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CHARACTERISTICS OF ELECTRICAL CURRENT RELAXATION IN MONOCRYSTAL $TlInSe_2$

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This study investigates the volt-ampere characteristic of the $TlInSe_2$ compound at various temperatures, as well as the relaxation of electric current across different voltages (corresponding to varying electric field intensities) at a temperature of 300 K. From the volt-ampere characteristic analysis, we calculated the concentration of free charge carriers, mobility, and trap concentration, yielding values of $n_0 = 5.45\cdot10^{18}$ cm^{-3}, $\mu = 3.03\times10^{-3}$ cm^2/V·s, and $N_t = 5.2\cdot10^{10}$ cm^{-3}, respectively. The dependence of electrical conductivity on temperature revealed local energy levels with activation energies of 0.2 eV and 0.53 eV in the $TlInSe_2$ crystal. It was found that at low, constant voltages (where the electric field intensity $E < 25$ V/m), relaxation processes occur within the $TlInSe_2$ compound, leading to a decrease in current due to charge accumulation. An increase in current was observed at higher voltages (where $E > 25$ V/m). This increase was attributed to the injection of charge carriers from the contacts, the discharge of charges accumulated near the contact under the influence of the electric field, and the partial discharge of electron centres.
Keywords: *Volt-ampere characteristics; Current relaxation; Current intensity; Electric field intensity; Charge carriers; Local energy levels*
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1. INTRODUCTION

Studying the structure and physical properties of materials used in electronics and spintronics under external influences is essential. These external factors, which include conditions experienced during the use of these materials in various fields like space technologies, have prompted extensive recent research. This research focuses on understanding functional materials that are subjected to various conditions, such as temperature, pressure, and radiation [1-6]. Chalcogenide semiconductors play a significant role among functional semiconductors. Investigating their structure and physical properties is critical for developing converters that can operate effectively under diverse conditions. Consequently, both the structural components and electronic processes of these materials are being explored. It has been established that physical properties are influenced by the type, stoichiometric amounts, and valence of the anions and cations present [7-11]. The generation and capture of charge carriers occurring at local levels within the forbidden zone of semiconductor materials significantly influence the performance of devices built from these materials, including their electrical characteristics. As a result, the dark current can vary over time based on the voltage applied to the crystal. In some instances, even with a constant voltage, the current may either increase or decrease, a phenomenon known as current relaxation. This relaxation depends on the nature, concentration, and energy state of the local energy levels within the studied crystals. The changes in the charges accumulated in these local levels, which are created by various structural defects within the forbidden zone of the crystal, lead to current relaxation. Additionally, the nature of this relaxation is influenced by the voltage applied to the system. Current relaxation and the accumulation of electric charge occur due to the presence of local levels in the forbidden zone, which are caused by inhomogeneities, mixtures, and other factors in semiconductor materials. Investigating current relaxation in semiconductor materials offers valuable insights into the kinetics of the local levels situated in the forbidden zone.

The semiconductor compound $TlInSe_2$ belongs to the group of ternary $A^{III}B^{III}C_2^{VI}$ compounds and has a chain-like defective structure [12,13]. According to electronographic and X-ray crystallographic analyses, the $TlInSe_2$ compound is a structural-coordinated analogue of TlSe, obtained by substituting Tl^{3+} ions with In^{3+} ions [14]. The interest in these compounds is due to their small crystal structure size, high concentration of defects (10^{17} cm^{-3}), distinct electrical and photoelectric properties, and sharp anisotropy of physical properties in various crystallographic directions [15-17]. $TlInSe_2$ crystals are applied in various electronic technology fields, such as in X-ray detectors, tensor resistors, and more. Despite the available information about the electrical and photoelectric properties of $TlInSe_2$ crystals, as well as the effect of radiation on their physical properties, the kinetics of current flow in chain-like crystals and the kinetic parameters of trapping centres have not been thoroughly studied [18]. Therefore, studying the current relaxation in $TlInSe_2$ crystals, which affects their electrical and photoelectric properties, is of particular interest.

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This work aims to investigate the relaxation processes in $TlInSe_2$ crystals, to study the relationship between electrical conductivity and accumulated charges in the semiconductor, and to clarify the causes of the occurring physical processes. The volt-ampere characteristics of $TlInSe_2$ compounds and the current relaxation at various constant voltages were studied to achieve this.

2. EXPERIMENT

$TlInSe_2$ single crystals were grown using the directed crystallisation method as described in [12]. These crystals exhibit a tetragonal structure, with the following lattice parameters: a = 8.06 Å, c = 6.822 Å. Parallelepiped-shaped samples with dimensions of 6.66×0.84×0.43 mm (Sample No. 1) and 5.2×1.56×0.53 mm (Sample No. 2) were prepared from the obtained crystals. The silver paste was used as the contact material. After making the contacts, the samples were kept at room temperature for 24 hours to dry. The contacts were found to be ohmic.

