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ABSTRACT

of the dissertation for the degree of Doctor of Philosophy

THE EFFECT OF LANTHANOIDE ELEMENTS (Dy, Eu) AND IONIZING RAYS ON THE ELECTROPHYSICAL PROPERTIES OF $TlInSe_2$ CRYSTALS

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Applicant: Nurana Alishir Verdiyeva

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The dissertation work was performed at the Ganja State University of the Republic of Azerbaijan and partially at the Institute of Physics Problems of Baku State University.

Scientific supervisors: Doctor of Physicsal and Mathematics Science, Professor
Mantig Bahadur Jafarov

Official opponents: Doctor of Physical Siences, Associate Professor
Abdullayev Nadir Allahverdi

Doctor of Physicsal and Mathematics, Professor
Madatov Rahim Salim

Doctor of Physicsal Sciences, Professor
Abdullayev Adil Polad

Dissertation Council ED 1.14 of Supreme Attestation Commission under the President of the Republic of Azerbaijan operating at Institute of Physics of the Ministry of Sciences and Education

Chairman of the Dissertation council:
Academician
Arif Mammad Hashimov

Scientific secretary of the dissertation council:
Doctor of Physics Sciences, Associate Professor
Rafiga Zabil Mehdiyeva

Chairman of the scientific seminar:
Doctor of Physics and Mathematics Sciences, Associate Professor
Talat Rzagulu Mehdiyev

GENERAL DESCRİPTİON OF WORK

Relevance of the research topic. The current development of Electronics has set itself the goal of obtaining and studying new multi – component semiconductor materials. Among these materials, compounds of type $A^{III}B^{III}C_2^{VI}$ with a lanthanoid element composition can be noted. A number of compositions, which include rare earth elements, are considered promising materials in the development of various thermoelectric converters. Various properties are observed when lanthanoid additives are introduced into compounds of type $A^{III}B^{III}C_2^{VI}$. These include luminiferous, thermoelectric, thermomagnetic, etc. we can show the properties. This is explained by the fact that in the internal electronic structure of lanthanoid atoms there are electrons at the level of. In electrons at this level, transitions f→d→s easily occur. Based on such transitions, the stable electron level of $4f_0$, $4f_7$, $4f_{14}$ in lanthanoid-containing compounds further increased interest in the study of these materials. Electrons at levels 5d6s affect the change in the kinetic properties of the composition of the compound. The 4F electrons are held tightly inside the atom, protected by the electrons holding the 5s and 5p layers. Therefore, 4f electrons have a small effect on chemical bonds and behave like free ions at the first approximation.

İn recent years, both scientific and practical interest in the study of compounds in such layered and chain structures has increased significantly. The emerging interest in these materials is due to the fact that the rays of the spectrum of devices created on their basis - infrared, visible and X-ray-have enormous sensitivity in the fields.

At temperatures above room temperature, all rare earth elements, with the exception of the elements Lu and La, have the property of strong paramagnetism. The reason for this is the presence of spin and orbital magnetic moments in the 4F layers of these elements, which are not filled. At low temperatures, some of the lanthanoids become antiferromagnetic, while the yththrium subgroup lanthanoids and their various compounds (Dy, Er, Ho, Tb,

Tm) have ferromagnetic properties at very low temperatures. The Dy^{3+} ion contains unpaired electrons, and as a result of the action of these unpaired electrons, the magnetic interaction increases, as a result of which the magnetization increases even more.

Based on the data in the region of Minimum magnetic sensitivity, we can say that the greater the diamagnetism, the greater the Holl coefficient and the lower the concentration of load carriers, holes, which coincides with the data on the paramagnet. The magnetic properties of dysprozium and its compounds give impetus to the widespread use of the element in radio engineering and electronics. Its high melting point allows special alloys of these elements to be used in stainless steels and nuclear control devices. The Dy element is integrated into the magnets to increase the thermal stability and corrosion resistance of the magnets, which further increases the demand for the Dy element Thus, the use of Dy in this work is associated, first of all, with an attempt to create a thermoelectric transducer controlled by an external magnetic field. The study of Dy, Eu defect in the created thermoelement is due to its wide application in the production of thermoelectric generators, Integrated Microelectronics, as well as as a magnetic element of functional nanoelectronics. The Eu ions embedded in the crystal lattice of some compounds create the conditions for the formation of intense fluorescence, and the wavelength of the emitted light depends on the degree of oxidation of the Eu ions. Eu^{3+} emits light at wavelengths of 613 and 618 nm, which correspond to an intense red color, practically regardless of the substance in its crystal lattice. On the contrary, the Eu^{2+} ion, on the other hand, is highly dependent on the structure of the crystal lattice of the main substance, an example of which is that in the case of barium-magnesium aluminate, the wavelength of the emitted light is 447 nm and is blue in color.

The ternary compound $TlInSe_2$ belongs to the group of semiconductors with a chain crystal structure and is a structural analogue of thallium selenide, in which trivalent thallium ions (Tl) are replaced by trivalent indium ions (In^{3+}). Trivalent indium ions In^{3+} are surrounded by four selenium ions and form negatively

charged chains ($In^{3+}Se^{2-}$) along the tetragonal Z axis. These chains are connected by monovalent thallium ions $T1^{+}$. The forces between atoms within the chains are strong, covalent, and between the chains weaker forces of the ionic type are realized. This is why our sample breaks down easily. These crystals have a layered structure and p-type conductivity. Optical properties, absorption edge structure, steady-state photoconductivity, capture processes, interest in this material is caused not only by its fundamental properties, but also by possible practical applications. Typically, thallium chalcogenide semiconductor compounds with the general formula TlBxZ (B = In, Ga; Z = Se, Te, S) have an interesting phase transition at low temperatures.

In these materials, the Seebeck effect allows the direct conversion of thermal energy into electrical energy. Thus, thermoelectricity has attracted a lot of attention due to its potential to harvest huge amounts of waste heat. However, before widespread industrial application, it is desirable to significantly improve the energy conversion efficiency. The efficiency of TE materials is determined by the dimensionless figure of merit $ZT= (S^2\sigma/\kappa)T$, where T is the temperature, and S, σ and κ are the Seebeck coefficient, electrical conductivity and total thermal conductivity provided by electrons and phonons, respectively. Because these transport properties are closely interrelated, improving the efficiency of technologically useful devices is challenging. Among the various optimization strategies that have been developed, size limitation has been identified as an effective mechanism for reducing lattice (phonon) thermal conductivity by increasing the phonon scattering rate. Confinement effects in an electronic system, such as in superlattices and quantum wells, lead to an increase in the Seebeck coefficient by increasing the electron density of states, thereby increasing the power factor $PF = S^2\sigma$.

