

STUDY OF ELECTRICAL CONDUCTIVITY AND DIELECTRIC PROPERTIES OF $\text{TlGa}_{1-x}\text{InSe}_2$ SOLID SOLUTIONS IRRADIATED WITH γ -QUANTUMS

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The electrical and dielectric properties of $\text{TlGa}_{1-x}\text{InSe}_2$ solids in a large temperature and frequency range, as well as the effect of γ -rays at a dose of 75 Mrad on them, were explained by means of possible mechanisms. Based on the results of the study of the temperature dependence of the electrical conductivity of $\text{TlGa}_{1-x}\text{InSe}_2$ ($x=0; 0.1; 0.2$) solid solutions before and after irradiation with different doses in the temperature range of 100...300 K, the temperature and frequency intervals of the presence of jump conductivity were determined. During the study of the temperature dependence of the electrical conductivity of solid solutions, the existence of two parts in the change of the specific conductivity was shown. In the studied solid solution samples, there is a sharp increase in the dielectric constant at a certain critical temperature. Above room temperature, the experimental points of $\ln(\epsilon)$ are characterized by a straight line as a function of $1/T$, which is a characteristic property of ionic conductivity.

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INTRODUCTION

Solid metal chalcogenides are a promising class of materials for detecting high-intensity radiation due to their wide range of semiconducting properties and the potential for diverse stoichiometries and crystal structures. Thallium-containing metal chalcogenides are particularly noteworthy because they can achieve high mass densities, which in turn enhance the material's absorption coefficient for γ -rays. A distinctive feature of solid solutions is the structural disorder caused by irregular occupancy of lattice sites in the crystal matrix, while maintaining ideal crystallographic symmetry. This phenomenon, termed "translational disorder", arises from the exchange of anions and cations, for instance, through substitution that retains the lattice type depending on the size of the replacing atom. During this process, the arrangement of atoms within the lattice sites is changed, disrupting the translational order of the crystal and ultimately resulting in a disordered system [1–3].

Modifications to the composition of semiconductor solid solutions affect their photoelectric, dielectric and optical properties, allowing for precise data acquisition. Additionally, altering the composition influences the levels within the band gap of the semiconductor, which defines the material's electronic structure. This, in turn, governs the electronic behavior and properties of the semiconductor, a critical factor in the development of devices.

These studies further suggest that the solid solutions under investigation exhibit structural imperfections or disorder. Notable features in the energy spectrum of disordered substances are observed: within these substances, the relaxation mechanisms of dielectric properties are revealed, alongside distinctive characteristics in charge transport processes [2–7].

OBJECT AND METHODOLOGY OF THE RESEARCH

The $\text{TlGa}_{1-x}\text{InSe}_2$ solid solution was synthesized by Bridgman-Stockbarger method. Comprehensive studies of dielectric permeability and electrical impedance were conducted within the frequency range of $25 \cdot 10^6$ Hz, employing the E7-28 wide-range immittance measurement device

RESULTS AND DISCUSSION

A) DIELECTRIC PERMITTIVITY OF $\text{TlGa}_{1-x}\text{InSe}_2$ SOLID SOLUTIONS IRRADIATED WITH γ -QUANTA

The observed temperature-dependent behavior of the dielectric constant, $\epsilon(T)$ in bulk materials samples is likely attributed to the movement of ions along defects, a characteristic feature of superionic conductors. According to literature, materials exhibiting superionic conductivity display not only an exponential increase in electrical conductivity but also a similar exponential rise in dielectric permittivity as temperature increases. At high temperatures, the dielectric constant can be several times higher than its value at lower temperatures.

Fig. 1 illustrates the temperature dependence of the dielectric constant, $\epsilon(T)$, for $\text{TlGa}_{1-x}\text{InSe}_2$ ($x=0; 0.1$). In the studied bulk samples, a sharp increase in the dielectric constant is observed at a specific critical temperature. This sharp rise in dielectric permittivity with increasing temperature is explained by the predominance of ionic conductivity at temperatures above the critical threshold, as confirmed by the temperature dependence of the dielectric constant [2–4, 13–14].

A change in the slope of $\ln(\epsilon)$ versus $(1/T)$ above the threshold temperature, where a significant shift in the dielectric constant occurs, indicates the ionic nature of the dielectric behavior. This change is attributed to the

diffusion of Tl^+ ions into the voids within the $TlGa_{1-x}InSe_2$ ($x=0; 0.1; 0.2; 0.3$) sublayers of the bulk material.

The change observed in the temperature dependence of $\varepsilon(T)$ for the bulk materials of the $TlGa_{1-x}InSe_2$ system ($x = 0; 0.1; 0.2; 0.3$) is a result of a phase transition caused by the disruption of the Tl sublayer. This conduction behavior is typical for superionic conductors. The $\varepsilon(T)$ dependence in bulk material samples is likely related to the movement of ions along the defects.

