Growth and Transport properties of the quaternary chalcogenidesTI$_2$GaInTe$_4$ compounds

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Abstract: The preparation and electrical properties of the quaternary chalcogenide TI$_2$GaInTe$_4$ crystals is reported for the first time in this work. Measurements of electrical conductivity and Hall coefficient were performed over the temperature range (213-568 K). The crystals obtained by a modified Bridgman technique for crystal growth had p-type conductivity with a hole concentration of 5.369 x 10^{12} cm^{-3} at room temperature. Conductivity and Hall mobility at 300 K were evaluated as 0.08033 Ω^{-1} cm^{-1} and 93500 cm^2/V.sec respectively. The energy gap width was calculated and found to be 0.88 eV. The scattering mechanisms of the carrier are checked over the whole investigated range. Furthermore the diffusion coefficient, relaxation time and diffusion length of holes are estimated.
[Jazi Abdullah Mohammed Abdulwahed. **Growth and Transport properties of the quaternary chalcogenides TI$_2$GaInTe$_4$ compounds.** *Life Sci J* 2014;11(4):109-113]. (ISSN:1097-8135). http://www.lifesciencesite.com. 14

Keywords: Crystal growth, TI$_2$GaInTe$_4$, DC electrical conductivity, Hall coefficient

1- Introduction

In the past three decades, significant increase in chalcogenide semiconductor has been shown by various worker, because of their interesting physical properties as well as their wide technological application [(1)]. The appearance and vigorous development of many modern branches of technology stimulate continuous searching for an investigation of new promising materials possessing a wide spectrum of properties than the already known and partially tested materials. One of the trends in semiconductor materials science is developing new functional materials is the increasing complexity of the investigated systems and of the compounds that form in these system. Determination of the application potential for new semiconducting materials in solid state physics requires the growth and examination of physical properties of the materials. One of the methods for modifying their physical properties is the production of solid solutions of these semiconductors. In the recent years an improving interest has been given to the physical properties of the chain-like structured materials such as TlInTe$_2$ and TlGaTe$_2$ crystals. Some of the structural, electrical and optical properties of the TlInTe$_2$ and TlGaTe$_2$ crystals has been reported [(2-15)]. The experimental studies carried out on TlGaTe$_2$ and TlInTe$_2$ demonstrate that investigations for the physical properties of the TlGaTe$_2$ - TlInTe$_2$ system, should be rewarding in view of the production of a new candidate materials for electronic devices Tl$_2$GaInTe$_4$ compound crystallizes in chain-like structure with TlGaTe$_2$ and TlInTe$_2$ components having the ratio 1:1. In spite of its importance in technological applications as a candidate material for optical devices and also for the information of its basic physical properties. So far very little information on the physical properties of this compound [(16-18)]. However, the electrical transport properties of the new semiconductor compound Tl$_2$GaInTe$_4$ crystals have not been studied at all. The original purpose of the present work was to prepare Tl$_2$GaInTe$_4$ and to carry DC electrical conductivity and Hall coefficient in wide range of temperature of the new chain-like crystal Tl$_2$GaInTe$_4$ for the first time. So the present investigation was done to be the first in the literature and to complete our plan concerning investigation of the physical properties of different $A^{111}B^{V1}$ semiconductor compounds.

