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ABSTRACT

of the dissertation for the degree of Doctor of Philosophy in Physics

THE EFFECT OF POINT DEFECTS CAUSED BY ADDITIVES IN THE TIInTe₂ COMPOUND ON THE RADIATION RESISTANCE OF THE CRYSTAL

Specialty: 2225.01--Radiation materials science

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GENERAL DESCRIPTION OF THE WORK

Relevance of the subject and degree of development: The complex study of physical processes in semiconductor compounds and the study of the effect of ionizing rays on these processes has always been of scientific and practical importance and has attracted the special attention of researchers. Therefore, acquiring new materials, purposefully managing their physical properties, and developing methods to increase resistance to external influences and ionizing radiation remain crucial areas of scientific research. The topic of this dissertation is both scientifically and practically relevant as it addresses these significant scientific issues.

TlGaTe₂ and $TIInTe_2$ crystals, which are promising representatives of the TlA^{III}C^{VI}₂ compounds (A–Ga, In; C–S, Se, Te) with a layered-chain structure, were taken as the object of research in the dissertation work. One of the materials that combines the mentioned properties is the TlInTe₂ compound. The TlInTe₂ crystal crystallizes in the TISe structure type (I4/mcm) and have the following lattice parameters: a=8.494 Å; c=7.181 Å; c/a=0.845; Z=4; d=7.36 g/cm³. The chemical bonding in TlInTe₂ crystals is ionic-covalent. A study of the electrical, photoelectric, and optical properties of this class of semiconductor compounds revealed numerous interesting characteristics. These properties depend on the crystal structure, the type of chemical bonds between the constituent components, and the degree of disorder in the crystal lattice (stability of ions).

Semiconductor monocrystals, (one of the characteristic properties of crystals with a chain structure is the anisotropy of conductivity) substances with a chain structure that have anisotropic properties in different crystallographic directions, are of special importance. This feature observed in crystals can allow the distribution of defects in the crystal lattice, the development of new types of converters, memory elements and tunnel diodes.

Although it is practically important to obtain semiconductor compounds of this type and create new functional semiconductor devices based on them, the disordered distribution of defects in the band gap does not allow for purposeful control of crystal parameters. To practically solve the mentioned issue, a special technological regime has been developed for the purpose of placing additive atoms of different nature and ionic radius in the octahedral space of TlGaTe₂, TlInTe₂ crystals. The application of the mentioned method can make it possible to control the electrophysical properties of the obtained crystals and to increase their resistance to ionizing rays.

The purpose of the research is to determine the mechanism of influence of point defects caused by additional atoms (Te, Bi, Si) and γ -quanta in TlGaTe₂, TlInTe₂ compounds on the structural properties of the crystal, current flow mechanisms, radiation resistance and to develop practical recommendations based on these findings.

In accordance with the goal set in the dissertation, the following issues were resolved:

– Synthesis of TlGaTe₂ and TlInTe₂ compounds and the growth of single crystals by selecting the appropriate technological regime;

 Determination of the solubility region of Te, Bi, Si additives in TlGaTe₂ and TlInTe₂ compounds, construction of phase diagrams for TlGaTe₂–Te, TlInTe₂–Te, TlInTe₂–Bi, TlInTe₂–Si systems;

- Study of the effect of additive atoms (Te, Bi, Si) on the electrical conductivity of TlInTe₂ crystal;

– Investigation of the impact of additive atoms (Te, Bi, Si) on the Volt-Ampere characteristics of TlInTe₂ crystal;

- Examination of the influence of gamma rays on the electrical conductivity of TlInTe₂ crystals doped with Te, Bi, Si atoms;

– Analysis of the influence of gamma rays on the Volt-Ampere characteristics of TlInTe₂ crystals doped with Te, Bi, Si atoms;

- Study of the impact of additive atoms (Te, Bi, Si) and gammaquanta on the conductivity anisotropy in TlInTe₂ crystal;

Research object and methods:

Differential thermal analysis (DTA) was conducted using the HTP-75 low-frequency thermographic device within the temperature range of 293–1100 K. Heating and cooling curves were recorded during the process, with a temperature measurement error not exceeding ± 1 K.

A "Perkin Elmer STA 6000" thermal analysis device with $\pm 2\%$ calorimetric accuracy was employed to record small thermal effects. Experiments were carried out in an argon atmosphere within the temperature range of 293–600 K.

X-ray phase analysis of system alloys was performed on ground powders after homogenizing annealing in a vacuum. Diffraction patterns of the studied alloys were obtained using Cu–K_{α} radiation on the XRD D8 ADVANCE device in constant shooting mode (I = 40 mA, V = 40 kV) within the angle range 4°<B<35°. The temperature dependence of the lattice parameter values of solid solutions was studied in the temperature range of 300–900 K using the high-temperature chamber of the XRD D8 ADVANCE device. The error in determining parameters was ±0.001 Å.

Microstructural analysis of alloys was conducted using an MMP-4 metallographic microscope with $150-300^{x}$ magnification. During the analysis, a diluted K₂Cr₂O₇+H₂SO₄ mixture was used for etching the surface of polished samples, with an etching period of 1–2 minutes.

The specific gravity of the studied samples was measured using the pycnometric method, employing pycnometers with a volume of 5.0 cm³. "ChDA" brand toluene was used as a filler.

The electrical conductivity of single crystal samples was studied at a frequency of 100 kHz within the temperature range of 100–600 K. Investigations were conducted in two crystallographic directions (001 and 110) using an E7–25 digital immittance measuring device. Electrical contacts to the samples were applied using silver paste.

The intensity of the electric field applied to the crystals was within the ohmic region of the Volt-Ampere characteristic, with the voltage not exceeding 1 V. Volt-Ampere characteristics and electrical conductivity of TlGaTe₂, TlInTe₂, and TlInTe₂–(Te, Bi, Si) solid solutions were studied within the temperature range of 100–300 K. Samples were irradiated with γ -quanta from a ⁶⁰Co source at radiation doses of 0.01–5 MGy.

