

## Study of Electrical Properties of TlInSe Layered Single Crystal

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**Abstract:** Single crystals layered compound TlInSeS were grown by modified Bridgman method. The crystals were identified by X-ray diffraction. In the present paper we describe and analyze conductivity and Hall effect measurements, performed on TlInSeS single crystals over the temperature range 148-558 K. The Hall Effect measurements revealed the extrinsic p-type conduction in the low temperature range of investigation. The dependence of the Hall coefficient conductivity, Hall mobility and charge carrier concentration on temperature were presented graphically. The analysis of the temperature dependent electrical conductivity and carrier concentration reveal that the acceptor level is located at 0.028 eV above the valence band of TlInSeS. From the obtained experimental data, the main characteristic parameters of the crystals have been estimated. Energy gap and acceptor concentration were 0.727 eV and  $1.305 \times 10^{14} \text{ cm}^{-3}$  respectively. The anomalous behavior of  $\ln \mu$  and  $\ln T$  in the low temperature range was checked.

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### 1. Introduction

Determination of the application potential for new semiconducting materials in solid state physics requires the growth and the examination of physical properties of the materials. Some binary and ternary layer-chain structured semiconductors such as the group III chalcogenides (InSe, GaSe, GaTe, TlInSe<sub>2</sub>, TlInS<sub>2</sub>, TlGaSe<sub>2</sub> etc.) have been explored thoroughly and proved already their applicability in solid state physics, for instance in optoelectronic devices [1]. One of the methods for modifying their physical properties is the production of solid solutions of these semiconductors. The experimental studies carried out on TlInS<sub>2</sub> and TlInSe<sub>2</sub> demonstrate that investigations for the physical properties of the TlInS<sub>2</sub>-TlInSe<sub>2</sub> system should be rewarding in view of the production of a new candidate material for optical devices in the visible range. Quaternary thallium chalcogenide TlInSeS is formed from the TlInS<sub>2</sub>-TlInSe<sub>2</sub> system and has a direct band gap of 2.328 at 10 K [2]. X-ray powder diffraction was used to characterize the TlInSeS crystals. The parameters of the monoclinic unit cell [3] were found to be  $a=0.72850$ ,  $b=0.45380$ ,  $c=0.78357$  nm and  $\beta=106.22^\circ$ . The lattice of TlInSeS crystals consists of two dimensional layers arranged parallel to the (001) plane. Each layer is oriented perpendicular with respect to the previous one. There is interlayer bonding between Tl and Se (S) atoms whereas the bonding between In and Se (S) atoms is an interlayer type. Infrared reflection and Raman spectroscopy were studied [4,5]. In view of possible optoelectronic device application in the visible region, a great deal of attention has been devoted to the study of the

photoelectric, optical and electrical properties of thallium chalcogenides [6-10]. Information about the trapping centers and their distributions in undoped TlInSeS layered single crystals by thermally stimulated current measurements were published [11]. The photoluminescence (PL) spectra of TlInSeS crystals have been investigated [12].

In spite of its importance in technological application as a candidate material for optical devices, and also for the understanding of its basic physics, so far very little information on the physical properties of this compound. To our knowledge, there is no information about the DC electrical conductivity and Hall coefficient and its temperature dependence. Hence, we report the results of the electrical conductivity and Hall coefficient of TlInSeS layered single crystal in the temperature range 148–558 K. As a result of this study we were able to calculate most of the physical parameters of this compound. To the best of the authors knowledge, the data presented in this work have not been reported before.

### 2. Experimental procedures

#### 2.1 Material and sample preparation

TlInSeS single crystals were grown from the melt by a modified Bridgman technique from a stoichiometric melt of starting materials sealed in evacuated ( $\approx 10^{-6}$  Torr) and carbon coated silica tubes (15 mm in diameter and about 25 cm in length) with a tip at the bottom. All the starting materials used were of extra pure elements (99, 9999 %). To prevent the ampoule from exploding, it was heated in a temperature gradient furnace, so that the sulphur

