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CONDUCTIVITY OVER LOCALIZED STATES OF THE SYSTEM OF (TlInSe₂)_{1-x}(TlGaTe₂)_x SOLID SOLUTIONS EXPOSE γ -IRRADIATION

R.M. Sardarly, O.A. Samedov, N.A. Aliyeva, A.P. Abdullayev,
F.T. Salmanov, A.A. Orucova

Institute of Radiation Problems, of ANAS

sardarli@yahoo.com

Abstract: The temperature dependences of electrical conductivity over localized states in the solubility region of mixed crystals of compositions (TlInSe₂)_{1-x}(TlGaTe₂)_x at $x = 0, 0.1, 0.2, 0.8, 0.9,$ and 1.0 are studied. It is established that hopping conductivity with a variable hop length over localized states near the Fermi level occurs in this system. The conductivity activation energy is determined; and the density of states in the vicinity of the Fermi level, their dispersion, localization radius, and average carrier hop distance for all compositions are estimated; and the concentration dependences of the calculated parameters are plotted.

Keywords: hopping conductivity, localization radius, Fermi level.

1. Introduction

The peculiarity of the development of modern nanoelectronics and microelectronics is the tendency to use fundamentally new physical phenomena based on the metaproperties of these materials associated with the low-dimensional character of this structure as well as with nanodimensional effects. Metaproperties are observed mainly in disordered heterogeneous systems, in composite materials with mesoscopic structures, in solid solutions with violated translational order, etc.

The peculiarity of the solid-solution structures is the fact that disorder appears in these systems while maintaining ideal crystallographic symmetry due to violations of the periodicity in filling sites of the crystalline matrix (translational disorder), in the orientation of electron spins (spin disorder), and when some crystalline sublattices turn out to be disordered and others remain ordered (site disorder), etc.

Semiconductor crystals TlGaTe₂ and TlInSe₂ belong to the class of compounds of Group A^{III}B^{III}C₂^{VI}, which crystallize in the tetragonal space group (structural type TlSe). Previously [1–8], we studied the temperature dependences of the electrical conductivity $\sigma(T)$ and permittivity $\varepsilon(T)$ for TlInSe₂ and TlGaTe₂ crystals. We found switching and memory effects, superionic conductivity, the presence of giant dielectric relaxation, and investigated the complex impedance spectra. It is shown that the electrical properties of TlInSe₂ in the region of ionic conductivity are determined by the hops of thallium ions and the frequency dependences of the real and imaginary parts of the complex permittivity (Cole–Cole curves) were investigated. Along with great scientific interest in crystals of the A^{III}B^{III}C₂^{VI} – family associated with structural features and instability of the crystal lattice to external effects, these materials turn out to be promising objects when fabricating photoelectric converters, tensor resistors, and detectors of X-ray and neutron radiation [9].

In this article, we present the results of studying the electrical conductivity of (TlInSe₂)_{1-x}(TlGaTe₂)_x solid solutions of compositions $x = 0, 0.1, 0.2, 0.8, 0.9,$ and 1.0 in the temperature range 90–300 K. According to our thermo graphic studies, the system forms a continuous series of solid solutions at these compositions; as for compositions $0.2 < x < 0.8,$ a eutectic-type solution

with only one melting point, at which the solidus and liquidus lines overlap, is observed. The target of these studies is to investigate the features of conductivity of the $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ system at $x = 0, 0.1, 0.2, 0.8, 0.9, 1.0$ (in the solubility region), as well as to perform analysis of the conductivity in terms of the Mott approximation [10, 11].

2. Experimental

The samples of the $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ solid solution ($x = 0, 0.1, 0.2, 0.8, 0.9, 1.0$) were synthesized from corresponding components taken in stoichiometric ratios in evacuated quartz cells. Single crystals were grown by the modified Bridgman method. The freshly cleaved samples prepared for study, in which axis “c” of the crystal was oriented in the cleavage plane, were rectangular and about 0.5mm thick. Indium contacts were deposited onto the samples. The ohmic of the contacts was controlled before each measurement.

