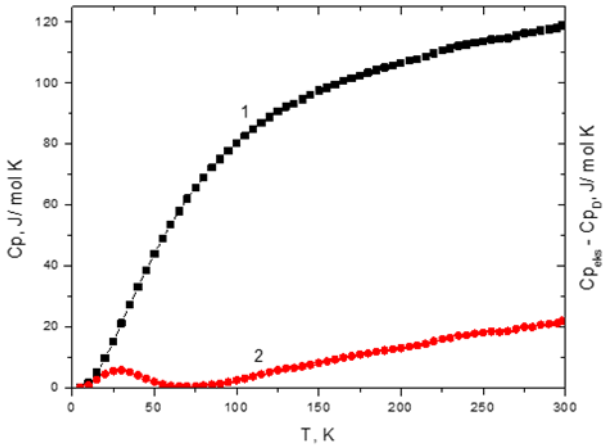

Fig.3. Temperature Dependencies of heat capacity.

Fig.3 shows the temperature dependence of heat capacity (1) and (2) additional heat capacity of $Cu_{1.04}Fe_{1.12}Te_{1.84}$ compound ($C_{pexp} - C_{pD}$). C_p and C_{Pexp} are experimental value of heat capacity and C_{pD} are calculated lattice values of heat capacity according to Debye model. The $C_p(T)$ linear section covers the temperature range of 65-125K. The $\Delta C_p(T)$ curve (1) undergoes discontinuity at antiferromagnetic temperature T_N=65K and spin-glass transformation.

Fig.3 clearly shows the phase transitions ΔC_P=0 from the dependence of $\Delta C_p(T)$ antiferromagnetic-paramagnetic (T_N=65K) and spin-glass state (T_g=55K).

To see these phase transitions, the temperature dependence of the heat capacity versus temperature is shown in figure 4(a).

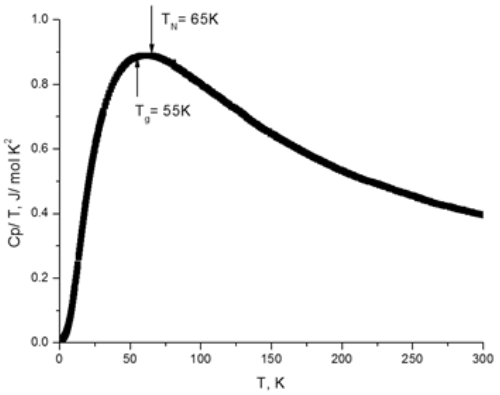

Fig.4. Temperature dependence of the ratio of heat capacity to temperature.

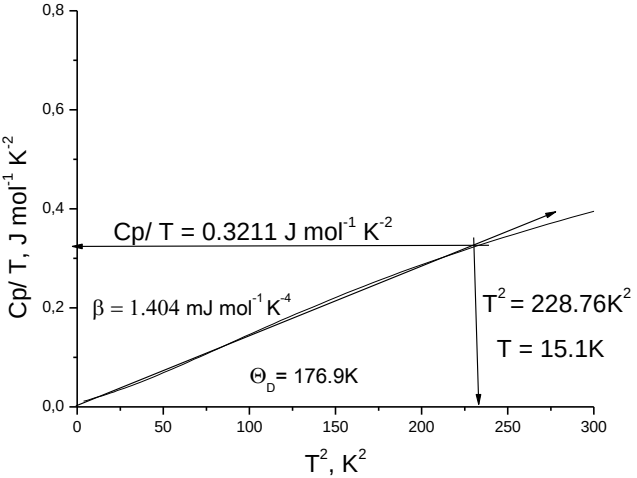

Fig. 4a. Dependence of the ratio of heat capacity to temperature on the square of temperature (Debay's cubic law).

In the temperature dependence of the heat capacity $\Delta C_p(T)$ it is spread close to the phase transition temperature and it determines the identity between the second type of phase transitions and critical

phenomena. Near the magnetic phase transition temperature, due to the development of fluctuations, the homogeneous phase is separated into a large number of spin "groups" that form a magnetically dispersed system. The temperature change at the phase transition reduces the dispersion and the system becomes homogeneous. The presence of this dispersion in the phase transition region leads to scattering of the maximum in the temperature curve of the heat capacity. The analysis of the results obtained at low temperatures clearly shows this. At low temperatures, the kinetic energy is small, so the behavior of the $C_p(T)$ and C_p/T dependences will be completely different. In the low kinetic energy region, the temperature dependence of C_P/T was established (Fig.4) and phase transitions due to small changes in potential energy in the curve were visible. Taking into account the unusual nature of the magnetic phase transition in $Cu_{1.04}Fe_{1.12}Te_{1.84}$, the temperature dependence of C_p/T, $C_{Pexp}(T)$ is established on the basis of experimental data.

Fig.4 and 4a clearly demonstrate the unusual (diffusion, scattering and fluctuation) nature of the phase transition.

The results of experimental data processing according to the method described in this work are presented in fig.4a, where $n_0=4$ is the number of atoms in the molecule of the studied crystal, $3k_BN=R_g$ is the gas constant, N is Avogadro's number, k_B is Boltzmann's constant, Θ_D is the Debye temperature. The figure (Fig. 4a) shows the dependence of C_p/T on T^2 (Debay's cubic law). Within $T \ll \theta_D$

$C_{\text{lattice}} \rightarrow \beta \cdot T^3 = 12\ R\pi^4/5 \cdot (T/\theta_D)^3$ (1)

Debye temperature from experimental data:

$\Theta_D = (1942.7\ n_0/\beta)^{1/3}$ (2)

where $n_0=4$ is the number of atoms in the molecule of the studied crystal, calculated by the formula $\beta=1.452 mJ \cdot mol^{-1}K^{-4}$. $\Theta_D=176.9K$ is the Debye temperature at $T \rightarrow 0K$. This indicates that the structural anisotropy in the compounds does not affect the heat capacity behavior below 15.1 K (Fig. 4a).

We used the characteristic equation for layered crystals with a linear part of the temperature dependence to determine the anisotropy parameter (η) and the two-dimensional Debye temperature (Θ_2). From Fig.4b, it is easy to evaluate $\eta=0.309$ and $\Theta_2=122.2K$ $C_P/3n_0k_BN=$ -

0.447 at T=0 and η=0.309, and $\Theta_2=(1 - \eta)\cdot\theta_D$=122.2K where $\eta=|\Delta C_p/(1+\Delta C_p)|$.

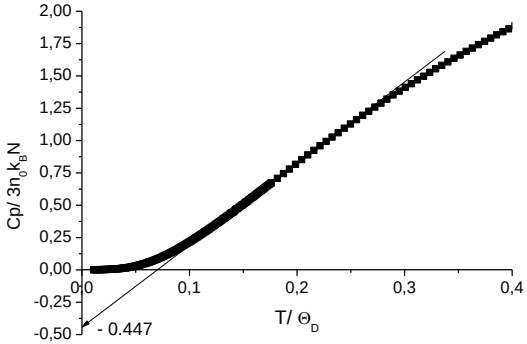

Fig.4b. Temperature dependance of heat capaticy in $C_P/3n_0k_BN - T/\Theta_D$ coordinates.

The following conclusions are drawn from the above:

1. $Cu_{1.22}Fe_{1.10}Te_2$ (a), $Cu_{1.04}Fe_{1.12}Te_{1.84}\approx Cu_{1.13}Fe_{1.22}Te_2$ (b) and $Cu_{1.15}Fe_{1.23}Te_2$ (c) single crystals with P4/nmm tetragonal structure were grown.