condensed at the cold end and slowly reacted with the heated elements at the hot end. The ampoule was kept at temperature higher than the melting point for 10 hours to ensure homogenization. The growth was achieved by lowering the ampoule from the hot side of the furnace at 1053 K, to the cold side at 703 K at a speed of 1.6 mmh<sup>-1</sup>. In the cold zone, the crystal cooled down slowly within a couple of days. The resulting ingots had no cracks and voids on the surface. The time required for this process was about 17 days. The crystals obtained have a layered structure (red in color) showed good optical quality and the freshly cleaved surfaces were mirror-like. The samples were identified by means of X-ray analysis. The X-ray diffraction analysis confirmed that TlInSeS compound have a monoclinic structure with the lattice parameters a=1.111, b=1.062 and c=1.592 nm. The samples for measurement were taken from the middle part of the ingots, by the razorblade. The freshly cleaved platelets (along the cleavage planes) were mirror-like. That is why no further polishing and cleaning treatments were required. Details for crystal growth are described elsewhere [13].

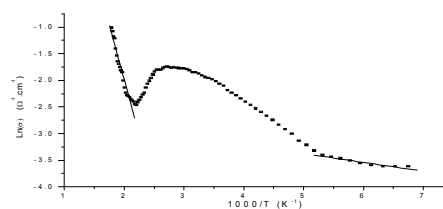
## 2.2. Electrical conductivity and Hall effect measurements.

Parallelepiped samples with mean dimensions 9.4×2.8×1.6 mm<sup>3</sup> and mirror surfaces were prepared for electrical measurements. These were performed in a vacuum cryostat in the temperature interval 148 to 558 K using a specimen container evacuated to 10<sup>-3</sup> Torr. Electrical conductivity and Hall coefficient were measured by a DC compensation method. Electrical measurements were made with the aid of silver paste contact. These contacts were Ohmic in the range of the applied voltage. The Ohmic nature of the contact was checked by recording the current-voltage characteristics. A study of the Hall effect was carried out in a static magnetic field employing a direct current. A magnetic field of 0.5 Tesla was employed for Hall coefficient measurement. The current direction was parallel and the field direction was perpendicular to the cleavage plane. In order to avoid thermogalvanomagnetic effects [14], several measurements were carried out for temperature values by reversing the direction of the current and the magnetic field. Welded copper and constantan wires (Ø 0.1 mm) served as junctions for measuring thermocouples. The temperature was varied above room temperature up to 558 K, with the help of electrically insulated heater. Temperatures below room temperature up to 148 K were achieved using liquid nitrogen. The measured Hall voltage was corrected for the finite ratio of the sample length to width according to calculated correction factors [15]. The apparatus and procedure of measurements are

mainly the same as those described in previous work [16].

## 3. Results and discussion

The result presented in the figures are that for TlInSeS single crystal with room temperature conductivity equal to 1.43×10<sup>-5</sup> Ω<sup>-1</sup> cm<sup>-1</sup>. Fig. 1 shows the temperature dependence of electrical conductivity. The complete temperature range can be subdivided into three regions: below transition, transition region and above transition. These curves are quite similar to semiconductor behaviour. These regions are clearly shown in fig. 1; with increasing temperature, the electrical conductivity increased slowly. Secondly, it reached the transition region at 190 K, then passed through a minimum and rose again. This pattern of change in the electrical conductivity is due to the appearance of impurity and intrinsic conductivity respectively and to the variation of hole mobility and concentration with temperature. The fall in the electrical conductivity was due to a decrease in mobility, since the carrier density in this temperature region remained practically constant (N<sub>A</sub>-N<sub>D</sub> = constant), until the intrinsic region was reached. At temperatures above the transition point, the conductivity rose rapidly. The temperature dependence exhibited a transition from a region of lower slope to one of higher slope. The transition region stretches from 190 to 460 K. The slopes of the curve increased with increasing temperature, and were higher at higher temperature due to carriers being excited from the extended state of the valence band into the conduction band. The width of the forbidden zone as calculated from the slope of the curve in the high temperature region was found to be 0.727 eV.