Despite the controversy over the widespread use of this compound due to the toxicity of Tl, $TlInSe_2$ can be considered as a model system with a quasi-one-dimensional phonon structure. Due

to the small number of atoms in the crystallographic unit cell, this compound is amenable to theoretical modeling using density functional theory with moderate computational effort. The crystalline substructure of $TlInSe_2$ consists of chains of trivalent In atoms having covalent bonds with Se atoms located along the crystallographic axis. The weaker ionic bonding of the monovalent octahedrally coordinated Tl atoms ensures that these chains are weakly coupled to each other. These structural properties place $TlInSe_2$ in a class of nanoscale materials with quasi-one-dimensional phononic properties, which are expected to have beneficial consequences for lattice dynamics by limiting phonon propagation and thereby reducing thermal conductivity. Thus, $TlInSe_2$ is considered to have the potential for a very high quality factor $ZT > 2$ at $T < 500$ K, exceeding the ZT of PbTe or PbSe with ZT values of ~1.8. Even this small review emphasizes the very high relevance of the research proposed in this work.

Obtaining new crystals based on triple compounds $TlInSe_2$, $TlInS_2$ and their physical properties are of practical importance. The newly formed compounds obtained also crystallize in tetragonal syngony. Such crystals are used as detectors of near-infrared radiation, X-ray, gamma, neutron radiation, solar energy converters, thermoelements, converters with memory, etc. it is considered promising materials in the development of devices such as

The lack of practical significant work devoted to the influence of ionizing Rays on the electrophysical properties of solid solution crystals $TlIn_{1-x}Dy_xSe_2$ and $TlIn_{1-x}Eu_xSe_2$, the study of the effect of these rays on the change in the electrophysical, thermoelectric and thermal properties of the crystal is of wide practical interest.

Object and subject of research. The object of the study is monocrystals $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0,01; 0,03; 0,05), alloyed with lanthanoid atoms (Dy, Eu).

The subject of the study was the improvement of electrophysical properties of semiconductor materials, determination of Physico –Chemical mechanisms realized in

stabilization of property indicators and their control, in terms of electron –ion level and structural changes.

Research aims and objectives: The purpose of the dissertation is $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) by the action of lanthanoid (Dy, Eu) atoms and ionizing gamma (γ) rays.%) determining the mechanisms of influence of defects of various nature formed in monocrystals on the patterns of formation of the crystalline structure, thermal conductivity and electronic phenomena and determining the possibilities of their application.

Research methods. X-ray diffraction, constant current, differential scanning calorimetry (SSC) stationary photoconductivity, stationary thermal conductivity, Holl radiation source Co_{60} isotope, ferrosulfide methods were applied while studying the effect of ionizing radiation (γ) on electrophysical properties of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) monocrystals doped with lanthanoid atoms. The $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) monocrystals were irradiated in the radiation research facility -60, consisting of Co_{25} isotope. During the experiments, layered monocrystalline samples with a width of 4-5 mm, a length of 10-15 mm and a thickness of 1-2 mm were used for electrical measurements $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05). Samples of cylindrical shape were used in thermoelectric measurements and determination of thermal conductivity coefficient. Before the action of ionizing radiation, silver contacts were applied under special conditions to the surface of the crystals-research objects, electrophysical studies were carried out, checking the conductivity and quality of the contact.

The main scientific propositions defended:

1) observing anomalies in temperature dependences of conductivity (T), thermoelectric driving force in $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) single crystals;

2) determination of the effect of rare earth element atoms (Dy, Eu) on the electrical conductivity of $TlInSe_2$ crystals, thermoelectric driving force;

3) Determination of the mechanism of heat conduction in $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) single crystals:

4) determining the effect of radiation on the mechanism of electrical and thermal conductivity of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05)

5) determination of scattering mechanism in $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) solid solution crystals in different temperature range;

Scientific novelty of the research.

1. The electrical and thermoelectric properties of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) solid solution crystals with a layered and chain structure strongly depend on the concentration of defects that cause local changes in the arrangement of atoms.

2. There are shallow and deep local energy levels in the energy band of $TlInSe_2$ solid solution crystal. Their charge-discharge rate depends on the applied electric field.

3. Conductivity at temperatures below 200 K is related to transitions from additional levels in the forbidden zone, acceptor levels located at an energetic distance ΔE from the ceiling of the valence band act as a "trap" for specific charge carriers, and the "capture" of these levels in the temperature range where the Fermi levels enter this zone is happening.

4. Crystallization of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0,01; 0,03; 0,05) quadruple compound solid solutions in tetragonal syngonia was found.

5. Irradiation with rare earth element atoms and γ-quanta added to the $TlInSe_2$ solid solution crystal has a sharp effect on both the numerical value of the substance's thermal conductivity and its temperature dependence. phonons are involved.

Theoretical and practical significance of research. The results obtained in the dissertation can be widely used in the development of electronic converters, nanoelectronics, memory elements, systems for collecting electrical and optical information. It was also shown in the dissertation that $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0,01; 0,03; 0,05) solid solution crystals can be used as an effective material in tenso-, thermo-, photoconverters that can work in a wide temperature range (300-600 K).

Approval and application. Reports on the general provisions of the dissertation were made from time to time at the physics department of GSU. The content of the dissertation was published in 22 scientific works, including 11 articles and 11 conference materials in national and foreign journals. Seven of the articles were published abroad, in journals included in international bases, and two in journals recommended by the AAK. Four of the articles are single-authored. The main results of the dissertation work were published in the following conferences:

Baku State University, Institute of Physics Problems. Ministry of Agriculture of the Russian Federation. Sankt –Petersburg Gosudarstvenny Agrarny Universitet, Nauchnoe objekcije razvitiya АПК в условия импортозамещения. Сборник научных трудов. (-Санкт – Петербург, part 1,-2017, -ст.535-537). Baku Engineering University, I International Scientific Conference of Young Researchers, (-Baku, May 05-06, -2017, -p. 123-125, 125-127). Ganja State University, International Scientific Conference, Current problems of modern natural and economic sciences. (-Ganja, part V, -04-05 May, -2018, -p.38-41, 265-268). ASU. Prospects of development of the food and textile industry in Azerbaijan and future tasks. II Republican Scientific-Practical Conference. (–Baku, -April 25, 2018, -p. 136-142). Sumgayit State University, Current Issues of Applied Physics and Energy, Materials of the International Scientific Conference. (–Sumgait, -24-25 May, -2018, -ст.228-231). Ganja State University, Ganja International Scientific Conference, Current problems of modern natural and economic sciences. (-Ganja, part IV, -03-04 May, -2019, -p. 7-10). Azerbaijan University of Technology, Current problems of food and light industry, International scientific practical conference. – (Ganja, -2019. Pg. 211-212). III International scientific-practical conference European Research Forum, Russian Federation МЦНП, Новая наука, -Petrozavodsk, (-April 19, -2021, article 73-79).