2. Superparamagnetic locking temperatures T_L=358K (a), T_L=360K (b) and T_L=370K (c) and Neel temperature T_N=65K were determined for single crystals of $CuFeTe_2$(a, b and c). Magnetic moments parallel and perpendicular to the (001) plane and paramagnetic Curie temperature were calculated for $CuFeTe_2$(a, b and c) type chalcopyrite AFM single crystals:

a) $\mu_{eff}\leq 0.7\mu_B$ in a magnetic field of 100÷1000Oe for $CuFeTe_2$(a, b and c);

b) For $CuFeTe_2$(a, b and c) $\mu_{eff}\|\approx 2.0\div 2.6$ and $\mu_{eff}{}^{\perp}=2.0\div 2.30\mu_B$ in 10÷50kOe magnetic field, as well as paramagnetic Curie temperature $\Theta_P\|$ and $\Theta_P{}^{\perp}$ is negative.

It was determined that iron (Fe^{3+}) is trivalent in all $CuFeTe_2$ single crystals (a, b and c).

3. It is shown that $CuFeTe_2$ single crystals (a, b and c) exhibit a spin-glass state at freezing temperatures T_g=35, 50 and 55K, respectively.

4. It was determined that Fe^{3+} cations are located between two Te^{2-} anions, and the magnetic moments of the spins are directed parallel to the (001) plane, which is opposite to each other (antiferromagnetic). The magnetic structure of these crystals: Cu^{+}-Te^{2-}-Fe^{3+}-Te^{2-} consists of four "clusters" located opposite each other.

5. It was established that the temperature dependences of electrical conductivity Ϭ(T) and thermo-EMF S(T) of $CuFeTe_2$ (b) single crystal can be divided into four regions:

a) temperature range 77÷121K, semiconductor region: ΔE_a=0.008eV (a), ΔE_a=0.002eV (b) and ΔE_a=0.002eV (c);

b) temperature range 155÷267.5K - where spin wave density (SDS) exists (metal domain): ΔE_a=0.013eV (a), ΔE_a=0.008eV (b) and ΔE_a=0.013eV (c), where (a), (b) and (c) are the activation energies of compressed single crystals;

The difference between the activation energies before and after the semiconductor-metal transition is the energy spent on the dissolution of this phase transition. This difference shows values of 0.005eV, 0.006eV and 0.011eV for chalcopyrite-type $CuFeTe_2$(a, b and c), respectively. This is the energy spent on the breakdown of the semiconductor-metal phase transition, and the duration of this transition increases with the increase of iron ions.

c) temperature range of 289÷360K - SDW is observed here;

d) 360÷390K temperature range (after T_L locking temperature) – being in superparamagnetic state.

6. Anomaly in the metal-semiconductor (bivalent copper and iron) transition temperature region, degeneration and disintegration of SDW from the thermo-EMF S(T) temperature dependence of the compound $CuFeTe_2$ (b), as well as the temperature dependence of the magnetic susceptibility χ(T) by reproducing T_L The locking temperature of is determined.

7. $CuFeTe_2$ at temperature 77÷120K corresponds to a hopping charge transport mechanism with variable hopping length R_L=2.1nm at 77K and R_L=2.3nm at 120K. The density of localized states near

the Fermi level was determined in $CuFeTe_2$ $N_F=5.3 \times 10^{21} eV^{-1} cm^{-3}$. The average hopping distance calculated in $CuFeTe_2$ was $R_{av}=2.2nm$, and the energy spread of localized states near the Fermi level was $\Delta E=0.01eV$. The value of ΔE corresponds approximately to the average value of the activation energy of the hopping in $CuFeTe_2$, which is $\Delta W=0.005eV$. The approximate concentration of localized states in the $CuFeTe_2$ band gap was $Nt=5.3x10^{19} cm^{-3}$.

8. Experimental studies have shown that magnetic interactions (Cu^{+}-Te^{2-}-Fe^{3+}-Te^{2-}) are antiferromagnetic "clusters". It should be noted that the magnetic structure of these chalcopyrite single crystals of type $CuFeTe_2$(a, b and c) consists of eight formula units - 8($CuFeTe_2$) Z=8, so Z=1 in the crystal structure. Fe^{3+} cations are located between two Te^{2-} anions, and their magnetic spins are directed parallel to each other (antiferromagnetically) parallel to the (001) plane.

9. In this case, one of the two Te^{2-} 3p valence electrons can be transferred to the half-filled layer of the Fe^{3+} transition metal ion (with five 3d electrons), and according to Hund's rule, its spin will be antiparallel to the spin of five d-electrons. The spin of the second remaining electron in tellurium ions will be antiparallel to the spin of the first electron (according to the Pauli exclusion principle). The same relationship with the antiparallel arrangement of unpaired electrons can occur with another metal ion (Fe^{3+}) on the opposite side of the tellurium ion.

10. The detected spin-glass ($T_g=55K$) and antiferromagnetic ($T_N=65K$) phase transitions are characterized by the deviation of the heat capacity in the dependence of C_p/T on T, characteristic of quasi-two-dimensional materials.

11. Anisotropy parameter $\eta=0.309$. The three-dimensional (3D) Debye temperature $\Theta_D=176.9K$ and the two-dimensional (2D) Debye temperature $\Theta_2=122.2K$ are found.

$TlFeX_2$ triple compounds are 3d-compounds compared to other groups of matter and have extremely different electrical properties. $TlFeX_2$ triple compounds belong to the class of anisotropic magnetic semiconductors. Single crystals of $TlFeS_2$ and $TeFeSe_2$ were grown by the modified Brigman-Stockbarger method. The conducted

structural studies showed that the received $TlFeS_2$ crystals have a chain structure and the parameters of the crystal lattice take the following values: a=1.1643, b=0.5306, c=0.6802nm; β=116.75º, space group C2/m. TlFeSe2 unit cell parameters a=1.202, b=0.55, c=0.713nm; A monocline with β=118.52º crystallizes in syngony.

The samples for electrical measurements were pressed into $TlFeS_2$ and $TlFeSe_2$ crystal fibers under pressure and took the shape of a parallelepiped with dimensions of $12.5x5.0x1.3mm^3$ and $12.5x5.0x2.3mm^3$, respectively. Pressed ($P=10^3kg/cm^2$) samples were then annealed at 450K. Ohmic contacts are created by electrolytic deposition of copper. Electrical conductivity (Ϭ) and thermo-EMF coefficients (S) of TlFeS2 and TlFeSe2 samples were measured by four probe methods in the temperature range of 85÷400K [78, 154, 166] .

At the Neel temperature, a small deviation of the conductivity from the temperature dependence of the electrical conductivity Ϭ(T) is observed, which is explained by the change in the activation energy of the hopping process. It is clear from the experimental results that the conductivity of the $TlFeS_2$ sample at T<200K and for the $TlFeSe_2$ sample above 250K does not depend on the temperatures. The temperature-independent conductivity can be related to the collapse of the antiferromagnetic order of these crystals (T_N=196K and 290K, respectively)[3].

From the thermo-EMF S(T) temperature dependence of $TlFeS_2$ compound, there is an inversion of sign at Neel temperature T_N=196K, and for $TlFeSe_2$ at 290K. The effect on the formation of the Sp-d interaction consists in the change of the sign of the temperature dependence of the thermo-EMF and the appearance of a negative S(T) section at high temperatures. A further increase in temperature leads to the depletion of this previously completely filled subzone. The latter leads to an increase in the number of current carriers through holes in a given d-subsystem. At the same time, carriers in the other d-subsystem, whose energies shift down, also increase. Magnetic studies of $TlFeS_2$ and $TlFeSe_2$ compounds show an antiferromagnetic phase transition below T_N=196K for $TlFeS_2$ single crystals and T_N=290K for $TlFeSe_2$. Magnetic susceptibility measurements for

single crystals of $TlFeS_2$ and $TlFeSe_2$ in the temperature range of 2÷400K showed the behavior characteristic of quasi-one-dimensional antiferromagnets. These magnetic studies are consistent with neutron diffraction and EPR measurements. $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ solid solution study of temperature dependence of conductivity and thermo-EMF coefficient is shown in fig.5.

