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Experimental Investigation of Electrical Conductivity and Hall Effect on Quaternary Thallium Sulphide $Tl_4InGa_3S_8$ Crystals

Jazi Abdullah Mohammed Abdulwahed

Physics Department, Umm Al-Qura University College in Qunfudah-Female, KSA
Email: *jaabdulwahed[at]uqu.edu.sa*

Abstract: ***A modified Bridgman technique has been used to grow the $Tl_4InGa_3S_8$ crystal. By measuring the Hall Effect and electrical conductivity at the same time in the $Tl_4InGa_3S_8$ sample in the range of 273–573 K, the conductivity type, carrier concentration, carrier mobility, diffusion coefficient, mean free time, and diffusion length for the majority carriers were identified. Our study was conducted in the 273–573 K temperature range. Our samples are n-type conducting, according to the investigation. With a Hall coefficient of 0.00327 m^3/c, the room-temperature electron concentrations were discovered to be 3.4 x 10^{10} m^{-3}at room temperature. Also, the Hall mobilities of $Tl_4InGa_3S_8$ are determined to be 24.79 m^2/V.sec at room temperature. $Tl_4InGa_3S_8$'s band gap has been calculated to be 0.2 eV, while the depth of the donor center was calculated to be 0.18 eV. This investigation is the first on this compound.***

Keywords: Quaternary semiconductors, $Tl_4InGa_3S_8$ crystal, electrical conductivity

1. Introduction

Due to their intriguing physical characteristics and wide range of potential applications, amorphous and crystalline chalcogenide semiconductors have been studied for a long time [1]. Quaternary layered-structured semiconductors enable band gap and lattice constant adjustments simultaneously, boosting radiant efficiency across a wider range of wavelengths. The group of layered semiconductors is contained in the quaternary compound of $Tl_4InGa_3S_8$ crystals. These crystals can be created by switching out one-fourth of the gallium ions in $TlGaS_2$ crystals for indium ions [2, 3]. Periodic two-dimensional layers parallel to the (001) plane make up the $Tl_4InGa_3S_8$ lattice. Ga (In) and S atoms are bonded via intralayer bonding, while Tl and S atoms are interlayer bound in the crystal. With the use of transmission and reflection measurements in the 400–1,100 nm range and the analysis of absorption data, the optical properties of $Tl_4InGa_3S_8$ crystals were examined. Measurements of transmission and reflection in the 400–1,100 nm range were taken in order to investigate the optical characteristics of $Tl_4InGa_3S_8$ crystals. The analysis of the absorption data yielded band gap values of 2.40 and 2.61 eV for the indirect and direct transitions, respectively [4]. The analysis's findings have shown the activation energies to be 13, 44, and 208 meV [5]. The focus of the current work was the investigation of the electrical conductivity and Hall effect of the relatively unexplored $Tl_4InGa_3S_8$ due to the high interest in the layered- and chain-structured $TlIn_xGa_{1-x}S_2$ chalcogenide systems for their potential applications in optoelectronic devices. The primary goals of this work are to present and discuss the findings related to electrical conductivity and the Hall effect on $Tl_4InGa_3S_8$ crystals in order to obtain the main physical parameters for finding a suitable possibility for electronic application.

2. Experimental

The Travelling Solvent Method (TSM), a modification of the Bridgman technique, was used to prepare $Tl_4InGa_3S_8$ crystals. Details of the new hydrolic pulling mechanism and the three-zone furnace have already been published [6]. This method was utilised for producing the samples for this study using stoichiometric amounts of pure (6N) elements (Aldrich Mark) in a sealed, evacuated (10^{-6} Torr) silica tube. After melting, the mixture was homogenised at T_m = 1075 K [7] for 24 hours. To improve the contents' mixing, the ampoule was shaken repeatedly during this process. The ampoule was then lowered at a rate of 1.4 mm/h, followed by 10 hours of holding, to drop the temperature. According to published data [6], the product ingot exhibited a black colour with a metallic lustre after cooling to room temperature. The sample was made in the shape of a rectangle and prepared for research on electrical conductivity and the Hall Effect. The sample's dimensions after polishing were 10 x 3 x 2.5 mm^3. In order to prevent a Hall voltage drop, the sample's length was three times wider than it was long. The DC Hall measurements were performed in accordance with ASTM-F67 guidelines and remarkably resembled those previously disclosed [8]. Under a vacuum of 10^{-3} Torr, electrical conductivity and the Hall effect were measured. With the aid of an electrically insulated heater, for measurements above room temperatures. The low temperature measurements were made using liquid nitrogen. The sample temperature was measured using a calibrated copper constant-temperature thermocouple. A GMW electromagnet type 5403 with a DTM-133 digital Tesla meter was used to provide a 0.55 Tesla intermediate magnetic field. In a unique cryostat, the conductivity and Hall coefficient were measured using a traditional DC-type measurement device. By reversing the current and magnetic field directions and obtaining the necessary averages, the Hall voltage was determined. Measurements across a broad temperature range are performed with the newly developed cryostat [9]. As an ohmic contact, silver paste was employed. The current-voltage characteristics were recorded in order to confirm the connections' ohmic nature.

