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THE EFFECT OF GAMMA QUANTA ON THE IONIC AND ELECTRON CONDUCTIVITY OF $TlInS_2$(10%C) COMPOUND

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Abstract: The electrical properties of the $TlInS_2$(10%C) crystal in a constant electric field were studied. In a constant electric field, the electrical conductivity decreases depending on time and remains unchanged after a certain value of time. The relaxation time τ and the diffusion coefficient D were determined from the kinetics of the electrical conductivity (σ(t)). It has been shown that with the increase of the absorption dose, the relaxation time decreases accordingly, and the value of the diffusion coefficient increases. It is also shown that the numerical value of the ion share in the electrical conductivity increases and the electron share decreases depending on the radiation doses. Experimental results show that the ionic conductivity of the $TlInS_2$(10%C) crystal increases from 63% to 78.5% at radiation doses of 0÷0.8MGy.

Keywords: ionic conductivity, electron conductivity, relaxation time, diffusion coefficient, activation energy, radiation dose, gamma quanta

1. Introduction

As is known, several ferroelectric and incommensurate phase transitions are observed in the $TlInS_2$ compound at temperatures lower than room temperature, which refers to $A^3B^3C_2^6$ type semiconductor compounds [1-3]. For the first time, phase transitions were observed in $TlGaSe_2$ and $TlInS_2$ crystals at temperatures above 300K, and it was shown that the electrical conductivity in these phases is both ionic and electronic [4, 5]. Also, in [6], anomalies were observed in the temperature dependence of dielectric permeability ε(T) in $TlInS_2$<5%C> compound at the temperatures of T=370K, T=415K, and T=532K. It has been shown that the experimental points in the ln(ε) dependence of the ionic conductivity are collected on a straight line and the activation energies have been calculated. The results obtained for tgδ(T) show that the peaks of tgδ(T) regularly shift to higher temperatures and the value of tgδ decreases as the frequency of the measuring field increases. The activation energy of jumps (ΔE=0.24 eV), its oscillation frequency $\nu = 8 \cdot 10^{12}$ Hz was determined.

In [7], the temperature dependence (ε(T)) of the dielectric constant of $TlInS_2$<10%C> crystal was studied. It is shown that the experimental points of the ln(ε) dependence are collected on a straight line, which is typical for the case of ionic conductivity, and the activation energies are determined *($\Delta Ea_1 = 0.54eV$, $\Delta Ea_2 = 0.32eV$, $\Delta Ea_3 = 0.22$ eV)*. The activation energy of the jump (ΔE=0.24eV), and its oscillation frequency $\nu = 8 \cdot 10^{12}$ Hz were determined from the frequency dependence of the loss-angle tangent, tgδ(T). At the same temperature, the permittivity in the direction of "c" axis of the $TlInS_2$<10%C> crystal is 34.5 times higher than

that of the $TlInS_2$ crystal. It has also been shown that the numerical value of permittivity increases at radiation doses of 0÷0.8MGy and the temperature range related to ionic conductivity widens. In the presented work, the electrical properties of the $TlInS_2$(10%C) crystal were studied in a constant electric field at a temperature range of 300-600K and at doses of 0÷0.8MGy.

2. Methods of the experiment

$TlInS_2$<10%C> single crystals were grown by the Bridgman-Stockbarger method. 5×2×2 mm samples were used for measurements. Anisotropy of electrical properties was not observed on the (001) plane, therefore, the electrodes were placed on the surface of the crystals in the direction perpendicular to the layers. The silver paste was used as a contact. The complex permittivity and impedance were measured with an E7-20 AC bridge in the frequency range of 25-106 Hz using a copper-constantan thermocouple at a step of 0.1 K/min.

3. Experimental Results and Discussion

Electron and ion concentrations of conductivity are determined using Wagner's polarization method. The Wagner method is based on the following phenomena. In solid electrolytes with ionic or electron-ion charge carriers, changes in electrical conductivity depending on time are observed in measurements in a constant electric field. This phenomenon is based on the formation of the polarization process in the crystal in the measuring cell. That is, a double electrical layer is formed at the sample/electrode interface. Since charge-carrying ions are trapped at the sample/electrode boundary of the gradient electrode, the mobile ions accumulate on the negatively charged electrode due to the effect of the electric field and form a concentration gradient in the volume of the crystal.

The presence of a concentration gradient of positive charges leads to the creation of a diffusion current of ions in the opposite direction of the drift current. Since the diffusion and drift currents compensate each other in the stationary state, only electrons pass through the crystal. Thus, it is possible to find the ratio of electron and ion accumulation from the time dependence of electrical conductivity.