**Fig.1. Electrical conductivity of TlInSeS as a function of temperature**

The temperature dependence of the conductivity can be expressed in the form

$$\sigma = \sigma_0 \exp\left(\frac{-\Delta E_g}{2KT}\right)$$

Where  $\sigma_0$  is the pre-exponential factor and  $\Delta E_g$  is the width of energy gap. The calculated energy gap

width is smaller than that reported in the literature. This value contrasts with data of other authors [2,3]. Not only our results contradict to those obtained by other authors, but also the published values for  $\Delta E_g$  disagreed with each other. We may attribute the discrepancy in the values of  $\Delta E_g$  partially to the presence of a large number of intrinsic defects that affects strongly the motion of the scattering of current carriers and phonons. On the other hand it is thought that the technology used to grow this crystal may influence its physical properties. In this paper, we tried to elucidate this confusion, but more experimental data were necessary to explain this contradiction. In the extrinsic region the electrical conductivity increases slowly with temperature due to the fact that the carrier concentration, in this region is determined by the number of ionized acceptors liberated from the impurity level. From this region the ionization energy was calculated indicating the acceptor centre lay at 0.028 eV. The temperature dependence of the Hall coefficient for TlInSeS is positive in the entire range of investigation. This indicates that the compound TlInSeS is an excellent p-type semiconductor. The Hall coefficient at room temperature was evaluated as  $R_H=4.786 \times 10^4 \text{ cm}^3/\text{C}$ . We can see from the figure at the beginning of the curve the Hall coefficient shows a less rapid dependence on temperature. The sample exhibits a considerable fall of the Hall coefficient when temperature is increased up to the transition region in which the experimental  $\text{Ln}R_H$  versus  $10^3/T$  curve deviates from linearity. In the intrinsic conduction region the Hall coefficient fall rapidly. Determination of the energy gap and ionization energy from Hall data is possible by plotting the relation between  $\text{Ln}R_H T^{3/2}$  and  $10^3/T$  as shown in fig.3. In the temperature region in which the conductivity is predominately intrinsic, the forbidden band was estimated to be 0.727 eV. The depth of the acceptor centre was determined from the region in which conductivity is predominantly due to impurity atoms and was found to be 0.028 eV. These values are in good agreement with the values obtained from the temperature dependence of electrical conductivity. Combination of the Hall measurements and the electrical conductivity data were used to study the temperature dependence of the mobility of the charge carriers. The nature logarithm of  $R_H \sigma$  against  $\text{Ln}T$  is plotted in figure 4. This plot results in a straight line slope which allows the determination of the exponent. It was found that the exponent  $n$  in the relation  $\mu \sim T^n$  below 190 K is 0.23, whereas in the high temperature range ( $T > 460\text{K}$ ), the mobility decreases according to the low  $\mu \sim T^{-2.94}$ . This dependence indicates that phonon scattering is the mechanism responsible for

this mobility behaviour in the high temperature range. The highest measured value of the hole mobility at 148K is  $9408.793 \text{ cm}^2/\text{V}\cdot\text{sec}$ . The fall in the mobility below  $7267.517 \text{ cm}^2/\text{V}\cdot\text{sec}$  as the temperature decreases, implies that other scattering mechanisms become more important but the small value of  $n$  are unusually compared with those obtained for impurity scattering in other semiconductors. However the variation of mobility with temperature in these defected semiconductors has not been previously reported, there is still insufficient experimental data to throw a clear light upon this behaviour. This may be associated with the presence of high density of stoichiometric vacancies and the creation of defects. The exact nature of defects in these semiconductors remains uncertain, but from structural considerations and also by analogy with III-V compounds, vacancies and anti-site defects are likely to play important role. Stoichiometric cation vacancies present are themselves not neutral, but their presence is responsible for easy restoration of radiation ejected atoms into lattice sites across low energy barriers. The room temperature value of the mobility was found to be  $6864.25 \text{ cm}^2/\text{V}\cdot\text{sec}$ .