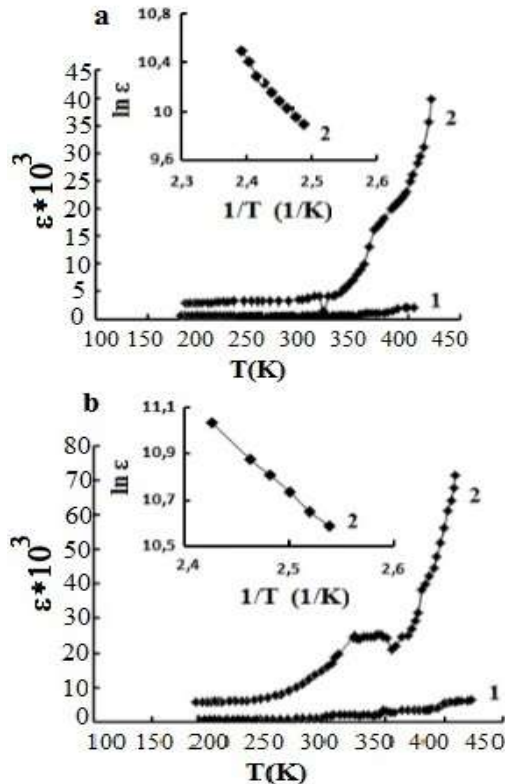


Fig. 1. Temperature dependence ($\varepsilon(T)$) curve of the dielectric constant of the $TlGa_{1-x}InSe_2$ solid solution that was not irradiated and irradiated at a dose of 0.75 MGy: a – $x=0.1$, b – $x=0.2$; 1 – 0 MGy, 2 – 0.75 MGy

Furthermore, the anomaly observed in the temperature dependence near room temperature is attributed to the disordering of the Tl^+ sublayer, which leads to a reduction in the dielectric constant at low frequencies. Consequently, the significant increase in the dielectric constant at low frequencies in bulk materials of the $TlGa_{1-x}InSe_2$ system ($x=0; 0.1; 0.2; 0.3$) is linked to the ion polarization mechanism caused by the weak interaction of thallium ions.

As previously noted [6, 9–11], impedance spectroscopy is the most appropriate method for studying the electrophysical processes that occur during metal contact with ion-conducting materials.

The experimental results suggest that, in addition to band-to-band conduction, hopping conductivity is also observed, indicating that the crystals possess a disordered structure. This signifies a mixed and disordered crystal structure, where irregularities in the lattice lead to distortions and the mixing of atomic sequences. These irregularities contribute to the

existence of hopping conductivity and the determination of the material's electrical conductivity properties. This conductivity is estimated based on localized levels over a broad temperature range in the compounds studied.

These findings also suggest that the solid solutions under investigation exhibit structural imperfections or are disordered materials.

B) EFFECT OF γ -RAYS ON SUPERIONIC CONDUCTIVITY OF $TlGa_{1-x}InSe_2$ SOLID SOLUTIONS

The article investigates the ionic conductivity of $TlGa_{1-x}InSe_2$ ($x = 0; 0.1; 0.2; 0.3$) materials in the temperature range of 100...450 K under γ -irradiation.

In addition, the temperature-dependent electrical conductivity ($\sigma(T)$) of the bulk $TlGa_{1-x}InSe_2$ ($x = 0; 0.1; 0.2; 0.3$) materials was studied both before and after irradiation at a dose of 0.75 MGy. The comparative results are illustrated in the Fig. 2 [13–15].

As shown in the figure, the irradiated samples at a dose of 0.75 MGy exhibit a noticeable increase in electrical conductivity at temperatures above a certain critical value. This increase indicates that ionic conductivity becomes dominant at temperatures above this critical threshold. One of the key indicators of ionic conductivity is the linear relationship between $\ln(\sigma \cdot T)$ and $1/T$ within the temperature range where ionic conductivity is present, as detailed in Fig. 2. Based on the experimental data, the dependence of $\ln(\sigma \cdot T)$ on $1/T$ in the temperature range of ionic conductivity follows the linear law for the studied bulk materials of $TlGa_{1-x}InSe_2$ ($x = 0 \dots 1.0$).

Dependence of activation energies of $TlGa_{1-x}InSe_2$ solid solutions (at $x = 0.1, 0.2, 0.3$) on composition and radiation dose

Composition	Dose, MGy	E_a , eV
$TlGa_{1-x}InSe_2$ ($x=0.1$)	0	0.16
	0.75	0.11
$TlGa_{1-x}InSe_2$ ($x=0.2$)	0	0.15
	0.75	0.07
$TlGa_{1-x}InSe_2$ ($x=0.3$)	0	0.13
	0.75	0.08

Based on this equation, the activation energy values for $TlGa_{1-x}InSe_2$ ($x = 0; 0.1; 0.2; 0.3$) were determined and analyzed depending on their variation. The concentration and radiation dose are detailed in Table.