Main Provisions of the Defense:

1. The phase diagrams of $TIInTe_2$ -Te, $TIInTe_2$ -Bi, $TIInTe_2$ -Si systems were established, the solubility regions of Te, Bi and Si additives in $TIInTe_2$ crystals were determined, and the parameters of the obtained solid solutions were calculated.

2. Ionic conductivity was observed in temperature dependences of electrical conductivity $\sigma(T)$ of TlInTe₂ single-crystal samples doped with Te, Bi and Si in the (110) and (001) crystallographic directions within the temperature range of 300-600 K.

3. The Volt-Ampere characteristics of doped TlInTe₂ (Te, Bi, Si) crystals irradiated with γ -quanta at radiation doses of 0.01–5 MGy were studied in the (*110*; 001) crystallographic directions within the temperature range of 100–300 K, and the effect of radiation defects on the current flow mechanism was determined.

4. The specific electrical conductivity of doped TlInTe₂ (Te, Bi, Si) crystals irradiated with γ -quanta at radiation doses of 0.01–5 MGy was studied in different crystallographic directions (*110*; 001) within the temperature range of 100–300 K. The depths of energy levels were calculated.

5. The joint effect of additive atoms (Te, Bi and Si) and γ quanta on the anisotropic properties and radiation resistance of the TlInTe₂ crystal was determined, and the issues related to the purposeful control of the electrophysical properties of this compound were investigated.

The scientific innovations of the research are followings:

1. Phase diagrams for $TIInTe_2$ -Te, $TIInTe_2$ -Bi, $TIInTe_2$ -Si compounds were studied in the concentration range of 0–10 at.% to determine the solubility regions of Te, Bi and Si additives in $TIInTe_2$ crystals. It was found that Te, Bi, Si atoms in the $TIInTe_2$ compound have a solubility range of 5.0 at.%.

2. It was discovered that the specific resistance in the TIInTe₂ compound decreases by $\sim 10^3$ units with the inclusion of Te atoms in the (001) crystallographic direction and by 5 times in the (110) crystallographic direction. The introduction of a Bi atom into the crystal increases the specific resistance in the (110) crystallographic direction ~ 8 times, and the introduction of a Si atom increases it by ~ 12 times.

3. From the temperature dependences of electrical conductivity $\sigma(T)$ of TlInTe₂ samples doped with Te, Bi, and Si in the (110) and (001) crystallographic directions within the temperature range of 300–600 K, it was determined that the anomalous high conductivity observed at temperatures above T>300 K is due to sublattice disruption of the thallium cation at the "critical temperature"

4. It was determined that the defects formed in the TlInTe₂ crystal without additives at doses of D<0.5 MGy are of donor nature and predominantly distributed in the (001) crystallographic direction. At doses of D>0.5 MGy, the defects formed are of acceptor nature, with distribution predominating in (110) crystallographic direction.

5. It was determined that the p–TIInTe₂ crystal has high radiation resistance in the (001) crystallographic direction, and the excess of Te atoms in the stoichiometric ratio reduces the radiation resistance in the (110) crystallographic direction. It is possible to control the properties of the p-TIInTe₂ +4% Te crystal irradiated with γ -quanta by the interaction of defect-dopant interaction, radiation resistance and anisotropic properties of the crystal.

Practical significance of research.

The parameters and radiation resistance of different types of converters and rectifiers created based on doped $TlGaTe_2$ and $TlInTe_2$ crystals can be effectively controlled.

Approbation and application of the research:

The results of the dissertation were presented at the following conferences: "7th International conference MTP–2021: Modern trends in physics" Baku (2021); "XXVI International Scientific and Technical Conference on Photoelectronics and Night Vision Devices" Moscow (2022); "Modern problems of theoretical & experimental chemistry" Baku (2022); "Republican scientific conference dedicated to the 60th anniversary of Sumgayit State University" Sumgayit (2022);

Fourteen scientific works related to the dissertation were published, including nine articles and five conference materials in both national and international scientific journals.

Name of the organization where the dissertation work was performed:

The presented dissertation work was performed in the "Materials Science" laboratory of the Institute of Physics of the Ministry of Science and Education of the Republic of Azerbaijan and in the "Radiation Physics of Semiconductors" laboratory of the Institute of Radiation Problems.

Structure and volume of the dissertation:

The dissertation consists of an introduction, four chapters, conclusions, and a list of references. The work includes 42 figures, 6 tables, and a total of 183200 characters.

MAIN CONTENT OF THE RESEARCH

The introduction justifies the relevance of the dissertation topic, outlines its purpose, scientific innovation, practical significance, and main provisions.

Chapter I examines the electrophysical properties of thalliumbased chalcogenides, included in the class of $A^3B^3C_2^6$ group compounds. It reviews literature data on the effects of gamma rays on these properties and discusses the theoretical basis of superionic conductivity.

Chapter II details the synthesis process of the studied materials, the determination of the solubility of additives, and the method for growing single crystals. The chapter also describes the devices used to study the electric and dielectric properties, their operating principles, and methods of irradiation with ionizing rays. Alloys of TlGaTe₂ and TlInTe₂ systems doped with Te, Bi, Si were synthesized in evacuated Stepanov quartz ampoules using a direct single-temperature synthesis method at 1.0 at.% intervals. The initial components were polycrystalline TlGaTe2, TlInTe2, and Te, Bi, Si additives. During the synthesis, the furnace temperature was raised to T=1150 K at a rate of ~150 K/h, maintained at this temperature for ~2 hours, then cooled to room temperature at a rate of ~100 K/h. To reduce pore formation, crystallization of the alloys was performed using 50 Hz frequency vibration. The alloys were then heated for two weeks at T=470 \pm 5 K to reach equilibrium. The equilibrium state was verified using X-ray phase analysis. The purity of the elements used was no less than 99.999%.