Carrier density versus reciprocal temperature measurements indicates acceptor level at 0.02 eV as shown in figure 5. At low temperatures and indeed (148-190 K) in TlInSeS the carrier concentration is determined by the number of ionized acceptors and the variation of the carrier concentration is quite slow. Since, the TlInSeS sample exhibited an intrinsic behavior above 460 K, then the expected value for the intrinsic concentration will be given as

$$p_i = 2 \left( \frac{2 \pi K}{h} \right)^{3/2} (m_p^* m_n^*)^{3/4} T^{3/2} \exp \left( \frac{-\Delta E_g}{2 K T} \right)$$

Where symbols have their usual significance and the energy gap width as deduced from this relation is 0.727 eV. The carrier concentration at room temperature as calculated from the  $R_H$  curve is  $1.305 \times 10^{14} \text{ cm}^{-3}$ . The diffusion coefficient is related to the mobility of charge carriers, its value for holes can be deduced as  $D_p=177.78 \text{ cm}^2/\text{sec}$ . The relaxation time as well as the diffusion length for holes were evaluated to be  $3.88 \times 10^{-7} \text{ sec}$  and  $8.305 \times 10^{-3} \text{ cm}$  respectively.

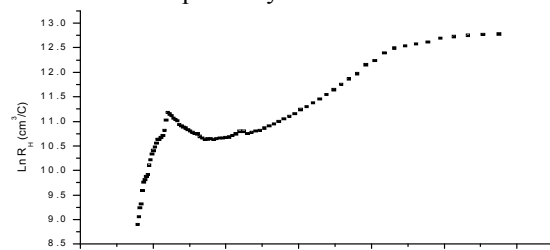
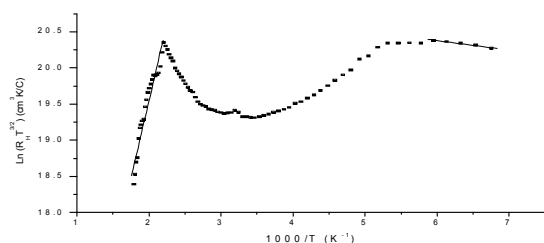
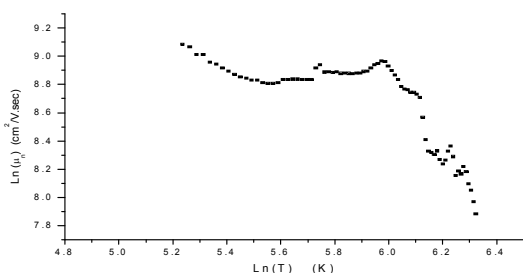


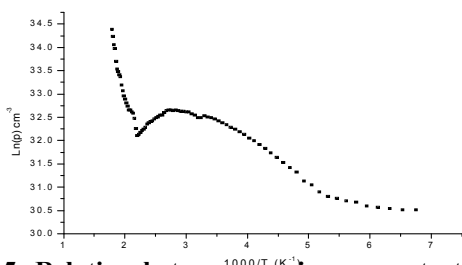
Fig.2. Variation of Hall coefficient with temperature for TlInSeS single crystal



**Fig.3. Relation between  $R_H T^{3/2}$  and  $10^3/T$  of TlInSeS single crystals**



**Fig.4. Temperature dependence of charge carrier mobility of TlInSeS**



**Fig.5. Relation between carrier concentration in TlInSeS and temperature**

#### 4. Concluding remarks

Thallium indium sulphur selenide single crystal was grown from the melt by a special modified Bridgman technique. Crystal perfection was checked by means of the x-ray diffraction technique. Measurements of electrical conductivity and Hall effect between 148 and 558 K were carried out on TlInSeS. The study was carried out with the current parallel to the c-axis and the magnetic field perpendicular to the c-axis. From the electrical conductivity and Hall effect measurements, the band gap of p-type TlInSeS and the depth of the impurity level was determined to be 0.727 eV and 0.028 eV respectively. The present investigations is the first one on electrical conductivity and Hall coefficient of

TlInSeS lead to the determination of the variation of carrier concentration and mobility with temperature. The variation of mobility with temperature shows small value of temperature exponents in the low temperature range. This anomalous behavior can not be understood by the usual theory of semiconductors. An attempt to explain this unusual character by assuming localized phonons due to stoichiometric vacancies are expected to play an important role in the behaviour TlInSeS while additional contribution may be presented due to creation of defects.

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