In $TlGa_{1-x}InSe_2$ ($x = 0; 0.1; 0.2; 0.3$) samples, the significant increase in electrical conductivity observed at temperatures above room temperature can be explained by the sharp rise in the population of highly mobile Tl^+ ions [7, 11]. This increase leads to a phase transition into the superionic state, where ions exhibit high mobility, significantly contributing to the overall conductivity of the material.

The obtained data indicate a clear change in the electrical conductivity mechanisms within the investigated bulk materials. At temperatures below room temperature, electronic conductivity predominates. However, as the temperature exceeds room temperature, the sharp increase in electrical conductivity due to stepwise mechanisms far surpasses

that of ionic conductivity. This transition is attributed to a phase transition resulting from the breakdown or melting of the Tl sublayer in $\text{TlGa}_{1-x}\text{InSe}_2$ ($x=0\ldots 1.0$) bulk materials.

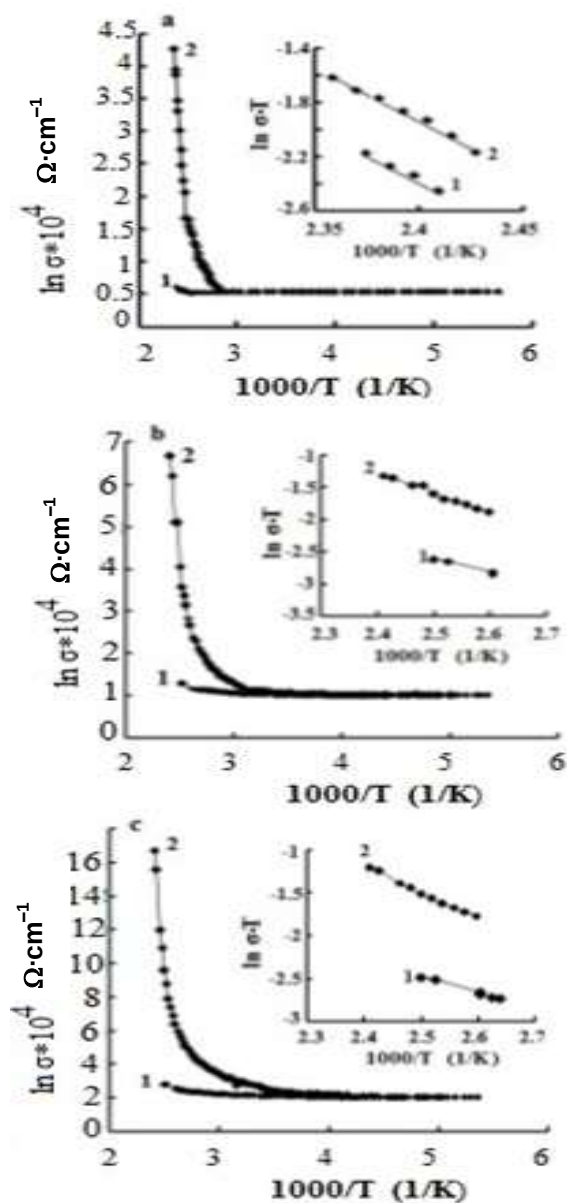


Fig. 2. Temperature dependence of the electrical conductivity of $\text{TlGa}_{1-x}\text{InSe}_2$ solid solutions that was not irradiated and irradiated at a dose of 0.75 MGy: a – $x=0.1$, b – $x=0.2$, c – $x=0.3$; 1 – 0 MGy, 2 – 0.75 MGy

CONCLUSIONS

The electrical conductivity and dielectric constant of $\text{TlGa}_{1-x}\text{InSe}_2$ solid solutions irradiated with γ -quanta were examined in the temperature range of 100...450 K and frequency range of 25·10⁶ Hz. The presence of characteristics associated with the transition of the crystal to the superion state was identified in the temperature dependence of the electrical conductivity ($\sigma(T)$) at temperatures above 300 K. A mechanism explaining the nature of the observed conductivity was proposed, demonstrating that the observed ionic conductivity is linked to the diffusion of Tl^+ ions through vacancies in the thallium sublattice.