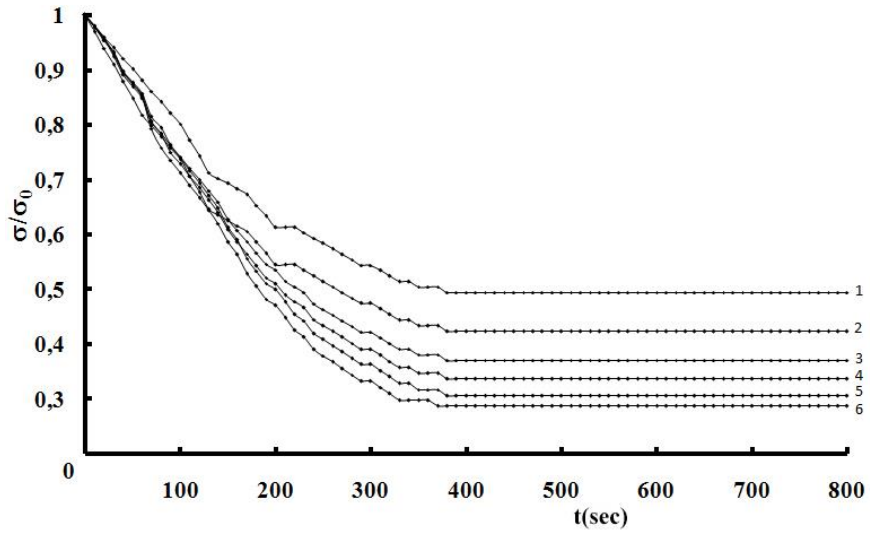

Fig. 1. Kinetics of electrical conductivity for $TlInS_2$(10%C) crystal:1-350K; 2-400K; 3-470K; 4-500K; 5-550K; 6-600K

Figure 1 shows the time dependences of the electrical conductivity of the $TlInS_2$(10%C) crystal in a constant electric field at different constant temperatures (400K, 450K, 500K, 550K, 600K). As can be seen from Figure 1, the electrical conductivity initially decreases exponentially and remains unchanged after a certain time. The non-linear drop in electrical conductivity occurs rapidly at relatively high temperatures.

The time dependence of the current in the constant electric field is due to the mutual compensation of the volume charges near the phasing electrodes.

The time dependence of the current density is determined by the second-order linear differential equation and [8] can be obtained based on the continuity condition. If we accept that the number of quasi-stable states is higher than the number of charge carriers, and if we do not take into account the inhomogeneous field of volume charges, the solution of this equation will be simplified.

It is also necessary to assume equal distribution of *q* charges in the sample with thickness *d* at t=0 and the complete impenetrability of the electrodes for charge carriers. When the depth of the diffusion zone $2\sqrt{Dt} \geq d$ (D-diffusion coefficient) exceeds the thickness of the sample, an exponential dependence can be obtained for the current density in unidirectional motion in a large time range [9,10].

$$j=j_0 \exp\{-\pi^2 Dt/d^2\} \quad (1)$$

According to (1), the experimental results can be written as a decreasing exponent.

$$\sigma=\sigma_{bgr} + \sigma_0 \exp\{-t/\tau_{SC}\} \quad (2)$$

where σ_{bgr} describes other clades that are time-independent in charge transport processes. τ_{SC} is a time parameter that characterizes the accumulation of charge carriers in the electrode region and is determined by the diffusion coefficient and the thickness of the sample according to expression (1) ($\tau_{SC}= d^2/(\pi^2 D)$).