Chapter III presents the results of physical-chemical studies to determine the solubility of Te, Bi, and Si additives in $TIGaTe_2$ and $TIInTe_2$ compounds. It was established that by creating pre-planned point defects in these compounds, the type of conductivity, specific conductivity, and dielectric permeability can be purposefully controlled.

The growing interest in the study of $A^3B^3C_2^6$ group compounds and solid solutions is related to the prospects of using these materials, notably the discovery of ionic conductivity at temperatures close to room temperature.

The type of atoms included in the lattice structure was chosen based on their chemical activity, value of ionic radius and coordination number. At this time, the chemical activity of the selected atom should not exceed the chemical activity of the lattice atoms, otherwise the lattice atoms will be displaced from the composition and the substitutional solid solution will be formed. In addition, the value of the ionic radius and the coordination number of the atom included in the lattice should allow it to be placed in the selected polyhedron without deforming the elementary lattice.

Phase diagrams for TlGaTe₂–Te, TlInTe₂–Te, TlInTe₂–Bi, TlInTe₂–Si combinations were established and the results of physical-chemical study of the alloys were shown.

The analysis of system alloys was performed using differential thermal (DTA), X-ray phase (RPA), and microstructural (MSA) analyses. Figure 1 depicts fragments of phase diagrams for the TlGaTe₂–Te and TlInTe₂–Te systems, showing that the solubility range of tellurium in these ternary compounds is 5.0 at.%.

According to the results of MSA, the alloys of the TlGaTe₂–Te, TlInTe₂–Te system with a concentration of 1.0–5.0 at.%Te are single-phase and are solid solutions of TlGaTe_{2+x} and TlInTe_{2+x}.

After examining the X-ray diffractograms of the TlInTe₂–Te system alloys, it was found that the lattice parameters of the samples show a slight change compared to TlInTe₂ crystals (a=8.494 Å, c=7.181 Å). The powder diffraction pattern of solid solution polycrystals containing 4.0 at.% Te indicates the following lattice parameters: a=8.4850 Å, c=7.1924 Å. Thus, a decrease in the a-

parameter of the elementary lattice, and a slight increase in the cparameter were observed.



Figure 1. Fragments of phase diagrams for TlGaTe₂–Te (a) and TlInTe₂–Te (b) systems studied in the concentration range of 0–10.0 at.%. In the figure, the single-phase state of the alloys in the Te concentration field is marked with white circles, and the two-phase states with black circles.

RF analyses were conducted on TlGaTe₂–Te and TlInTe₂–Te alloys with 3.0, 4.0, and 5.0 at.% Te concentrations, showing slight changes in lattice parameters with increasing tellurium concentration in the α -solid solutions. For the alloy containing 5.0 at.% Te, the lattice parameters calculated from the 004, 006 and 400 index reflections are: TlGaTe_{2.04} a=8.476 Å, c=6.985 Å and TlInTe_{2.04}, a=8.482 Å, c=7.192 Å.

To assess the solubility of bismuth in $TIInTe_2$ crystals, the $TIInTe_2$ -Bi phase diagram was studied in the concentration range of 0–10 at.% Bi using physicochemical analysis methods.

In the electrochemical activity of metals, Bi ranks after hydrogen, its chemical activity is lower than that of thallium and indium, in the case of Bi^{3+} , the ionic radius is 1.2 Å. Bismuth ions can occupy positions with coordination number 5, 6 and 8 in the lattice structure. Thus, according to a preliminary assessment, the additive atoms we introduced can be located in the octahedral spaces between the thallium atoms in the TlGaTe₂ and TlInTe₂ elementary lattice.

To study the TlInTe₂–Si system, alloys containing 0–10.0 at.% Si with an interval of 1.0 at.% were synthesized. Since the chemical activity of Si is weaker than the Tl and In cations contained in the studied ternary compound, substitution and displacement cannot occur in the TlInTe₂ compound. Considering that the ionic radius of Si⁴⁺ is 0.4 Å in 4-coordination and 0.54 Å in 6-coordination, it is highly likely to dissolve in TlInTe₂ compound.

Microstructure analysis of polished samples confirmed that alloys containing 1.0-5.0 at.% Si are homogeneous and belong to the solid solution region based on the TIInTe₂ compound.

Samples with a height of 5–7 mm along the c axis were cut from the grown single crystal. Contacts with silver paste were applied to the surface of the crystals in the (001) and (110) crystallographic directions. The VA characteristics of the samples were studied, the type conductivity was determined, the electrical conductivity was measured within the temperature range of 100–370 K, and the values of activation energy (E_a) and specific resistance (ρ) were calculated.

The study revealed significant differences in electrophysical properties and indicators of samples doped with Te, Bi, and Si atoms compared to TlInTe₂ single crystals. Unlike TlInTe₂ single crystal, samples doped with Bi and Si exhibited n-type conductivity. The inclusion of Te atoms in the lattice reduced the specific resistance in the (001) crystallographic direction by ~10³ times and in the (110) crystallographic direction by ~5 times. Introduction of Bi and Si atoms increased the specific resistance in the (110) crystallographic direction by ~8 and ~12 times, respectively.

The study of Volt-Ampere characteristics of $TIInTe_2$ and $TIInTe_{2.04}$ crystals in (001) and (110) crystallographic directions at 300 K showed different behavior. It was determined that the

dependence of $I \sim U^n$ in the TIInTe₂ sample taken in the (001) crystallographic direction is linear (n=1) and is characterized as an state without traps. In the (110) crystallographic direction, the ohmic region (J~U) is observed in the Volt-Ampere characteristic up to a certain value of the voltage, and the quadratic region (J~Uⁿ, n>1) in the subsequent increase of the value of the field. The linear part is observed in the field range of 1–11.5 V/cm, and the quadratic region is observed in the field range of 11.5–13.0 V/cm. The increase of the current value depending on the transition voltage from the ohmic region to the quadratic region occurs as a result of the ionization of acceptor-type defects.

