

EFFECT OF GAMMA IRRADIATION ON THE DC CONDUCTIVITY OF $\text{TlIn}_{0.9}\text{Se}_{1.8}\text{Ga}_{0.1}\text{Te}_{0.2}$ IN THE LOCALIZED STATES REGIME

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Abstract: The aim of this study is to investigate the effect of gamma irradiation on the hopping conductivity parameters of the $\text{TlIn}_{0.9}\text{Se}_{1.8}\text{Ga}_{0.1}\text{Te}_{0.2}$ compound. The DC conductivity of this solid solution was studied in the 100–300 K temperature range before and after gamma irradiation (250 kGy). The temperature intervals associated with localized-state conduction were identified for both conditions. Within the Mott model, the hopping conductivity parameters were determined for both unirradiated and irradiated samples. Irradiation increases the density of localized states (N_F) and trap concentration (N_t) near the Fermi level while reducing the energy difference (ΔE) and hopping length (R). Irradiation at 250 kGy was found to broaden this temperature range, which is attributed to the formation of radiation-induced defects.

Keywords: DC conductivity, solid solution, gamma irradiation, hopping conduction.

1. Introduction

Semiconductor materials belonging to the $\text{A}^{\text{III}}\text{B}^{\text{III}}\text{C}_2^{\text{IV}}$ family are highly suitable for practical applications, attracting considerable research interest due to their highly anisotropic crystal structure and lattice instability under external influences. These materials are promising candidates for the fabrication of photoelectric converters, voltage-measuring devices, and X-ray and neutron radiation detectors [1–4]. Among these, the chain-structured compounds TlInSe_2 and TlGaTe_2 crystallize in the tetragonal system with lattice parameters of $a = 8.075 \text{ \AA}$ and $c = 6.847 \text{ \AA}$ for TlInSe_2 , and $a = 8.420 \text{ \AA}$ and $c = 6.865 \text{ \AA}$ for TlGaTe_2 [5–10].

In recent years, the ternary chain-structured semiconductor crystals TlInSe_2 and TlGaTe_2 have been the subject of extensive research. In previous studies [11–15], the authors investigated the temperature dependences of the electrical conductivity $\sigma(T)$ and dielectric permittivity $\varepsilon(T)$ in TlInSe_2 and TlGaTe_2 crystals. The existence of transformation and memory effects, superionic conductivity, and giant dielectric relaxation has been demonstrated, and complex impedance spectra have been analyzed. It has been shown that the electrical properties of TlInSe_2 and TlGaTe_2 in the ionic conductivity region are determined by the hopping of thallium ions, and the frequency dependences of the real and imaginary parts of conductivity have been studied. Beyond their significant scientific interest due to structural features, highly anisotropic crystal structure, and lattice instability under external influences, $\text{A}^{\text{III}}\text{B}^{\text{III}}\text{C}_2^{\text{IV}}$ family crystals are also promising

materials for the development of photoelectric converters, strain gauges, and detectors [16–20]. Sheleg et al. investigated the conductivity and temperature dependence of TlInSe₂ and TlGaTe₂ single crystals under alternating electric fields [10]. A sequence of phase transitions has been observed in the low-temperature region of TlInSe₂ and TlGaTe₂. The expansion of this class of semiconductor materials, as well as the modification and control of their physical parameters, has been a key focus of research in recent years.

In [11], the electrical and photoelectric properties of TlInSe₂ single crystals—considered promising for X-ray dosimetry - as well as the effects of lithium-ion intercalation on these crystals were investigated. In [12], the effects of Ag, Cu, and Au dopants on the electrical properties of TlInSe₂ single crystals were studied. In recent years, considerable attention has been devoted to exploring anion–anion and anion–cation substitution in ternary compounds and to the systematic investigation of the properties of the resulting solid solutions [17–25]. It is of interest to study the physical properties of anion–cation substitution solid solutions formed on the basis of the tetragonal structure of semiconductor compounds such as TlInSe₂ and TlGaTe₂.

In [26], the authors prepared alloys of the TlInSe₂–TlGaTe₂ system and carried out a comprehensive study of their physicochemical properties. It has been shown that the system exhibits a quasi-binary eutectic phase diagram, in which the components have limited solid-state solubility: the solubility of TlGaTe₂ in TlInSe₂ is 30.0 mol%, while the solubility of TlInSe₂ in TlGaTe₂ is 25.0 mol%. The eutectic point in the system occurs at 55.0 mol% TlGaTe₂ and 973 K [26–30].