2- Experimental set up

2-1- Material and sample preparation

Tl$_2$GaInTe$_4$ single crystals were grown using modified Bridgman method from TlGaTe$_2$ and TlInTe$_2$ polycrystalline compounds having stoichiometric starting materials. The purity of the materials used 99.9999%. The silica tube was washed with pure alcohol and distilled water, then coated with a thin layer of graphite to prevent contamination of the charge on the internal surface of the ampoule. The silica tube has a constructed sharp end at the bottom to facilitate seeding in the growth process. Extreme care was taken in the transfer of material to maintain the accuracy to the level of µg / g of material. The material were weighed by high sensitive rate and then transferred carefully to the ampoule which previously was weighted empty, and then re-weighing the ampoule after the development of the materials to make sure the weight of the starting material. The appropriate amount was first sealed in evacuated quartz ampoule. Tl$_2$GaInTe$_4$ single crystals were grown by slight freezing in sealed quartz ampoule evacuated to about (~10^{-6}mbar).In the first procedure the tube was placed in a three-stage tube furnace, in which controlled temperature gradient was maintained. The ampoule is allowed to move with a constant rate of 1.5 mm/h through the stationary furnace. The movement of the ampoule was very gently and show with the aid of the hydraulic mechanical system. At least two weeks growth are needed to obtain $Tl_2GaInTe_4$ single crystal , details about the apparatus and method of modified Bridgman technique reported in previous work(19). The resulting ingots (grey-black in color) showed good optical quality. The X-ray powder diffraction technique was used to identify the crystalline nature of the $Tl_2GaInTe_4$ compound. For this purpose, a philips PW140 diffractmeter with a monochromatic cuk$_\alpha$ radiation λ=1.54049Å at scanning speed of 0.0.2˚θ /s were used. The x-ray diffraction pattern of the sample is illustrate in figure (1).

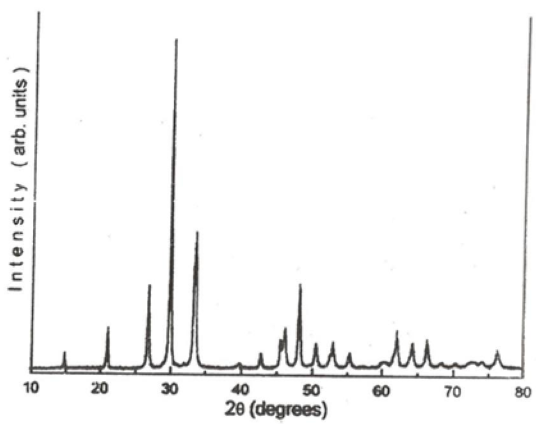

Fig (1): x-ray diffraction pattern for $Tl_2GaInTe_4$

2-2-Measurement technique

Single crystal samples of $Tl_2GaInTe_4$ were prepared by gently cleavage from the product with the aid of a razor blade. Typical dimensions of the crystals suitable for electrical measurement were 8.5 x 2.5 x1.5 mm^3. The sample had a length three times its width. This aspect ratio is useful to avoid a Hall voltage drop. The measured Hall voltage was corrected for the finite ratio of the sample length to width according to calculated correction factors(20). The electrical conductivity and Hall effect measurements were measured using a DC four probe method .The sample was placed in an evacuated Pyrex cryostat(21). The cryostat works as a liquid nitrogen container and is supported with electric heaters for low and high-temperature measurements respectively. The magnetic field value in the experiment was 0.5 T using a GMW electromagnet model 5403. The electrical conductivity and Hall coefficient were measured by a DC compensation method. Ohmic contact was made with the aid of silver paste. The ohmic nature of the contact was confirmed by I-V characteristic. These contact were ohmic in the range of the applied voltage. Details of the experimental procedures and apparatus have been published(22).

3-Results and Discussion

Fig (2) illustrates the variation of σ against 10^3/ T. The curve is divided into three parts: starting from the low temperatures, the first part (213-283K) represents the extrinsic conduction range where the carrier concentration is mainly determined by the number of ionized acceptors. This is naturally occurs as a result of the liberation of ionized acceptors and their transition from the impurity level. Accordingly σ increases slowly. In this range the following formula describes the relation between σ and T:

$$\sigma = \sigma_o \exp(-\Delta E_a/2KT) \qquad (1)$$

where σ_o is pre-exponential factor and K is the Boltzmann's constant. From the above relation we could calculate the impurity ionization energy ΔE_a, it was 0.32 eV. The second region (283-423 K), represents the transition region where the behavior of σ is governed by the behavior of both the charge carrier concentration and their mobility. In this region the small increase in the electrical conductivity is due to the increase in the hole concentration. Above 423 K the intrinsic conduction appears (as seen from the figure) where σ sharply increases. This predicts that both electrons and holes contribute in the conduction at this high temperature range. The following equation is used to estimate the value of the energy gap.