In the TIInTe_{2.04} crystal, in the (001) crystallographic direction, as in the sample without additives, the dependence of I~Uⁿ is linear (n=1) and is characterized as an state without traps. It was shown that by adding Te atom, the distribution of defects in the (001) crystallographic direction in the TIInTe₂ crystal is regulated, and as a result, the conductivity of the crystal increases.

In the VA characteristic of $(TIInTe_2)_{0.95}Bi_{0.05}$ crystal in the (001) crystallographic direction, the ohmic region (J~U) up to the field value of 0–0.68 V/cm was detected. In the (110) crystallographic direction, a sharper voltage increase (J~Uⁿ, n>1) is observed at the field value of 0-32.5 V/cm. Unfortunately, it was not possible to measure the VA characteristic at high field values due to sample heating.

Also, the Volt-Ampere characteristic of $(\text{TIInTe}_2)_{1-x}\text{Si}_x$ sample was studied. It was shown that the dependence of I(U) in the (*110*) crystallographic direction of the Si-doped TIInTe₂ crystal is linear, which is related to the thermal ionization of the defect level. In contrast, the VA characteristic in the (*001*) crystallographic direction is exponential. Such a nature of dependence is related to the transition of free charge carriers between local regions.

In chapter III, the temperature dependences of electrical conductivity of TlInTe₂ samples and those doped with Te, Bi, Si in (001), (110) crystallographic directions within the temperature range of 100–370 K were studied.

The temperature dependence of electrical conductivity $\sigma(T)$ of TlInTe₂ and TlInTe_{2.04} samples in (001), (110) crystallographic directions was studied. From the $\sigma(T)$ dependence curves of the TlInTe₂ crystal, the values of the activation energies E_a were determined: 0.35 eV in the (001) crystallographic direction, and 0.12 eV in the (110) crystallographic direction.

The $\sigma(T)$ dependence in the TlInTe₂ crystal in the (001) crystallographic direction exhibits additional conductivity and consists of two parts. In the temperature range of 165–250 K, conductivity does not depend on temperature, while in the range of 250–300 K, conductivity increases with temperature.

The introduction of a Te atom is shown to increase the conductivity of the crystal, and the additive Te atom creates an acceptor-type energy level in the TIInTe₂ crystal.

For the $(TIInTe_2)_{1-x}Bi_x = 0.05$ composition, the $\sigma(T)$ dependence in the (001) crystallographic direction in the temperature range of 165–307 K is linear and does not depend on temperature. This behavior is attributed to the high number of energetically close defect levels formed due to the introduction of the bismuth atom into the crystal.

The introduction of a Bi additive resulted in a decrease in the electrical conductivity of the crystal in the (110) crystallographic direction by one order, with an activation energy of $E_a=0.5$ eV. This decrease in electrical conductivity is attributed to the compensation of acceptor levels and the formation of donor levels, leading to a change in the type of conductivity in the crystal.

The addition of a Si atom into the TIInTe₂ crystal resulted in a weak change in the value of electrical conductivity in both crystallographic directions, although the conductivity in the (001) crystallographic direction relatively decreases. By adding a silicon additive atom to the TIInTe₂ crystal, it regulates the distribution of defects in both crystallographic directions, and as a result, the conductivity of the crystal partially changes. In the (TIInTe₂)_{1-x}Si_x x=0.05 crystal, the conductivity is replaced by electron conductivity.

Chapter III also presents the results of the study of the temperature dependences of electrical conductivity $\sigma(T)$ of TlGaTe_{2.04} and Tl1nTe_{2.04} single crystals in the (*110*) and (*001*) crystallographic directions in the temperature range of 300–600 K. (Figure 3). The tests were performed using an E7–24 digital impedance meter, with the applied electric field intensity corresponding to the ohmic region of the I~f(U) characteristic and the voltage not exceeding 1 V.

It was found that the temperature dependences of the electrical conductivity $\sigma(T)$ of the TlGaTe₂ crystal exhibited jumps in both crystallographic directions ((001) and (110)) due to the transition to the superionic phase. The transition in the (001) crystallographic direction occurs at a lower temperature than in the (110) crystallographic direction, with a conductivity change in the (001) direction being three orders of magnitude higher.

In the $\sigma(T)$ dependence of the TlGaTe_{2.04} crystal, the transition to the superion phase is observed only in the (*110*) crystallographic direction (Figure 2). A comparison of the $\sigma(T)$ curves of TlGaTe₂ and TlGaTe_{2.04} crystals in the (*110*) crystallographic direction shows that the transition temperature to the ion-conducting phase is 5 K higher in the TlGaTe_{2.04} crystal. Note that a similar observations is made in the $\sigma(T)$ dependences of TlInTe₂ and TlInTe_{2.04} crystals.



Figure 2. Temperature dependences of the electrical conductivity of $TIGaTe_2$ (a) and $TIGaTe_{2.04}$ (b) crystals at a measurement frequency of 100 kHz: in the (001) crystallographic direction (curve 1), in the (110) crystallographic direction (curve 2).

It was determined that in $\sigma(T)$ dependences of the TlGaTe₂ crystal, jumps were observed in both crystallographic directions due to the transition to the superion phase. The transition observed in the (001) crystallographic direction occurs at a lower temperature than in the (110) crystallographic direction, and the conductivity change in the (001) crystallographic direction is three orders of magnitude bigger.