In [31–37], the authors investigated the temperature dependence of the electrical conductivity of Tl-based compounds in the temperature range of 100–300 K and found that charge transport in a constant electric field occurs via hopping conduction with a variable hopping length along localized states near the Fermi level. The parameters of transient conductivity were calculated, their variation with concentration was determined, and additional factors contributing to the localization of energy states in solid solutions were also considered.

This study investigates the conductivity properties of TlIn_{0.9}Se_{1.8}Ga_{0.1}Te_{0.2} solid solutions within the 90–300 K temperature range to evaluate the effects of a 250 kGy gamma-ray dose.

2. Crystal Growth and Experimental Methods

TlIn_{0.9}Se_{1.8}Ga_{0.1}Te_{0.2} compound was synthesized using a two-temperature method from high-purity elemental components (Tl 99.999%, In 99.9999%, Se 99.99%, Ga 99.999%, Te 99.99%). The synthesis was carried out in quartz ampoules purified to 10⁻³ Pa by direct alloys of constituent elements taken in stoichiometric amounts corresponding to the required solid solution composition. The completeness of the synthesis, the homogeneity of the resulting composition, and its properties were monitored using differential thermal analysis and X-ray phase analysis (DTA and X-ray analysis). The synthesized ingots were crushed into powder, loaded into quartz ampoules, evacuated, and sealed. The ampoules were subsequently placed in a vertical two-zone furnace with independently controlled temperature zones for single-crystal growth by the Bridgman method. During the growth process, the ampoule was moved through the temperature gradient zone at a rate of 0.15 mm/h. Homogenization of the resulting solid solutions was achieved by cooling to 700 K in the lower zone of the furnace, after which the furnace was turned off and allowed to cool to room temperature.

Samples of the TlIn_{0.9}Se_{1.8}Ga_{0.1}Te_{0.2} solid solutions were synthesized from the constituent elements taken in stoichiometric ratios in purified quartz ampoules. Single crystals were grown using a modified Bridgman method. Freshly cleaved samples prepared for the study, with the crystallographic *c*-axis oriented in the cleavage plane, had a rectangular shape and a thickness of

approximately 0.5 mm. Silver contacts were applied to the samples, and their ohmic behavior was verified prior to each measurement.

Electrical conductivity measurements were carried out using a digital immittance meter (E7-25). The measurements were performed in a nitrogen cryostat under quasi-stationary continuous heating (cooling) conditions at a rate of approximately 0.1 K/min, employing the four-probe method. The samples were cut perpendicular to the tetragonal c -axis. Irradiation was conducted at room temperature using a Co^{60} source.

3. Results and discussion

In the present study, the DC conductivity properties of the $\text{TlIn}_{0.9}\text{Se}_{1.8}\text{Ga}_{0.1}\text{Te}_{0.2}$ solid solution, irradiated with gamma quanta at initial and various doses, were investigated over the temperature range of 110–300 K.

Figure 1 presents the temperature dependences of the electrical conductivity of the $\text{TlIn}_{0.9}\text{Se}_{1.8}\text{Ga}_{0.1}\text{Te}_{0.2}$ solid solution in the unirradiated state and after gamma irradiation at a dose of 250 kGy over the temperature range of 110–300 K. As can be seen, the electrical conductivity increases with increasing irradiation dose in this compound.

As can be seen from Figure 1, the $\sigma(T)$ dependence exhibits an exponential character in the temperature ranges of 151–221 K (0 kGy) and 143–237 K (250 kGy). In this region, electrical conduction is dominated by thermally activated charge carriers in the allowed band [35–38]. This behavior of steady-state conductivity at low temperatures suggests that charge transport in the studied samples within the temperature range of 139–241 K occurs via hopping conduction with variable hopping lengths along localized states near the Fermi level [39–43]. The hopping conduction mechanism is characterized by low mobility of charge carriers, since hopping occurs due to weak overlap of the wave-function tails of adjacent acceptor levels [42].

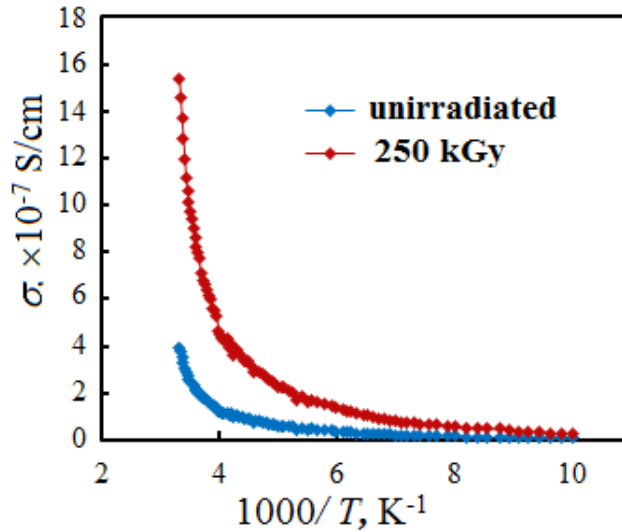


Fig. 1. Temperature dependences of the electrical conductivity of the $\text{TlIn}_{0.9}\text{Se}_{1.8}\text{Ga}_{0.1}\text{Te}_{0.2}$ compound irradiated with gamma quanta at different doses: 1 – 0 kGy, 2 – 250 kGy.