The electrical conductivity $\sigma(T)$ was measured by the four-probe method in the geometry perpendicular (σ_{\perp}) to the tetragonal axis “c” in a nitrogen cryostat in the mode of quasi-steady continuous heating (cooling) of the crystal with a rate of ~ 0.1 K/min. Measurements were performed in the temperature range of 90–300 K. The electrical conductivity was studied using an E7-25 digital impedance meter.

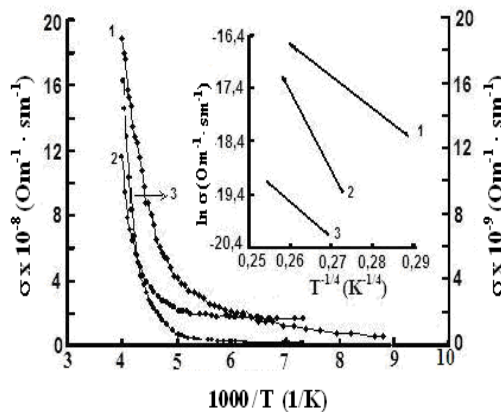


Fig. 1. Temperature dependence of the specific conductivity for $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ compositions (curve 1— $x = 0$, (2)—0.1, and (3)—0.2). The sample dependence in Mott coordinates is presented in the inset.

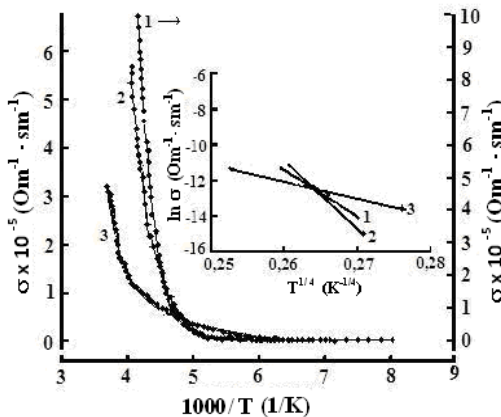


Fig. 2. Temperature dependence of the specific conductivity for $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ x compositions (curve 1— $x = 1$, (2)—0.9, and (3)—0.8). The sample dependence in Mott coordinates is presented in the inset.

3. Results of measurements

Figures 1 and 2 show the temperature dependences of the specific electrical conductivity of the system of $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ solid solutions, where $x = 0, 0.1, 0.2$ (Fig. 1) and $x = 0.8, 0.9, 1$ (Fig. 2), presented in Arrhenius coordinates. Investigations were performed in a variable electric field in the frequency range $\nu=10\text{--}10^6$ Hz. The dependence $\sigma_{ac}\sim f^{0.8}$, which is characteristic of the mechanism of conductivity over localized states in the vicinity of the Fermi level, is established. According to [10], this dependence is determined by the following expression:

$$\sigma_{ac}(f) = \frac{\pi^3}{96} e^2 k T N_F^2 a^5 f \left[\ln \left(\frac{\nu_{ph}}{f} \right) \right]^4 \quad (1)$$

where e is the elementary charge, k is the Boltzmann constant, N_F is the density of states in the vicinity of the Fermi level, $a = 1/\alpha$ is the localization radius, α is the decay constant of the wave function of a localized charge carrier $\psi = e^{-\alpha r}$, and ν_{ph} is the phonon frequency.

Studies of the conductivity of crystals of the $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ solid solution, where $x=0, 0.1$, and 0.2 ; and $x= 0.8, 0.9$, and 1 ; in the solubility region in variable electric fields showed that the deviation from the dependence $\sigma_{ac}\sim f^{0.8}$, which is characteristic of the hopping-conduction mechanism [10,11] over localized states in the vicinity of the Fermi level, is observed in a region of comparatively low frequencies.