In the $\sigma(T)$ dependence of the TlGaTe_{2.04} crystal, the transition to the superion phase is observed only in the (*110*) crystallographic direction (Figure 3 b). Comparison of the $\sigma(T)$ curves of TlGaTe₂ and TlGaTe_{2,04} crystals in the (*110*) crystallographic direction shows that the transition temperature to the ion-conducting phase is 5 K higher in the TlGaTe_{2,04} crystal. Note that a similar picture is observed in the $\sigma(T)$ dependences of TlInTe₂ and TlInTe_{2,04} crystals.

In the temperature dependence of the dielectric constant $\varepsilon(T)$, there is an increase in the value of the dielectric constant at a temperature above 300 K, and a sharp jump is observed at a temperature of 498 K, and the crystal becomes ionic conductive.

Thus, two anomalies are observed in the $\varepsilon(T)$ dependence of the $(TIInTe_2)_{1-x}Bi_x = 0.05$ sample in the (001) crystallographic direction. The first anomaly appears in the temperature range of 450–500 K, when the value of the dielectric constant increases by an order of magnitude.

The second anomaly occurs at 567 K, when the dielectric constant increases by four order of magnitude. It was determined that the bismuth atoms introduced into the TIInTe₂ crystal lattice affect the transition temperature (*Ti*) of the crystal to the ion-conducting phase, increasing it by 69 K in the (001) crystallographic direction.

Conductivity anomalies at the corresponding temperatures in the temperature dependence curves of the electrical conductivity of the studied samples: are more than two order of magnitude for the TIInTe₂ crystal, and four order of magnitude for the sample containing (TIInTe₂)_{1-x}Bi_x x=0.05.

A similar anomaly is observed in the $\sigma(T)$ dependence measured at a frequency of 100 kHz for the sample with $(TIInTe_2)_{1-x}Bi_x x=0.05$ composition. At this time, the conductivity increases by four orders of magnitude at 567 K in the (001) crystallographic direction and at 572 K in the (110) crystallographic direction and the crystal becomes ion conductive.

Note that anomalies in the $\varepsilon(T)$ and $\sigma(T)$ curves in both samples are also observed in the (*110*) crystallographic direction. This anomaly occurs at a temperature of 485 K for the TlInTe₂ crystal, and 572 K in the sample containing (TlInTe₂)_{1-x}Bi_x x=0.05. Comparing the Ti values of the phase transition temperature in the (001) and (110) crystallographic directions, it is known that doping with bismuth has a strong effect on the dielectric properties. The phase transition temperature of Ti increases by 69 K in the (001) crystallographic direction, and by 87 K in the (110) crystallographic direction.

It has been shown that bismuth additives in the sample containing $(TIInTe_2)_{1-x}Bi_x x=0.05$ increase the conductivity in the (001) crystallographic direction and change the conductivity type in the TIInTe₂ crystal. Bismuth additives increase the conductivity anisotropy $\rho \perp / \rho \parallel$ in the sample of $(TIInTe_2)_{1-x}Bi_x x=0.05$ more than 10^3 times compared to the crystal without additives.

The obtained results are of practical importance, they can be used in obtaining ion conductors with controlled properties and materials with high electrical conductivity anisotropy.

In the fourth chapter, the effect of γ -irradiation on the charge carrying mechanism in TlInTe₂, TlInTe₂–Te, Bi, Si solid solutions was studied and the obtained results were compared with the materials in the scientific literature.

Radiation defects caused by γ -rays in semiconductor materials are of special interest. Because unlike heavy particles, when irradiating samples with γ -rays with an energy of 1.2 MeV, simple defects such as vacancies and internodal atoms (Frenkel pairs) are formed.

The chapter IV presents the VA characteristics of TlGaTe₂, TlInTe₂, TlInTe₂–Te, Bi, Si solid solutions at 300 K, within the voltage range of 0-5 V in (001) and (110) crystallographic directions (Şəkil 3).



Figure 3. VA characteristics of TlInTe₂ crystal at 300 K in different crystallographic directions: a– (001) crystallographic direction, b– (110) crystallographic direction.
1–D=0; 2–D=0.01 MGy; 3–D=0.1 MGy; 4–D=0.5 MGy; 5–D=2.5 MGy; 6–D=5 MGy

It was determined that the $I \sim f(U)$ dependence of the TlInTe₂ crystal in the (001) crystallographic direction before irradiation is linear, with the current increasing proportionally to the applied voltage and obeys the $I \sim U^n$ law. These results are interpreted in accordance with Lampert's law. The obsserved exponential dependence of the current in the $I \sim f(U)$ relationship for the TlInTe₂ crystal is attributed to the thermal ionization of the local levels in the band gap and the current generated by charge carriers injected from the contact.

The effect of γ -quanta on the VAC in the TlInTe₂ crystal was investigated. It was determined that radiation defects formed during irradiation at doses of 0.01–0.5 MGy cause a decrease in the current in the (001) crystallographic direction of the TlInTe₂ crystal, and an increase in the current at doses of 0.5–5 MGy. In the TlInTe₂ crystal, in the (110) crystallographic direction, the current decreases after irradiation at doses of 0.01–0.5 MGy, and the current increases at doses of 0.5–5 MGy. Based on the observed results, it can be said that the nature of the defects formed during irradiation depends on the value of the absorption dose. Thus, since the defects formed at low radiation doses are anionic in nature, they create a donor level, while at high doses, they are cationic in nature, so the conductivity of the crystal increases.

VA characteristics in TlInTe₂ crystals doped with Te atoms were studied before and after irradiation in different crystallographic directions.

It was determined that when an electric field is applied to the TlInTe₂–Te crystal irradiated at doses of 0.1-2.5 MGy, a sharp decrease in the current indicates that the defects are mainly distributed in the (001) crystallographic direction and that the resulting defects are of donor nature.

