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AC CONDUCTIVITY OF TISe0.5S0.5 COMPOUND IRRADIATED WITH GAMMA QUANTA

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Abstract: The electrical conductivity, complex dielectric constant, and dielectric loss angle of a $TlSe_{0.5}S_{0.5}$ solid solution sample were examined before and after exposure to 100kGy and 250kGy of gamma radiation, both at room temperature and in a variable electric field. Several parameters were then computed.

Keywords: gamma radiation, AC conductivity, relaxation time.

1. Introduction

The literature has established that TIS and TISe semiconductors belong to the A³B⁶ class and feature a chain structure. The crystal structure of TISe exhibits tetragonal symmetry, with a space group of D_{4h}^{18} -14/mcm, lattice parameters of a=b=8.020 Å, c=6.7910 Å, and Z=4. TIS monocrystals are isostructural with TISe and also crystallize in tetragonal symmetry [1-4]. The structural parameters of tetragonal TIS are a=b=7.785 Å, c=6.802 Å, Z=8 D_{4h}^{18} -14/mcm [2, 3, 5].

The discovery that compounds in the A^3B^6 class exhibit superionic conductivity near room temperature has sparked a recent surge in interest in their investigation [6-10]. In recent years, the demand for ion-conducting materials has grown, driven by the successful application of lithium-ion-based devices in electronics, energy, and industry. Since the operation of these devices relies on electrochemical processes at the electrode-ion-conducting interface and within the crystalline electrolyte, studying ion transport properties in these systems is crucial [2, 11–13].

Since Tl-based compounds are photosensitive, the optical properties of crystals and solid solutions of these compounds have also been studied [2, 3, 14–16].

A thorough examination of the literature reveals that TISe and TIS crystals have been extensively investigated at temperatures below room temperature, making them suitable for practical applications. Thus, these compounds are particularly well-suited for the preparation of infrared-sensitive pyroelectric receivers operating over a wide temperature range [3–5, 17–19].

Recently, various electrophysical characteristics of TISe and TIS crystals have been investigated at temperatures below room temperature. This study examined several electrical properties of TIS and TISe solid solutions at room temperature in a variable electric field [4–12, 17–19].

2. Experimental and methods

The studied TIS and TISe-based $TISe_{1-x}S_x$ solid solution mono crystals were grown using the Bridgman-Stockbarger method. After synthesis, the substance is placed inside the quartz

ampoule and deaerated. After determining the cultivation and brewing temperature, the temperature of the melting zone in the electric furnace is set according to the selected temperature and stabilized automatically using a thermal regulator (RIF-101). After 2-3 hours of stabilization, the Pt–Pt/Rh thermocouple connected to the 2-coordinate recording device is released into the furnace at a rate of 0.2 cm/min, starting from point "0" to the end of the brewing zone. The temperature of the brewing zone was maintained at $\approx 60\%$ of the melting temperature of the substance being grown. Once the temperature and necessary gradient in the zone are established, the quartz ampoule (filled with the material to be grown) is connected to the driving mechanism and inserted into the furnace. After stabilizing for 1.5–2 hours, the container is moved, initiating the crystallization process.

Conductivity measurements were carried out using the four-probe method with a digital impedance E7-25 device. Electrical conductivity was measured in the frequency range of 25-10⁶ Hz at room temperature (300 K). The measurements were conducted in a quasi-stationary continuous heating mode within a nitrogen cryostat at a rate of ≈ 0.1 K/min, both parallel and perpendicular to the crystallographic "c" axis. The applied electric field amplitude did not exceed 1 V/cm. After preliminary measurements of tg δ (T) and ϵ (T), the samples were exposed to γ - irradiation from a standard ⁶⁰Co source. The irradiation dose was incrementally increased in each sample through sequential gamma exposures of 100 and 250 kGy [6, 9, 10, 12, 17–19].

3. Experimental results and discussion

In this study, the frequency dependence of the real (ϵ') and imaginary (ϵ'') parts of the dielectric permittivity, the tangent of the loss angle (tg δ), and the ac-conductivity (σ_{ac}) of the TlSe_{0.5}S_{0.5} crystal exposed to different doses were measured at room temperature in the frequency range of 25–10⁶ Hz. The sample was prepared as plates with a thickness of 2800 µm and a surface area of 32.3 · 10⁻² cm². Measurements were carried out at room temperature (300K), and the applied field amplitude did not exceed 5 V/cm.