Starting from the boundary frequency (f_b), the conductivity follows the dependence $\sigma_{ac}\sim f^{0.8}$, and the concentration dependence of the boundary frequency linearly shifts into the low-frequency region (at values of $x = 0, 0.1$, and 0.2) and into the high-frequency region (at $x = 0.8, 0.9$, and 1) compared with average solution components (see Fig. 3).

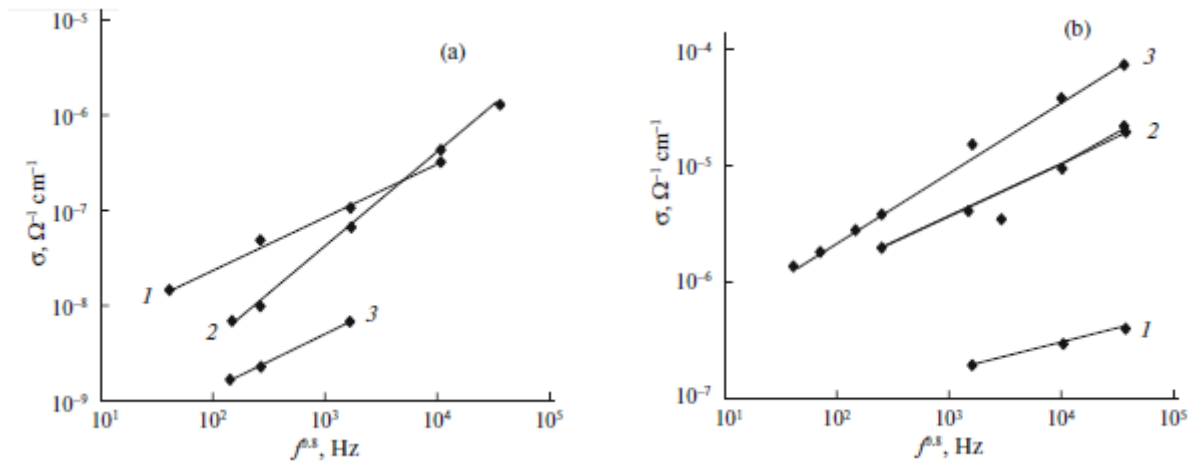


Fig. 3. Frequency-dependent conductivity of crystals of the $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ solid solution: (a) (1) $x = 0$, (2) 0.1 , and (3) 0.2 ; (b) (1) $x = 1$, (2) 0.9 , and (3) 0.8 . Measurements are performed at 200 K.

The dependence $\sigma_{ac} \sim f^{0.8}$ is observed both for marginal solid-solution components TlInSe_2 and TlGaTe_2 and for all compositions of the $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ solid solution, where $x = 0, 0.1$, and 0.2 ; and $x = 0.8, 0.9$, and 1.0 . Figure 3(b) shows the dependence of the boundary frequency (f_b) on the composition for the solid solution under study, above which, the dependence $\sigma_{ac} \sim f^{0.8}$ is fulfilled.

The presence of two segments of varying specific conductivity is characteristic of the dependences $\sigma_{\perp}(T)$ for all solid-solution compositions. The high temperature branch of the presented dependence (172–240 K) had an exponential character. The conductivity of thermally excited impurity charge carriers in the allowed band is dominant in this temperature range. A decrease in temperature is accompanied by a rapid decrease in the concentration of impurity carriers, i.e., at temperatures below 172 K, $\sigma_{\perp}(T)$ are the regions of impurity-carrier freezing. The gentle variation in the slope of curve $\sigma_{\perp}(T)$ in coordinates $\log\sigma(1/T)$, as well as the dependence of conductivity on the frequency of the measuring field that we observed previously, are characteristic signs of the hopping conductivity over localized states, which is observed for all crystals of Group $A^{III}B^{III}C_2^{VI}$ [12– 16]. Greater sensitivity of the conductivity to a variation in temperature is characteristic of the studied solid solutions.

