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# ABSTRACT

of the dissertation for the degree of Doctor of Philosophy

## OBTAINING CRYSTALS $TlA_{1-x}M_xS_2$ ($Se_2$) (A-In, Ga; M-Dy, Er, Yb; x=0÷0.3) THEIR ELECTRICAL AND OPTICAL PROPERTIES

Speciality: 2220.01 – Physics of semiconductors

Field of science: Physics

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## GENERAL DESCRIPTION OF WORK

**Relevance and development of the topic;** The search for crystals with new physical parameters is the result of the development of nanotechnology and optoelectronic technology. The achievements of semiconductor and quantum electronics are related to the search for complex semiconductor materials and the development of high-precision devices based on them, which are suitable for computing, control, automation tools and other production areas. Among semiconductor crystals, layered and chain crystals have a special place, which differ in their sharp anisotropy of physical properties along the crystallographic axes. In recent years, interesting properties have been discovered in the electrical, photoelectric and optical properties of these substances, as well as in their practical applications. However, to date, their potential has not been fully explored.

Triple $TlMC_6{}^2$ thallium chalcogenide-type compounds are quasi-two-dimensional systems with a chain, layered structure. In compounds belonging to this class, their long-period common proportional and non-proportional structures, as well as consistent phase transitions have been identified. Although there are many articles devoted to the study of such crystals, their study remains relevant. With the application of these materials, instrument making, computing and other industries have developed. A number of studies show that these triple-chain and layered crystals are used to make photoelectric converters, spectrum analyzers, strain gages, and X-ray detectors, etc.

It is important to expand the class of layered semiconductors and create new materials based on them. The search for semiconductor materials with predetermined properties, as well as the study of the effect of various impurities on these compounds, is also of interest. For this reason, the acquisition of new crystals obtained with the participation of REEs (rare earth elements), the main component of which is $TlGaS_2(Se_2)$, $TlInS_2(Se_2)$, is of great practical importance in terms of managing the physical parameters of the obtained crystals over a wider range.

**Object and subject of research;** Large homogeneous solid solutions of $TlGa_{1-x}Dy_xSe_2$, $TlGa_{1-x}Er_xSe_2$, $TlIn_{1-x}Yb_xS_2$ (x=0; ÷ 0.03) obtained on the basis $TlGaS_2(Se_2)$, $TlInS_2(Se_2)$ crystals and the effect on the electrical and optical properties of rare earth elements in small concentrations in such compounds.

**Objectives and tasks of the research;** Obtaining new crystals with the presence of rare earth elements (Dy, Er, Yb), the main composition of which is $TlGaS_2(Se_2)$, $TlInS_2(Se_2)$ crystals, determination of the mechanism of current relaxation in them, transfer of charge carriers in constant and variable electric field, as well as is to determine the mechanisms of influence on dielectric and optical properties.

The following issues have been resolved to achieve the set goal:

- Complex study of physical and chemical properties of system alloys $TlA_{1-x}M_xS_2(Se_2)$ (A - In, Ga; M – Dy, Er, Yb; x=0÷0,03) and construction of state diagrams of systems;

- Synthesis and cultivation of single crystals of solid solutions of $TlA_{1-x}M_xS_2(Se_2)$ (A - In, Ga; M – Dy, Er, Yb; x=0÷0,03) system by selecting the optimal mode, determination of technological parameters;

- Investigation of the obtained crystals by X-ray phase (XRPA), differential-thermal (DTA) analysis methods;

- Investigation of the electrical conductivity of crystals of the $TlA_{1-x}M_xS_2(Se_2)$ (A - In, Ga; M – Dy, Er, Yb; x=0÷0,03) system in a constant and variable electric field;

- Investigation of dielectric properties of system crystals $TlA_{1-x}M_xS_2(Se_2)$ (A - In, Ga; M – Dy, Er, Yb; x=0÷0,03);

- Investigation of relaxation processes in crystals of $TlGa_{1-x}Dy_xSe_2$ (x=0,01, 0,03) system;

- Investigation of optical properties of crystals of the system $TlA_{1-x}M_xS_2(Se_2)$ (A - In, Ga; M – Dy, Er, Yb; x=0÷0,03);

**Research methods;** The Bricman-Stockbarger method was used to obtain the crystals. The crystals were tested by DTA and XRPA methods. The dielectric and electrical properties of the obtained crystals were determined by the bridge method using 1920

Precision LCR Meter IET Labs, Inc. (USA) devices in the alternating electric field. Relaxation of current depending on time was measured by determining VAC in a constant electric field.

Optical absorption and emission spectra were determined using UTREX helium creostat and MDR-6 monochromator.

**The main provisions of the defense;**

1. Detection of the presence of $TlA_{1-x}M_xS_2(Se_2)$ (A - In, Ga; M (NTE) – Dy, Er, Yb) compounds based on DTA, XRPA analysis and the effect of rare earth elements on the parameters of these crystals.

2. Regularities of change of dielectric permittivity, tangent of dielectric loss angle under the influence of rare earth elements in the obtained $TlA_{1-x}M_xS_2(Se_2)$ (A - In, Ga; M– Dy, Er, Yb) crystals.

3. The jumping electricalconductivity of the obtained $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) crystals, relaxation processes in $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) crystals, effective mobility of charges due to injection caused by deep traps in contact according to the relay mechanism of charge carrying, transfer of depth centers, contact capacity of samples, area of charges placement in samples, determination of physical parameters such as the time of passage of the carriers passing through the sample.

4. Occurrence of charge carrying in $TlA_{1-x}M_xS_2(Se_2)$ (A - In, Ga; M– Dy, Er, Yb) crystals in the temperature range of 100-300 K with a jump of localized states near the Fermi level.

5. Construction of state diagrams by studying solid solutions of $TlAS_2(Se_2)$-$TlMS_2(Se_2)$ (A-In, Ga; M-REEs) systems solid solutions and determination of the formation of the congruent melting compound in 1:1 ratio of components in the system. Determination of the width of the forbidden band and the position of the exciton peaks due to the energy dependence of the photon of the direct and indirect transitions by analyzing the absorption spectra of the obtained compounds at a temperature of 80-300 K.

**Scientific novelty of the research;**

1. Based on DTA, XRPA analyzes, $TlGa_{1-x}Er_xS_2$ (x=0; 0.001; 0.005, 0.01), $TlIn_{1-x}Yb_xS_2$ (x=0; 0.001; 0.005, 0.01), $TlGa_{1-x}Dy_xSe_2$

(x=0; 0.01; 0.03) compounds was detected and lattice parameters were determined.