Figure 1 shows the frequency dependence of the electrical conductivity for the TlSe_{0.5}S_{0.5} crystal both before and after exposure to radiation at T = 300 K, with doses of 100 and 250 kGy. Compared to the unirradiated sample, the TlSe_{0.5}S_{0.5} crystals exposed to gamma radiation at doses of 100 kGy and 250 kGy exhibit a weaker frequency dependence (curves b and c in Figure 1). This change in $\sigma(lgv)$ dependence of the TlSe_{0.5}S_{0.5} crystal irradiated at doses of 100 kGy and 250 kGy is attributed to radiation annealing, which leads to the "treatment" of initially uncontrolled defects in the thallium sulfide crystal.



Fig. 1. Frequency dependence of the permittivity of a $TlSe_{0.5}S_{0.5}$ sample, unirradiated (a) and irradiated with γ -rays (b - 100 kGy; c - 250 kGy).

The $\sigma(lgv)$ curves, obtained from the experimental results for unirradiated and irradiated TlSe_{0.5}S_{0.5} crystals at doses of 100 and 250 kGy (curves a, b, c in Figure 1), can be divided into two parts. At room temperature, a slight increase in the frequency dependence of the conductivity is observed in the low- frequency range v < 10³ Hz and adheres to the law $\sigma_{ac} \sim f^{0.6}$.

Charge transport mechanisms must be considered when a variable electric field is applied to the capacitor plates by a dielectric with a disordered structure. In this case, electrical conductivity occurs through several mechanisms, depending on temperature and frequency range [4–12]: transport of charge carriers beyond the mobility limit in delocalized states; hopping transport of excited charge carriers in localized states near the mobility limit; hopping transport of charge carriers over localized states; and primarily ionic charge transfer (superionic conductivity). At high frequencies, up to 10^6 Hz, charge carriers are transferred through a delocalized state. In disordered systems, this behaviour is characteristic of the hopping mechanism of charge transport [5, 11, 14] and [17]. Electrical transport processes in a variable field with a wide distribution of hopping distances are also considered [10–14, 17].

Figure 2 shows the frequency dependence of real (ϵ ') and imaginary (ϵ ") parts of the dielectric constant for TlSe_{0.5}S_{0.5} crystals. Measurements were conducted at T = 300 K and radiation doses D = 0; 100 and 250 kGy. As the frequency of the applied electric field increases, both (ϵ ') and (ϵ ") decrease. As shown in Figure 2, ϵ ' decreases with increasing frequency in the range of 25–10³ Hz, and at higher frequencies ($\nu > 10^3$ Hz), ϵ ' becomes weakly dependent on frequency. In this frequency range, a stronger dispersion is observed in the imaginary part of the dielectric constant.



Fig. 2. Frequency dependence of the real (ε') and imaginary (ε'') parts of the complex dielectric constant of the TlSe_{0.5}S_{0.5} crystal, unirradiated (a) and irradiated with γ -rays (b - 100 kGy; c - 250 kGy).

The observed characteristics indicate the presence of dispersion, manifested by a decrease in the values of the components of the complex dielectric constant as the frequency of the electric field increases.

The frequency dependence of the imaginary part of the complex permittivity (ϵ ") typically reaches a maximum in dielectrics exhibiting relaxation polarization due to activation processes. However, as seen from Figure 2, both ϵ components decrease monotonically with increasing frequency.

Furthermore, the curves in Figure 2 show that after irradiation, the maximum values of both the real and imaginary parts of the complex dielectric constant decrease.



Fig. 3. Frequency dependence of the dielectric loss angle tangent of a $TlSe_{0.5}S_{0.5}$ sample, unirradiated (a) and irradiated with γ -rays (b – 100 kGy; c – 250 kGy).