Based on the obtained results, it can be said that during irradiation at a dose of 2.5 MGy, γ -quanta weaken the In-Te bond, and therefore the conductivity of Tl-ions increases, as a result, conductivity decreases in the (001) crystallographic direction, and increases in the (110) crystallographic direction.

The effect of gamma rays on the VA characteristics of Bidoped TIInTe₂ crystal taken at 300 K temperature in different crystallographic directions was also studied.

It was shown that the conductivity of the TIInTe₂<Bi> crystal decreases due to the formation of a donor-type energy level during irradiation at a dose of 0.01 MGy. Conductivity increases after irradiation at a dose of 0.1 MGy. At further doses of irradiation, 0.5– 5 MGy, the conductivity of the TIInTe₂<Bi> crystal begins to decrease again. The reason for this is the increase in the concentration of donor-type defects with an increase in the radiation dose. The reason for the increase in the conductivity of the crystal at a dose of 0.1 MGy compared to the initial sample is the self-annealing of the radiation defects created during the irradiation. That is, the concentration of donor-type defects decreases due to the recombination of the generated Frenkel pairs. Thus, it was determined that the conductivity of the TIInTe₂<Bi> crystal decreases in the (001) crystallographic direction during gamma-quantum irradiation. In the (110) crystallographic direction at radiation doses of 0.01-5 MGy, the conductivity of the TlInTe₂ <Bi> crystal decreases at low values of the electric field intensity applied to the crystal (103 V/cm), and increases at high fields. Based on the obtained results, it can be said that since the energy of the applied field is lower than the height of the potential barrier, the concentration of charge carriers participating in conduction decreases. At high applied fields, the height of the potential barrier decreases due to the effect of the field, and the conductivity of the crystal increases as a result of the increase in the concentration of charge carriers participating in the conductivity.

The results obtained from the analysis of the VA characteristics of the investigated crystals irradiated at the selected radiation dose show that the complexes formed as a result of the interaction of structural defects with radiation defects cause a change in conductivity in the crystallographic direction.

In the fourth chapter of the thesis, the Volt-Ampere characteristics of the $(TIInTe_2)_{1-x}Si_x$ sample, irradiated at doses of 0.01–5 MGy, in the (001) and (110) crystallographic directions were studied.

At a radiation dose of 0.01 MGy, the value of the current decreases sharply and the observed quadratic region shifts to the high voltage region. The decrease in the current indicates that the radiation defects formed during irradiation are donor in nature. An increase in current is observed at radiation doses of 0.1 and 0.5 MGy. With increasing dose (2.5; 5 MGy), the current increases from the initial value and the quadratic region shifts to the lower voltage region. The analysis of the obtained results shows that the change of the current depending on the radiation dose depends on the nature of the generated radiation defects. In the TIInTe₂+5.0 at.% Si crystal in the (*110*) crystallographic direction, the value of the current increases from the initial value at the radiation doses of 0.01–5 MGy. The obtained results show that the increase of the current in the (*110*) crystallographic direction is due to the acceptor nature of the generated radiation defects.

In the IV chapter of the dissertation, the temperature dependence of the specific electrical conductivity of the

TlInTe₂+4%Te crystal at different radiation doses $(0.01\div 5 \text{ MGy})$ is given in Figure 4. The excess of Te atom in the TlInTe₂ crystal does not change the character of the curve in the dependence of $\lg \sigma \sim f(1/T)$, but the conductivity value partially increases. In the dependence of $\lg \sigma_1 \sim f(1/T)$, after 0.01 MGy irradiation, the conductivity increases sharply and an acceptor-type energy level with activation energy of $E_a=0.14$ eV is formed. At the radiation dose of D=0.5 MGy, a sharp decrease in conductivity occurs as a result of the formation of a donor-type level with activation energy $E_a=0.30$ eV. With the subsequent increase in radiation, a sharp increase in conductivity is observed.



Figure 4. Temperature dependences of specific electrical conductivity of TIInTe₂–Te (a,b) crystal irradiated at different radiation doses (0.01÷5 MGy): a–(001) crystallographic direction, b– (110) crystallographic direction 1–D=0; 2–D=0.01 MGy; 3–D=0.1 MGy; 4–D=0.5 MGy; 5–D=2.5 MGy; 6–D=5 MGy

In the TlInTe₂–Te crystal, after irradiation with doses of 0.01-2.5 MGy, the electrical conductivity in the (110) crystallographic

direction decreases. The reason for this is the predominance of donor-type defects, that is, the formation of donor-type levels. After irradiation with a dose of 5 MGy, the conductivity in the dependence of $\sigma(T)$ does not change depending on the temperature. Such a dependence is due to the presence of tellurium additive in the TlInTe₂ crystal and the high number of energetically close defect levels formed due to the effect of radiation defects.

Figure 5 presents the temperature dependence of the anisotropy of the specific electrical conductivity in $TlInTe_2$ and $TlInTe_2$ –Te crystals at radiation doses of $0.01\div 5$ MGy.



Figure 5. Temperature dependences of the anisotropy of specific electrical conductivity of TlInTe₂ (a) and TlInTe₂–Te (b) crystals irradiated at different radiation doses (0.01÷5 MGy).
1–D=0; 2–D=0.01 MGy; 3–D=0.1 MGy; 4–D=0.5 MGy; 5–D=2.5 MGy; 6–D=5 MGy

It was determined that the dependence of $\lg \sigma_{\parallel}/\lg \sigma_{\perp} \sim f(1/T)$ is exponential. From $(\lg[\sigma_{\parallel}/\sigma_{\perp}] \sim f(1/T)$ dependence, it was determined that the conductivity anisotropy in the *p*-TIInTe₂ crystal within the temperature range of 160–300 K varies between values of $-0.5 \div +0.5$, and this is related to the weak anisotropy of the effective mass of charge carriers. However, within the temperature range of 200–250 K, a partial increase of the $\sigma\perp$ -component is observed, which is related to the irregular distribution of electroactive defects in the crystal lattice. After irradiation with γ -quanta at a dose of 0.01 MGy, a sharp increase of $\sigma\perp$ -component of the conductivity is observed within the temperature range of 160-300 K compared to σ_{\parallel} . In the subsequent doses of radiation, the $\sigma\perp$ -component of conductivity is observed to decrease compared to σ_{\parallel} . At the radiation dose of D=2.5 MGy, the ratio of $\lg\sigma_{\parallel}/\lg\sigma_{\perp}$ approaches unit, that is, the anisotropy of the system decreases sharply.