2. It was determined that the electrical conductivity of solid solutions $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) is of a jumping nature. The inclusion of dysprosium atoms in the matrix of the $TlGaSe_2$ crystal significantly changes the values of the real and imaginary part of the complex dielectric permittivity, as well as the parameters of the jumping conductivity. On the other hand, the relaxation currents occurring in different electric fields in the crystals of $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03), the hysteresis of the VAC and the accumulation of charges are suitable for charge carriers by the relay mechanism, the location of disconnected charge carriers in the forbidden zone is based on the transfer of energy through deep energy levels.

3. The state diagram of $TlGaSe_2$-$TlErSe_2$ system was constructed and it was determined that up to 6 mol% solid solution was formed on the basis of $TlGaSe_2$ at room temperature. Charge carrying in the temperature range of 100-300 K occurs with a jump of localized conditions near the Fermi level. An increase in temperature and detection by Er ions increases the value of the density of localized states near the Fermi level, decreases the energy scattering $\Delta E$, and changes the value of the concentration of local levels.

4. $TlInS_2$-$TlYbS_2$, $TlInS_2$-$TlErS_2$ systems were studied and state diagrams were constructed and it was determined that the congruent melting compounds $Tl_2InYbS_4$, $TlIn_2ErS_4$ were formed in these systems in the ratio of 1: 1. By analyzing the absorption spectra at a temperature of 80-200 K, the width of the forbidden zone of compounds obtained from the dependence of the direct and indirect transitions on the energy of the photon was determined. Compared to $TlInS_2$, the width of the forbidden zones of these compounds is reduced.

5. The state diagram of $TlGaS_2$-$TlErS_2$ system was constructed and it was determined that $TlErS_2$ is soluble at room temperature up to 4 mol% on the basis of $TlGaS_2$ and $TlGaS_2$ is soluble up to 2 mol% on the basis of $TlErS_2$. The optical absorption spectra of solid solutions $TlGa_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) were studied and it

was determined that at low temperatures x = 0.001, the direct transition exciton is formed.

**Theoretical and practical significance of the research;** Compounds of the triple thallium chalcogenide $TlMC_6^2$ type are quasi-dimensional systems with a layered, chain structure, and the study of electrical and optical properties of small concentrations of rare earth elements in $TlA_{1-x}M_xS_2(Se_2)$ type crystals obtained in the presence of REEs based on them showed that the parameters of these crystals can be easily controlled. It can also be used in instrument making, computing and other industries. The obtained materials can be used as an active element in a wide range of spectral optical radiation detectors, spectrum analyzers, strain gages, X-ray detectors.

The results of the dissertation were published in national and foreign scientific journals, presented at the following conferences and symposiums:

1."Abstracts of 12th European Conference on Solid State Chemistry- ECSSC XII" (Germany, 2009)

2. Scientific and technical progress and modern aviation "(Baku-2009),

3. ."17th International Conference on Ternary and Multinary Compounds- ICTMC 17" (Baku, 2010),

4. "Actual Problems of Physics, VI Republican Scientific Conference" (Baku, 2010); 5. "Scientific Conference of Postgraduate Students of the Azerbaijan National Academy of Sciences" (Baku 2010),

6. Proceedings of the XII International Conference "Opto-nanoelectronics, nanotechnology and microsystems" (Ulyanovsk, 2011),

7. "Scientific Conference of Doctoral Students of the Azerbaijan National Academy of Sciences" (Baku 2011),

8. "International Conference on Nanotechnologies and their application in technology" (Baku 2011),

9. "Labor of the XXII International Scientific and Technical Conference, school of young specialists and installation of photoelectronics and night vision devices." (Moscow, 2012),

10. “International Symposium on Crystal Physics-2013” (Moscow, 2013),

11.“Book of Abstracts of 19th International Conference on Ternary and Multinary Compounds ” (Niigata. Japan, 2014),

12. International conference "Fundamental and applied questions of physics" (Tashkent 2017)

13. Ministry of Education of the Republic of Azerbaijan, Sumgayit State University. “Actual issues of Applied Physics and Energy” (Sumgayit 2018)

14. “International conference and school dedicated to the 100th anniversary of Academician HB Abdullayev” (Baku 2018);

15. Republican Scientific Conference on Actual Issues of Personnel Training in Energy Specialties (Sumgayit 2019),

16. “Modern Materials and Advanced Manufacturing Technology MMAMT-2019” (Saint-Petersburg);

17.VIII International Conference "Deformation and Fracture of Materials and Nanomaterials" (Moscow 2019).

18. Eighth International Conference “Crystal Physics and Deformation Behavior of Advanced Materials” (Moscow 2019).

19. Proceedings of the All-Russian Scientific and Technical Conference with International Participation "Actual Problems of Condensed Matter Physics" (Grozny 2020)

20. Proceedings of the International Conference "Fundamental and Applied Questions of Physics" (Tashkent 2020).

**Name of the organization where the dissertation work is carried out;** The work was performed at the Institute of Physics of Azerbaijan National Academy of Sciences, laboratory “Cystallophysics”.

**Structure and scope of the dissertation.** Dissertation work-172410 marks; It consists of an introduction, four chapters, a conclusion, and references. Introduction - 35732 characters, Chapter I - 48074 characters, Chapter II - 26391 characters, Chapter III - 36166 characters, Chapter IV - 21556 characters, results - 4491 characters. The work is commented on 188 pages and contains 65 figures and 22 tables. The list of cited literature includes 149 works, including the author's personal articles.

## CONTENT OF THE WORK

**The introduction** substantiates the relevance of the topic of the dissertation, indicates the purpose of the work, scientific novelty, practical significance, provides information about the main provisions, the degree of approbation, publications, as well as briefly explains the main content of the work by chapters.

**In the first chapter of** the dissertation the literature on the electrophysical and optical properties of thallium-based layered and chain-type chalcogenides and crystals obtained on the basis of this group of compounds and chemical interactions in systems $A^3B^3C^6_2$ – $A^3B^3C^6_2$ was collected and analyzed.

**Chapter II of the** dissertation describes the method of synthesis and cultivation of single crystals of $TlA_{1-x}M_xS_2(Se_2)$ (A - In, Ga; M – Dy, Er, Yb) systems, describes the methodology for the study of physicochemical and physical properties. Rare earth elements with double and triple thallium chalcogenide are a new class of semiconductor materials that attract the attention of researchers.