The name of the organization where the dissertation work was performed: Ganja State University, partly Institute of Physics Problems of BSU

The structure and scope of the dissertation. The submitted dissertation consists of 20244 marks in the introduction, 57627 marks in chapter I, 32478 marks in chapter II, 69862 marks in chapter III, 26450 marks in chapter IV and 3267 marks in the results, in total 209928 marks. The dissertation consists of 163 pages, including 126 pages of text (209928 characters), 16 images, 26 graphs and 3 tables.

To achieve the set goal, the following issues were resolved:

$TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) solid solutions were synthesized and their single crystals were grown by Bridgeman-Stockbarger method.

- $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) single crystals were exposed to radiation with different doses:

- To determine the mechanism of temperature dependence of electrical conductivity, Hall effect, thermo-electric motive force and thermal conductivity of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) solid solution crystals:

- The mechanism of phonon scattering in $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) single crystals and the mechanisms of formation of defects of various natures were determined;

- The mechanism of the effect of defects of various nature on the nature of heat and electrical conductivity in $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) monocrystals caused by γ-radiation was determined:

CONTENTS OF THE WORK

In the introduction, the relevance of the dissertation and its application in technology are justified, the purpose of the conducted research, the scientific innovation and practical importance of the work are indicated, the main propositions defended, the degree of approval, published scientific works are given, and the main content of the dissertation is briefly explained according to the chapters.

The first chapter of the dissertation is dedicated to the investigation of the electrophysical properties of the $TlInSe_2$ solid solution crystal, which is included in the class of type compounds, and literature data. This chapter provides general information about the electrophysical properties of type compounds, lanthanoid-based solid solution crystals, and their electronic structure. In this chapter, literature data on the mechanism of formation of defects of different nature in type $A^{III}B^{VI}$ crystals were also analyzed.

The methods of growing and synthesis of single crystals of $TlInSe_2$, $TlInTe_2$ and $TlGaTe_2$ triple compounds have been investigated in the literature, and the zone melting method was considered the most effective as an effective growing method.

The temperature dependence of the specific resistance of the $TlInTe_2$ layered semiconductor both perpendicular to the layers and in the direction of the layers was studied, and the activation energy of the additional conductivity was determined[1]. The localization radius and state densities near the Fermi level were determined. This chapter provides information on the existence of various types of specific (point and macroscopic) defects in A^3B^6 type semiconductor compounds and the mechanism of formation of various radiative defects and additive levels in these materials. It is always relevant to study the methods of formation of defects, both specific and created by certain methods, existing in such compounds.

As a result of the effect of radiation rays, a certain irregularity is observed in the crystal structure of the semiconductor. This irregularity depends on the nature of the collision of particles with atoms and causes the excitation of atoms or the change of the coordinate center. It is also clear from the research conducted on $TlInSe_2$ crystals doped with different percentages of rare earth element atoms that acceptor and donor type radiation defects are formed in the crystal during γ-irradiation, but the degree of compensation increases in p-type $TlInSe_2$

[1] Керимова, Э. М., Мустафаева С. Н., Исмаилова П. Г., Керимов Р. Н., Ахмедова Г. М. Электрические и термоэлектрические свойства $TlCoS_2$ $TlCoSe_2$ // Матер.респ.научн.конф. «Неорганическое материалов-едение», -Баку: БДУ. -2002, -ст.39.

crystals as donor levels are predominant[2]. Data on the effect of radiation rays on the physical properties of semiconductor elements were analyzed[3][4]

The second chapter of the dissertation work is the methodological part. In this chapter, the physicochemical properties of type solid solution crystals are investigated. Here, information is provided on the acquisition of crystals of different compositions with Eu and Dy rare earth element atoms, their gamma quantum irradiation, the study of electrophysical, thermoelectric and thermal properties of the obtained crystals, and measurement methods. In this chapter, the determination of the main parameters of semiconductors, their calculation methods, basic electrical circuits, research devices, and the method of irradiation of samples with γ-rays were investigated in detail.

$TlInSe_2$ type compounds included in the class of type compounds and the analysis of the physico-chemical properties of the solid solutions obtained based on them, the growing technology of the studied crystals was analyzed and the possibility of obtaining solid solution crystals of different composition by replacing the indicated compounds with anions and cations was investigated[5].

In literature data, the nature of the chemical effect in the $TlGaSe_2$ – $TlInSe_2$ system was analyzed and the possibility of anion and cation substitutions in the crystal lattice was considered. Here, the method of synthesis and production of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0,

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[2] Мадатов Р. С. Влияние ионизирующего излучения на механизм электропроводности монокристаллов Особенности механизма токопрохождения в кристаллах $TlInSe_2$ /Р.С.Мадатов, А.И. Наджафов., М. Р. Газанфаров // Перспективные материалы, -2012, №3, -ст.1-5.

[3] Зарбалиев М. М. Особенности электрических свойств твердых растворов $TlIn_{1-x}Yb_xS_2(Te_2)$ // Физика. -1999. Т.5. №3. -с.26-29.

[4] Мадатов, Р.С., Эффект переключения в монокристаллах TlInS2 облученных гамма квантами / Р. С. Мадатов, А. И. Наджафов, М. А. Мамедов, В. С. Мамедов // AMEA-nın Xəbərləri. Fizika-riyaziyyat və texnika elmləri seriyası,- 2008, № 2, -с. 64-68.

[5] Зарбалиев, М. М. Теплопроводность твердых растворов системы $TlIn_{1-x}Yb_xS_2$ // РАН, Неорганич. Материалы. -2000. Т.36.№ 5. -с.619-623

0,01; 0,03; 0,05) solid solution single crystals by Bridgman-Stockbarger method is shown in detail and the device is described. At the same time, the method of irradiating samples with gamma rays and the way of creating radiation defects were analyzed. The device used to measure the electrical conductivity and thermoelectric driving force of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) crystals is reported. A description of the principle structure of the cryostat used to measure the electrical and thermoelectric properties of the samples from nitrogen temperature to 600 K is given.