3. Results and Discussion

The impact of temperature on electric conductivity σ was measured in $Tl_4InGa_3S_8$ crystals, as depicted in Fig. 1, from 273 to 573 k. The curve displays typical behaviour for semiconductors. It should be noticed that three distinct zones can be seen in the curve in Fig. 1. Due to the fact that the number of ionised impurities released from the impurity level determines the carrier concentration in this location, the electrical conductivity σ rose slowly with temperature, starting at a low temperature. The relationship between temperature and electrical conductivity may be as follows in the region of low temperatures:

$$\sigma = \sigma_o \exp(-\Delta E_a / 2 K_B T) \quad (1)$$

Where ΔE_a is the ionisation energy of the donors, and σ_o is the preexponential factor. This is seen between the temperatures of 273 and 416 K, and ΔE_a was calculated to be 0.18 eV right below the conduction band, according to the calculation of the ionisation energy from this region. At 300 K, the value of σ is 7575 Ω^{-1} m^{-1}.

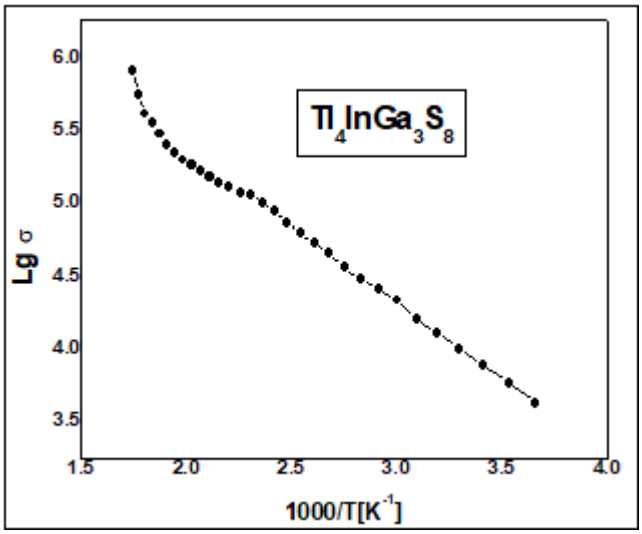

Figure 1: $Tl_4InGa_3S_8$Crystal electrical conductivity as a function of temperature

The second region stands for the transition region, where the behaviour is controlled by the mobilities and concentrations of charge carriers. The exhausted area, which spanned from 416 to 500 K, is characterised by relatively rapid exponential increases in electrical conductivity. In the high-temperature region above 500 K, a linearly increasing rate of conductivity is seen. This demonstrates that conduction at this high temperature range is a result of both electrons and holes. This temperature range obeys the following relationship:

$$\sigma = \sigma_o \exp(-\Delta E_g / 2 k_B T) \quad (2)$$

This formula yields an energy gap of ΔE_g = 0.21 eV. This number suggests that our crystals have a very narrow energy gap. In the current work, the computed energy gap width is lower than that which has been published in the literature [4]. However, the energy gap (0.21 eV) and data from the literature (0.21 eV)[5] are in excellent agreement. We can partially explain the discrepancy in ΔE_g values by the existence of numerous intrinsic defects that have a significant impact on the motion of the scattering of phonons and current carriers. On the other hand, it is hypothesised that the process employed to generate these crystals may have an impact on their physical characteristics. We attempted to clarify this misunderstanding in this study, but additional experimental data were required to fully understand this discrepancy. This work was expanded to cover the effect of temperature on the Hall coefficient R_H as shown in Fig. 2, because the Hall effect is significant and beneficial for the calculation of many physical parameters. This was carried out across a wide temperature range (273–573 K). It is clear from the measurements of the Hall coefficient that the Hall coefficient of $Tl_4InGa_3S_8$ is negative over the whole temperature range under examination. In reasonable agreement with the findings of other published data [10–11], this suggests that the substance is an n-type semiconductor. At room temperature, the Hall coefficient was calculated to be 0.00327 m^3/c. By displaying the relationship between Ln $R_H T^{3/2}$and 10^3 /T as shown in Fig. 3, one can calculate the energy gap and ionisation energy from Hall data using the following relationship:

$$R_H T^{3/2} = C \exp(-\Delta E_g / 2 k_B T) \quad (3)$$

The forbidden band width was calculated to be ΔE_g = 0.2 eV in the temperature range where intrinsic conductivity predominates. In a region where impurity atoms are mostly responsible for the conductivity, the depth of the donor centre was calculated and found to be 0.18 eV. These results and the conductivity data are in good agreement with other published data.