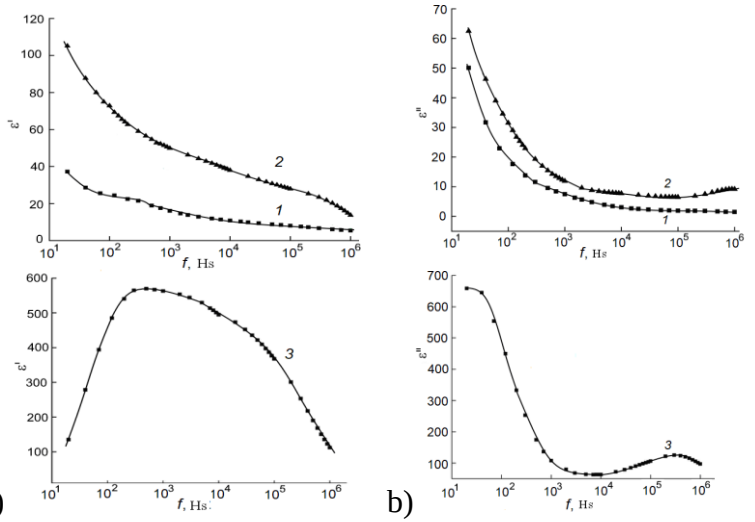
The chemical interactions occurring in the $TlInSe_2$-$TlYbSe_2$ and $TlInS_2$-$TlErS_2$ systems were studied by differential thermal (DTA), X-ray phase (XRPA) analysis methods. It was found that a solid solution of 12 mol% $TlYbSe_2$ is formed on the basis of $TlInS_2$ at room temperature. The non-variant eutectic point corresponds to a composition of $(TlInSe_2)_{0,75}(TlYbSe_2)_{0,25}$ and a temperature of 980 K. In the $TlInSe_2$-$TlYbSe_2$ system, 1:1 ratio of $Tl_2InYbSe_4$ components is formed[1], which has the congruent melting point of 1175K. This compound crystallizes in tetragonal syngonium, parameters a=8.14 Å, c=6,72Å In a solid solution of $(TlInS_2)_{1-x}(TlYbS_2)_x$ in the crystal lattice of $TlInSe_2$ 3 valet In ions are replaced by Yb ions with a large radius.

[1] Seidov, F.M. Investigation of the Interaction of $TlInSe_2$ with $TlYbSe_2$ and the electrical properties of $Tl_2InYbSe_4$ crystals / F.M.Seidov, E.M.Kerimova, N.Z. Gasanov, R.G.Veliyev, K.M. Huseynova, // GESJ: Physics, -2019. No.2(22), ISSN 1512-1461, p.12-17.

Phase equilibrium of $TlGaSe_2$-$TlErSe_2$ and $TlGaS_2$-$TlErS_2$ systems was studied on the basis of DT, XRP, MS studies. In the $TlGaSe_2$-$TlErSe_2$ system, the nonvariant eutectic point corresponds to a composition of $(TlGaSe_2)_{0,85}(TlErSe_2)_{0,15}$ and a temperature of 950K. $TlErSe_2$ on the basis of $TlGaSe_2$ is soluble up to 10 mol% at eutectic temperature. At room temperature, the solubility of $TlErSe_2$ on the basis of $TlGaSe_2$ is 6 mol%. It was determined that up to 6 mol% of solid solution based on TlGaSe2 is formed at room temperature. X-ray studies revealed that $TlGaSe_2$<Er> single crystals have a monoclinic structure and the following values were obtained for the lattice parameters: a=10,744, b=10,773Å, c=15,623Å, □=100,04º, z=16. It has been shown that the small amount of Er impurity does not affect the lattice parameters.

**In the third chapter of** the dissertation the frequency and temperature dependences of electrical conductivity and dielectric properties of In the third chapter of the dissertation the frequency and temperature dependences of electrical conductivity and dielectric properties of $TlA_{1-x}M_xS_2$ (A - In, Ga; M (REEs) - Dy, Er, Yb) single crystals are presented. The relaxation processes of these crystals in a constant electric field are also described. It is known from the literature that $TlGaSe_2$ and $TlInS_2$ single crystals can be used as an active element in various semiconductor devices due to their layered structure and physical anisotropy. Thus, despite the extensive study of the $TlGaSe_2$ crystal, $TlGaSe_2$ samples containing a rare earth element have been little studied. Based on this, we can expect a change in the physical properties of the $TlGaSe_2$ crystal, which is replaced by rare earth elements.

The effect of the increase in the concentration of Dy on the dielectric characteristics of solid solutions $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) and the mechanism of action of the transfer mechanism in the radio frequency range of the alternating electric field have been studied. The frequency dependence of the true part of the complex dielectric permittivity (έ) of samples $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) was studied (Figure 1,a).

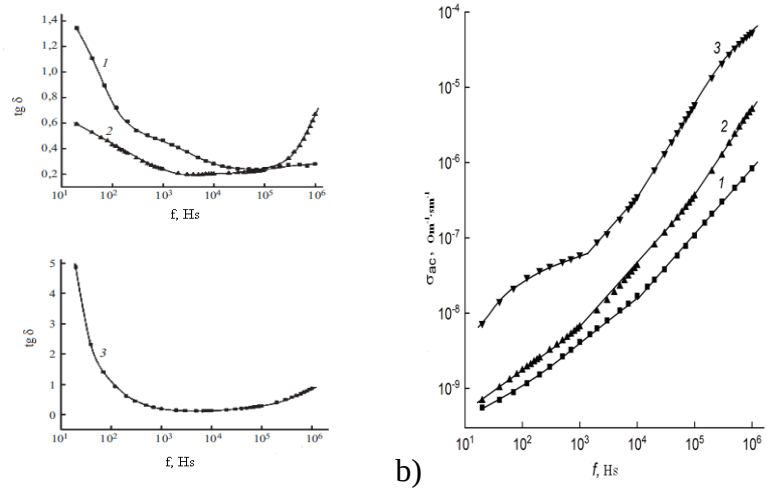



a) b)

Figure 1. Frequency dependence of the real (a) and imaginary(b) part of the dielectric permittivity of $TlGa_{1-x}Dy_xSe_2$ $x$ = 0 (*1*); 0.01 (*2*) and 0.03 (*3*) crystals at temperature T = 298 K.

It has been shown that for the $TlGaSe_2$ and $TlGa_{1-x}Dy_xSe_2$ (x=0.01) samples, a decrease in the value of the true part of the dielectric permittivity (έ) is observed in the entire measured frequency range. In the example of $TlGa_{1-x}Dy_xSe_2$ (x=0.03), έ increases in the frequency range of 20-500 Hz, and then sharply decreases to a value of 1 MHz. An increase in Dy in crystals significantly increases the value of the true part of the dielectric permittivity (έ).