For electrical measurements, $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ samples have a parallelepiped shape: 1 - single crystal 14.48x1.4x1.26mm^3 and 2 - pressed single crystal 10.9x5.27x1.0mm^3, 3 - rubbed and pressed 8.7x5.09x1.14mm^3.

Contacts to the head and foot planes with the crystals are created by electrolytic deposition of copper. The temperature dependences of electrical conductivity (б) and thermo-EMF (S) of the obtained samples were measured with an accuracy of up to 3% by the four-probe method in the temperature range of 80÷355K. The amplitude of the external constant electric field applied to the studied samples during electrical measurements corresponds to the ohmic region of VA [1, 60, 85, 87]. Experience shows that at low concentrations (up to 10%) of substitutional cations, the resistance of solid solutions usually increases. This may indicate that at low concentrations the substituted cations cannot be evenly distributed in the crystals, resulting in local violations of the crystal symmetry. This can lead to a decrease in the free path of electrons and, accordingly, their mobility.

With the increase in the concentration of the substituted gallium cation (x) in solid solutions, the mechanism of interaction between the dopant centers, expressed in practice by conductivity, is activated. As the temperature rises, the internal conduction mechanism gradually begins to take effect, and the dependence lnб=f(T) becomes exponential.

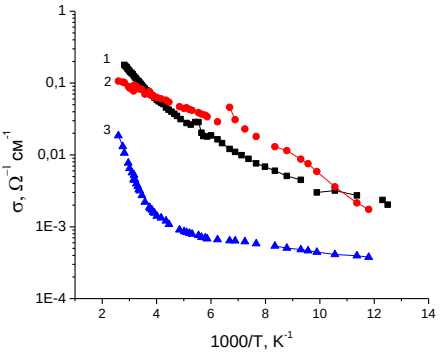

Fig. 5. $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ sample
Ϭ(1000/T) of electrical conductivity from temperature dependencies; 1 - single crystal ($Ϭ_{mon}$), 2 – along the a axis pressed single crystal ($Ϭ_p$) and 3 –rubbed and pressed. ($Ϭ_{ovul.p}$).

It was calculated from Fig. 5 that the activation energy (ΔE_a) of $TlFe_{0.975}Ga_{0.025}S_2$ samples in the paramagnetic region in the temperature range above the Neel temperature: 1 - single crystal ΔE_{amon}=0.083eV, 2 - pressed ΔE_{ap}=0.103eV -$\Delta E_{aovul.p}$=0.337eV.

In the magnetic ordered region at a temperature lower than T_N, the activation energy was: 1 – ΔE_{amon}=0.137eV for single crystal, 2 – ΔE_{ap}=0.063eV for pressed single crystal and 3 – $\Delta E_{arub.p}$=0.02eV for rubbed and pressed single crystal. The differences of these activation energies were 1 – 0.054, 2 – 0.04 and 3 – 0.317eV, respectively, which was spent on energy dissipation in the AFM phase transition.

Fig. 6 shows the temperature dependence of thermo-EMF S(T) of $TlFe_{0.975}Ga_{0.025}S_2$ solid solution.

1 - single crystal S_{mon}, 2 - pressed S_p and 3 - rubbed and pressed $S_{rub.p}$, get different values at Neel temperature (T_N). Thermo-EMF at the temperature of the magnetic phase transition of the $TlFe_{0.975}Ga_{0.025}S_2$ solid solution at the Neel temperature takes the following values:

1 – single crystal S_{mon}=0 μV (T_N=150K),

2 - pressed S_p=44μV (T_N=180.5K) and

3 – rubbed and pressed $S_{rub.p}$=24.8μV (T_N=180.5K). These results indicate that mechanical motions affect the antiferromagnetic phase transition.

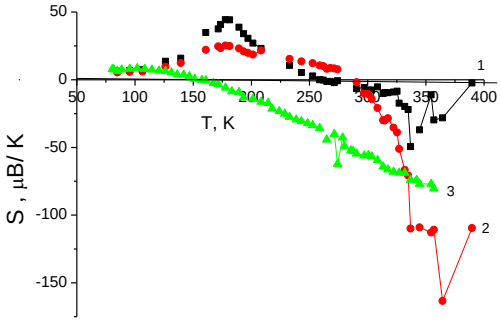

Fig. 6. Thermo-EMF S(T) of samples from temperature dependencies $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$; 1 - single crystal ($S_{mon}$), 2 – single crystal pressed along the a axis (S_p) and 3 – rubbed and pressed ($S_{rub.p}$).

At the Neel temperature ρ(T), the resistance has a small break, which is explained by the change in the activation energy of the hopping process. The experimental results show that the conductivity of the $TlFeS_2$ sample at T<200K does not depend on temperature. The temperature-independent conductivity may be related to the breakdown of the antiferromagnetic order (T_N=196 K) of this crystal.

At T_N=196K, the sign inversion occurs in the S(T) dependence of the compound $TlFeS_2$. The influence of the Sp-d interaction on the formation of the temperature dependence of thermo-EMF consists of a change of sign and the formation of a negative part in the S(T) dependence at high temperatures. The subsequent increase in temperature leads to the emptying of the lower zone, which was previously completely filled. The latter causes the number of current carriers to increase with holes in a given d-subsystem. At the same time, the number of current carriers in another d-subsystem, whose

energy shifts downward, also increases. Magnetic studies of $TlFeS_2$ and $TlFeSe_2$ compounds show an antiferromagnetic phase transition below temperatures of T_N=196K for $TlFeS_2$ single crystals and T_N=290K for $TlFeSe_2$. Magnetic susceptibility measurements of $TlFeS_2$ and $TlFeSe_2$ single crystals in the temperature range of 2-400 K, which are characteristic of quasi-one-dimensional antiferromagnets showed the behavior. These magnetic studies are in agreement with neutronographic and EPR measurements.

The temperature dependence of electrical conductivity at the value of three-layer solid solution $(TlFeS_2)_{1-x}(TlGaS_2)_x$, x=0.025 σ(1000/T) is characterized by monotonically decreasing activation energy with decreasing temperature. This behavior of conductivity in a solid solution at temperatures of 80÷180K (state of magnetic order) is characteristic of the variable-length hopping mechanism of charge transport, when the current is transferred by charge carriers located in localized states near the Fermi level. In this regard, the temperature dependence of the reconstructed conductance in Mott coordinates $lg(\sigma)=f(T^{-1/4})$ was also proved.

The density of localized states near the Fermi level was estimated as $N_F=2.9x10^{19}eV^{-1}cm^{-3}$. In this case, the value of a_L=1.4nm was taken from binary GaS for the localization radius. $N_F=1.7x10^{18}eV^{-1}cm^{-3}$ for $TlFeS_2$. In the studied $(TlFeS_2)_{1-x}(TlGaS_2)_x$ samples, the hopping length (R_L) of charge carriers at different temperatures of 80÷180K was calculated. In $TlFe_{0.975}Ga_{0.025}S_2$ at temperatures of 80÷180K R_{av}=6.2nm, the ratio $R_{av.}/a_L$=4.4. Note that $R_{av.}$=10.9 nm in $TlFeS_2$. For $(TlFeS_2)_{1-x}(TlGaS_2)_x$ (where x=0.025) solid solution, the energy dispersion of trap states near the Fermi level was ΔE=0.07eV, and the concentration of deep traps was $Nt=2x10^{18}$ cm^{-3}. (Fig. 7).