Sample preparation and measurement method of electrophysical quantities were analyzed. The single crystallinity, structure and chemical composition of the obtained crystals were checked by X-ray phase analysis. It was determined that the single crystals have a tetragonal structure and the parameters of the crystal lattice were - a=8.062 A^0, c=6.820 A^0. The conductivity type of the crystals was determined according to the sign of thermo-e.h.g. and it was determined that they have p-type conductivity. Based on the Hall effect and electrical conductivity, the concentration and specific resistance of free charge carriers were determined. These parameters were ~$5\cdot10^{11}$ cm^{-3}, ~10^5-10^7 Om·cm, respectively.

Since the surface of the crystals is smooth-mirror, there was no need for additional chemical and polishing.

The third chapter of the dissertation work is the empirical part. In this chapter, the results obtained from the study of electrical and thermoelectric properties of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) system compounds, their investigation and explanation are given.

It is known from the literature that with an increase in the relative amount of lanthanoid atoms in the composition, the parameters of the elementary lattice increase linearly up to a certain limit. This is due to the fact that the ionic radius of lanthanoid atoms (Dy, Eu) is larger than the ionic radius of indium.

However, the tetragonal syngonia corresponding to the initial component $TlInSe_2$ compound is preserved in the obtained composition, and the number of atoms in the elementary lattice does not change. The conducted studies once again confirm $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) systems have a $TlIn_{1-x}M_xSe_2$ (M=Dy,

Eu) (x=0, 0,01; 0,03; 0,05) solid solution region at room temperature in the range of 0 ÷ 11 mol percent $TlInSe_2$.

Figures 1 and 2 show the temperature dependences of electrical conductivity of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) solid solutions, and Figure 3 shows the temperature dependences of the Hall coefficient for the $TlIn_{1-x}Dy_xSe_2$ crystal.

At low temperatures (80-450 K), additive conductivity is observed, and at temperatures above ~500 K, specific conductivity is observed. The width of the forbidden zone (activation energy) was determined for the appropriate composition according to the trends of the temperature dependence of the conductivity. The values of the forbidden zone width determined by the trends of the high-temperature parts of both the electrical conductivity and the Hall coefficient agree with each other within the experimental error. At low temperatures (in the temperature range of 80-450 K), the conductivity increases with small trends. Such a dependence is related to the additional conductivity created in the sample and has a quasi-metallic character. Starting from a certain temperature, a relative decrease in electrical conductivity is observed, and this decrease is felt more sharply with the increase of lanthanoid atoms in the composition. In the subsequent increase in temperature, a region of specific conductivity with an exponential increase in electrical conductivity is observed. At sufficiently high temperature values, the thermal energies of charge carriers provide a better opportunity for them to cross the forbidden zone. It was determined from the temperature dependence of the Hall coefficient that this observed dependence is in good agreement with the literature data on the temperature dependence of electrical conductivity. In the temperature range of 300-500 K, the concentration of free charge carriers remains practically unchanged, and due to the tendency of its dependence in the high temperature region, the calculated value of the width of the forbidden zones is consistent with the experimental error plot.

Based on the temperature dependence of the Hall effect, it can be said that charge carriers are mainly scattered from acoustic phonons. In the studied temperature range, the temperature dependence of conductivity changes according to the $T^{3/2}$ law, which corresponds to

the scattering of charge carriers from long-wave acoustic phonons. Transitions occurring in the crystal in the temperature range of 100-450 K arise from additional levels located in the forbidden zone. After a certain temperature, the transitions from this level "exhaust". Greater energy, i.e. higher temperature, is required for re-transitions to occur.

From the temperature dependence of the Hall coefficient, it is clear that starting from about ~ 450 K, the Hall coefficient (R-in) increases with increasing temperature. In this case, when indium atoms are replaced by lanthanide atoms, the increase in the Hall coefficient shifts to a higher temperature region. In all cases, the increase of the Hall coefficient continues up to a certain temperature, after which a sharp decrease is observed. In other words, the Hall coefficient passes through the maximum, and as the relative amount of lanthanoid atoms in the composition increases, the maximum of the Hall coefficient (R) shifts towards the high temperature region. An increase in the Hall coefficient occurs due to a decrease in concentration.

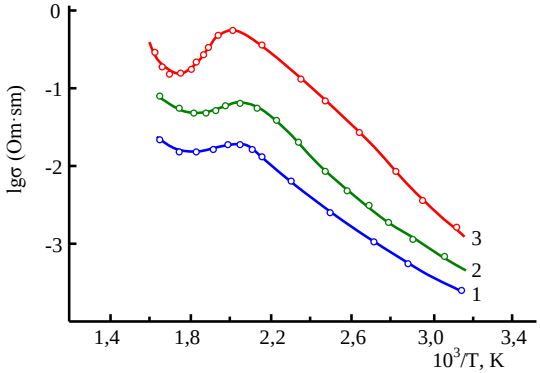

Figure 1. Temperature dependences in the coordinates of the electrical conductivity of the $TlIn_{1-x}Dy_xSe_2$ (x: 0.01; 0.03; 0.05) solid solution crystal

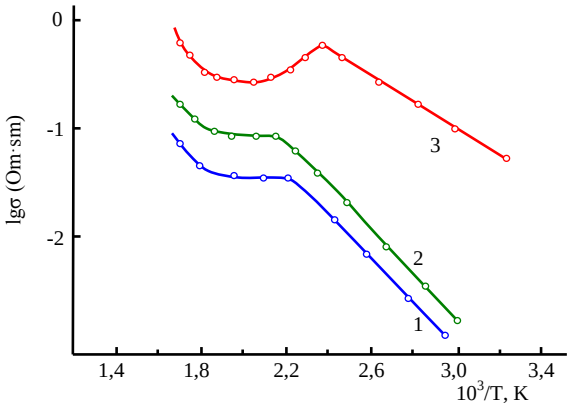

Figure 2. Temperature dependences in coordinates of electrical conductivity of $TlIn_{1-x}Dy_xSe_2$ solid solution crystal (x: 0.01; 0.03; 0.05)

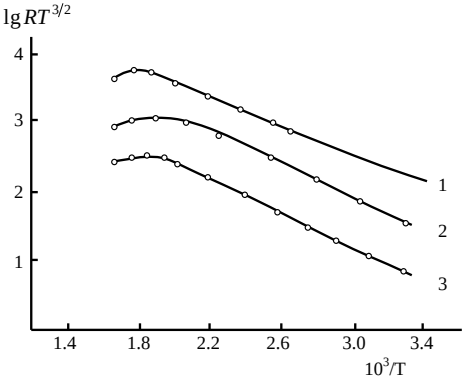

Figure 3. Temperature dependences of the Hall coefficient in coordinates for the $TlIn_{1-x}Dy_xSe_2$ crystal (x: 0.01; 0.03; 0.05).