The frequency dependence of the imaginary part of the complex dielectric permittivity (ε") in solid solutions of $TlGa_{1-x}Dy_xSe_2$ has also been found to be relaxation (Figure 1, b). This dependence is characterized by a hyperbolic decrease, which is associated with a loss of conductivity. At high frequencies, the value of the dielectric loss angle tangent (tgδ) tends to increase, which indicates the presence of relaxation losses. The inclusion of a Dy atom in the $TlGaSe_2$ crystal causes a significant increase (Figure 2,a).




a) b)

Figure 2. Frequency dependence of the tangent angle of dielectric loss (a) and conductivity (b) in $TlGa_{1-x}Dy_xSe_2$ $x = 0$ (*1*) x=0.01 (2) and 0.03 (3) crystals at temperature T = 298 K.

In this study, we also studied the frequency dependence of the AC conductivity of solid solutions $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03)[2] (Figure 2, b). For $TlGa_{1-x}Dy_xSe_2$ solid solutions, the $\sigma_{ac}$ value is higher than for $TlGaSe_2$ crystals. For the $TlGaSe_2$ crystal, two parts were observed in the $\sigma_{ac}(f)$ dependence. In the first state, the dependence $\sigma_{ac} \sim f^{0.6}$ was observed, and then increased by ($f \geq 10^4$) $\sigma_{ac} \sim f^{0.8}$. For solid solutions of $TlGa_{1-x}Dy_xSe_2$, three parts were observed in the $\sigma_{ac}(f)$ dependence. Initially, $\sigma_{ac} \sim f^{0.5\text{-}0.6}$ , which was later replaced by $\sigma_{ac} \sim f^{0.8}$. As the frequency value increased to 1 MHz, $\sigma_{ac} \sim f^{1,2}$ was replaced by the superlinear part. Thus, it is shown that the conductivity obeys the law $\sigma_{ac} \sim f^{0.8}$, which indicates that the charge carriers change with a jump near the Fermi level. The parameters of $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) solid solutions were calculated within the Mott approximation. Experimental values of $\sigma_{ac}$ (f) for solid solutions $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) were calculated

[2] Mustafaeva, S.N., Guseynova, K.M., Asadov, M.M. Dielectric parameters of (1–x)$TlGaSe_2$□xDy single crystals in AC electric fields // Metal Science and Heat Treatment, -2020. V.62, No 1-2, -p.30-34.

according to Mott's theory of density of localized states ($N_F$) near Fermi levels (Table 1).

Table 1.

Parameters calculated for $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) solid solutions within the Mott approximation.

| $TlGa_{1-x}Dy_xSe_2$ | $\Box f$, hs | $N_F$, $10^{18}$ $eV^{-1}sm^{-3}$ | τ, s | $R$, Å | $\Delta E$, eV |
|---|---|---|---|---|---|
| $x = 0$ | $10^4$–$10^6$ | 1.98 | $10^{-6}$ | 234 | $1.9\times10^{-2}$ |
| $x = 0.01$ | $10^3$–$10^5$ | 2.97 | $10^{-5}$ | 273 | $8\times10^{-3}$ |
| $x = 0.03$ | $10^3$–$10^4$ | 7.14 | $10^{-4}$ | 312 | $2.2\times10^{-3}$ |

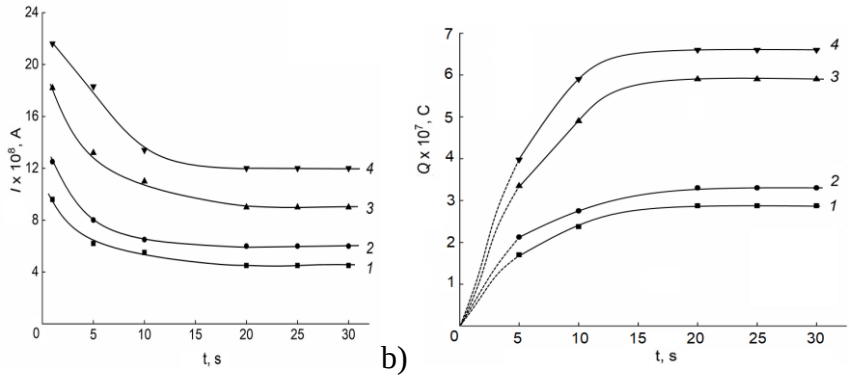
As the concentration of Dy in solid solutions of $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) increases, the density of localized states near the Fermi level increases. According to the theory of bounce conduction in solid solutions $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03), the value of the bounce distance (R) is about 8-9 times greater than the average distance between localized charge carriers. In solid solutions of $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03), changes in the density of localized energy levels near the Fermi level were evaluated[3] and it was shown that, as the concentration of Dy increases, the energy level $\Box E$ of solid solutions of $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) narrows, the density of localized states near the Fermi level increases, the average distance and time spent increase. Thus, it was found that the inclusion of Dy in the matrix of the $TlGaSe_2$ crystal modifies its physical properties.

Relaxation phenomena of layered single crystals $TlGa_{1-x}Dy_xSe_2$ (x=0,01; 0,03) were studied and the mechanism of current flow in the sample in Ag-$TlGa_{1-x}Dy_xSe_2$-Ag system was clarified. It was found that relaxation processes occur in a constant electric field at a temperature of T = 85 K due to significantly accumulated

[3] Mustafaeva, S.N., Huseynova, K.M., Asadov, M.M. Frequency and temperature dependences of the physical properties of (1-x)$TlGaSe_2$ · xDy solid solutions // Applied Solid State Chemistry, -2018. **№** 2, -p. 59 – 64.

charges on the $TlGa_{1-x}Dy_xSe_2$ crystal. In the voltage range of 100-250 V (Figure 3, a) , the current decreases with time, then becomes stationary. This is also observed with the hysteresis curve.

The VAC of the samples Ag-$TlGa_{1-x}Dy_xSe_2$-Ag (x=0,01; 0,03) is given (Figure 3). Two areas are observed here: 1~F and $J \sim F^2$ the linear and the quadratic region. This is due to the reduction of current relaxation due to the accumulation of charges. When the external voltage is eliminated and the contacts are short-circuited, a discharge current flows in the circuit in the opposite direction of the external voltage current. The amount of charges collected was also calculated here (Figure 3, b). The thickness of the accumulated charge was determined by knowing the contact capacity. (x = 0,01; 0,03) Relaxation currents in different electric fields, hysteresis of VAC and the charge accumulation correspond to the relay mechanism of cargo transportation, where the disconnected charge carriers from these contacts are based on conduction through deep energy levels located in the forbidden zone.




a) b)

Figure 3. Dark current relaxation in Ag–$TlGa_{0.99}Dy_{0.01}Se_2$–Ag single crystals (I) (a - L=10 mkm ) *U*, V: *1* – 100; *2* – 150; *3* – 200; *4* – 250**.** *T* = 85 K. b) time dependence of collected charges