During the experiments, the samples were placed in a cryostat, and the air inside the cryostat was evacuated to a pressure of 10^{-2} Pa. The width of the forbidden band of the sample was determined from the temperature-dependent graph of electrical conductivity at a constant voltage in the $TlInSe_2$ crystal, yielding a value of 1.23 eV.

In the study of current relaxation at a constant voltage, the ends of the sample were short-circuited after each measurement, and measurements were taken after 1 hour.

3. RESULTS AND DISCUSSIONS

Several theories explain how charge carriers injected from contacts in semiconductor materials contribute to electrical conductivity. According to these theories, charge carriers generate current in semiconductors either through the conduction band or via a hopping mechanism involving deep energy levels within the forbidden band. Consequently, the band structure of semiconductor materials is studied both theoretically and experimentally [19-23]. The physical properties of semiconductors are largely determined by the energy levels of electrons and ions. Therefore, understanding the formation of electronic processes and the electrical properties in these materials is essential [24-27].

Figure 1 illustrates the volt-ampere characteristic for the $TlInSe_2$ compound at a temperature of 300 K, displaying both the increasing voltage (curve 1) and decreasing voltage (curve 2) directions. From the graph (curve 1 in Fig. 1), it is evident that at very low voltages (300 K), the current intensity shows a sublinear dependence on voltage. This is followed by a linear region, after which a quadratic dependence is observed. When the voltage is increased to 25 V (electric field intensity of 3.75×10^3 V/m) and then reduced, the graph exhibits slight deviations above the initial curve, resulting in hysteresis-like behaviour. When the voltage is lowered to 10 V, the curve obtained during the decreasing voltage measurement closely resembles that of the increasing voltage direction, indicating that the hysteresis effect is practically negligible.

In subsequent measurements, raising the voltage to higher levels reached the point of complete trap filling, causing both the sample and contacts to heat up and leading to failure. Consequently, studies at high voltages were not conducted. Figure 2 presents the voltage-current characteristic obtained at various temperatures for the $TlInSe_2$ compound. As the temperature decreases, the sublinear region expands, the linear portion shortens, and the transition voltage from the linear to quadratic part shifts toward higher voltage values. At relatively high voltage levels, the exponent n varies between 2 and 5, indicating that the current intensity follows the relation $I\sim(U^n)$.

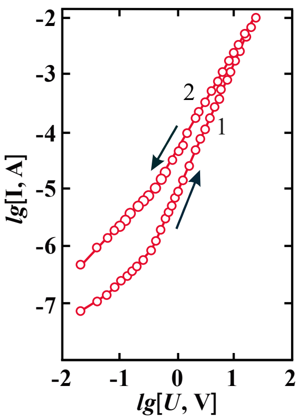

Figure 1. Volt-ampere characteristics in the direction of increasing (curve 1) and decreasing (curve 2) voltage in a $TlInSe_2$ single crystal at a temperature of 300 K

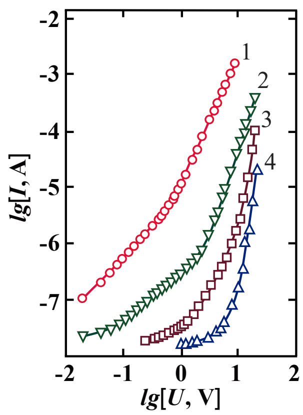

Figure 2. Volt-ampere characteristics of the $TlInSe_2$ compound at different temperatures. 1-300 K, 2- 250 K, 3- 198 K, 4-132 K

The graph depicting the temperature dependence of current intensity at a constant voltage in the TlInSe2 crystal reveals local energy levels with activation energies of 0.2 eV and 0.53 eV within the forbidden zone. Based on these results and existing literature, the conductivity mechanism in the $TlInSe_2$ crystal can be explained as follows:

According to the theory of space charge-limited currents, in semiconductors with trapped charge carriers, the current flows through the semiconductor primarily due to the equilibrium charge carriers when the electric field is small. As the applied electric field increases, the number of charge carriers entering the semiconductor from the contacts also rises. If the time ($t = L/\upsilon$) that equilibrium charge carriers spend travelling through the semiconductor (where L is the distance between contacts and υ is the drift velocity of these carriers) exceeds the lifetime of the charge carriers entering from the contacts, then the current flowing through the sample behaves in an ohmic manner.

$$j = en_0\mu\frac{U}{L}. \quad (1)$$

In this context, j represents the current density, n_0 is the equilibrium charge carrier concentration, μ denotes carrier mobility, U is the applied voltage, and L is the distance between the contacts.