In the subsequent increase in temperature, additional levels are gradually filled, and when the average kinetic energy of charge carriers is greater than the width of the forbidden zone ($kT>\Delta E$), the transition of electrons to the conduction zone occurs from those levels. This effect, in turn, causes a decrease in the Hall coefficient and, accordingly, an increase in the concentration of charge carriers and conductivity.

The study of thermoelectric motive force (thermo-e.h.g.) is of great importance in the study of the physical properties of a solid body and its temperature dependence. Studying the temperature dependence of thermo-e.h.g. is one of the important issues in order to make an opinion about the temperature dependence of the concentration of charge carriers. Thermo-e.h.g. should decrease with the increase of the temperature when the concentration of charge carriers increases by the exponential law. During determination of thermo-e.h.q. of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0,01; 0,03; 0,05)single crystals, it was determined that the characteristics of their dependence are practically the same for both crystals. The dependence obtained for both crystals is linear. These results prove that the concentration increases with the increase in temperature, and that the investigated substances are semiconducting in nature.

The main efforts in thermoelectric materials science are aimed at finding the material with the highest value of the dimensionless parameter ZT. It was determined from the temperature dependence of the thermal conductivity of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,01; 0,03; 0,05) single crystals that the rare earth elements added to the $TlInSe_2$ crystal affect the overall thermal conductivity of the crystal. It was found that the value of thermal conductivity decreases during the effect of Dy and Eu atoms on the $TlInSe_2$ crystal. This leads to an increase in the ZT coefficient, which means that the thermoelectric property of the single crystal is strengthened.

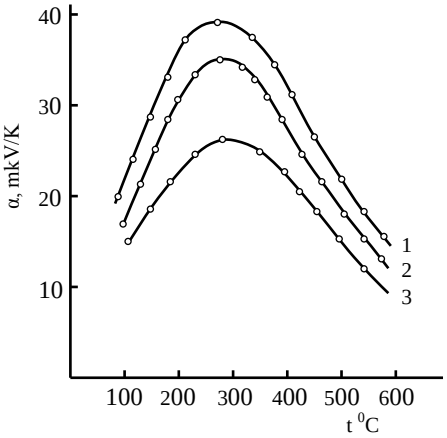

Figure 4. Temperature dependence of the thermoelectric driving force of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0, 0,05) solid solution crystal. 1-$TlInSe_2$, 2- $TlIn_{0.95}Dy_{0.05}Se_2$, 3- $TlIn_{0.95}Eu_{0.05}Se_2$.

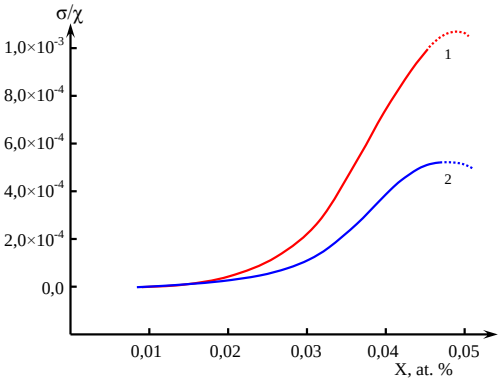

Figure 5. Determination of ZT coefficient of $TlIn_{1-x}Eu_xSe_2$ (1) and $TlIn_{1-x}Dy_xSe_2$ (2) (x=0.01; 0.03; 0.05 at. %) crystals

The property of magnetism depends on the magnetic order and temperature conditions as an elemental property in matter. Dysprosium lanthanoid is paramagnetic. It has high magnetic strength. It exhibits ferromagnetism at temperatures below 85K, and helical antiferromagnetism and paramagnetism at temperatures above 300K.

Determining the mechanism of carrier scattering in semiconductors is related to certain theoretical difficulties. But taking into account certain conditions, the issue can be simplified. In the case that the electron gas does not crack, the conductivity at partially high temperatures is strongly dependent on the temperature. The temperature dependence of the observed law of carrier mobility corresponds to the scattering of charge carriers from acoustic phonons.

Theoretically calculated values of some basic semiconductor parameters of $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x=0,01; 0,03; 0,05) solid solution crystals with different atomic percentages of lanthanoid element atoms are given in the following table.

Table

Some semiconductor parameters of solid solutions containing $TlIn_{1-x}M_xSe_2$ (M=Dy, Eu) (x= 0,01; 0,03; 0,05)

Ingredients	$\Delta\varepsilon_g, eV$ $\lg\sigma = f(10^3/T)$	$\Delta\varepsilon_g, eV$ $\lg RT^{3/2} = f(10^3/T)$	α V/K 500 K
$TlIn_{0,99}Dy_{0,01}Se_2$	1,20	1,19	730
$TlIn_{0,97}Dy_{0,03}Se_2$	1,12	1,09	657
$TlIn_{0,95}Dy_{0,05}Se_2$	1,10	1,06	600
$TlIn_{0,99}Eu_{0,01}Se_2$	1,17	1,17	672
$TlIn_{0,97}Eu_{0,03}Se_2$	1,15	1,14	585
$TlIn_{0,95}Eu_{0,05}Se_2$	1,10	1,02	500

Figure 6 shows the experimental volt-ampere characteristics of a thin layer of $TlIn_{0.97}Dy_{0.03}Se_2$ crystal at different temperatures. The volt-ampere characteristic is linear when the contact to the sample is ohmic. But when the external field applied to the sample

increases, the contact becomes non-ohmic. This fact arising from the external electric field is associated with the following effects[6][7]:

a) Due to the effect of the generated strong electric field, the current density increases sharply due to the increase in the concentration of charge carriers flowing between the electrodes. b) Another reason for the non-linearity in the volt-ampere characteristic is the presence of seizure centers in the forbidden zone. Seizure centers can be element atoms of various nature injected into the crystal - inclusions, defects, dislocations and other types of lattice defects.