The physical parameters characterizing the electron poses flowing in the studied crystals were determined[4].

$$\mu_f = \frac{L^3 I_c}{U^2 C_c d_c}\ ,\ \tau = \frac{L^3}{\mu_f d_c U},\quad t_t = \frac{\tau \cdot d_c}{L}$$

Using these formulas, the effective mobility for $TlGa_{0,99}Dy_{0,01}Se_2$ at 85 K is $\mu_f=2{,}7\cdot10^{-8}$ $sm^2$/ Vsan, and for $TlGa_{0,97}Dy_{0,03}Se_2$ it is $\mu_f=8{,}8\cdot10^{-8}$ $sm^2$/Vsan. The contact capacity of the sample is $C_c=2{,}4\cdot10^{-9}$ F, the area of charge accumulation in the crystal is $d_c=6{,}7\cdot10^{-5}$ cm, the contact charging constant is $\tau$=5.5 s, the time of the charges passing through the sample is $t_t$=0,37 s has been identified.

Dielectric and electrical conductivity of solid solutions $TlGa_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) were studied by the resonance method. The frequency dependence of the dielectric permittivity of $TlGa_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) solid solutions at room temperature has been studied, as the frequency of the measuring field increases, a decrease in the value of the true part of the dielectric permittivity (ε') is observed. The true part of the total complex dielectric permittivity (ε') of all studied solid solutions of $TlGa_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) is characterized by weak dispersion at all frequencies studied.

In solid solutions $TlGa_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) the value of ε' decreases as the rate of cation substitution increases. This is due to the decrease in the polarization of the $Er^{3+}$ replacement crystal in the $Ga^{3+}$ cation field due to the low electronegativity of $Er^{3+}$ (1.24) compared to $Ga^{3+}$ (1.81). This is due to the location of Er atoms in Van-der Vaals gaps between the $Ga_4S_{10}$ layers in the $TlGaS_2$ lattice.

Comparative analysis of the values of $TlGa_{1-x}Er_xS_2$ x=0 and 0.001 shows that the values of the imaginary part (ε") and the tangent of the dielectric loss angle change approximately one order of

[4] Мустафаева, С.Н., Гусейнова, К.М., Асадов, М.М. Релаксац ия тока в монокристаллах $TlGa_{1-x}Dy_xSe_2$ (x=0.01; 0.03) // ФТТ, -2020. т.62, в.7, -с.1022-1027

magnitude, in contrast to the real part of the dielectric permittivity (ε') and at frequencies $5\times10^4 \div 3.5\times10^7$ Hz. significant dielectric dispersion is observed. The increase in the amount of erbium in solid solutions $TlGa_{1-x}Er_xS_2$ has a significant effect on the dielectric properties at $f = 5\times10^4$ Hz: in the crystal $TlGaS_2$ at frequency $f = 5 \times 10^4$ Hz, ε ' ~23.5 and in solid solution $TlGa_{0,99}Er_{0,01}S_2$, ε' ~3.6. In other words, as the amount of erbium increases, ε' decreases by about 6.5 times, and tgδ increases in the range of 0.017 ÷ 0.184. In solid solutions of $TlGa_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) the value of the density of localized states near the Fermi level is $N_F=5,9\cdot10^{18} \div 1,1\cdot10^{19} eV^{-1}sm^{-3}$, the average value of transition time from one localized state to another. was $\tau=5\cdot10^{-8} \div 10^{-7}$s, the duration of the jump between localized states was R=77÷81 Å, the energy scattering of localized states was ΔE=0,11÷0,15 eV, and the concentration of localized states responsible for AC conductivity $N_t = 8.8 \times 10^{17} \div 1\times10^{18}$ $sm^{-3}$.

The dielectric properties of solid solutions $TlGa_{1-x}Er_xSe_2$ (x=0, 0.001, 0.005, 0.05, 0.01) and the charge carrying process in a alternating electric field were studied. A decrease was observed for the samples studied in the entire measured frequency range (έ). In $TlGa_{1-x}Er_xSe_2$ (x=0; 0,001; 0,005; 0,05, 0,01) crystals, the frequency increases with increasing έ in the range of 20 ÷ 500 Hz, then decreases sharply to 1 MHz. An increase in erbium in crystals significantly increases the value of the true part of the dielectric permittivity (έ)

The frequency dependence of the AC conductivity of solid solutions of $TlGa_{1-x}Er_xSe_2$ (x=0; 0,001; 0,005; 0,05, 0,01) was also studied. It was found that the conductivity in solid solutions $TlGa_{1-x}Er_xSe_2$ in the frequency range $6\cdot10^6 \div 3,5\cdot10^7$ Hz also occurs on the basis of the mechanism of bounced charge carrying at localized levels near Fermi levels. It was found that in solid solutions based on $TlGaSe_2$, depending on the amount of Er, the concentration of localized states and their energy scattering increases, but the conductivity, dielectric permittivity, time spent on jumps and the distance between jumps decreases.

This chapter also presents the results of the study of the temperature dependence of the electrical conductivity of the $TlErSe_2$ single crystal. It was found that an increase in temperature from 111 K to room temperature leads to ≈ 5 fold increase in the conductivity of the $TlErSe_2$ crystal. The temperature dependence of the electrical conductivity of the $TlErSe_2$ single crystal was performed for various samples. As the temperature increases, the value of electrical conductivity increases and σ(T) is semiconductor for the $TlErSe_2$ crystal. The reason for the exponential increase in electrical conductivity at high temperatures is due to the formation of specific conductivity. The width of the forbidden zone of the $TlErSe_2$ crystal was determined from the slope of the curves $lg\sigma=f(10^3/T)$ and $E_g$= 1.40eV was obtained.

In the third chapter of the dissertation, the dielectric properties of solid solutions of $TlGa_{1-x}Er_xSe_2$ (x=0; 0,005) were also studied at different frequencies in the temperature range of 150-300 K. It was found that the value of dielectric permittivity (ε) for a solid solution of $TlGa_{0,995}Er_{0,005}Se_2$ increases with increasing temperature. As with the dielectric permittivity in a solid solution of $TlGa_{0,995}Er_{0,005}Se_2$, the value of electrical conductivity increases with increasing temperature. The absolute value of electrical conductivity depends on the frequency of the measuring field, which is more pronounced at low temperatures. As the value of electrical conductivity increases with frequency, several orders of magnitude increase. Conductivity at low temperatures is practically independent of temperature. In the high temperature region, the conductivity increases exponentially with the law $\sigma\sim e^{1/T}$. This shows that the electrical conductivity at high temperatures depends mainly on the concentration of the main charge carriers. The values of the tangent of the dielectric loss angle of a solid solution of $TlGa_{0,995}Er_{0,005}Se_2$ increase with increasing temperature compared to the crystal of $TlGaSe_2$. This increase can be attributed to the increase in the concentration of free charge carriers.