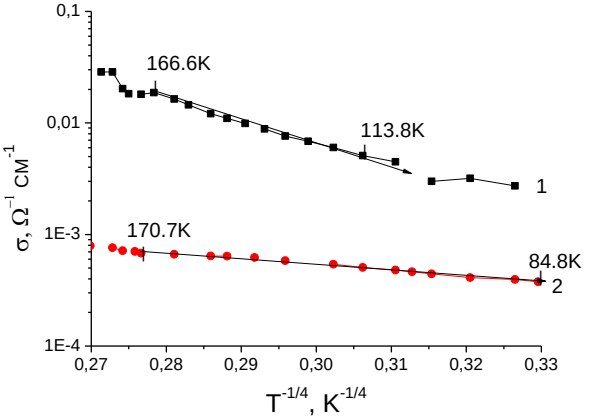

Fig.7. Temperature dependence of the conductivity of the $TlFe_{0.975}Ga_{0.025}S_2$ $\sigma(T^{-1/4})$ sample in the Mott coordinate: 1-single crystal, 2-pressed single crystal.

In the temperature range of 128-178 K, it was determined that the layered single crystal of $(TlFeSe_2)_{0.5}(TlGaSe_2)_{0.5}$ solid solution has hopping conductivity in a constant electric field along natural layers. Localized near the Fermi level $N_F=2.8x10^{17}eV^{-1}cm^{-3}$ states have variable hopping length conductivity and their dispersion $\Delta E=0.13eV$, $R_{av.}=23.3nm$ and concentration of deep traps $Nt=3.6x10^{16}cm^{-3}$.

In ferromagnets, antiferromagnets, and ferrimagnets, the spins of the atoms and their associated magnetic moments are strictly ordered in the absence of excitation. The excited state of the magnetic system is due to the deviation of the spin from the equilibrium state. Due to the interaction between atoms, such deviation is not localized, but propagates in the form of a wave in the magnetic medium. In the $\rho(T)$ curve of $(TlFeSe_2)_{0.5}(TlGaSe_2)_{0.5}$ solid solution at $T_{his}=109\div120$ K, a temperature hysteresis is observed, which is a transition to the incommensurate phase at $T_1\approx117.2K$ ($TlGaSe_2$) and $T_C\approx114K$- is also associated with a proportional phase transition.

It is characterized by a monotonically decreasing activation energy with decreasing temperature in the lower part of the conductivity (σ) 1000/T dependence curve. Such conductance

behavior at low temperatures from 80 K to 180 K for sulfur and up to 300 K for selenium in $TlFeS_2$, $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$, and $(TlFeSe_2)_{0.5}(TlGaSe_2)_{0.5}$ suggests a charge transport hopping mechanism. is typical for In this case, the current is carried by charge carriers localized near the Fermi level. This was also shown by the temperature dependence of the reconstructed conductance in Mott coordinates $lg\sigma=f(T^{-1/4})$.

The following conclusions can be drawn from the above:

1. We found that the temperature dependence of the reconstructed conductivity $\sigma(T)$ in Mott coordinates $lg\sigma=f(T^{-1/4})$ of $TlFeS_2$ can be used to estimate the jump distances at different temperatures. Thus, R_L=10.7nm at T=230K, R_L=11 1nm at T=203K. In the indicated temperature interval, the average jump distance is R_{av}=10.9 nm, and the ratio is R_{av}/a_L=8, that is, the average hopping distance significantly exceeds the average distance between charge carrier localization centers. The energy distribution of trap states close to the Fermi level was estimated from the R_{av} values of the $TlFeS_2$ sample: 1 - ΔE=0.005eV and 2 - ΔE=0.007eV.

2. It is shown that the temperature dependences of electrical conductivity $\sigma(T)$ and thermo-EMF S(T) of $TlFeS_2$ compound have anomalies. Thus, during the antiferromagnetic phase transition at T_N=196K, a break is observed in the temperature dependences of the electrical conductivity, and in the thermo-EMF dependences, a sign inversion from p-type to n-type is observed.

The calculated magnetic phase transition dissipation energy for pressed $TlFeS_2$ is 0.129 eV.

3. It was calculated that the tangent of the inclination angle of the dependence established in Mott coordinates in $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ solid solution is equal to $T_0=2.3x10^6$K, which allows us to estimate the density of localized states near the Fermi level. Localized Fermi states (N_F) are equal to $N_F=1.7x10^{18}eV^{-1}cm^{-3}$ for a single crystal and $N_F=2.9x10^{18}eV^{-1}cm^{-3}$ for a pressed single crystal. It was calculated that the charge carrier jump length (R_L) of the $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ samples studied at different temperatures in the temperature range of 80÷180K is 6.8nm at T=80K

and 5.6nm at T=180K. Thus, the average hopping distance of $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ in the temperature range of 80÷180K (antiferromagnetic regularity) was R_{av}=6.2 nm. The value of R_{av} was 4.4 times higher than the ratio of the average distance between the localization centers of charge carriers to the localization radius, that is, R_{av}/a_L=4.4. The value of the activation energy difference between the magnetically ordered and paramagnetic phases, which is 0.106eV and 0.013eV, is the energy spent on the dissolution of the magnetic phase transition of the single crystal and pressed solid solutions $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$.

4. Thermo-EMF S(T) of $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ solid solution is shown in temperature dependence, which undergoes sign inversion from p-type to n-type at S_{mon}=0μV in a single crystal at T_N=150K. A pressed single crystal at T_S=255K has S_p=0μV and a pressed single crystal rubbed at T_S=284K has $S_{rub.p}$=0μV. These values of Thermo-EMF show that the narrow energy band created by the p-type electrons of the sub-band dissipates with increasing temperature. In the 150÷180K region, the relative temperature dependence of electrical resistances $\rho/\rho_m(T)$ of $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ samples: pressed resistance $p/\rho_m(T)$ and rubbed pressed resistance $\rho_{rub.p}/\rho_m(T)$ have anomalies related to antiferromagnetic phase transition.

5. It was determined that the density of localized states close to the Fermi level was calculated from the temperature dependence of $lg\sigma=f(T^{-1/4})$ in samples of $TlFeSe_2$ compound, which was $N_F=3.3x10^{18}eV^{-1}cm^{-3}$. The average jump distance in TlFeSe2 compound was Ror=10.4nm. With the increase of temperature, the thermo-EMF coefficient increased slightly, reached its maximum value at Neel temperature T_N=290K, then decreased to zero at T=340K, after which there was a sign inversion from p-type to n-type. At T=400K, thermo-EMF (S) reaches -20 μkV/K. The calculated magnetic phase transition breakdown energy for pressed $TlFeSe_2$ is 0.129 eV.

6. Based on the temperature dependence of electrical conductivity $lg\sigma=f(T^{-1/4})$ in the layered single crystal of $TlGa_{0.5}Fe_{0.5}Se_2$ solid solution in the range of 128÷178K, it is determined that a variable-length hopping conductance occurs in a constant electric field near

localized states near the Fermi level along its natural layers has been done. Near the Fermi level, the density $N_F=2.8x10^{17}eV^{-1}$ cm^{-3}, their spread $\Delta E=0.13eV$, the average distance of jumps Ror=23.3nm, as well as the concentration of deep traps in this single crystal is $Nt=3.6x10^{16}$ cm^{-3}.

7. It was determined that in the $(TlFeSe_2)_{0.5}(TlGaSe_2)_{0.5}$ solid solution, there is a temperature hysteresis in the temperature dependence of the electrical conductivity $\sigma(T)$ at the temperature $T_{his}=109\div120K$, which indicates that the $TlGaSe_2$ compound enters the incommensurate phase at $T\approx117.2K$ and At $T_S\approx114K$, it is due to a phase transition to a commensurate magnetoelectric phase.