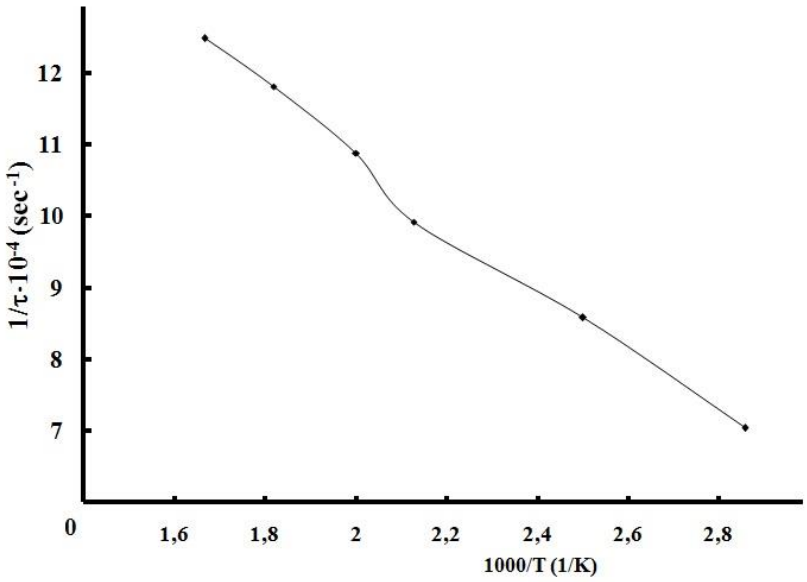

Fig. 2. Dependence of relaxation time of $TlInS_2$(10%C) crystal on 1/T

As can be seen from Fig.2, the relaxation time of $TlInS_2$(10%C) crystal decreases as the temperature increases. As a result of the calculations, it was shown that the polarization of volume charges occurs in the infra-low frequency range depending on the temperature (~10^{-4}Hz ÷10^{-3}).

Figure 3 shows the temperature dependence of the diffusion coefficient of the $TlInS_2$(10%C) crystal. As can be seen from Figure 3, the diffusion coefficient varies in the range of 4÷7·10^{-10} (m^2/sec) depending on the temperature, and the value of the diffusion coefficient increases with the increase in temperature. That is the concentration of ions involved in ionic conductivity increases with the increase in temperature.