It was found that the value of the dielectric constant of solid solutions of $TlGa_{1-x}Er_xSe_2$ decreases with increasing frequency. Such a course of dielectric constant in the studied solid solutions is due to

the presence of relaxation polarization at low frequencies and causes dielectric loss at low frequencies. The tgδ(T) curves of the dielectric loss angle show the maximums of semiconductors, and the maximum spreads slightly as the frequency of the measuring field increases from 1 kHz to 1 MHz. Which are located in the temperature range, where the value of the dielectric constant increases sharply.

For $TlGaSe_2$ and $TlGa_{0,995}Er_{0,005}Se_2$ crystals, a decrease is observed in the frequency dependence of the true part (ε`) of the complex dielectric permittivity of $TlGa_{1-x}Er_xSe_2$ (x=0; 0,005) solid solutions at different temperatures. The increase in temperature, as well as the impurity of Er, significantly increases the value of the true part of the dielectric permittivity (ε`).

The frequency dependence of the AC conductivity of $TlGa_{1-x}Er_xSe_2$ (x=0; 0,005) crystals was studied. For $TlGa_{0,995}Er_{0,005}Se_2$ solid solution, the $\sigma_{ac}$ value is higher compared to $TlGaSe_2$ crystal. There are three parts to the $\sigma_{ac}(f)$ dependence curves of a solid solution of $TlGa_{1-x}Er_xSe_2$ (x=0; 0,005): in the first case $\sigma_{ac} \sim f^{0.6}$ was observed, and then $\sigma_{ac}(f)$ increased by $\sigma_{ac} \sim f^{0.8}$ ($10^2 \div 10^4$ Hz). . When the frequency value increased to 1 MHz, $\sigma_{ac} \sim f$ was replaced by 1.2. The obeying of the law $\sigma_{ac} \sim f^{0.8}$ obtained by us indicates the occurrence of a jumping conductivity of localization states near the Fermi level.

$\sigma_{ac} \sim f^{0.8}$ dependence is observed in the crystals studied in the frequency range $10^2$-$10^4$ Hz. For solid solutions of $TlGa_{1-x}Er_xSe_2$ (x=0; 0,005), according to Mott's theory, the density of localized states near the Fermi level ($N_F$), the length of the jump (R), and the energy scattering ΔE of localized states near the Fermi level were calculated. The increase in temperature and the impurity of erbium increase the density of localized states near the Fermi level, narrow the energy scattering $\square E^5$, and change the value of the concentration of local levels. Thus, it was found that when erbium is added to a $TlGaSe_2$ crystal, its physical properties are modified.

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[5] Hüseynova, K.M. $TlGa_{1-x}Er_xSe_2$ (x=0; 0,005) kristallarının elektrik və dielektrik xarakteristikaları // AMEA Xəbərlər, fizika və astronomiya, -2019. cild XXXIX, №5. -S.155-159.

**Chapter IV of** the dissertation presents the results of the study of the optical properties of single crystals $TlA_{1-x}M_xS_2$ (A - In, Ga; M (REEs) – Dy, Yb, Er).

The results of the study of the optical properties of solid solutions obtained on the basis of the system $TlGaSe_2$-$TlErSe_2$ are given. The temperature dependence of the forbidden zone of the solid solution $TlGa_{1-x}Er_xSe_2$ (x=0; 0,001) in the temperature range 77-300K was studied. By analyzing the absorption spectra of a solid solution of $TlGa_{1-x}Er_xSe_2$ (x=0; 0,001), the energy of the direct transitions from the energy dependence of $(\alpha\hbar\omega)^2$ (α-absorption coefficient) on the photon (ħω) was determined and the width of the forbidden band ($E_g$) of these crystals was calculated. A comparison of the absorption boundary of $TlGaSe_2$ and $TlGa_{0,999}Er_{0,001}Se_2$ crystals revealed that an exciton band was observed near the direct transition boundary in the solid solution of $TlGa_{0,999}Er_{0,001}Se_2$ at low temperatures. In the temperature range 77-140K, the temperature coefficient of the forbidden zone of the $TlGaSe_2$ crystal is $(-3{,}1\cdot10^{-4}$eV/K), for a solid solution of $TlGa_{0,999}Er_{0,001}Se_2$, this value $(-6{,}1\cdot10^{-4}$eV/K) is twice as different; The value of the absorption coefficient of solid solutions $TlGa_{1-x}Er_xSe_2$ is significantly higher than that of $TlGaSe_2$.

In solid solutions $TlGa_{1-x}Er_xSe_2$ (x=0; 0,001), $dE_g/dT=2{,}1\cdot10^{-4}$eV/K in the temperature range of 140-300K. The shift of the forbidden zone to the long-wavelength region is 60 meV in a solid solution of $TlGa_{0,999}Er_{0,001}Se_2$ relative to $TlGaSe_2$. At a temperature of 77K, the width of the forbidden band for the $TlGaSe_2$ crystal is $E_g$=2,192 eV, and for the solid solution $TlGa_{0,999}Er_{0,001}Se_2$, it is $E_g$=2.132 eV. It can be concluded that when the Ga atoms in the $TlGaSe_2$ single crystal are replaced by 0.001% Er atoms, the width of the forbidden band decreases.

This chapter also studies the optical absorption spectra of $TlGa_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) crystals.

It is known that the absorption limit of $TlGaS_2$ single crystal is formed by the direct exciton zone at temperatures up to 180 ÷ 200K. In addition, the temperature coefficient of the forbidden zone, which is rare in $TlGaS_2$ semiconductors, has a positive sign. All this has

aroused interest in the study of the optical absorption limit of solid solutions $TlGa_{1-x}Er_xS_2$ (x=0÷0,01) obtained on the basis of $TlGaS_2$ semiconductor compounds. Studies show that there is a difference in the structure of the absorption boundary of solid solutions of $TlGa_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01): at low temperatures the absorption spectrum is observed with the formation of an exciton belonging to the direct transition at x = 0.001; This is not observed in $TlGa_{0,995}Er_{0,005}S_2$ and $TlGa_{0,99}Er_{0,01}S_2$ crystals.