8. It is shown that the transition inversion of thermo-EMF from p to n-type in temperature dependence of thermo-EMF S(T) of $(TlFeS_2)_{0.95}(TlGaS_2)_{0.05}$ antiferromagnet at temperature $T_S=327K$ and ρ (at $T_N=190.9K$ A break occurs in the dependence of T). Thus, for $(TlFeS_2)_{0.9}(TlGaS_2)_{0.1}$ and $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ antiferrimagnets, thermo-EMF inversion from p-type to n-type at $T_S=150K$ and $T_N=$ at $\rho(T)$, respectively breakdown occurs at temperatures of 176.1K and $T_N=150K$.

9. It was determined that the sign is positive (p-type) in the thermo-EMF S(T) temperature dependence of $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ solid solution from 80K to T_N. This shows that the studied samples have p-type conductivity in the specified temperature range.With increasing temperature, inversion occurs in thermo-EMF for single crystals of solid solution at Neel temperature $T_N=150K$. $T_N=180.5K$ for a pressed single crystal of this solid solution along the a axis and a rubbed pressed single crystal of the solid solution. The sign inversion of thermo-EMF for pressed and rubbed pressed single crystals occurs at 255K and 284K, respectively. Thermo-EMF up to 355K after Neel temperature of all three samples of $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ solid solution is n-type.

In the **fourth chapter**, the results of the research of electrical and thermoelectric properties of low-size $TlCrS_2$, $TlCrSe_2$, $TlCoS_2$, $TlCoSe_2$ and $(TlGaS_2)_{0.95}(TlCoS_2)_{0.05}$ solid solution in the temperature range of 77-400K were presented.

X-ray analysis of $TlCrS_2$ and $TlCrSe_2$ was performed with CuK_α radiation on a DRON-3M diffractometer at room temperature.

The synthesized composition of $TlCrS_2$ and $TlCrSe_2$ crystallizes in hexagonal syngony (space group R3m). Lattice parameters: for $TlCrS_2$ a=0.3538nm, c=2.1962nm, c/a ~ 6.207, Z=3 and ρ_x=6.705 g/cm^3 and for $TlCrSe_2$ a= 0.36999nm, c=2.26901nm, c/a 6.133, Z =3 and ρ_x=6.209 g/cm^3. Electrical properties and thermo-EMF studies of $TlCrS_2$ and $TlCrSe_2$ layered compounds in the temperature range of 77÷400K showed that these compounds have p-type semiconductor character. It became clear that the activation energy for $TlCrS_2$ in the region of ferromagnetic regularity is ΔE_a=0.008eV, and ΔE_a=0.006eV for $TlCrSe_2$. In the paramagnetic temperature region, the activation energy for $TlCrS_2$ and $TlCrSe_2$ is ΔE_a=0.018eV and ΔE_a=0.018eV, respectively. These compounds are ferromagnets with Curie temperatures T_C=125K for $TlCrSe_2$ and T_C=140K for $TlCrSe_2$, and effective magnetic moments μ_{eff}=3.59 and μ_{eff}=3.71μB, respectively. The dependence of Lgσ on $T^{-1/4}$ for the $TlCrS_2$ sample in the temperature range of 80÷300K is given in Mott coordinates. A graph of temperature dependence of conductivity in $T^{-1/4}$ to Lgσ coordinates was constructed for $TlCrS_2$ compound in the range of 80÷190K. The value of T_0=6.3x10^{4}K was determined from the slope of the dependence of Lgσ on $T^{-1/4}$. From the experimentally found value of T_0, the density of localized states near the Fermi level in $TlCrS_2$ N_F=8.2x10^{19}eV^{-1}cm^{-3} was determined. In this case, the value of a_L=3.3nm was taken for the localization radius. The hopping distance in $TlCrS_2$ crystal at different temperatures was determined. So at T=80K the value is R_L=6.6nm and at T=190K R_L=5.3nm ie. The average hopping distance (R_{av}) in $TlCrS_2$ was 6.0nm. The R_{av} value is almost twice as high as the average distance between charge carrier localization centers in $TlCrS_2$. After determining the hopping distance at different temperatures (R_L) for the $TlCrS_2$ compound, we determined the energy distribution of trap states near the Fermi level: ΔE=0.027eV. It is precisely in the energy band ΔE in the band gap of the $TlCrS_2$ crystal that charge transfer occurs. At this time, the average value of the activation energy of hoppings in $TlCrS_2$ determined in the temperature range of 80÷190K was ΔW=0.026eV. The concentration

of localized states (Nt) responsible for charge transfer in $TlCrS_2$ at constant current was also calculated: $Nt=N_F x \Delta E=2.2x10^{18} cm^{-3}$.

The temperature dependence of thermo-EMF in the hopping conductance region of $TlCrS_2$ crystal was analyzed. The thermo-EMF coefficient of materials in the field of influence of hopping conductivity with variable hopping length ($\log б \sim T^{-1/4}$) can also be characterized by linear asymptotics related to Mott's formula. With increasing temperature, a linear increase in the thermo-EMF coefficient is observed in metallic materials during charge transfer in an energy gap of k_BT close to the Fermi energy (E_F). This is true only if $k_BT<<E_F$. It follows that the thermo-EMF coefficient at T=0 is also zero. In contrast to semiconductor materials, the temperature dependence of the thermo-EMF coefficient (S) in metallic materials in the region of semiconductor hopping conductivity has the form $S(T)=A+BT$ (1), where B is the thermo-EMF temperature coefficient. That is, the extrapolation of the curve S(T) to T=0 does not pass through zero. It is also possible that $S(T)\approx const$ in the hopping region. Thus, from the analysis of theoretical models of thermo-EMF and experimental data, it follows that three types of regularities are possible to describe thermo-EMF in the hopping conduction region: $S(T)\sim T^{1/2}$, S(T))~T and $S(T) \approx const$. These formulas were tested in semiconductors ($TlMX_2$, where M=Fe, Cr; X=S, Se) in the study of the thermo-EMF coefficient in the region of hopping charge carrier transfer.

In the temperature dependence of the thermo-EMF S(T) in the $TlCrS_2$ compound, at low temperatures, hopping charge transport occurs, while the thermo-EMF increases linearly with increasing temperature. The experimental results of the compound $TlCrS_2$ correspond to the formula (1). The temperature coefficient of thermo-EMF was $B=0.2\mu V/K^2$. I mean. The low-temperature thermo-EHQ coefficient in TlCrS2 is determined by the formula S(T)=(0.2T-13)μV/K.

Synthesis of $TlCoS_2$ and $TlCoSe_2$ was carried out by the interaction of primary components with high purity in quartz ampoules created under vacuum up to 10^{-3} Pa pressure. The diffractograms of $TlCoS_2$ and $TlCoSe_2$ samples are identical and the

parameters of the elementary core based on hexagonal syngony: for $TlCoS_2$ a=0.3726nm, c=2.2510nm, Z=3 и ρ=6.026g/cm^3 and for $TlCoSe_2$ a=0.3747nm, c=2.2472nm and Z =3. Due to the large ratio c/a=6.04, it can be assumed that $TlCoS_2$ is a quasi-two-dimensional ferrimagnet.

The temperature dependences of the inverse magnetic susceptibility χ-1(T) of $TlCoS_2$ and $TlCoSe_2$ compounds both have a hyperbolic shape, which is a sign of ferrimagnetism. The paramagnetic Curie temperature (Θ_P) was determined by extrapolating the χ^{-1}(T) dependence to the temperature axis. The Curie temperature was T_C ~ 120K for $TlCoS_2$ and T_C ~ 75K for $TlCoSe_2$. From the χ^{-1}(T) dependence, the experimental values of the effective magnetic moment were calculated as $\mu_{eff.}$=4.6μB for $TlCoS_2$ and μ_{eff}=4.85μB for $TlCoSe_2$. The theoretical value of the effective magnetic moment calculated taking into account the pure spin value of the trivalent cobalt ion (Co^{3+}) is 4.9μB. Good agreement is observed between the experimental and theoretical results for $TlCoSe_2$. Curie temperatures of T_C~120K for $TlCoS_2$ and T_C~75K for $TlCoSe_2$ were found from magnetic measurements, which are consistent with literature data.