Such a high sensitivity of the specific conductivity to a variation in temperature makes use of the studied crystals as sensitive temperature sensors or in systems for temperature control and stabilization promising.

Since the observed feature manifests itself in the entire system of solid solutions, we can vary the temperature region of highest sensor efficiency. $\sigma_{\perp}(T)$ in the temperature range $172 < T < 240$ K in Mott coordinates. It is seen from Figs. 1 and 2 that the experimental points are well straightened in the mentioned coordinates. This fact is evidence in favor of the fact that charge transfer perpendicularly to the tetragonal axis “c” in crystals of compositions $(TlInSe_2)_{1-x}(TlGaTe_2)_x$ ($x = 0; 0.1; 0.2; 0.8; 0.9; 1.0$) in the mentioned region is performed by the hopping conduction of charge carriers over localized states lying in a narrow energy region near the Fermi level. In this case, the electrical conductivity is described by the known Mott relation [10], and disregarding the frequency of the measuring field compared with phonon frequencies ($\sim 10^{12}$ Hz), we have

$$\sigma \sim \exp\left[-\left(\frac{T_0}{T}\right)^{1/4}\right], \quad T_0 = \frac{\beta}{kN_F a^3} \quad (2)$$

Here, N_F is the density of localized states in the vicinity of the Fermi level, a is the localization radius of the state in the vicinity of the Fermi level, k is the Boltzmann constant, and β is a number, which depends on the problem dimension.

Thus, impurity charge carriers rapidly freeze in the allowed band as the temperature decreases and, due to this fact, carrier hops over separate impurity states without activation into the allowed band start to play the largest role in electrical conductivity. The hopping-conduction mechanism is characterized by low carrier mobility since carrier hops are performed over weak overlaps of the tail parts of wave functions of close acceptor levels [10]. However, it is seen from Figs. 1 and 2 that the conductivity over localized states exceeds the band conductivity. This peculiarity of the conductivity can be explained if we take into account the fact that conductivity over localized states is carried out by carriers at impurity levels, while the contribution of band conductivity is substantially suppressed due to the freezing of carriers from the valence band.

According to the hopping-conduction model [10, 11], a noticeable deviation of the conductivity from linearity in coordinates $\log \sigma(1/T)$ should be observed at temperatures below $T_x \approx T_D/2$, where T_D is the Debye temperature. The Debye temperature for these crystals can be estimated by the phonon spectra [17] $T_D \approx 290$ K (200 cm^{-1}). It is accepted in the model under consideration that charge transfer at temperatures above the Debye temperature is performed by the thermal activation of carriers by means of multiphonon inelastic collisions. The hopping-conduction mechanism with a variable hop length, which leads to a temperature dependence of the form $\sigma = A \exp[(T_0/T)^{1/(1+d)}]$, where $d = 3$ for a three-dimensional system and $d = 2$ for a two-dimensional system, is dominant in the low-temperature region ($T < T_D/2$).

Calculated parameters of conductivity in the Mott approximation for $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ solid solutions ($x = 0, 0.1, 0.2, 0.8, 0.9, \text{ and } 1.0$) at 200 K

Composition	$N_F, \text{eV}^{-1} \text{cm}^{-3}$	$R, \text{\AA}$	$\Delta E, \text{eV}$	N_t, cm^{-3}	τ, s
TlGaTe_2	1.44×10^{18}	138	0.061	8.78×10^{16}	1.12×10^{-8}
$(\text{TlInSe}_2)_{0.1}(\text{TlGaTe}_2)_{0.9}$	4.44×10^{18}	94	0.0617	2.74×10^{17}	2.11×10^{-9}
$(\text{TlInSe}_2)_{0.2}(\text{TlGaTe}_2)_{0.8}$	5.2×10^{18}	90	0.062	3.22×10^{17}	1.54×10^{-9}
$(\text{TlInSe}_2)_{0.8}(\text{TlGaTe}_2)_{0.2}$	1.03×10^{18}	139	0.1	1.03×10^{17}	7.55×10^{-8}
$(\text{TlInSe}_2)_{0.9}(\text{TlGaTe}_2)_{0.1}$	7.3×10^{17}	149	0.095	6.94×10^{16}	1.67×10^{-7}
TlInSe_2	4.09×10^{17}	189	0.084	3.44×10^{16}	3.28×10^{-7}