We also investigated the condition of the exciton peak in the temperature range 77 ÷ 200K for $TlGa_{0,999}Er_{0,001}S_2$. For a given composition, the positive sign of the temperature coefficient of maximum of the exciton band is maintained, and if we take into account that the binding energy of the exciton is very weakly dependent on temperature, it can be concluded that, the width of the forbidden zone of the $TlGa_{0,999}Er_{0,001}S_2$ compound increases with increasing temperature. It should be noted that the displacement of the exciton peak to the short-wave region is large due to the 0.1% replacement of the Ga atom in the $TlGaS_2$ single crystal with Er atoms, a rare earth element, which is 90 meV. In this case, the temperature coefficient of the exciton peak increases significantly ($1{,}9\cdot10^{-4}$eV/K for $TlGaS_2$, and $3{,}1\cdot10^{-4}$eV/K for $TlGa_{0,999}Er_{0,001}S_2$).

The results of the study of the optical properties of solid solutions $TlIn_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) are given[6]. It was shown that the absorption limits of $TlInS_2$ and $TlIn_{1-x}Er_xS_2$ (x=0,001; 0,005; 0,01) single crystals were similar and formed in the direct transitions with the formation of excitons. It has been shown that the concentration of erbium atoms affects the energy state of the transitions. Thus, at T=120K, the exciton peak corresponding to (n=1) for $TlIn_{0,99}Er_{0,01}S_2$ shifted 25 meV toward the low-energy region relative to $TlInS_2$. Due to the dependence of the optical

[6]Gasanov, N.Z., Kerimova, E.M., Seidov, F.M., Asadov, Yu.G., Huseynova, K.M. $TlIn_{1-x}Er_xS_2$ (x=0; 0,01) Solid Solutions and Their Optical Properties // Book of Abstracts of 19th International Conference on Ternary and Multinary Compounds (ICTMC-19). Niigata. Japan. September 1-5, -2014, P1-003, P.76.

absorption coefficient (□) at 120K on the energy of the photons, the width of the forbidden band for a solid solution has been determined. A study of the temperature dependence of the absorption coefficient in the range of 80–200K showed that for all the studied components, the exciton peak shifts toward the longitudinal region as the temperature increases. From the temperature dependence of the maximum corresponding to the exciton band of the $TlIn_{1-x}Er_xS_2$ single crystal, it was shown that the exciton peak above 200K becomes the stage and then disappears. In the $TlInS_2$ crystal, the mean temperature shift of the forbidden zone was $\partial Eg/\partial T = 5{,}6\cdot 10^{-4} eV/K$ in the range of 80-240 K. As a result, the temperature coefficient decreases in part due to the replacement of indium atoms with erbium atoms in the $TlInS_2$ crystal, for example, $\partial Eg/\partial T = 5{,}4\cdot 10^{-4} eV/K$ is for $TlIn_{0,99}Er_{0,01}S_2$.

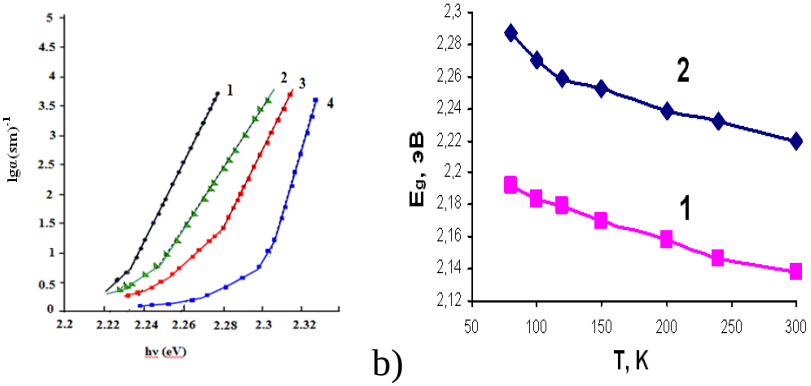
The results of the study of the optical absorption spectrum of solid solutions $TlIn_{1-x}Yb_xS_2$ (x=0; 0,001; 0,005; 0,01) in the temperature range 77 ÷ 300K are given in this chapter[7]. It has been shown that as the temperature decreases, the absorption boundary shifts to the higher energy side, and an absorption band is observed due to the transition of electrons directly to the exciton levels at the specific absorption boundary in the temperature range 77□200 K. The energy of the exciton peaks was determined and based on this, the temperature coefficient of sliding of the exciton in the temperature range 77□200 K was calculated. This ratio is negative, as observed in the $TlInS_2$ crystal. As a result of the replacement of In ions in the $TlInS_2$ crystal with Yb ions, a rare earth element, the temperature coefficient of the exciton peak shift varies compared to the main crystal matrix: so that, for the $TlInS_2$ crystal in the range of 77□200 K, $dE^{ek}/dT=-5{,}1\cdot 10^{-4}$eV/K, while for the solid solution $TlIn_{0,995}Yb_{0,005}S_2$, $dE^{ek}/dT=-2{,}0\cdot 10^{-4}$eV/K which is also significantly

[7] Hüseynova, K.M. $TlInS_2$-$TlYbS_2$ sisteminin fiziki-kimyəvi analizi $TlIn_{1-x}Yb_xS_2$ monokristallarinin alınması və optik xassələr // Azərbaycan Milli Elmlər Akademiyası Aspirantlarınıın Elmi Konfransı, Bakı: – 2010. -s. 23-26

less. The width of forbidden band the solid solution $TlIn_{0,995}Yb_{0,005}S_2$ decreased by 50meV compared to the $TlInS_2$ crystal: $E_g$=2,590eV for the $TlInS_2$ crystal at 77K, while for the solid solution $TlIn_{0,995}Yb_{0,005}S_2$ it is 2.540. Given that the binding energy of the exciton varies slightly, but the replacement of In atoms by 0.5% with Yb atoms, a rare earth element, results in a significant reduction in the width of the forbidden band.

This chapter also presents the results of the study of the optical properties of $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01) solid solutions in the temperature range of 77-300K. For $TlGaSe_2$ and $TlGa_{0,99}Dy_{0,01}Se_2$ single crystals, the width of forbidden band ($E_g$) of the straight line part from the (hν) dependence curve of the energy of the incident ray $(\alpha h\nu)^2$ was found by extrapolation (Figure 4, a).

Exciton formation is not observed near the absorption limit in solid solution $TlGa_{0,99}Dy_{0,01}Se_2$ at low temperatures. In the range of 80÷120K, the temperature coefficient $dE_g/dT$ is □ $-3\cdot10^{-4}$ eV/K for the $TlGaSe_2$ crystal, and □ $-7\cdot10^{-4}$ eV/K for the solid solution $TlGa_{0,99}Dy_{0,01}Se_2$, ie it differs by about 2 times.




a) b)

Figure 4. a ) The dependence of the absorption coefficient on the photon energy at different temperatures for a solid solution of $TlGa_{0,99}Dy_{0,01}Se_2$. 1- T=300 K, 2- T=240 K, 3-180 K, 4- T=83 K. b) Temperature dependence of the forbidden zone of $TlGaSe_2$ (1) and $TlGa_{0,99}Dy_{0,01}Se_2$ (2).