In order to study the nature of the magnetic phase transition in $TlCoS_2$, the heat capacity of this compound was studied. The thermodynamic functions of the solid state (energy, entropy, volume, etc.) remain in continuum when passing through the second type of phase transition point. At the same time, the derivatives of these functions (perceptivity, heat capacity, etc.) undergo a jump at this critical point. For this reason, the maximum of the anomaly in the non-magnetic properties of the magnet, such as the heat capacity, can be used to estimate the state of the magnetic transition. Thus, the small peak in the heat capacity of cobalt-thallium sulfide at a temperature equal to 118 K is characteristic of the weak phase transition of the second type corresponding to the Curie point. This maximum is due to the sharp temperature dependence of the magnetization of the ferrimagnet sublattice, at which temperature the collapse of the magnetic order leads to the absorption of the main part of the energy. However, the value of the observed heat capacity anomaly and the

determined magnetic entropy (~8J/(mol K)) in $TlCoS_2$ are smaller than expected for ferrimagnetic ordering. Presumably, this may be due to the presence of close order in temperature ($T > T_C$). This consists in the maintenance of magnetic order at temperatures above the critical point due to the interaction of spins at close distances. In this case, the exchange forces are sufficient to create regularity at close distances, creating a cluster of ordered spins (magnetization fluctuation).

The entropy of the $TlCoS_2$ layered ferrimagnet, the small value of the ratio $\Delta S/R=0.02<\ln 2$, indicates that this phase transition belongs to the second type of displacement transitions.

The temperature dependence of the conductivity of $TlCoS_2$ and $(TlGaS_2)_{0.95}(TlCoS_2)_{0.05}$ solid solution is typical for metals, while semiconducting behavior occurs for $TlCoSe_2$. In $TlCoS_2$, $(TlGaS_2)_{0.95}(TlCoS_2)_{0.05}$ solid solution and $TlCoSe_2$ compounds, the activation energy in the temperature region with magnetic order is $\Delta E_a = 0.009$ eV, $\Delta E_a = 0.002$ eV and $\Delta E_a = 0.005$ eV, respectively. The activation energy for these ferrimagnets in the paramagnetic temperature range is $\Delta E_a = 0.116$ eV, $\Delta E_a = 0.005$ eV and $\Delta E_a = 0.014$ eV, respectively.

The following conclusions can be drawn from the above:

1. It was determined that the activation energies ΔE_a=0.008eV for $TlCrS_2$ and $TlCrSe_2$ from the temperature dependence of electrical conductivity $\sigma(T)$ were calculated ΔE_a=0.006eV in the ferromagnetic magnetic ordered region (before the transition). Also, in the paramagnetic region (after phase transition) ΔE_a>0.018eV and ΔE_a<0.018eV for $TlCrS_2$ and $TlCrSe_2$, respectively. The calculated breakdown energy of magnetic phase transitions for pressed $TlCrS_2$ and $TlCrSe_2$ is 0.01eV and 0.012eV, respectively. For these compounds, the resistivity at 100K is ρ=134.8Omcm and ρ=0.1025Omcm, and at 300K it is ρ=48.2Omcm and ρ=0.1482Omcm, respectively.

2. It has been shown that ferromagnets $TlCrS_2$ (T_C=116.8K) and $TlCrSe_2$ (T_C=138K) have p-type conductivity in all investigated temperature ranges. These compounds are ferromagnets with Curie temperature T_C=125K for $TlCrS_2$ and T_C=140K for $TlCrSe_2$ with

effective magnetic moments μ_{eff}=3.59 and μ_{eff}=3.71μB, respectively. The theoretical value of the effective magnetic moment calculated taking into account the purely spin value of the trivalent chromium ion (Cr^{+3}) magnetic moment is 3.87μB.

3. It was shown that the slope of S(1/T) dependence for $TlCrS_2$ compound in the temperature range of 338÷370K was ΔE_S=0.06eV. The activation energy values obtained from the temperature dependence of the thermo-EMF coefficient (ΔE_S=0.06eV) and electrical conductivity (ΔE_σ=0.05eV) are consistent with each other. While the temperature coefficient of the activation energy of conduction is equal to $\gamma=2.06x10^{-4}$ eV/K, the temperature coefficient of the optical gap (β) in the $TlCrS_2$ sample is equal to β=2, $\gamma=4.12x10^{-4}$eV/K.

4. We determined that the value of $T_0=6.3x10^4$K found from the temperature dependence of conductivity $lg\sigma \sim T^{-1/4}$ allows us to calculate the density of localized states near the Fermi level in $TlCrS_2$, which is equal to $N_F=8.2\ x10^{19}eV^{-1}cm^{-3}$. The hopping distance was determined at different temperatures. The hopping distance at T=80K is R_L=6.6nm, at T=190K R_L=5.3nm. Thus, the average hopping distance (R_{av}) in $TlCrS_2$ is 6.0nm. The R_{av} distance is twice the average distance between charge carrier localization centers in the $TlCrS_2$ compound.

5. It was determined that the resistance (ρ) of antiferromagnet $TlCr_3Se_5$ at 100K temperature was 23.65Om cm, and at 300K it was equal to 1.674Om cm. Thermo-EMF of $TlCr_3Se_5$ compound remained p-type in the entire temperature range. The calculated magnetic phase transition breakdown energy for pressed $TlCr_3Se_5$ is 0.007eV.

6. It is shown that the resistivity of $TlCoS_2$ and $TlCoSe_2$ at 100K temperature is ρ=0.053Omcm and ρ=0.0246Omcm, and at 300K it is ρ=0.58Omcm and ρ=0.0448Omcm, respectively. For $TlCoS_2$, at a temperature of T_C=230K (T_C=109.9K), the thermo-EMF sign changes from p-type to n-type. $TlCoSe_2$ (T_C=140K) n-type ferrimagnet in the entire temperature range. The calculated magnetic phase transition breakdown energy for pressed $TlCoS_2$ and $TlCoSe_2$ is 0.007eV and 0.009eV, respectively.

In the **fifth chapter**, the electrical and thermo-EMF properties of $TlNiS_2$ and $TlNiSe_2$ low-dimensional compounds in the temperature range of 77÷400K were discussed.

$TlNiS_2$ and $TlNiSe_2$ samples synthesized by the interaction of highly pure primary elements (Tl, Ni, S and Se) were subjected to X-ray phase analysis. X-ray phase analysis showed that these crystals have hexagonal syngony and lattice parameters: a=1.228nm, c=1.932nm, ρ=6.900g/cm^3 for $TlNiS_2$ and a=1.260nm, c=1.984nm for $TlNiSe_2$.

It has been shown that the activation energy of $TlNiS_2$ and $TlNiSe_2$ samples in the temperature range before the metal-dielectric phase transition is ΔE_a=0.012eV and ΔE_a=0.004eV, respectively. In the temperature range after the metal-dielectric phase transition, the values of ΔE_a=0.006eV and ΔE_a=0.2eV are obtained for $TlNiS_2$ and $TlNiSe_2$ samples, respectively. The specific resistance of $TlNiS_2$ and $TlNiSe_2$ samples at 100K is ρ=201Om cm and ρ=0.0032Om cm, and at 300K, ρ=0.25 Ohm cm and ρ=0.0357Om cm, respectively.

In the ρ(T) and S(T) curves of the $TlNiS_2$ samples, the metal-dielectric phase transition at the Curie temperature TC=240K occurs with a sharp hop in the electrical resistance $\rho/\rho_{room} \sim 10^3$ and thermo-EMF $S/S_{room} \sim 2x10^2$, and the transfer of charge carriers with a hop happens. The observed break in the temperature dependence of ρ(T) of $TlNiSe_2$ at temperature T_N=120K and the inversion of the thermo-EMF S(T) sign indicate a metal-dielectric phase transition in this compound, which is characteristic of 3d-transition metals.