As can be seen from the graphical dependence, there are linear and non-linear regions in the volt-ampere characteristic (VAX) in a wide temperature range for the studied sample at electric field values of 10÷103 V/cm. In the volt-ampere characteristic of a thin layer of $TlIn_{0.97}Dy_{0.03}Se_2$ solid solution single crystal, there are ohmic ($I\sim U$), quadratic ($I\sim U^2$) and intervals with a sharp increase in current ($I\sim U^3$). Part I observed in the volt-ampere characteristic corresponds to the ohmic part ($I\sim U$), part II is quadratic ($I\sim U^2$) and part III corresponds to a sharp increase in current ($I\sim U^3$). In the characteristics extracted at different temperatures, as the temperature increases from nitrogen temperature to 600 K, the transition threshold voltage from linear dependence to quadratic dependence in the characteristic decreases. The reason for this change is the change in the probability of trapping charge carriers injected into the crystal. The observed dependence is related to the decrease in conductivity of the samples. The variation of the current in the volt-ampere characteristic of $TlIn_{0.97}Dy_{0.03}Se_2$ -single crystal in the temperature range of 80 ÷ 600 K depending on the external field is explained by the Lampert theory following the Child-Langmuir law. The reason for the

[6] Нифтиев Н. Н. Электрические свойства слоистых монокристаллов $FeGaInS_4$ // ФТП, 2004, т.38, в.5, с.522-523.

[7] Керимова Э. М., Мустафаева С. Н., Керимов С. Н., Гаджиева Г. А. Фото и рентгенопроводимость твердых растворов $(TlGaS_2)_{1-x}$ $(TlInSe_2)_x$ // Неорган. материалы, 1999, т.35. №11. с.1313-1314.

occurrence of this effect in the studied crystal is due to the formation of new trapping centers in the forbidden zone with the increase in temperature and the change in the probability of trapping the injected charge carriers. Additional levels, dislocations and defects of various nature in the crystal lattice belong to this type of capture centers. The observed fact is that in the temperature range of 80 ÷ 600 K, the volt-ampere characteristic of $TlIn_{0.97}Dy_{0.03}Se_2$ - single crystal. According to Lampert's theory, the current in the low temperature region is determined by charge carriers generated due to thermal ionization, and at high temperatures, by injected charge carriers. The transition voltage from the ohmic part to the quadratic part changes with the temperature change in the range U_{kech}=8-15 V. These voltage values can be calculated using Lampert's theory from the volt-ampere characteristics. Using Lampert's theory, the concentration of electrons (n_0), mobility of charge carriers (μ) and concentration of traps (Nt) in equilibrium at nitrogen temperature were calculated according to Lampert's theory. Accordingly, the values of $n_0 = 5 \cdot 10^{10}$ cm^{-3}, μ=1.9 $V \cdot m \cdot sec^{-1}$, $N_t = 1.5 \cdot 10^{17}$ cm^{-3} were obtained for these quantities.

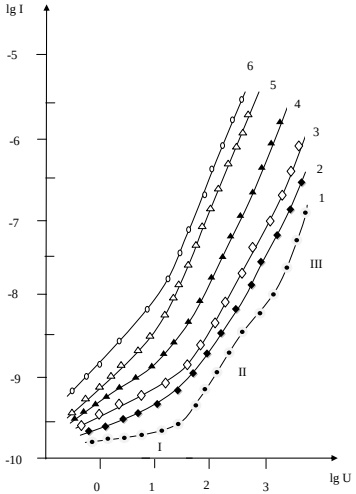

Figure 6. Volt-ampere characteristics of $TlIn_{0.97}Dy_{0.03}Se_2$ crystal at different temperatures. 1-100 K; 2-200 K; 3-300 K; 4-400 K; 5-500 K; 6-600 K

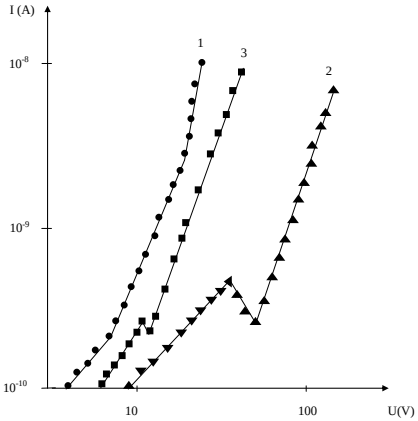

Figure 7. Volt-ampere characteristics of the Ag-$TlIn_{0.97}Dy_{0.03}Se_2$ –Ag structure (1-unirradiated, 2, 3 after 24 and 240 hours of γ-quantum irradiation, respectively).

Figure 7 shows VAX dependences at 300 K temperature for unirradiated $TlIn_{0.97}Dy_{0.03}Se_2$ and Ag –TlInSe2 – Ag structure irradiated with 50 krad γ-quanta. In the figure, curve 1 is not irradiated, and curves 2 and 3 are the volt-ampere characteristics of the irradiated $TlIn_{0.97}Dy_{0.03}Se_2$ sample after 24 hours and 240 hours, respectively. It was found that after a certain period of time after irradiation, the dependences of VAX shift to the left and after 240 hours it approaches the characteristic of the non-irradiated sample (curve 3 in the figure). It can be seen from the graphical dependences that as the intensity of the external electric field increases, the probability of capturing electrons also increases, and at a certain value of the field, not a quadratic dependence, but an N-shaped dependence is observed. Such a dependence of the current intensity on the voltage indicates that the current gradually decreases during the formation of domains in the sample.

As a result of the conducted studies, it was determined that the charge transport in the fields $E<10^2$ V/cm in the square region of VAX of irradiated $TlIn_{0.97}Dy_{0.03}Se_2$ monocrystals is related to monopolar injection, as in the case of primary crystals, and the rate of filling and emptying of traps increases with the increase of the

radiation dose. The heat-field ionization observed in the region of sharp increase of the current is due to the effect of γ-quanta and is observed at high values of the electric field ($E>10^3$ V/cm). This is due to the increase in the concentration of donor-type centers as a result of radiation.

Figure 8 and Figure 9 show volt-ampere characteristics for solid solution crystals. It can be seen from the figure that all the studied structures have a conversion property, and as the relative amount of dysprosium and europium element atoms in the composition increases, the value of the threshold voltage decreases.

The observed conversion phenomena can be explained on the basis of electrothermal and thermal mechanisms. In the electronic mechanism of conversion, it is necessary to take into account double injection, tunneling of electrons through the potential barrier, impact ionization of valence electrons and additional atoms, and the Pool-Frenkel effect. In the electro-thermal mechanism of the conversion, the transition from a state of low conductivity to a state of high conductivity depends on the applied electric field and temperature. In the field of conversion, the electronic mechanism of conversion plays a key role. The main drawback of the switching elements is the instability of the threshold voltage. Various thermal treatment methods are used to increase stability.