As the applied voltage (or external field intensity) increases, the number of charge carriers injected from the metal contacts into the semiconductor also increases. These carriers gradually penetrate a larger area of the semiconductor. Once the charge carriers have filled the entire area of the semiconductor sample, the lifetime of the equilibrium charge carriers becomes shorter than that of the injected charge carriers. Consequently, the amount of current flowing through the semiconductor becomes limited by these charges. In this scenario, the current density can be expressed as follows:

$$j = \frac{9}{32\pi}\varepsilon\mu\frac{U^2}{L^3}. \quad (2)$$

Based on the transition voltage (U') value from the ohmic to the quadratic part:

$$n_0 = \frac{9\varepsilon U'}{32\pi e L^2}. \quad (3)$$

The equilibrium charge carrier concentration in the sample was determined using formula (3). From this calculation, we found that the equilibrium charge carrier concentration in the $TlInSe_2$ crystal is $n_0 = 5.45\cdot10^{18}$ cm^{-3}. We can now use this value in formula (1).

$$\mu = \frac{jL}{en_0U} = \frac{IL}{Sen_0U}. \quad (4)$$

The mobility value of the load carriers was calculated using formula (4), resulting in a value of $\mu = 3.03\cdot10^{-3}\,\frac{\text{cm}^2}{\text{V}\cdot\text{s}}$.

The volt-ampere characteristic provides information about the concentration of traps in the crystal by determining the transition voltage (U') from the ohmic region to the quadratic part:

$$N_t = \frac{CU'}{eLS}. \quad (5)$$

Using the formula (5), the calculated value of $N_t = 5.2\cdot10^{10}$ cm^{-3} (where e is the elemental charge, L is the distance between contacts, S is the contact area, and C is the electric capacity of the sample).

When studying the electrical properties of the $TlInSe_2$ compound, it was observed that the electric current relaxes at a constant voltage. Unlike other similar compounds, samples of $TlInSe_2$ do not always show a decrease in current intensity over time at low constant voltage values.

At an electric field intensity of E = 1.92 V/m *(U* = 0.1 V*)*, the current intensity initially increases over 120 seconds, then decreases for 60 seconds, before consistently rising again. At E = 3.83 V/m (U = 0.2 V), the current intensity decreases for the first 60 seconds, followed by a steady increase. At E = 19.2 V/m (U = 1 V), the current intensity decreases for the first 120 seconds, then increases briefly for 60 seconds, changes slowly for a while, and ultimately increases at a steady rate (see Fig. 3.a).

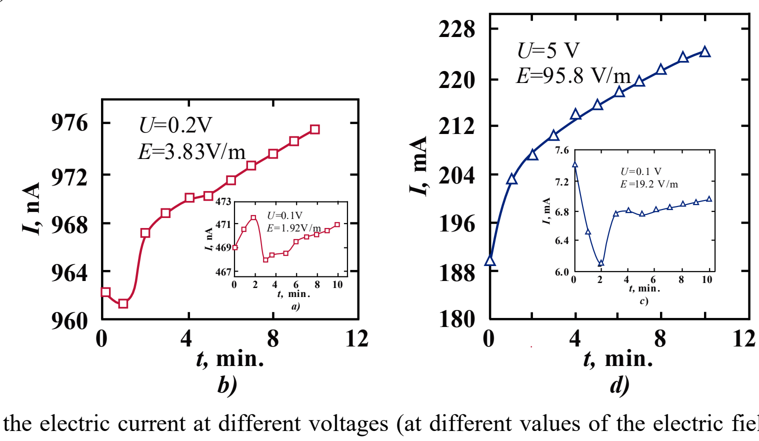

Figure 3. Relaxation of the electric current at different voltages (at different values of the electric field intensity) in the $TlInSe_2$ compound at a temperature of 300 K. *a)* E = 1.92 V/m, *b)* E = 3.83 V/m, c) E = 19.2 V/m, d) E = 95.8 V/m.

When the electric field intensity exceeds 25 V/m at constant voltage, only an increase in current intensity is observed over time. Initially, the rate of this increase is significant, but it gradually slows down, indicating that the current rises more slowly as time goes on (see Fig. 3.b). As the electric field intensity increases, the rate of increase in current intensity also rises.

Figure 4 illustrates the time dependence of the electric charge accumulated in the $TlInSe_2$ crystal at a voltage of 1 V (E = 19.2 V/m). The graph indicates that the rate of charge accumulation gradually decreases as the duration of the constant electric field applied to the sample increases.

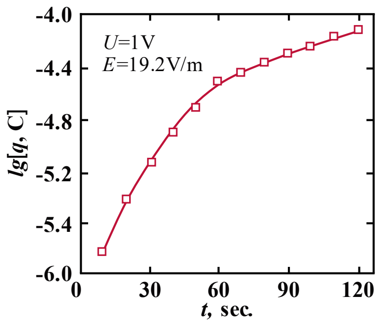

Figure 4. Time dependence of the electric charge accumulated in a TlInSe2 crystal under a constant voltage of 1 V.