The observed facts show that in the p-TIInTe₂ crystal, the radiation defects generated at initial doses during irradiation with γ -quanta are of donor nature - they are caused by the occupation of the V_{TI} vacancy by internodal *Tl* atoms and increase the anisotropy of conductivity. At higher doses of radiation, the value of the σ_{\parallel} -component changes in the opposite direction due to the increase in the concentration of the acceptor level as a result of the dissociation of the resulting complexes [V_{TI} – Tl_i].

Also, in the dissertation, the temperature dependence of the anisotropy of the specific electrical conductivity of the TlInTe₂-Te crystal was studied and the obtained results were explained in the fourth chapter. It is shown in the dependence of $\lg[\sigma_{\parallel}/\sigma_{\perp}] \sim f(1/T)$ in the TIInTe₂-Te crystal that the introduction of Te-doped atom leads to a partial increase of the anisotropy $lg[\sigma_{\parallel}/\sigma_{\perp}]$ at temperatures T<250 K. From the analysis of the curves, it was determined that the excess of Te atom partially fills the octahedral void in the crystal lattice and creates an additional potential barrier for the free movement of Tl⁺ ions in the (001) crystallographic direction. In $\lg[\sigma_{\parallel}/\sigma_{\perp}]$, the σ_{\parallel} -component sharply increases with respect to σ_{\perp} at the radiation dose of 0.01 MGy, and partially increases at the dose of 0.1 MGy. Compared to the non-irradiated sample, a sharp decrease in conductivity is observed at a dose of 0.5 MGy, and an increase is observed at a dose of 2.5 MGy. Radiation defects generated during irradiation have a stimulating effect on the redistribution of excess Te atoms, and as a result, defects are arranged. For this reason, the conductivity increases in the (001) crystallographic direction. As a

result of the formation of the $[V_{TI}-Te_i]$ complex at high radiation doses, the σ_{\parallel} -component of conductivity decreases sharply. At a dose of 5 MGy, a decrease in the conductivity anisotropy is observed.

As a result of the influence of radiation, the formation of radiation defects, their migration, and the formation of complexes with the presence of additive and cation atoms are observed. Due to the dissociation of complexes at high radiation doses, the concentration of point defects increases, and the anisotropy of conductivity decreases.

It was determined that it is possible to control the distribution of defects in the crystallographic direction by the influence of γ -quanta. Thus, in the p–TlInTe₂<Te> crystal, in which more than stoichiometric Te atoms have been inserted at high radiation doses, the compensation of the initial defects occurs as a result of the interaction of radiation defects with additive atoms, as a result, the conductivity of the crystal decreases. At low radiation defects is less than structural defects. The analysis of the obtained results shows that the excess of additive atoms in the crystals with a chain structure allows controlling the anisotropic properties of the materials and increasing the radiation resistance of the crystals.

According to the research results, it can be said that γ -quanta and the excess of Te atom provides an opportunity to purposefully control the conductivity anisotropy in the TIInTe₂-Te crystal.

The fourth chapter presents the results of the study of the electrical conductivity of the $TIInTe_2$ crystal doped with Bi atoms within the temperature range of 100–300 K.

The temperature dependence of electrical conductivity in different crystallographic directions of $TIInTe_2$ and $TIInTe_2$ –Bi crystals irradiated at doses of 0; 0.01; 0.1; 0.5; 2.5 and 5 MGy was studied and depicted in Figure 6.



Figure 6. Temperature dependence of specific electrical conductivity in TlInTe₂–Bi (a,b) crystal irradiated at different radiation doses (0.01÷5 MGy): a–(001) crystallographic direction; b–(110) crystallographic direction . 1–D=0; 2–D=0.01 MGy; 3–D=0.1 MGy; 4–D=0.5 MGy; 5–D=2.5 MGy; 6–D=5 MGy

It was determined that in the sample with $(TIInTe_2)_{1-x}Bi_x$ x=0.05 composition within the temperature range of 165–307 K, the $\sigma(T)$ dependence before and after irradiation is linear in the (001) crystallographic direction and no change is observed depending on the temperature. Such a dependence is explained as a high number of of energetically close defect levels formed as a result of bismuth doping in the TIInTe₂ crystal.

In the crystal with $(TIInTe_2)_{1-x}Bi_x x=0.05$ composition, there is a change in the nature of the curves in the $lg\sigma \sim f(1/T)$ dependence in the (*110*) crystallographic direction. In the $lg\sigma \sim f(1/T)$ dependence, no change is observed within the temperature range of 150–200 K, and an increase is observed in the high temperature region. The excess of Bi atom in the TIInTe₂ crystal does not change the nature of the two-part curve observed in the $lg\sigma \sim f(1/T)$ dependence, but the value of the conductivity partially increases. After the radiation dose in the direction of $lg\sigma \sim f(1/T)$, there is a decrease in the conductivity. At 0.01 and 0.1 MGy radiation doses, the nature of the initial curve is almost preserved. At the radiation dose of D=0.5 MGy, 2.5 MGy and 5 MGy, the temperature dependence of conductivity is weak. At the radiation dose of D=0.01 MGy, the reduction of conductivity occurs as a result of the formation of a donor-type level with activation energy of E_a =0.40 eV.