In the fourth chapter of the dissertation, the mechanism of heat energy transfer in type crystals was investigated, types of heat conduction in semiconductors, research methods of heat conduction, the method of determining the thermal conductivity of $TlInSe_2$ crystal, the effect of lanthanoid atoms (Dy, Eu) of different atomic percentages and radiation on the heat conduction of $TlInSe_2$ solid solution crystal was investigated. During the study, the heat transfer coefficient was measured by the stationary method in the temperature range of 80-600 K, and the mechanism of heat transfer in this type of crystals was investigated. It was determined that the heat conduction in the investigated crystals occurs mainly with lattice phonons.

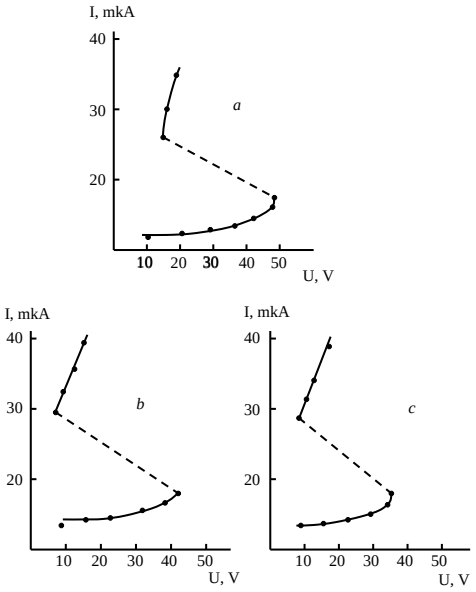

Figure 8. Volt-ampere characteristics of a solid solution crystal injected with dysprosium element atoms: *a)* $TlIn_{0.99}Dy_{0,01}Se_2$**, *b)*** $TlIn_{0,97}Dy_{0,03}Se_2$ **, *c)*** $TlIn_{0,95}Dy_{0,05}Se_2$**.**

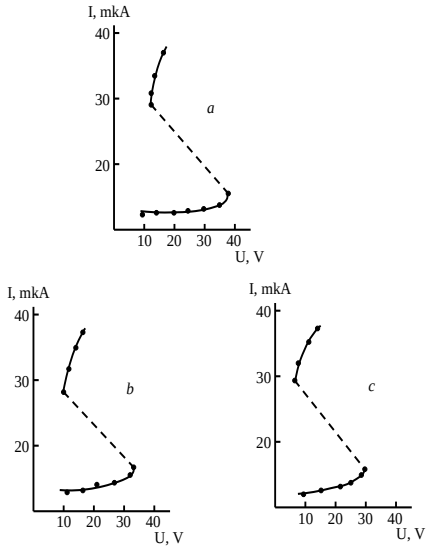

Figure 9. Volt-ampere characteristics of a solid solution crystal doped with atoms of the European element.

a) $TlIn_{0.99}Eu_{0,01}Se_2$**,**

b) $TlIn_{0,97}Eu_{0,03}Se_2$ **, *c)*** $TlIn_{0,95}Eu_{0,05}Se_2$**.**

The mechanism of formation of defects of different nature in $TlInSe_2$ solid solution crystals with different atomic percentages of Dy and Eu lanthanoid atoms, scattering mechanism of phonons from lattice nodes and defect centers of different nature was investigated. It was determined from the conducted research that a certain percentage of Dy and Eu rare earth element atoms added to the $TlInSe_2$ solid solution crystal affects both the numerical value of the thermal conductivity of the substance and the nature of the temperature dependence of the thermal conductivity coefficient. It was determined that Dy element atoms have a greater influence on both the numerical value of the heat transfer coefficient and the nature of the temperature dependence than Eu element atoms. It was found that the electronic heat conduction in both studied solid solution crystals is negligible and phonons are involved in the heat conduction.

Figure 10 shows the temperature dependence of the thermal conductivity coefficient in the temperature range of 80-600 K in the $TlInSe_2$ solid solution crystal (1 – parallel to the layers; 2 – perpendicular to the layers). Such a temperature dependence of the heat transfer coefficient confirms the temperature-dependent three-phonon scattering process in the studied crystal in the transport of thermal energy. It was determined that electronic thermal conductivity in the studied $TlInSe_2$ solid solution crystal is negligible and phonons are involved in thermal conductivity. In the mathematical calculations, values were obtained in the direction parallel to the layers at room temperature. The obtained values correspond to the information shown in the literature[8] Even if lanthanoid element atoms are included, since the electronic component of thermal conductivity is negligible (in the formulation), the temperature dependence of the thermal conductivity coefficient and also its numerical value once again confirm the transport of thermal energy by lattice phonons in the investigated crystals.

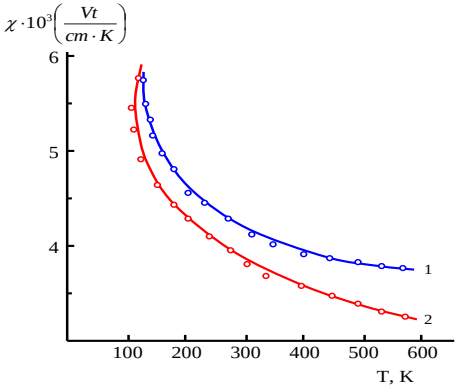

Figure 10. Temperature dependence of the thermal conductivity coefficient in the $TlInSe_2$ solid solution crystal (1 – parallel to the layers; 2 – perpendicular to the layers).

[8] Козловский С. Ю. О методах измерения электропроводности. ПТЭ. 1989. № 3, с. 110-113.

Figure 11 shows the temperature dependence of the thermal conductivity of the $TlIn_{1-x}Dy_xSe_2$ solid solution crystal. (x: 1-0; 2-0.01; 3-0.03; 4-0.05), temperature dependence of the thermal conductivity coefficient of $TlIn_{1-x}Eu_xSe_2$ solid solution crystal in Fig. 12 (x: 1-0; 2-0 .01; 3-0.03; 4-0.05) are given

For both crystals, with the increase in temperature, a decrease in the thermal conductivity coefficient is observed in a certain temperature interval. This decrease occurs according to the law in the temperature range of 80 – 300 K, then the decrease is partially weakened. The observed temperature dependence of the thermal conductivity corresponds to the three-phonon scattering mechanism in the crystal.

It should be noted that the temperature dependence of the thermal conductivity coefficient of the $TlInSe_2$ solid solution crystal containing different percentages of rare earth elements (Dy, Eu) corresponds to the characteristic dependence for crystals crystallized in the TlSe structural type. When replacing In ions with Dy and Eu ions in the composition, the value of the total heat transfer coefficient in the crystal decreases.