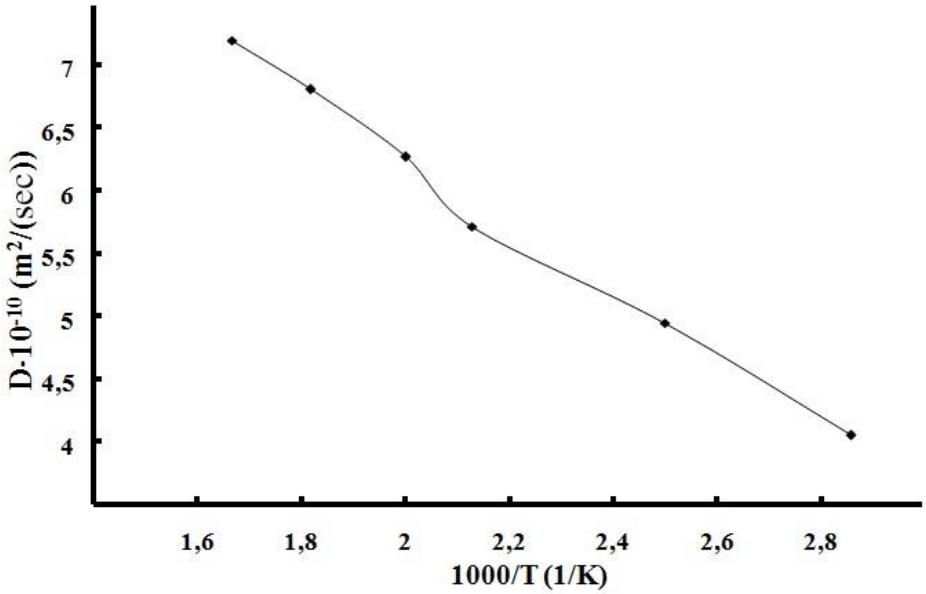

Fig.3. Dependence of diffusion coefficient on 1/T in $TlInS_2$(10%C) crystal

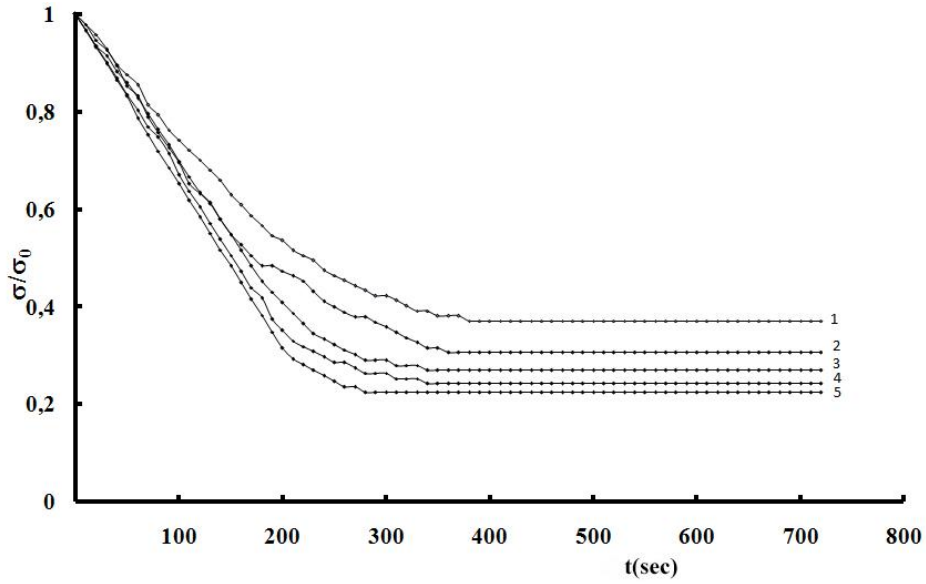

Fig. 4. Kinetics of electrical conductivity of $TlInS_2$(10%C) crystal irradiated with γ-rays at T=470K: 1-0; 2-0.2 MGy; 3-0.4MGy; 4-0.6MGy; 5-0.8MGy

Figure 4 shows the kinetics of the electrical conductivity of the $TlInS_2$(10%C) crystal irradiated with γ-rays at the temperature T = 470K in the dose range of 0÷0.8MGy. As can be seen from the figure, as the radiation dose increases, the share of electrons in the total conductivity decreases, and, as a result, the share of ions increases. Thus, the experimental results show that the ion conductivity of the $TlInS_2$(10%C) crystal increases from 63% to 78.5% at radiation doses of 0÷0.8MGy.

Table 1

$D_{absorp.dose}$(MGy)	τ (sec)	D_{diffus} $(\frac{m^2}{sec})$
0	1008.79	$5.7 \cdot 10^{-10}$
0.2	842.61	$6.8 \cdot 10^{-10}$
0.4	761.158	$7.6 \cdot 10^{-10}$
0.6	704.402	$8.2 \cdot 10^{-10}$
0.8	669.797	$8.6 \cdot 10^{-10}$

The dependence of relaxation time and diffusion coefficient on absorption dose is given in Table 1. As can be seen from the table, with the increase in the absorbed dose, the relaxation time decreases and the value of the diffusion coefficient increases accordingly. In the $TlInS_2$(10%C) crystal, depending on the absorption dose (by increasing the absorption dose), the decrease of the relaxation time and the increase of the value of the diffusion coefficient are due to the increase in the concentration of ions.

As it is known, $TlInS_2$ crystals belong to the compounds of the $A^3B^3C^6_2$ group crystallizing in the tetragonal space group D_{4h}^{18} (structural type TlSe)[1-3]. The characteristic feature of $TlInS_2$ crystals is that they have In – S chains along the tetragonal c-axis of the crystal. The tetragonal axis is the optical axis. Univalent Tl^+ atoms are surrounded by octahedral S atoms. From crystal-chemical considerations, it can be assumed that the structure of $TlInS_2$(10%C) is the most favorable for the mobility of highly polarizable Tl^+ cations. According to the crystal-chemical parameters, additive carbon atoms can occupy the octahedral spaces between the InS_4 tetrahedra within the tetrahedral complex of the $TlInS_2$ monoclinic lattice [InS_{10}] (due to the small size of the carbon ions (0.2Å)), and as a result, the parameters of the $TlInS_2$<C> crystals will be different from the parameters of the initial crystals. In this case, the placement of an electropositive atom weakens the bond with sulfur and thallium atoms [7]. It is known that the degree of irregularity of crystals can be changed under the influence of γ-radiation. Under the influence of γ - quanta, radiation defects in the form of vacancies, atoms between nodes, as well as different types of defect complexes interacting with each other and with chemical additives, are formed in the crystal. The dominant role in these processes is played by ionization-type defects (charged defects) caused by irradiation with γ-quanta. That is, the reason for the increase in ion conductivity of the $TlInS_2$ crystal depending on the radiation dose is the increase in the concentration of ions.