It is seen from Figs. 1 and 2 that the experimental points of the specific conductivity of $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ solid solutions ($x = 0; 0.1; 0.2; 0.3; 0.8; 0.9, 1$) in Mott coordinates $\sigma = A \exp[(T_0/T)^{1/(1+d)}]$ at $d = 3$ lie on a straight line (measurements are performed upon the application of a measuring field perpendicular to the chains). The parameters of the conductivity calculated in the Mott approximation are tabulated. In terms of the model under consideration, the average carrier hop length R over localized states near the Fermi level for specified temperature T is found from expression $R = 3/8a(T_0/T)^{1/4}$. It follows from the presented expression that parameter R increases as the temperature decreases. This is accompanied by the emptying of local states in the forbidden band, and carrier hops over separate impurity levels without activation into the conduction band start to play the main role in charge transfer. This results in an increase in the probability of carrier hops to localization centre that are spatially more remote but closer energetically. Due to this fact, the activation energy of the hop decreases. Such character of the electrical conductivity is also called electrical conductivity with steadily decreasing hop activation energy.

ΔE is the width of the optimal energy band near the Fermi level, which encompasses almost the entire energy region of charge transfer at a specified temperature and is determined from the following relation:

$$\Delta E = 3/\pi R^3 N_F$$

The concentration of traps in the vicinity of the Fermi level for the solid solution under study, which are responsible for charge transfer over these localized states, was determined by the formula

$$N_t = N_F \Delta E$$

According to Mott, the average hop length (R) is determined by the following formula:

$$R = (1/2\alpha) \cdot \ln(v_{ph}/f), \quad (3)$$

where v_{ph} is the phonon frequency and α is the localization radius. Knowing R allowed us to determine the average hop time ($\tau^{-1} = v_{ph} \exp(-2\alpha R)$) of the carrier in the $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ solid solution ($x = 0, 0.1, 0.2, 0.8, 0.9, 1.0$) (last column of the table). When calculating R and τ , the value of v_{ph} was taken equal to 1012 Hz, while values $a = 30 \text{ \AA}$ for the TlInSe_2 crystal, $a = 30 \text{ \AA}$ for the TlGaTe_2 crystal, and $a = 20 \text{ \AA}$ for the solid solution $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ ($x = 0.1, 0.2, 0.8, 0.9$) were taken as the localization radii [1, 12, 14].

It is seen from Fig. 4 that the concentration dependence of the parameters of hopping conduction substantially varies in a mixed crystal in the solubility region. The density of states at the Fermi level (N_F) (Fig. 4a) increases in the solid-solution region compared with margin components TlInSe₂ and TlGaTe₂.

The energy dispersion of the states localized near the Fermi level (ΔE) broadens at $x = 0, 0.1, \text{ and } 0.2$ (the solution region on the side of TlInSe₂) and at values $x = 0.8, 0.9, \text{ and } 1$ (the solution region on the TlGaTe₂ side) (Fig. 4b). An increase in the trap concentration (N_t) (Fig. 4c) is observed near the Fermi level in the solid-solution region.

The hop length (R) over localized states in both concentration regions (in the solubility region) decreases compared with the margin components of the solid solution (Fig. 4c). According to relation (4), a decrease in the hop length can be caused by two circumstances, notably, (a) a decrease in v_{ph} in the solid solution and (b) an increase in the localization radius.