$$\sigma = \sigma_o \exp(-\Delta E_g/2KT) \qquad (2)$$

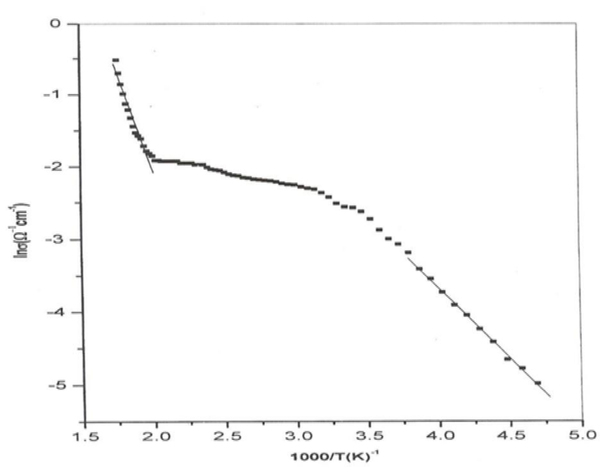

Fig (2): variation of conductivity with temperature for $Tl_2GaInTe_4$

It was found that ΔE_g equal to be 0.89 eV. The electrical conductivity at room temperature amounts to be as 0.08033 Ω^{-1} cm^{-1}. This pattern of the appearance of extrinsic and intrinsic type of conductivity respectively and to the variation of the carrier mobility and to the variation of the carrier concentration with temperature. In the intermediate region where the carrier density (N_A- N_D)= constant, until the intrinsic region is reached. The slopes of the curve increase with increasing temperature, and are higher at higher temperature because of the carriers being excited from the extended state of the valance band into the conduction band. As for the importance of the Hall effect measurement, the present investigation is extended to cover this unique phenomenon. In the same limited temperature range (213-568 K) the variation of the Hall coefficient against temperature was examined. In fig (3) the curve is divided mainly into:-

* low temperature part (from 213 up 278 K) which represents the case of the extrinsic conduction
* The high temperature part which appears between (328-568 K)

At room temperature R_H has a value of 1.269×10^6 cm^3/C.

From the measurement of the Hall coefficient it is evident that the sign of the Hall coefficient of $Tl_2GaInTe_4$ is positive over the entire range of investigation, indicating that the compound is a p-type semiconductor and the major contribution to conductivity by holes.

Fig (4) depicts the relation between $R_H T^{3/2}$ and $10^3/T$. From the figure we, also distinguished the two region at the same corresponding temperatures according to the relation.

$$R_H T^{3/2} \propto \exp(-\Delta E_g/2KT) \qquad (3)$$

Utilization of the figure and the above formula leads to estimation of values of the energy gap. In was found to 0.88eV, and the depth of the impurity level was computed. It was found to amount 0.31eV. Assessment of the forbidden width from this graph and found have a value close to that determined from electrical conductivity and that reported early[16] from the study of spectral distribution of the photocurrent.

A combination of the Hall measurements and electrical conductivity data were used to study the temperature dependence of the mobility of the charge carries.

Fig (5) shows the variation of μ_H as a function of temperature can be divided into two regions. The first one is that μ_H increases as the temperature grows till T=288 K.