References

1. A.A. Volkov, Y.G. Goncharov, G.V. Kozlov. Structural phase transitions in the $TlInS_2$ crystal // FTT, 1983, v. 25, ed. 12, p. 3583-3585
2. S.B. Vakhrushev, V.V. Zhdanova, B.E. Kvyatkovsky and others Incommensurable phase transition in $TlInS_2$ crystal // Letter to JETF, 1984, vol. 39, ed. 6, p. 245-247.
3. R.A. Aliev, K.R. Allahverdiyev, A.I. Baranov, I.R. Ivanov, R.M. Sardarly. Ferroelectricity and structural phase transitions in crystals of the $TlInS_2$ family // FTT, 1984, vol. 26, ed. 5, p. 1272-1276.
4. O.A. Samadov, O.Z. Alakparov, A.I. Najafov, S.F. Samadov, M.M. Guliyev, Kh.Z. Fatalizadeh, N.T. Mosumli, N.I. Huseynov. Dielectric and electrical relaxation in $TlInS_2$ crystals irradiated by γ- quanta. Journal of Radiation Research, vol.2, №1, 2015, p.11-17.
5. S.F. Samadov, O.A. Samadov, O.Z. Alakparov, M. Kulikz, A.I. Najafov, N.M. Mehdiyev and E. M. Huseynov. Dielectric and electrical properties of near-surface. International Journal of Modern Physics B Vol. 33, No. 27 (2019) 1950320 (7 pages)]
6. O.A. Samadov, O.Z. Alakparov, Kh.B. Orujova, A.I. Najafov, E.M. Huseynov, S.F. Samadov, I.A. Gabulov, N.M. Mehdiyev, Modern Physics Letters B, Dielectric permittivity and relaxation process investigation of C-doped $TlInS_2$ crystals. 2150325,2021
7. O.A. Samadov, Kh.B. Orujova, N.M. Mehdiyev, A.I. Najafov, S.F. Samadov, R.N. Mehdiyeva, T.I. Karimova, Dielectric properties of $TlInS_2$(10%C) COMPOUND irradiated with gamma quanta, Journal of Radiation Researches, vol.9, No.2, 2022, pp.22-27,Baku
8. Lydiard. Ionic conductivity of crystals. Publishing house IL, M. (1962). P. 222
9. M.D. Volnyansky, S.N. Plyaka, M.P. Trubitsin, Yahya A.Kh. Obydat, Ionic conductivity and volume-charge polarization processes in $Li_2Ge_7O_{15}$ crystals, FTT, 2012, vol. 54, ed.3, p.471
10. Features of impurity states of vanadium in lead telluride © A.I. Artamkin, A.A. Dobrovolsky, A.A. Vinokurov, V.P. Zlomanov, S.Y. Gavrilkin∗, O.M. Ivanenko∗, K.V. Mitzen∗, L.I. Ryabova, D.R. Khokhlov, FTP, 2010, volume 44, ed.12, p.1591

ВЛИЯНИЕ ГАММА-КВАНТА НА ИОННУЮ И ЭЛЕКТРОННУЮ ПРОВОДИМОСТЬ СОЕДИНЕНИЯ $TlInS_2$(10%C)

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Резюме: Электрические свойства кристалла $TlInS_2$ (10%C) изучались в постоянном электрическом поле. В постоянном электрическом поле электропроводность уменьшается во времени и остается неизменной через определенное время. Время релаксации τ и коэффициент диффузии D определяли из кинетики электропроводности (σ(t)). Показано, что с увеличением дозы поглощения соответственно уменьшается время релаксации и увеличивается значение коэффициента диффузии. Показано также, что в зависимости от доз облучения численное значение доли ионов в электропроводности увеличивается, а доля электронов уменьшается. Экспериментальные результаты показывают, что ионная проводимость кристалла $TlInS_2$(10%C) увеличивается с 63% до 78,5% при дозах облучения 0÷0,80 МГр.

Ключевые слова: ионная проводимость, электронная проводимость, время релаксации, коэффициент диффузии, энергия активации, доза излучения, гамма-кванты

$TlInS_2$(10%C) BİRLƏŞMƏSİNİN İON VƏ ELEKTRON KEÇİRİCİLYİNƏ QAMMA KVANTLARININ TƏSİRİ

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Xülasə: $TlInS_2$ (10%C) kristalının sabit elektrik sahəsində elektrik xassələri tədqiq olunmuşdur. Sabit elektrik sahəsində elektrik keçiriciliyi zamandan asılı olaraq azalır və zamanın müəyyən qiymətindən sonra dəyişməz qalır. Elektrik keçiriciliyinin kinetikasından ($\sigma(t)$) relaksasiya müddəti τ və diffuziya əmsalı D təyin olunmuşdur. Göstərilmişdir ki, udulma dozasının artması ilə, uyğun olaraq relaksasiya müddəti azalır və diffuziya əmsalının qiyməti artır. Həmçinin göstərilmişdir ki, şüalanma dozalarından asılı olaraq elektirik keçiriciliyində ion payının ədədi qiyməti artır və elektron payı azalır. Təcrübi nəticələr göstərir ki, $TlInS_2$(10%C) kristalının ion keçiriciliyi 0÷0,8MQr şüalanma dozalarında 63%-dən 78,5% -ə qədər artır.

Açar sözlər: ion keçiriciliyi, elektron keçiriciliyi, relaksasiya müddəti, diffuziya əmsalı, aktivləşmə enerjisi, şüalanma dozası, qamma kvantları