The following conclusions can be drawn from the above:

1. It was found that the metal-dielectric phase transition in hexagonal TlNiS2 occurs at T_S=240K and temperatures lower than T_S and is accompanied by a sharp jump in electrical resistance ($\rho/\rho_{room}>10^3$). Thermo-EMF S(T) phase occurs at the metal-dielectric phase transition temperature ($S/S_{room}>10^2$) and by hopping of charge carriers. From the dependences of the $TlNiSe_2$ sample, the metal-dielectric phase transition is accompanied by a ρ(T) bend at T_N=120K and an inversion of the thermo-EMF S(T) sign.

2. It is shown that the activation energy of pressed samples of $TlNiS_2$ and $TlNiSe_2$ in the temperature range up to TMD is ΔE_a=0.012 eV and

ΔE_a=0.004 eV, respectively. And ΔE_a=0.006eV and ΔE_a=0.2eV in the temperature range after the metal-dielectric phase transition for $TlNiS_2$ and $TlNiSe_2$ samples, respectively. The specific resistance of $TlNiS_2$ and $TlNiSe_2$ samples at 100K is ρ=201Om cm and ρ=0.0032Om cm, respectively, and at 300K it is ρ=0.25Om cm and ρ=0.0357Om cm, respectively. The calculated magnetic phase transition breakdown energy for pressed $TlNiS_2$ and $TlNiSe_2$ is 0.006eV and 0.196eV, respectively.

The **sixth chapter** discusses low-dimensional $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ solid solutions. Layered crystals are prototypes of two-dimensional materials in which the ions in the layer are bound by strong covalent and/or ionic bonds, and in the direction of joining the layers, in most cases, the bonding is of Van der Waals character. Such a relatively large difference in the forces of interaction between the elements forming it in the lattice leads to significant anisotropy of the properties, mechanical, electrical, optical, elastic, etc. Below, we will consider experimental data on structural transitions that allow us to understand the reason for the transition and the description of transitions based on Mott's theory.

Samples of $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ solid solutions were obtained by melting stoichiometrically weighed portions of pre-prepared $TlInSe_2$ and $TlGaTe_2$ elementary elements in brazed quartz ampoules under a vacuum of 10^{-3}Pa. To prepare $TlInSe_2$ and $TlGaTe_2$, brands of thallium, indium, gallium Tl-000, In-000, Ga-000, tellurium TB-3 and selenium OST-16^{-4} containing no more than 5% additives were used.

$(TlInSe_2)_{0.1}(TlGaTe_2)_{0.9}$, $(TlInSe_2)_{0.2}(TlGaTe_2)_{0.8}$, $(TlInSe_2)_{0.4}(TlGaTe_2)_{0.6}$ and $(TlInSe_2)_{0.6}(TlGaTe_2)_{0.4}$ samples were prepared for electrical measurements at constant current in such a configuration that the external constant let the electric field be directed along the chains of the crystal. Electrodes to the samples were created by electrolytic deposition of copper on the side faces of the crystals. The distance between the electrodes is L=9 mm. The intensity of the electric field applied to the crystals corresponds to the ohmic region of the voltammetric characteristic. Temperature dependences of conductivity (σdc) of $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ (where x=0.1, 0.2, 0.4

and 0.6) solid solutions at DC are reconstructed in both Arrhenius coordinates and Mott coordinates. Electrical measurements at direct current cover the temperature range 172–373K.

The $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ samples for AC electrical measurements were fabricated in the form of capacitors so that charge transfer occurs along the width of the crystal chains. Silver paste was used as electrodes. The thickness of the crystal is d ~ 0.45 mm. Dielectric coefficients of crystals $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ were measured by resonance method using TESLA BM 560 coumimeter. The frequency range of the alternating electric field is 5kHz÷35MHz. All dielectric measurements were performed at a temperature of 300K. Repeating the resonance condition was ±0.2pF on capacitance, ±1.0÷1.5 scale division on Q factor (Q=1/tgδ). In this case, the biggest deviation from the average values was 3÷4% for ε and 7% for tgδ.
Measurements of the physical properties of single crystals of $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ solid solutions with a tetragonal structure made it possible to determine the dielectric properties and their frequency dispersion, to determine the nature of dielectric losses and the mechanism of charge transfer in constant and alternating current. The density of localized states and energy dissipation, the average duration and length of jumps, as well as the concentration of deep traps responsible for conduction in direct and alternating current were evaluated. It has been established that in $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ solid solutions, hopping conduction occurs in states localized near the Fermi level in constant and alternating current. The dispersion of the dielectric constant in $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ has a relaxation character. The hyperbolic drop of the tangent of the dielectric loss angle with increasing frequency from 50 kHz to 35 MHz indicates direct conduction losses in $(TlInSe_2)_{1-x}(TlGaTe_2)_x$.

The following conclusions can be drawn from the above:

1. Single crystals of $(TlInSe_2)_{0.1}(TlGaTe_2)_{0.9}$ solid solution with tetragonal structure were measured, which made it possible to determine dielectric properties and their frequency dispersion, to determine the nature of dielectric losses. DC and AC charge transfer mechanism, density of localized states and energy dissipation, average

time of crystallization, concentration of deep traps responsible for DC and AC conductivity $(TlInSe_2)_{0.1}(TlGaTe_2)_{0.9}$ are evaluated.

It was found that localized charge carriers near the Fermi level occur in direct and alternating current in $(TlInSe_2)_{0.1}(TlGaTe_2)_{0.9}$ solid solution with jump conductance. The conductivity dispersion of $(TlInSe_2)_{0.1}(TlGaTe_2)_{0.9}$ solid solution has relaxation character.

2. The hyperbolic drop in the dielectric loss tangent with increasing frequency from 50 kHz to 35 MHz shows the loss of conductivity in $(TlInSe_2)_{0.1}(TlGaTe_2)_{0.9}$.

3. Crystals of $(TlInSe_2)_{0.2}(TlGaTe_2)_{0.8}$ solid solution with tetragonal syngony were grown. In constant electric field $(TlInSe_2)_{0.2}(TlGaTe_2)_{0.8}$ crystals, hopping conductivity is observed in localized states close to the Fermi level at temperatures of 100÷175K. The density of states localized near the Fermi level ($N_F=4\times10^{20}eV^{-1}cm^{-3}$) and their energy spread ($\Delta W=0.02eV$), activation energy ($\Delta E=0.018eV$) and average hopping distance ($R_{av}=3.7nm$) are estimated. In low-temperature $(TlInSe_2)_{0.2}(TlGaTe_2)_{0.8}$ solid solution, thermo-EMF charge carriers obeyed the regularity $S(T)=(86+1.14T)\mu V/K$, which is characteristic of the hopping conduction mechanism. With increasing temperature, the thermo-EMF becomes inversely proportional to temperature, when charge carriers excited in the allowed band begin to dominate the conduction. The temperature coefficient of the activation energy of conduction is determined to be $\gamma=1.86\times10^{-4}eV/K$.

4. It was determined that during the synthesis of $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ (where x=0.1, 0.2, 0.4 and 0.6) compositions, tetragonal system solid solutions are formed. The electrical and dielectric properties of the grown chain single crystals in constant and alternating current were studied.

5. It was determined that dielectric losses in $(TlInSe_2)_{0.4}(TlGaTe_2)_{0.6}$ samples are related to conductivity. In the frequency range f=3.2÷35MHz, their alternating current conductivity obeyed the regularity $\sigma_{ac} \sim f^{0.8}$, which is characteristic of the hopping mechanism of charge transport through localized states near the Fermi level.