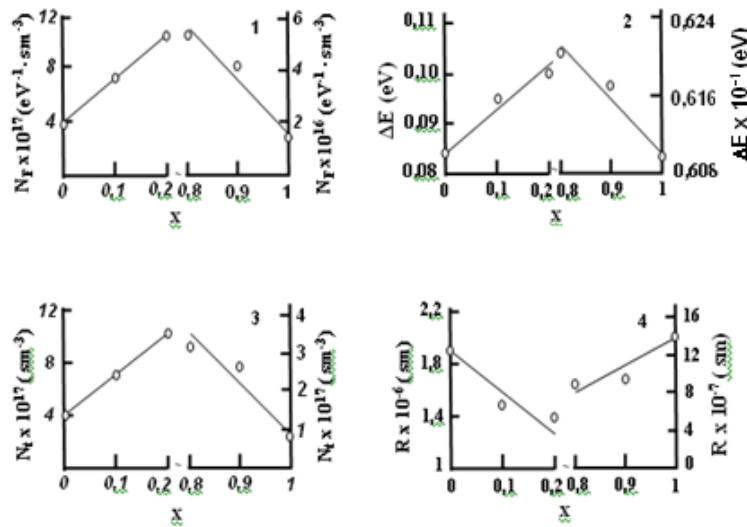


Fig. 4. Concentration dependences of (a) the density of electron states (N_F), (b) the energy dispersion of localized states (ΔE), (c) the concentration of deep traps (N_t), and (d) the hop length (R) of charge carriers over localized states near the Fermi level of the (TlInSe₂)_{1-x}(TlGaTe₂)_x solid solution ($x = 0, 0.1, 0.2, 0.8, 0.9, \text{ and } 1,0$).

The phonon frequencies of mixed crystals in the solubility region, as a rule [17], lie within the limits of frequencies of the margin components of the solid solution. Thus, we should not expect a decrease in the hop length over localized states associated with a decrease in phonon frequencies in the solid solution. In this case, a decrease in R upon the transition to the solid solution region can only be caused by an increase in the localization radius.

4. Results and discussion

It follows from the results of measurements that the hopping-conduction mechanism with a variable hop length is dominant in the studied crystals of (TlInSe₂)_{1-x}(TlGaTe₂)_x solid solutions ($x = 0, 0.1, 0.2, 0.8, 0.9, 1$). This is accompanied (as is seen from the table and Figs. 1–4) by a substantial variation in the parameters of hopping conduction in the solid solution compared with margin solid-solution components TlInSe₂ and TlGaTe₂. This fact is indicative of the occurrence of additional types of disorder, which cause the localization of electron states in the vicinity of the Fermi level. Revealing the nature of these disordered regions is essential when establishing the charge-transfer mechanism in a multicomponent solid solution.

The defect concentration in TlGaTe_2 and TlInSe_2 exceeds 10^{18} cm^{-3} . The high density of states near the Fermi level is attributed to the presence of such an amount of defects. The cause of structural imperfection may be the presence of a broad region of homogeneity of Group $\text{A}^{\text{III}}\text{B}^{\text{III}}\text{C}_2^{\text{VI}}$ crystals, which reaches 6–8 mol %. The segregation coefficient in the homogeneity region is smaller than unity; therefore, when growing single crystals, there is a large probability that the solid-solution composition deviates from stoichiometry.

This can in turn promote the appearance of numerous layer-joining defects, vacancies, and dislocations. Traps generated by various defects in crystals play the main role in charge-transfer phenomena. The activation energy, which determines the width of the energy band near the Fermi level, along which charge carriers can hop, represents the electrical conductivity of the crystal almost completely.

We can note the following additional causes of the appearance of defects in the case of the $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ solid solution, which lead to additional disorder and, consequently, to the appearance of new localized states with energies entering the range of values forbidden in an ideal crystal: (I) translational invariance of the crystal lattice is violated, i.e., equivalent sites of the crystal lattice turn out to be occupied by nonequivalent atoms; (II) the presence of microinclusions of other phases, for example, micronuclei of the phases TlSe , InSe , TlInTe_2 , etc. in the solid solution; (III) the content of antisite defects caused by mutual partial substitutions of cations In^{3+} , Ga^{3+} , Tl^{1+} , and Tl^{3+} in the solid-solution structure; (IV) the high probability of the occurrence of site disorder, which consists in the fact that one sublattice in a multicomponent solid solution turns out to be ordered (Tl^{1+} sublattice), while the atoms in the second sublattice (InSe_2 and GaTe_2 chains) chaotically substitute each other at sites of the chain. Without a doubt, the listed types of disorder introduce an additional contribution to the structural disorder causing the localization of electron states near the Fermi level.