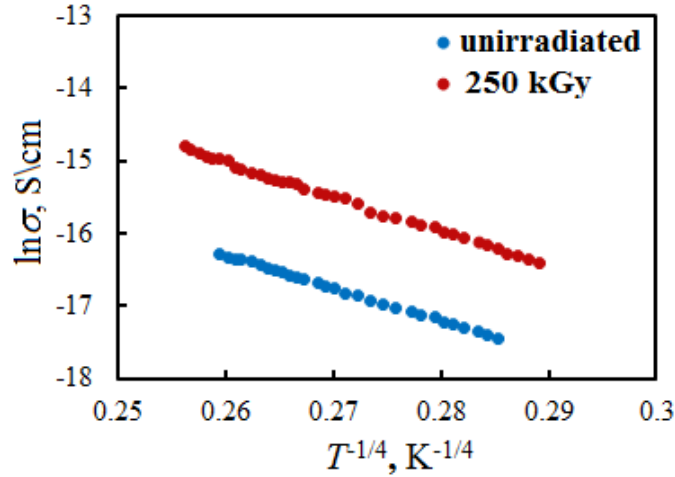


Fig. 2. Dependence of $\ln \sigma$ on $T^{-1/4}$ within the Mott approximation. 1 – 0 kGy, 2 – 250 kGy.

This conclusion is further supported by the linear dependence of $\ln \sigma$ on $T^{-1/4}$ [36-43]. Figure 2 presents the $\ln \sigma$ versus $T^{-1/4}$ dependence in Mott coordinates for the studied solid solutions in the temperature range $139 < T < 241$ K. As can be seen, this dependence is linear in the given temperature interval.

In this case, the electrical conductivity is described by the well-known Mott relation [42, 43], assuming that the frequency of the measuring field is negligible compared to the phonon frequencies ($\approx 10^{12}$ Hz).

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

where N_F – is the density of localized states at the Fermi level, a – is the localization radius of states near the Fermi level, k – is the Boltzmann constant, and β – is a numerical constant dependent on the dimensionality of the system.

The density of localized states (N_F) near the Fermi level can be estimated using the following formula.

$$N_F = \frac{16}{T_0 k a^3} \quad (2)$$

When calculating the N_F value, which represents the density of localized states near the Fermi level, a localization radius of the solid solutions of $a = 30 \text{ \AA}$ and a phonon frequency $\nu_{ph} = 10^{12}$ Hz were used [36-43].

Within the framework of the considered model, the average hopping length of charge carriers between localized states near the Fermi level at a given temperature T is given by the following formula:

$$R = \frac{3}{8} a \left(\frac{T_0}{T} \right)^{1/4} \quad (3)$$

It can be seen from this formula that the value of the parameter R increases with decreasing temperature. In this case, the localized energy levels in the forbidden gap become depleted, and the main contribution to electrical conductivity is provided by the hopping of charge carriers from individual dopant levels to the conduction band. As a result, the probability of charge carriers hopping to more distant localization centers that are energetically closer increases, which leads to

a decrease in the activation energy of hopping. This type of electrical conductivity is also referred to as conduction with monotonically decreasing hopping activation energy.

ΔE - The width of the optimal energy band near the Fermi level, which effectively encompasses the entire energy range of electrical transport at a given temperature, is determined by the following relationship: $\Delta E = 3/4\pi R^3 N_F$.

For the studied solid solution, the concentration of traps responsible for the electrical transport via localized states near the Fermi level was determined using the following formula: $N_t = N_F \Delta E$

The density of localized states (N_F), the concentration of deep traps (N_t), the energy width of localized states (ΔE), and the hopping length (R) were calculated within the Mott approximation for $\text{TlIn}_{0.9}\text{Se}_{1.8}\text{Ga}_{0.1}\text{Te}_{0.2}$ compounds before irradiation and after irradiation at different doses. The calculated values as a function of irradiation dose are presented in Table 1.