REFERENCES

1. R. Waser. Nanoionics-based resistive switching memories // *Nature Materials*. 2007, v. 6, p. 833-840.
2. A.M. Panich, R.M. Sardarly. *Physical Properties of the Low Dimensional A^3B^6 and $A^3B^3C^6$ Compounds*. Nova Science Publishers, NY, 2010, 310 p.
3. A.U. Sheleg, V.V. Shautsova, V.G. Hurtavy. Low-Temperature X-Ray Studies of TlInS_2 , TlGaS_2 and TlGaSe_2 single crystals // *Journal of Surface Investigation*. 2013, v. 7, N 6, p. 1052-1055.
4. F.T. Salmanov. Influence of γ -irradiation on superionic conductivity of $(\text{TlGaSe}_2)_{1-x}(\text{TlInSe}_2)_x$ solid solutions // *Journal of Radiation Researches*. 2019, v. 6, N 1, p. 5-10.
5. R.M. Sardarli, F.T. Salmanov, N.A. Aliyeva, and R.M. Abbasli. Impedance Spectroscopy of $(\text{TlGaSe}_2)_{1-x}(\text{TlInSe}_2)_x$ Solid Solutions in Radio Frequency Range // *Modern Physics Letters B*. 2020, v. 34, N 11, p. 2050113; <https://doi.org/10.1142/S0217984920501134>
6. A.F. Qasravi, N.M. Gasanly. Photoelectronic, optical and electrical properties of TlInS_2 single crystals // *Phys. Status Solidi A*. 2003, v. 199, p. 277-283.
7. G.E. Delgado, A.J. Mora, F.V. Perez. Crystal structure of the ternary semiconductor compound thallium gallium sulfide TlGaS_2 // *Physica B*. 2007, v. 391, p. 385-388.
8. R.M. Sardarly, F.T. Salmanov, N.A. Aliyeva, R.N. Mehdiyeva, S.M. Gakhramanova. A conductivity of superionic thallium sulfide crystals exposed to γ -irradiation // *Modern Physics Letters B*. 2021, p. 2150504-1-2050113-9.12; <https://doi.org/10.1142/S0217984921505047>
9. R.M. Sardarly, G.M. Ashirov, L.F. Mashadiyeva, N.A. Aliyeva, F.T. Salmanov, R.Sh. Agayeva, R.A. Mamedov, and M.B. Babanly. Ionic conductivity of the Ag_8GeSe compound // *Modern Physics Letters B*. 2022, v. 36, N 32n33, p. 2250171; <https://doi.org/10.1142/S0217984922501718>
10. R.M. Sardarly, O.A. Samedov, F.T. Salmanov, N.A. Aliyeva. Impact of γ -irradiation on dielectric and electric properties of TlInS_2 <V> crystals // *PAST*. 2019, N 2(120), p. 30-33.
11. R.M. Sardarly, O.A. Samedov, A.P. Abdullaev, F.T. Salmanov. Superionic Conductivity in One-Dimensional Nanofibrous TlGaTe_2 Crystals // *Japanese Journal of Applied Physics*. 2011, v. 50, N 5, p. 05FC09.
12. F. Mamedov, S. Nabieva, S. Melikova, G. Imanova, O. Tagiyev, E. Mansurova. Effect of annealing on thermoelectric properties of crystals $\text{Yb}_x\text{Bi}_{2-x}\text{Te}_3$ // *Modern Physics Letters B*. 2024, v. 38(12), p. 2450090; <https://doi.org/10.1142/S0217984924500908>
13. Sardarly Rauf, Sardarli Arzu, Salmanov Famin. Phase Transition in TlS , TlSe and TlInS_2 Crystals Caused by Nanoscale Defects // *International Journal of Theoretical and Applied Nanotechnology (IJTAN)*. 2018, v. 6, p. 5-10.
14. L.H. Ye, K. Hoang, A.J. Freeman, S.D. Mahanti, J. He, T.M. Tritt, M.G. Kanatzidis. First-Principles Study of the Electronic, Optical, and Lattice

ДОСЛІДЖЕННЯ ЕЛЕКТРОПРОВІДНОСТІ ТА ДІЕЛЕКТРИЧНИХ ВЛАСТИВОСТЕЙ ТВЕРДИХ РОЗЧИНІВ TlGa_{1-x}InSe₂, ОПРОМІНЕНИХ γ -КВАНТАМИ

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За допомогою можливих механізмів пояснено електричні та діелектричні властивості твердих тіл TlGa_{1-x}InSe₂ у широкому діапазоні температур і частот, а також вплив на них γ -випромінювання при дозі 75 Мрад. За результатами дослідження температурної залежності електропровідності твердих розчинів TlGa_{1-x}InSe₂ ($x=0; 0,1; 0,2$) до та після опромінення різними дозами в інтервалі температур 100...300 К встановлено температуру та визначено частотні інтервали наявності стрибкової провідності. При дослідженні температурної залежності електропровідності твердих розчинів було показано існування двох частин у зміні питомої провідності. У досліджуваних зразках твердого розчину спостерігається різке зростання діелектричної проникності при певній критичній температурі. Вище кімнатної температури експериментальні точки $\ln(\epsilon)$ характеризуються прямою лінією як функція $1/T$, яка є характерною властивістю іонної провідності.