5. Conclusions

We investigated the conductivity of the samples of $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ mixed crystals from the solubility region. The temperature and frequency ranges of occurrence of hopping conduction as well as the cut off frequency, above which conductivity over localized states is observed for all solid-solution compositions, are established. Mixed crystals are characterized by a disordered crystal lattice, which can be formed in a number of ways. Additional causes (compared with margin solid-solution components), which are responsible for the hopping character of the conductivity, are considered.

The parameters of localized states are estimated, notably, the density of states in the vicinity of the Fermi level and their energy dispersion, the concentration of deep traps, and average hop time and length are estimated. The concentration dependences of these parameters are plotted. Additional causes leading to the localization of energy states in the solid solution are considered.

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ПРОВОДИМОСТЬ ПО ЛОКАЛИЗОВАННЫМ СОСТОЯНИЯМ СИСТЕМЫ ТВЕРДЫХ РАСТВОРОВ $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ ОБЛУЧЕННЫХ γ – КВАНТАМИ

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Резюме: Изучены температурные зависимости электрической проводимости по локализованным состояниям, в области растворимости смешанных кристаллов составов $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ при $x=0; 0,1; 0,2; 0,8; 0,9; 1$. Установлено, что в этой системе имеет место прыжковая проводимость с переменной длиной прыжка по локализованным состояниям вблизи уровня Ферми. Определена энергия активации проводимости, оценены плотность состояния в окрестности уровня Ферми, их разброс, радиус локализации, среднее расстояние прыжков носителей для всех составов и построены концентрационные зависимости рассчитанных параметров.

Ключевые слова: прыжковая проводимость, уровень Ферми, радиус локализации.

γ -KVANTLARLA ŞÜALANMIŞ $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ SİSTEMİ BƏRK MƏHLULLARIN LOKALLAŞMIŞ HALLAR ÜZRƏ KEÇİRİCİLİYİ

**R.M. S rdarlı, O.A. S m dov, N. . liyeva, A.P. Abdullayev,
F.T. Salmanov, A.A. Orucova**

Xülasə: $(\text{TlInSe}_2)_{1-x}(\text{TlGaTe}_2)_x$ ($x=0; 0,1; 0,2; 0,8; 0,9; 1,0$) sistemi bərk məhlulları nümunələrinin elektrik keçiriciliyinin temperatur asılılığı tədqiq olunmuşdur. Müəyyən edilmişdir ki, tədqiq edilən bərk məhlullarında yükün daşınması Fermi səviyyəsinin yaxınlığında dar enerji zolağında yerləşən lokallaşmış hallar üzrə yükdaşıyıcıların sıçrayışlı keçiriciliyi vasitəsilə həyata keçirilir. Tədqiq olunan bərk məhlullar üçün lokallaşmış halların parametrlərinin: Fermi səviyyəsinin ətrafında lokallaşmış halların sıxlığı, dərin tələlərin konsentrasiyası, eləcə də yükdaşıyıcıların sıçrayışlarının orta müddəti və məsafəsinin qiymətləri hesablanmışdır. Hesablanmış parametrlərin qiymətlərinin konsentrasiyadan asılılıqları qurulmuşdur. Bərk məhlullarda enerji hallarının lokallaşmasına gətirən əlavə səbəblər nəzərdən keçirilmişdir.

Acar sözlər: sıçrayışlı keçiricilik, Fermi səviyyəsi, lokallaşma radiusu.