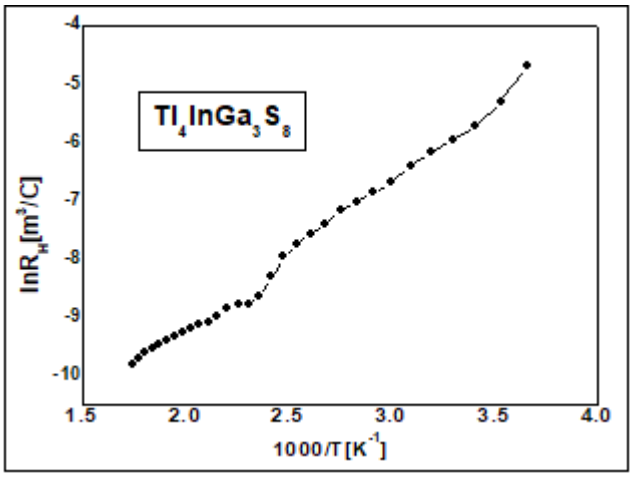

Figure 2: R_H for $Tl_4InGa_3S_8$ crystal and temperature dependency

Extrinsic conduction arises between 273 and 416 K, and intrinsic conduction starts between 462 and 573 K, according to the three sections of the curve, while the transition region is between 416 and 454 K.

In order to fully characterise the samples, Fig. 4 shows the ln μ_H against ln T as a plot. Regarding the significance of mobility data in the field of substances, particularly semiconductors, the current work focuses on examining how temperature affects free carrier mobility.

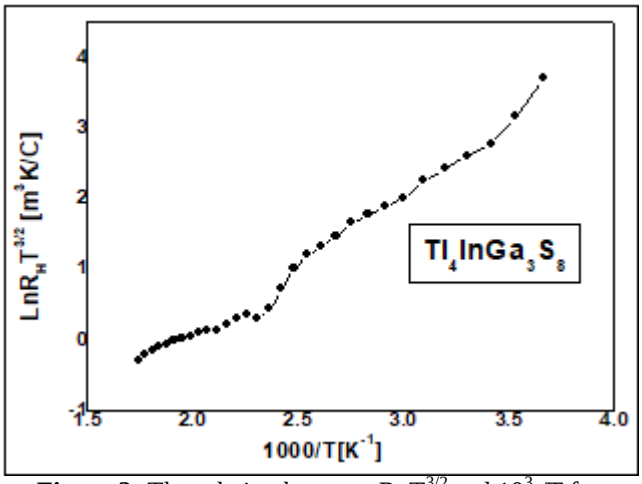

Figure 3: The relation between $R_HT^{3/2}$ and 10^3 /T for $Tl_4InGa_3S_8$ crystal.

The carrier mobility can be estimated by measuring both the resistivity and the carrier simultaneously. It is practicable to obtain details regarding the scattering mechanisms due to the temperature dependence and magnitude of the carrier mobility [12].The Hall mobility varies as a function of temperature according to the relation $\mu_H \alpha T^n$. As the fluctuation of μ_H as a function of temperature is seen in Fig. 4, it is evident that the variation in carrier mobility with temperature is more complicated. Three zones are distinguished by this curve. In the first portion of the 273–403 K temperature range, the Hall mobility rises as temperature rises, following a curve that corresponds $\mu_H \alpha T^{0.38}$. Such behaviour is typical of a charge carrier scattering mechanism with ionised impurities, which is thought to be acting in these $Tl_4InGa_3S_8$ crystals at a temperature (T<403 K). In the second, at the temperature range of 391 to 450 K, the transition zone likewise exhibits a sharp change in Hall mobility.