The presence of a sublinear portion in the volt-ampere characteristic at low values of electric field intensity at a temperature of 300 K—namely, a decrease in electrical conductivity—can be attributed to small energy (shallow) traps located in the forbidden zone close to the conduction band. The existence of these shallow traps is further confirmed by the observed decrease in current intensity over time when a constant small voltage is applied. Moreover, at low voltages (where electric field intensity $E < 25$ V/m), most of the charge carriers injected from the contacts are captured by these traps. Consequently, both the concentration of injected additional charge carriers decreases, and the electric field created by the trapped electrons hinders the movement of electrons involved in conduction. This leads to a reduction in current intensity over time at the applied constant voltage. In such a case, the current limit by space charges within the sample is significantly lower than in the absence of traps.

Simultaneously, electrons can escape from the traps to the conduction band due to thermal excitation. When the rate at which electrons are excited from the traps equals the rate at which they are captured, the quasi-Fermi level rises, the concentration of free electrons increases, and a stationary state is established. One reason for the increase in current intensity after a decrease in constant voltage in the $TlInSe_2$ compound could be related to the effective filling of the traps.

Additionally, the presence of a barrier at the boundary with the anode causes the accumulation of volume charges near the anode when current flows through the sample. This accumulation makes it difficult for electrons to pass through this region, increasing resistance. Consequently, the voltage applied to that section also rises. An increase in the voltage drop near the anode enhances the electric field intensity in that region, which lowers the height of the barrier. As a result, current intensity increases again due to both the discharge of charges accumulated near the contact under the influence of the field and the partial release of the centres that capture electrons.

At relatively high voltages (where electric field intensity $E > 25$ V/m), the effects of thermal excitation and the filling of traps by captured electrons overshadow the decrease in current due to the trapping of injected electrons. Thus, instead of decreasing, the current shows an increase. The upward shift of the graph obtained during the voltage decrease in Fig. 1, compared to the graph obtained during the voltage increase, further suggests that current intensity increases due to the filling of traps during current flow through the crystal. Notably, the current intensity does not decrease during the first 60 seconds at a constant voltage of $U = 0.1$ V; instead, it increases, potentially due to the transfer of charge carriers from shallow levels in the forbidden zone to the conduction band.

CONCLUSIONS

In the studied $TlInSe_2$ compound, it was determined that electronic relaxation processes occur at a stable voltage (or electric field intensity). The crystals of the $TlInSe_2$ compound exhibit defects, which contribute to current relaxation in their electrical conductivity. Consequently, electric charge accumulates in the shallow energy levels present within the crystal structure. At relatively high electric field values, the current intensity increases again due to the discharge of charges that had accumulated near the contact, the partial discharge of centres that capture electrons, and the injection of charge carriers from the contacts. Since the measurements were conducted at a temperature of 300 K, the influence of charge carriers on electrical conductivity through a jump mechanism, involving deep levels within the forbidden zone, can be considered negligible at this temperature.

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ХАРАКТЕРИСТИКИ РЕЛАКСАЦІЇ ЕЛЕКТРИЧНОГО СТРУМУ В МОНОКРИСТАЛІ $TlInSe_2$

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У цьому дослідженні досліджується вольт-амперна характеристика сполуки TlInSe2 при різних температурах, а також релаксація електричного струму при різних напругах (що відповідають різним інтенсивностям електричного поля) при температурі 300 K. З аналізу вольт-амперної характеристики ми розрахували концентрацію вільних носіїв заряду, рухливість і концентрацію пастки, що дало значення $n_0 = 5.45\cdot10^{18}$ cm^{-3}, $\mu = 3.03\times10^{-3}$ cm^2/V·s і $N_t = 5.2\cdot10^{10}$ cm^{-3} відповідно. Залежність електропровідності від температури виявила в кристалі $TlInSe_2$ локальні енергетичні рівні з енергією активації 0,2 еВ і 0,53 еВ. Виявлено, що при низьких постійних напругах (де напруженість електричного поля E < 25 В/м) у сполуці $TlInSe_2$ відбуваються релаксаційні процеси, що призводять до зменшення струму внаслідок накопичення заряду. Збільшення струму спостерігалося при вищих напругах (де E > 25 В/м). Це збільшення пояснюється інжекцією носіїв заряду з контактів, розрядом зарядів, накопичених поблизу контакту під дією електричного поля, і частковим розрядом електронних центрів.

Ключові слова: *вольт-амперна характеристика; поточне розслаблення; сила струму; напруженість електричного поля; носії заряду; місцеві рівні енергії*