Figure 3 shows the frequency dependence of the dielectric loss angle tangent for the TlSe_{0.5}S_{0.5} crystal at various temperatures, both before and after irradiation with doses of 100 kGy and 250 kGy. In both unirradiated and gamma-irradiated samples, the dielectric loss angle tangent at the dependence of tg $\delta(v)$ reaches a minimum at 10⁶ Hz and a maximum in the low-frequency range (10³-10⁴ Hz). This behaviour in the frequency dependence of the dielectric loss angle tangent suggests the presence of relaxation losses and current conduction in the crystals under study.

Furthermore, as shown in the figure, a decrease in the maxima of the frequency dispersion of the dielectric loss angle is observed following radiation exposure.

4. Conclusions

The relaxation time for the studied sample can be calculated from the maxima observed in the tg $\delta(v)$ curves of the TlSe_{0.5}S_{0.5} crystal. The calculated values are $\tau = 10^{-3}$ s before irradiation and $\tau = 10^{-4}$ s after irradiation.

References

- H. Hahn, W. Kleingler, Röntgenographische Beiträge zu den Systemen Thallium/Schwefel, Thallium/Selen und Thallium/Tellur, Z. Anorg. Chem., 1949, Vol. 260, No. 1–3, pp. 110–119, <u>https://doi.org/10.1002/zaac.19492600109</u>
- A.M. Panic, Electronic properties and phase transitions in low-dimensional semiconductors, *Phys. Condens. Matter*, 2008, Vol. 20, No. 29, Article ID: 293202, <u>https://doi.org/10.1088/0953-8984/20/29/293202</u>
- 3. R.S. Itoga, C.R. Kannewurf, Electrical resistivity, Hall effect and optical absorption in TIS, TlS_{0.5}Se_{0.5} and TlSe. *J. Phys. Chem. Solids*, 1971, Vol. 32, No. 6, pp. 1099–1110, https://doi.org/10.1016/S0022-3697(71)80168-3
- 4. R.A. Mammadov, F.T. Salmanov, AC conductivity of crystals of TlSe_{1-x}S_x (x = 0; 0.1; 0.2) solid solution, *Silk Road Int. Sci. Res. Conf.*, Dec. 8–9, 2022, LSU, Azerbaijan, ISBN: 978-625-7898-68-3, pp. 548–549, Available at: <u>https://www.silkroadcongress.co.uk/books</u>