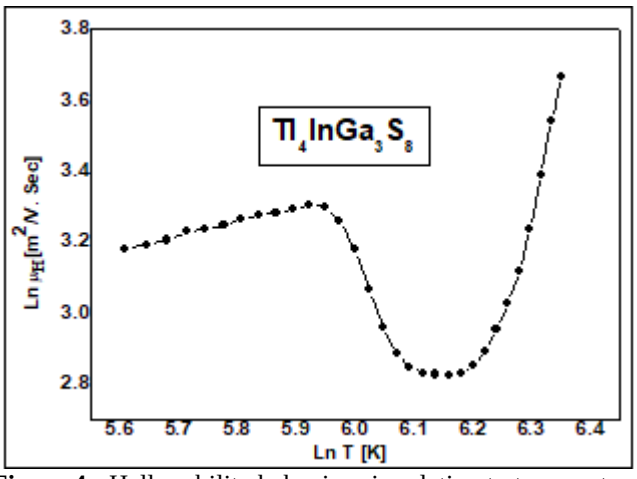

Figure 4: Hall mobility behaviour in relation to temperature

At high temperatures, the Hall mobility $\mu_H \alpha T^{5.5}$ was found to rise with temperature by relation in the third region, which covers the 493-573 K temperature range. The assumption that inclusion or crystalline imperfections may arise in the layer planes is made as a result of μ_H thermally triggered behavior [13-15]. The numerical value of n and its sign indicate that carriers' scattering mechanisms are unique to semiconductor materials. Both traditional theories of semiconductors and the idea of acoustic phonon scattering based on Schmid's model [16] cannot account for such abnormal behaviour. Although the fact that the mobility of these quaternary semiconductors varies with temperature has not yet been documented, there is insufficient experimental data to definitively explain this behaviour. In these examinations, the hole mobility for the $Tl_4InGa_3S_8$ sample at room temperature was found to be 24.79 m^2/ V.sec.

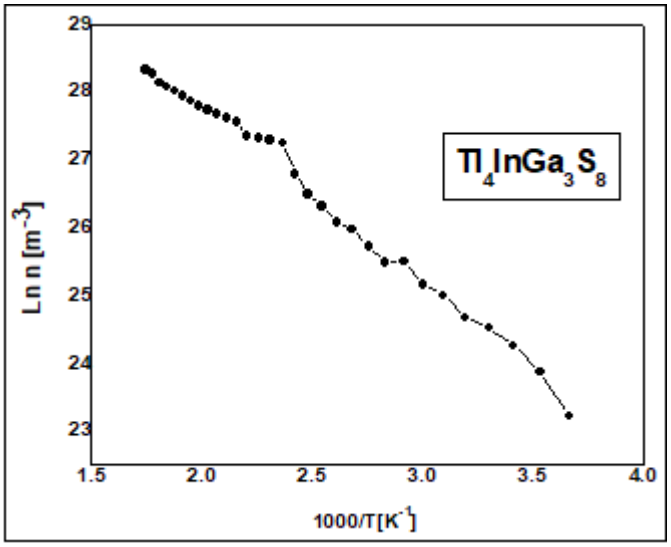

Figure 5: Carrier concentration for $Tl_4InGa_3S_8$ crystal changes with temperature

The temperature dependence of charge carrier concentration is shown in Fig. 5 so that one may analyse how the carrier concentration in this sample behaves with temperature. Using the relationship n = $1/R_He$, where n is electron concentration and e is electron charge, the charge carrier concentration was derived from the Hall data. This figure shows that whereas the concentration of carriers increases rapidly with temperature in the intrinsic zone, it slows down in the extrinsic region. From the slope of this curve's the energy gap in the high-temperature region is found to be ΔE_g = 0.2 eV. The electron concentration at room temperature is 3.4 x 10^{10} m^{-3}.

The diffusion coefficient is given as D = μ k_B T / q in the Einstein relation, where q is the charge on an electron, k_B is the Boltzmann constant, T is the temperature, and μ is the charge carrier mobility. The diffusion coefficient for electrons was calculated, and the result was 0.68 m^2 /s. Using the value for the electron mobility at room temperature and the supposition that the effective mass for electrons is equal to the rest mass, it was possible to calculate the mean free time (τ= m μ/e), which had a value of 14 x 10^{-11} seconds. Additionally, the $Tl_4InGa_3S_8$ specimen's electrons' diffusion length was estimated to be 98 x 10^{-7}m.

4. Conclusion

Over the temperature range of 273–573 K, the electrical conductivity (σ) and Hall coefficient (R_H) of the $Tl_4InGa_3S_8$ crystal have been studied. With the use of a modified Bridgman method, the crystal was grown. The investigation revealed that our samples were n-type.$Tl_4InGa_3S_8$'s band gap was 0.2 eV, while calculations revealed that the donor center's depth depth was 0.18 eV. Electrical conductivity,

Hall coefficient, and carrier concentration were all 7575 Ω^{-1} m^{-1}, 0.00327 m^3/c, and 3.4 x 10^{10} m^{-3}, respectively, at room temperature. At room temperature, it was discovered that the Hall mobility (μ_H) was 24.79 m^2/V.sec. Additionally, a graphic representation of Hall mobility's temperature dependence was provided. Our sample was found to have a narrow energy gap, which is used in many different recent applications in electronic devices.

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