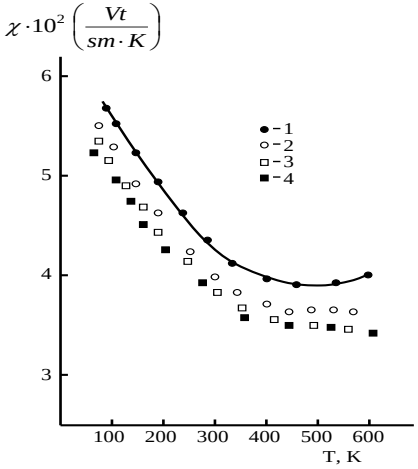

Figure 11. Temperature dependence of the thermal conductivity coefficient of $TlIn_{1-x}Dy_xSe_2$ solid solution crystal. (x: 1-0; 2-0.01; 3-0.03; 4-0.05).

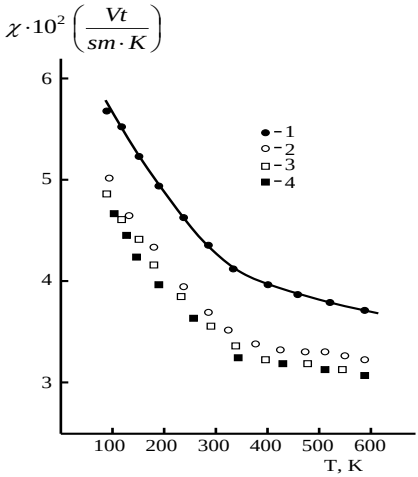

Figure 12. Temperature dependence of thermal conductivity coefficient of $TlIn_{1-x}Eu_xSe_2$ solid solution crystal. (x: 1-0; 2-0.01; 3-0.03; 4-0.05).

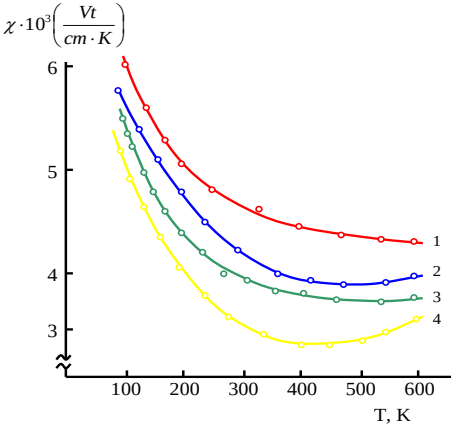

Figure 13. Temperature dependences of the thermal conductivity coefficient for the $TlInSe_2$ solid solution crystal. 1 – non-irradiated, 2 – irradiated with 50 kGray γ-quanta, 3 – 240 hours after irradiation, 4 – for samples irradiated with 100 kGray γ-quanta.

It has been established that when the $TlInSe_2$ crystal is irradiated with γ-quanta, radiation-stimulating processes occur as a result of activation of the migration of specific defects. The control of these processes allows to purposefully change the electrical parameters of crystals. Irradiation with rare earth element atoms and γ-quanta added to the $TlInSe_2$ solid solution crystal strongly affects both the numerical value of the substance's thermal conductivity and its temperature dependence. Acoustic phonons at low temperatures and optical phonons at relatively high temperatures are involved in heat conduction. Figure 13 shows the temperature dependence of the thermal conductivity coefficient for the $TlInSe_2$ solid solution crystal.

Since point defects cause additional scattering of phonons, the thermal conductivity of the crystal decreases with increasing temperature. The observed empirical facts agree well with the theoretical considerations.

Studies show that $A^{III}B^{V}$-type compounds are sensitive to irradiation with γ-quanta and some of their physical properties can be controlled by this method.

CONCLUSİON

1. Based on the research of differential, thermal, microstructure, X-ray structure and other properties, state diagrams of TlInSe2-TlDySe2, TlInSe2-TlEuSe2 systems were constructed in the temperature range of 100-600 K, and it was found that in these systems, new quadruple TlIn1-xMxSe2 in the ratio of 1:1 of the initial compounds (M=Dy, Eu) (x=0.01; 0.03; 0.05) compounds are formed. Both the quadruple compound and TlInSe2 solid solution crystals crystallize in tetragonal syngonia, and the elemental lattice parameters of the solutions increase as the relative amount of rare earth atoms in the composition increases.

2. The thermoelectric properties of TlIn1-xMxSe2 (M=Dy, Eu) (x=0.01; 0.03; 0.05) crystals with a layered and chain structure strongly depend on the concentration of defects that cause local changes in the arrangement of atoms. It has been shown that Eu is more effective than Dy in the preparation of thermocouples from TlIn1-xMxSe2 (M=Dy, Eu) (x=0.01; 0.03; 0.05) crystals. However,

if the magnetic property is used for the preparation of thermocouples, the effect of Dy is greater than that of Eu.

3. It has been shown that three-phonon scattering processes (U-processes) are involved in the transport of thermal energy in the studied solid solution crystals in the temperature range of 80-600 K. There are shallow and deep local levels in the energy band of crystals of TlIn1-xMxSe2 (M=Dy, Eu) (x=0.01; 0.03; 0.05) solutions, which are also filled - the rate of discharge depends on the electric field.

4. For TlIn1-xMxSe2 (M=Dy, Eu) (x=0.01; 0.03; 0.05), the activation energies for different compositions were determined and their values were obtained according to the trends of the high-temperature parts of the electrical conductivity and the Hall coefficient.

5. When the TlInSe2 crystal is irradiated with γ-quanta, radiation-stimulating processes occur as a result of the activation of the migration of specific defects. The control of these processes allows to purposefully change the electrical parameters of crystals. Irradiation with rare earth element atoms and γ-quanta added to the TlInSe2 solid solution crystal strongly affects the numerical value of the thermal conductivity of the substance and its temperature dependence. Acoustic phonons at low temperatures and optical phonons at temperatures above 300 K are involved in heat conduction.

6. It has been shown that the VAX-characteristics of thin-layer planar structures obtained by thermal evaporation in vacuum based on TlIn1-xMxSe2 (M=Dy, Eu) (x=0.01; 0.03; 0.05) crystal materials with a chain layered structure has a memory electrical conductivity characterized by negative differential resistance (S-shaped) dependence.

7. TlIn1-xMxSe2 (M=Dy, Eu) (x=0.01; 0.03; 0.05) combinations in a wide temperature range are promising materials for making memory converters, as well as thermoelectric converters that can work in the temperature range of 300-600 K. it is possible to prepare.

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Address: 131 H. Javid ave., AZ-1143, Baku

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