Table 1

Calculated parameters of electrical conductivity for $\text{TlIn}_{0.9}\text{Se}_{1.8}\text{Ga}_{0.1}\text{Te}_{0.2}$ compound within the Mott approximation at 225 K.

D (kGy)	N_F ($\text{eV}^{-1} \cdot \text{cm}^{-3}$)	R (Å)	ΔE (meV)	N_t (cm^{-3})
0	$4.43 \cdot 10^{17}$	182	88	$3.89 \cdot 10^{16}$
250	$6.21 \cdot 10^{17}$	167	80	$4.96 \cdot 10^{16}$

4. Conclusion

The DC conductivity of $\text{TlIn}_{0.9}\text{Se}_{1.8}\text{Ga}_{0.1}\text{Te}_{0.2}$ compound, irradiated with gamma quanta at a dose of 250 kGy, was studied in the temperature range of 100–300 K. The temperature ranges of conductivity were determined under local conditions before and after radiation exposure. It was shown that the temperature range of hopping conductivity expands with increasing radiation dose, from 151–221 K at 0 kGy to 143–237 K at 250 kGy. Within the framework of the Mott model, the parameters of hopping conductivity were calculated for both the unirradiated and irradiated samples. It was found that irradiation leads to an increase in the density of localized states (N_F) and the concentration of traps (N_t) near the Fermi level, whereas the energy difference (ΔE) and hopping length (R) decrease. The experimental results obtained are attributed to the formation of radiation-induced defects.

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ВЛИЯНИЕ ГАММА-ОБЛУЧЕНИЯ НА ПРОВОДИМОСТЬ ПОСТОЯННОГО ТОКА TlIn_{0.9}Se_{1.8}Ga_{0.1}Te_{0.2} В РЕЖИМЕ ЛОКАЛИЗОВАННЫХ СОСТОЯНИЙ

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Резюме: Целью работы является исследование влияния гамма-облучения на параметры прыжковой проводимости соединения TlIn_{0.9}Se_{1.8}Ga_{0.1}Te_{0.2}. Электропроводность постоянного тока данного твердого раствора изучалась в интервале температур 100–300 К до и после гамма-облучения с дозой 250 кГр. Были определены температурные области, соответствующие проводимости через локализованные состояния, как для исходных, так и для облученных образцов. В рамках модели Мотта рассчитаны параметры прыжковой проводимости. Установлено, что облучение приводит к увеличению плотности локализованных состояний (N_F) и концентрации ловушек (N_t) вблизи уровня Ферми, а также к уменьшению энергетического интервала (ΔE) и длины прыжка (R). Показано, что облучение дозой 250 кГр расширяет температурный диапазон прыжковой проводимости, что связано с образованием радиационно-индуцированных дефектов.

Ключевые слова: DC проводимость, твёрдый раствор, гамма-облучение, прыжковая проводимость.

TlIn_{0.9}Se_{1.8}Ga_{0.1}Te_{0.2} BİRLƏŞMƏSİNDƏ LOKALLAŞMIŞ HALLAR REJİMİNDƏ QAMMA ŞÜALANMASININ DC KEÇİRİCİLİYİNƏ TƏSİRİ

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Xülasə: Bu tədqiqatın məqsədi TlIn_{0.9}Se_{1.8}Ga_{0.1}Te_{0.2} birləşməsində qamma şüalanmanın sıçrayışlı keçiricilik parametrlərinə təsirini araşdırmaqdır. Bu bərk məhlulun DC keçiriciliyi 100–300 K temperatur intervalında şüalanmadan əvvəl və 250 kQr dozada qamma şüalanmadan sonra öyrənilmişdir. Hər iki hal üçün lokallaşmış hallar üzrə keçiriciliyə uyğun temperatur sahələri müəyyən edilmişdir. Mott modeli çərçivəsində həm şüalanmamış, həm də şüalanmış nümunələr üçün sıçrayışlı keçiricilik parametrləri hesablanmışdır. Müəyyən edilmişdir ki, şüalanma Fermi səviyyəsi yaxınlığında lokallaşmış halların sıxlığını (N_F) və tələlərin konsentrasiyasını (N_t) artırır, eyni zamanda enerji fərqi (ΔE) və tullanma uzunluğunu (R) azaldır. 250 kQr dozasında şüalanmanın bu temperatur intervalını genişləndirdiyi göstərilmişdir ki, bu da radiasiya ilə induksiya olunmuş defektlərin yaranması ilə izah olunur.

Açar sözlər: DC keçiriciliyi, bərk məhlul, qamma şüalanma, sıçrayışlı keçiricilik.