For a solid solution of $TlGa_{0.99}Dy_{0.01}Se_2$, the displacement of the width of forbidden band to the short-wave side is approximately 90 meV relative to the $TlGaSe_2$ crystal.

Thus, at a temperature of 80 K, $E_g$ = 2.192 eV for $TlGaSe_2$ crystal, and $E_g$ = 2.287eV for a solid solution of $TlGa_{0.99}Dy_{0.01}Se_2$. It has been found that the replacement of 1-2% gallium atoms in the $TlGaSe_2$ crystal with Dy atoms significantly increases the width of the band gap (Figure 4,b).

## Result

1. The presence of $TlDySe_2$, $TlErSe_2$, $TlYbS_2$, $TlTmS_2$ compounds was detected on the basis of DT, XRP and MS analyzes $TlGaSe_2$-$TlErSe_2$, $TlGaSe_2$-$TlDySe_2$, $TlInS_2$-$TlYbS_2$ the state diagram was constructed by synthesizing systems, physical and chemical analyzes were performed. Large-scale homogeneous solid solutions of $TlGa_{1-x}Dy_xSe_2$, $TlGa_{1-x}Er_xSe_2$ , $TlIn_{1-x}Yb_xS_2$ ( x=0; 0.01; 0.03) were grown by the Bricman-Stockbarger method and their lattice parameters were determined.

2. It was determined from the change of dielectric coefficient and conductivity of $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03) solid solutions in the frequency range f=20-$10^6$ Hz depending on the composition that the electrical conductivity is jumping. The inclusion of Dy in the matrix of the $TlGaSe_2$ crystal significantly changes the real and imaginary part of the complex dielectric permittivity, increases the value of the density of localized states near the Fermi level (1.98–7.14) × $10^{18}$ $eV^{-1}$ $cm^{-3}$, causes an increase the average distance of jumps and time between jumps.

3. By studying the relaxation phenomena in solid solutions $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01; 0,03), the mechanism of current flow of Ag-$TlGa_{1-x}Dy_xSe_2$-Ag system samples was clarified, it was found that relaxation processes occur in a constant electric field at T = 85 K due to significantly accumulated loads on the $TlGa_{1-x}Dy_xSe_2$ crystal. $TlGa_{(1-x)}Dy_xSe_2$ determined the relaxation currents in different electric fields, the hysteresis of VAC and the physical parameters characterizing the electronic processes in the crystal: The effective mobility of the carrying charge through the deep centers at 85 K is $\mu_f$=2,7·$10^{-8}$ $sm^2$/ Vsan for $TlGa_{0,99}Dy_{0,01}Se_2$ and $\mu_f$=8,8·$10^{-8}$ $sm^2$/Vsan

for $TlGa_{0,97}Dy_{0,03}Se_2$. The contact capacity of the sample is $C_c=2,4\cdot10^{-9}$ F, the area of charge accumulation in the crystal is $d_c=6,7\cdot10^{-5}$ cm, the contact charging constant is $\tau=5,5$ s, the time of charges passing through the sample is $t_t=0,37$ s has been identified.

4. From the change of dielectric coefficient and conductivity of $TlGa_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) solid solutions in the frequency range f=20-$10^6$ Hz depending on the composition, it was determined that the amount of erbium at the frequency $f = 5 \times 10^4$ Hz significantly affects the dielectric properties: so that the value of the true part of the dielectric permittivity (ε') decreases by about 6.5 times, while the value of the tangent of the dielectric loss angle (tgδ) increases from 0.017 to 0.184 .

5. In solid solutions of $TlGa_{1-x}Er_xSe_2$ (x=0; 0,001; 0,005; 0,05; 0,01) charge carrying in the temperature range of 100-300 K occurs with a jump of localized states near the Fermi level. The increase in temperature and the impurity of Er ions increase the value of the density of localized states ($N_F$) near the Fermi level, narrow the energy scattering ($\Delta E$), and change the value of the concentration of local levels ($N_t$). When erbium is added to $TlGaSe_2$ crystal, its physical properties are modified (25). By analyzing the absorption spectra of solid solutions of $TlGa_{1-x}Er_xSe_2$ (x=0; 0,001) in the temperature range 77-300K, the value of the width of the forbidden band was determined by the dependence of the direct and indirect transitions on the energy of the photon. It was found that when the Ga atoms in the $TlGaSe_2$ crystal under study were replaced by Er atoms, the width of the forbidden zone of the single crystal decreased.

6. From the analysis of the optical absorption spectra of solid solutions $TlGa_{1-x}Er_xS_2$ (x=0; 0,001;0,005; 0,01), it was determined that at low temperatures an exciton belonging to the direct transition with the composition x = 0.001 is formed. In this case, the temperature coefficient $\partial E_{eks}/\partial T$ remains positive, as in $TlGaS_2$. However, the exciton peak shifts to the shortwave region. This is due to the replacement of the Ga atom in the $TlGaS_2$ single crystal with Er atoms, a rare earth element.

7. By analyzing the absorption spectra of solid solutions $TlIn_{1-x}Yb_xS_2$, $TlIn_{1-x}Er_xS_2$ (x=0; 0,001; 0,005; 0,01) at a temperature of 77-300K, the value of the width of the forbidden band is determined by the dependence of the photon energy on the direct and indirect transitions. It has been shown that as the temperature decreases, the absorption boundary shifts towards the higher energy, and at the temperature 77□200 K, an absorption band is observed due to the transition of electrons directly to the exciton levels at the specific absorption boundary. The width of forbidden band the $TlIn_{0,995}Yb_{0,005}S_2$ compound decreased by 50 meV compared to $TlInS_2$. This means that in $TlInS_2$ there is a structural change in the zone structure in the substitution of In-Yb, Er .

8. By analyzing the absorption spectra of $TlGa_{1-x}Dy_xSe_2$ (x=0; 0,01) crystals in the temperature range 77 ÷ 300K, the value of the width of the forbidden band was determined due to the dependence of the direct and indirect transitions on the energy of the photon. While $E_g$ = 2.192 eV eV in the $TlGaSe_2$ crystal, it is 2.287eV in the solid solution of $TlGa_{0.99}Dy_{0.01}Se_2$. That is, the replacement of 1-2% gallium atoms with Dy atoms significantly increases the width of the forbidden band for $TlGaSe_2$ crystals.

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