6. The density (N_F) and energy distribution (ΔE) of these states $N_F=5.8\times10^{18}eV^{-1}cm^{-3}$, $\Delta E=29meV$, average time (τ) and distance (R_L) of hoppings, $\tau=7.4\times10^{-8}$ c is evaluated as and $R_{av}=14.1nm$, as well as the concentration of deep traps $Nt=1.7\times10^{17}cm^{-3}$, is responsible for the conductivity of $(TlInSe_2)_{0.6}(TlGaTe_2)_{0.4}$ crystals.

7. It was determined that in the temperature range of 291÷357K $(TlInSe_2)_{0.4}(TlGaTe_2)_{0.6}$ the thermo-EMF sign is positive, and at T>357K the thermo-EMF sign is inverted, that is, the conductivity of the crystal changes from p-type to n-type.

MAIN RESULTS

1. It was determined that single crystals of $Cu_{1.22}Fe_{1.10}Te_2$(a) and $Cu_{1.15}Fe_{1.23}Te_2$(v) grown by gas transport method and $Cu_{1.13}Fe_{1.22}Te_2$ (b) grown by directed crystallization method have a tetragonal structure in chalcopyrite-type compounds. A phase with symmetry D_{2h}^2=P4/nmm is formed. The main parameters of the crystal lattice for $CuFeTe_2$ (a, b, c) were calculated by radiographic research: a=0.399nm and c=0.617nm for $Cu_{1.22}Fe_{1.10}Te_2$(a), a=0.39749nm and c=0.6078 for $Cu_{1.04}Fe_{1.12}Te_2 \approx Cu_{1.13}Fe_{1.22}Te_2$ and also a=0.402nm, c=0.604nm and c/a=1.5 for $Cu_{1.15}Fe_{1.23}Te_2$(v), vol. v=0.09745nm3, density $\rho_{cal}=6.764g/cm^3$ and $\rho_{rentgen}=6.76g/cm3$.

2. It was determined that below the semiconductor-metal transition temperature (125K) of single crystals of chalcopyrite type $CuFeTe_2$(a, b, c) there is a break in the temperature dependence of conductivity $\sigma(T)$. This is explained by the change in the activation energy of the charge transport process with the average hopping length R_{av} = 2.2 nm. At the Neel temperature (T_N = 65K) and in the spin-glass state (T_g = 55K), the temperature dependences of $C_p(T)$ and $\chi(T)$ show anomalies related to the antiferromagnetic-paramagnetic phase transition and the spin-glass state. Spin density waves (SDW) are observed above 125K (semiconductor-metal transition temperature) and dissipate near ~300K. From the temperature dependence of magnetic susceptibility (magnetic field H = 0) $\chi^{ZFC}(T)$ and (H = 1kOe) $\chi^{FC}(T)$ expansion of magnetic susceptibility is observed, and the point where this temperature dependence intersects is called the closing temperature (T_c). Above the temperature T_c (Tq > 360K), the

temperature dependence of the magnetic susceptibility $\chi(T)$ changes characteristic of superparamagnets.

3. It was determined that the spin-glass state transition temperature Tg = 55, 40 and 35K decreases with an increase in the magnetic field of 1, 10 and 20 kOe in single crystals of $CuFeTe_2$(a, b and c). Effective magnetic moments of paramagnetic clusters have values of 100 ÷ 1000 Oe $\mu_{eff} \geq 0.7\mu_B$ in weak field and 10 ÷ 50 kOe $\mu_{eff} \sim 2.4\mu_B$ (μ_B – Boron magneton) in strong field. Paramagnetic Curie temperature (Θ_P) is negative.

4. It is shown that the mutual exchange between magnetic "clusters" in $CuFeTe_2$(a, b and c) single crystals (Cu^+ - Te^{2-} - Fe^{3+} = Te^{2-}) is antiferromagnetic. In this case, the iron ($Fe3^+$) cation is located between two tellurium (Te^{2-}) anions, and the magnetic moments of the spins are opposite to each other and are arranged parallel to the (001) plane. While $CuFeTe_2$ single crystals have Z = 1 for the crystal structure, it consists of eight formula units - 8 ($CuFeTe_2$) Z = 8. The anisotropy coefficient η = 0.309, three-dimensional Debye temperature Θ_D = 176.9K and two-dimensional Debye temperature Θ_2 = 122.2K were calculated from the temperature dependence of the heat capacity. .

5. It is shown that in the $TlFeS_2$ and $TlFeSe_2$ samples, the hopping conduction with variable length R_L occurs in the region with antiferromagnetic order. The average hopping distance in the temperature range of 80-180K is R_{av} = 10.9 nm and $R_{av}/a_L > 8$ (a_L-localization radius) for $TlFeS_2$ and R_{av} = 10.4 nm and $R_{av}/a_L < 8$ for $TlFeSe_2$ in the range of 85-250K, respectively.

6. It was determined that $(TlFeS_2)_{0.975}(TlGaS_2)_{0.025}$ solid solutions have a small refraction at the Neel temperature. The average jump distance R_{av}= 6.2 A conductance jump of nm occurs. The Neel temperature changes from 150K to 180K when the samples are pressed under pressure $P=1.5x10^3 kg/cm^3$.

7. It has been shown that changing the values of x = 0.025, 0.05, 0.075 and 0.01 in $(TlFeS_2)_{1-x}(TlGaS_2)_x$ solid solutions leads to changes in the Neel temperature and activation energy. It shows the redistribution of gallium (Ga) and iron (Fe) cations in the magnetic structure in the temperature range of 77-180K in these solid solutions.

8. It was shown that the average hopping length R_{av} along the natural layers of the layered single crystal of the solid solution $(TlFeSe_2)_{0.5}(TlGaSe_2)_{0.5}$ in the temperature range of 128-178 K. = 23.3 nm and R_{av}/a_L=16, a breakthrough transmittance occurs. $(TlFeSe_2)_{0.5}(TlGaSe_2)_{0.5}$ This in the temperature dependence of the specific resistance ρ(T) of the solid solution. In the temperature range = 109 - 120 K, temperature hysteresis associated with the proportional and non-proportional phases of $TlGaSe_2$ single crystal is observed.

9. It was determined that $TlCrS_2$ samples have a jump conductivity with an average jump length R_{av} = 10.9nm and R_{av}/ a_L > 8 in the temperature range of 100-180K. It was determined that the values of ΔE_a = 0.009 eV and ΔE_a = 0.005 eV were calculated for the activation energy of $TlCoS_2$ and $TlCoSe_2$ ferrimagnets in the region of magnetic order. The activation energies in the paramagnetic region for these ferrimagnets are equal to ΔE_a = 0.116eV and ΔE_a = 0.014eV, respectively.

10. It was determined that $TlNiS_2$ compound has variable hopping conductivity with average length R_L=3.0nm in localized cases near the Fermi level in a constant electric field. It is shown that the metal-dielectric phase transition in the hexagonal $TlNiS_2$ compound occurs at temperature T_C=240K, and at a temperature below this temperature, it is accompanied by a sharp hopping of electrical conductivity $\rho/\rho_{room}>10^3$, which is accompanied by the change of conductivity from p-semiconductor to metal. Thermo emf S(T) is $S/S_{room}>10^2$ at metal-dielectric(MD) phase transition temperature. In $TlNiS_2$ crystals, a break in the temperature dependence of the MD phase transition ρ(T) and an inversion of the sign of thermo emf at T_N=120K are observed.

11. It was established that dielectric loss in $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ (x=0.4 and 0.6) solid solutions occurs due to direct conduction. In the frequency range f=3.2÷35MHz, the indicated solid solutions obey the regularity of their alternating current conductivity $\sigma_{ac}\sim f^{0.8}$, which is characteristic of the hopping mechanism of charge transfer on localized states near the Fermi level.

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