Based on the obtained results, it was shown that during irradiation with γ -quanta, cation and anion type defects are formed in the crystal. Since the concentration of Frenkel-type defects formed in the initial doses of radiation is less than the concentration of structural defects, acceptor-type defects prevail. For this reason, the conductivity of the TlInTe₂ crystal increases at a radiation dose of 0.1 MGy. At the doses of 0.5 and 2.5 MGy, the conductivity decreases because the donor-type defects prevail.

The characteristic feature of the obtained results is that since the radiation defects formed during irradiation are of a structural defect nature, only their concentration changes, but the activation energy of energetic levels does not change.

Also, during the dissertation work, the temperature dependence of the specific electrical conductivity $\sigma(T)$ of the $(TIInTe_2)_{1-x}Si_x$ sample at different doses and in the (001) and (110) crystallographic directions within the temperature range of 100-350 K was studied and explained in the fourth chapter. Comparing the (TIInTe₂)_{1-x}Si_x sample with the nonirradiated sample, it was determined that the conductivity increases after irradiation with γ -quanta at doses of 0.01 MGy and 0.1 MGy. The slope of the curve does not change in the studied temperature range. At doses of 0.1 and 0.5 MGy of irradiation, the nature of the initial curve is almost preserved, but the conductivity decreases. At the subsequent radiation dose (5 MGy), the change in the conductivity is observed to be weak. The obtained results show that cation and anion type defects appear in the crystal during irradiation with γ -quanta. Since the concentration of Frenkel-type defects formed in the initial doses of radiation is less than the concentration of structural defects, acceptor-type defects prevail. For this reason, the conductivity of TlInTe₂ crystal doped with silicon increases at radiation doses of 0.01-0.1 MGy. At 0.5 and 2.5 MGy radiation doses, a partial decrease in the conductivity is observed due to the predominance of donor-type defects. At a radiation dose of 5 MGy, the change in the conductivity is observed to be weak.

The temperature dependence of the specific electrical conductivity of (TIInTe₂)_{1-x}Si_x crystal at different radiation doses (0.01÷5 MGy) in the (*110*) crystallographic direction was studied. Based on the experimental results, it was determined that after irradiating the studied crystal with γ quanta, the nature of the σ (T) dependence in the (*110*) crystallographic direction changes compared to the non-irradiated sample. After irradiation with a dose of 5 MGy, the conductivity in the σ (T) dependence does not change depending on the temperature. Such a nature of the dependence is due to the inclusion of silicon additive in the TIInTe₂ crystal and the high number of energetically close defect levels formed as a result of radiation defects after irradiation.

Results

During the implementation of the dissertation work, the following main results were obtained:

1. To determine the degree of dissolution of Te, Bi, and Si atoms in TlInTe₂ and TlGaTe₂ crystals, the phase diagram was studied within the concentration range of 0–10 at.%. It was established that Te, Bi, Si atoms in the TlInTe₂ compound have a solubility range of 5 at.%. The inclusion of Te, Bi and Si atoms in the (001) crystallographic direction decreases the specific resistance by ~10³ times. The inclusion of Te atoms reduces the specific resistance in the (110) crystallographic direction by 5 times, Bi - by 8 times, and Si - by ~12 times.

2. It was determined that the transition to the superion phase in the $\sigma(T)$ dependence of the TlInTe₂-Te(4at.%) crystal is observed only in the (110) crystallographic direction, attributed to the placement of Te atoms in the octahedral voids in the (001) crystallographic direction.

3. The activation energies of energy levels in (001) and (110) crystallographic directions in TlInTe2, TlInTe2–Te crystal were calculated. The values of these energies are 0.12, 0.35 eV and 0.10; 0.32 eV, respectively;.

4. Partial filling of the octahedral voids due to the introduction of excess Te atom into the TlInTe₂ crystal creates an additional potential barrier for the free movement of Tl⁺ ions in the (110)

crystallographic direction. Concequently, the conductivity anisotropy increases at T>250 K, and decreases at T< 250 K. Defects caused by irradiation with γ -quanta at 0.5–5 MGy partially compensate for additive and structural defects, as a result of which the anisotropy of conductivity does not depend on temperature.

5. The Volt-Ampere characteristics and specific electrical conductivity of TlInTe₂ and TlInTe₂–Te (4at.%) crystals irradiated with γ -quanta were studied within the temperature range of 100–300 K. It was determined that the defects formed in the TlInTe₂ crystal without additives at doses of D<0.5 MGy are of donor nature, predominantly distributed in the (001) crystallographic direction. At doses of D>0.5 MGy, the defects formed are of acceptor nature, with a predominant distribution in the (110) crystallographic direction.

6. It was determined that the complex defects-complexes are formed in p-TlInTe₂ crystals doped with Te, Bi, and Si additive atoms during γ -quanta irradiation and their distribution in (001), (110) crystallographic directions changes the anisotropic properties depending on the nature of the additive atom and the radiation dose.

7. It was determined that in p–TlInTe₂<Te> and n–TlInTe₂<Si> crystals, at radiation doses of 0.01-0.5 MGy, the conductivity decreases due to the predominance of donor-type defects in the (001) crystallographic direction. However, at doses of 0.5-5 MGy, conductivity increases as defects of acceptor nature become dominant.

8. It was determined that the radiation resistance of the p–TIInTe₂ crystal increases in the (001) crystallographic direction, while it decreases in the (110) crystallographic direction in the p–TIInTe₂<Te> crystal with an excess of Te atoms beyond stoichiometry. In the p– TIInTe₂+4% Te system irradiated with γ -quanta, the interaction of defect–dopant associations allows for controlling the radiation resistance and anisotropic properties of the crystal.

The main results of the dissertation work were published in the following articles and theses.

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