- 5. R.A. Mammadov, AC conductivity of $TlSe_{1-x}S_x$ (x = 0; 0.2; 0.4) solid solutions, *Young Researcher Sci. Pract. J.*, 2022, Vol. VIII, No. 2, pp. 18–22, Available at: <u>Issue page</u>, <u>PDF</u>
- R.M. Sardarly, O.A. Samadov, A.P. Abdullayev et al., Ionic conductivity and dielectric relaxation in TlGaTe₂ crystals irradiated with γ-quanta, *Semiconductors*, 2013, Vol. 47, pp. 696–701, https://journals.ioffe.ru/articles/4977
- R.M. Sardarly, A.P. Abdullayev, N.A. Aliyeva et al., Superionic conductivity of solid solutions (TlGaSe₂)_{1-x}(TlInS₂)_x, *Semiconductors*, 2018, Vol. 52, pp. 1111–1114, https://doi.org/10.1134/S1063782618100184
- R.M. Sardarly, G.M. Ashirov, L.F. Mashadiyeva et al., Ionic conductivity of the Ag₈GeSe₆ compound, *Mod. Phys. Lett. B*, 2022, Vol. 36, No. 32–33, Art. No. 2250171, https://doi.org/10.1142/S0217984922501718
- 9. R.M. Sardarli, N.A. Aliyeva, F.T. Salmanov, R.A. Mammadov, Ion conductivity of $TlSe_{1-x}S_x$ (x = 0.1) solid solutions irradiated with γ -quanta by the method of impedance spectroscopy, 2023, pp. 69–71, Available at: <u>https://www.researchgate.net/</u>
- R.M. Sardarly, N.N. Hajiyeva et al., Radiation-stimulated conversion to superionic state of TlSe and TlS crystals. *Probl. At. Sci. Technol.*, 2024, No. 4, pp. 23–28, <u>https://doi.org/10.46813/2024-152-023</u>
- R.M. Sardarly, O.A. Samedov, N.A. Aliyeva et al., Conductivity on the localized states of (TlInSe₂)_{1-x}(TlGaTe₂)_x solid solutions, *Semiconductors*, 2015, Vol. 49, No. 12, pp. 1704–1709, <u>https://journals.ioffe.ru/articles/42557</u>
- 12. N.A. Aliyeva, R.A. Mammadov, G.M. Rushanova, Effect of gamma quanta on electrical properties of $(TIS)_{1-x}(TISe)_x$ (x = 0; 0.1) solid solutions. 2023, pp. 168–169, Available at: <u>https://inis.iaea.org/records/wpxpc-bnv47</u>
- 13. O.A. Samadov, M.M. Zarbaliyev et al., Semiconductor thallium chalcoindianates and alloys with rare-earth elements, *J. Radiat. Res.*, 2018, Vol. 5, No. 2, pp. 234–241, Available at: <u>Issue page</u>, <u>PDF</u>
- 14. N.F. Mott, E.A. Davis, *Electronic processes in noncrystalline matter*, Moscow: Mir, 1982, 368 p., Available at: <u>https://books.google.az/</u>
- N.N. Gadzhieva, G.B. Akhmedova, R.A. Mammadov, Structures of HDPE/GaAs and HDPE/GaAs<Te> composites irradiated with gamma quanta. *Probl. At. Sci. Technol.*, 2024, No. 4(152), pp. 20–22, <u>https://doi.org/10.46813/2024-152-020</u>
- 16. R.M. Sardarly, F.T. Salmanov, N.A. Alieva, Optical transitions at the absorption edge in TlGaSe₂ and TlInS₂ crystals subjected to γ-radiation, *Opt. Spectrosc.*, 2019, Vol. 127, pp. 454–458, <u>https://doi.org/10.1134/S0030400X19090224</u>
- 17. R.M. Sardarly, O.A. Samadov et al., Conductivity over localized states in $(TIInSe_2)_{1-x}(TIGaTe_2)_x$ solid solutions under γ -irradiation, *J. Radiat. Res.*, 2016, Vol. 3, No. 1, pp. 5–13, Available at: Issue page, PDF
- 18. R.M. Sardarly, O.A. Samedov et al., Impact γ-irradiation effects on dielectric and electric properties of TlInS₂<V> crystals, *Probl. At. Sci. Technol.*, 2019, No. 120(2), pp. 30–33, Available at: <u>PDF</u>
- R.M. Sardarly, F.T. Salmanov, N.A. Aliyeva, R.M. Abbasli, Impedance spectroscopy of (TlGaSe₂)_{1-x}(TlInSe₂)_x solid solutions in RF range, *Mod. Phys. Lett. B*, 2020, Vol. 34, No. 11, Art. No. 2050113, <u>https://doi.org/10.1142/S0217984920501134</u>

АС-ПРОВОДИМОСТЬ СОЕДИНЕНИЯ ТІSe0.5S0.5 ОБЛУЧЕННОГО ГАММА-КВАНТАМИ

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Резюме: Электропроводность, комплексная диэлектрическая проницаемость и угол диэлектрических потерь образца твердого раствора TlSe_{0.5}S_{0.5} были исследованы до и после воздействия гамма-излучения 100 кГр и 250 кГр как при комнатной температуре, так и в переменном электрическом поле. Затем были рассчитаны несколько параметров.

Ключевые слова: гамма-излучение, АС-проводимость, время релаксации.

QAMMA KVANTLARLA ŞÜALANMIŞ TISe0.5S0.5 BİRLƏŞMƏSİNİN AC-KEÇİRİCİLİYİ

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Xülasə: Şüalanmadan əvvəl və 100 kGy; 250 kGy qamma kvantlarla şüalanmadan sonra TlSe_{0.5}S_{0.5} bərk məhlul nümunəsinin otaq temperaturunda və dəyişən elektrik sahəsində elektrik keçiriciliyi, kompleks dielektrik sabiti və dielektrik itki bucağı öyrənilmişdir və bir sıra parametrlərin qiymətləri hesablanmışdır.

Açar sözlər: qamma şüalanma, AC-keçiriciliyi, relaksasiya müddəti.