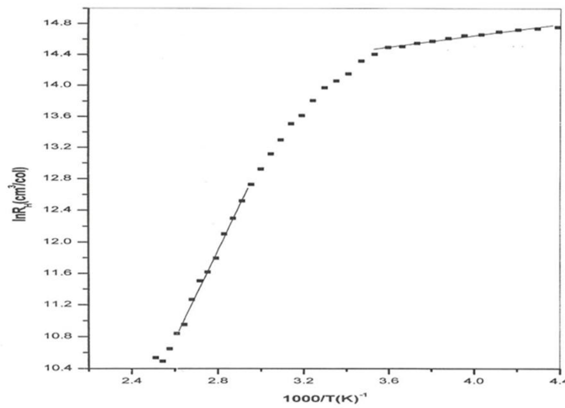

Fig (3): Relation between Hall coefficient and $10^3/T$ for $Tl_2GaInTe_4$

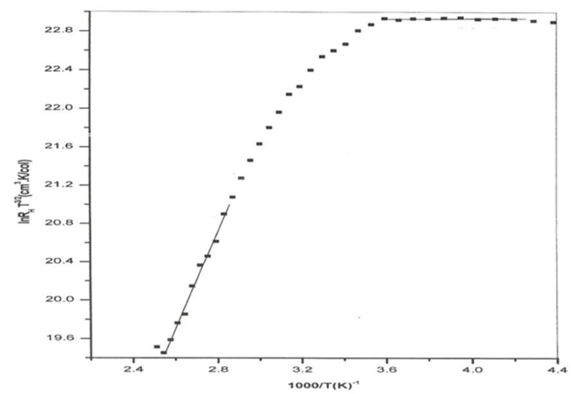

Fig (4): The relation between R_H $T^{3/2}$ and 10^3/ T for $Tl_2GaInTe_4$

The second part is that μ_H decreases. In the low temperature part (from 228 K up to 288 K) where the mobility has appositive temperature coefficient. The relation between μ_H and T obey the low $\mu \propto T^{6.95}$. High temperature part, the Hall mobility decreases with increasing temperature. In this region, μ_H decreases with increasing temperature according to the low $\mu \propto T^{-12.69}$. From this relation, it seems that the value of the exponent n in the relation $\mu \propto T^n$ is usually large compared with those obtained for impurity and lattice scattering in other semiconductors. However such behavior was observed earlier during the investigation of $TlGaTe_2$ [8], GaTe[23], Ga_2Se[24] and In_2Te_3[25]. This behavior in our opinion, may be associated with the presence of a high density of stoichiometric vacancies and the creation of defects. The room temperature value of the mobility equals 9.35 x 10^4 cm^2/V.

As a complementary part to the Hall work; the mode of variation of the charge carrier concentration against temperature was checked. They are calculated by the relation P=1/(e R), which are shown in fig (6).

The figure shows the remarkable increase of carrier concentration in the high temperature range, where the crystal exhibits an intrinsic behavior. The expected value for the intrinsic concentration can be given as follows:

$$n_i = 2\,(2\pi K/h^2)^{3/2}\,(m_p^* m_n^*)^{3/4}\,T^{3/2}\,\exp(-\Delta E_g/2KT) \qquad (4)$$

where m^*_p and m^*_n are the effective mass of holes and electrons, respectively. Using this formula (4), we can calculate the energy gap width of $Tl_2GaInTe_4$ to be 0.88 eV. This value estimated from the last work agree with those obtained from the published value [16]. The value of the charge carrier concentration at room temperature is 5.369×10^{12} cm^{-3}. Calculating the diffusion coefficient for holes yielded a value of 2421.69 cm^2/sec. Assuming the effective mass for holes is equal to the rest mass, and using the value for the hole mobility at room temperature, the relaxation time could be determined and was equal to 5.33×10^{-11} sec. Additionally, the diffusion length of holes in $Tl_2GaInTe_4$ specimen was 3.59×10^{-4} cm

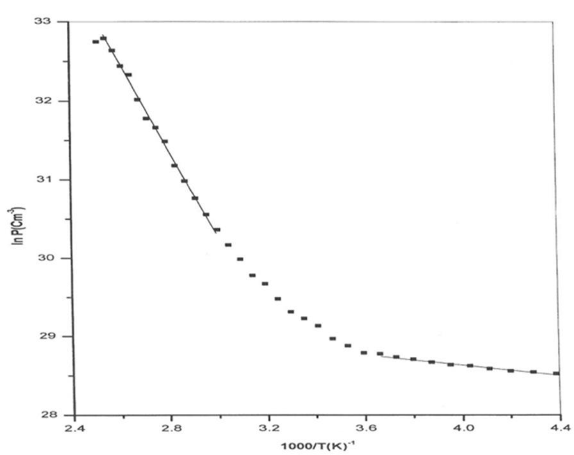

Fig (5): Variation of carrier concentration with temperature for $Tl_2GaInTe_4$

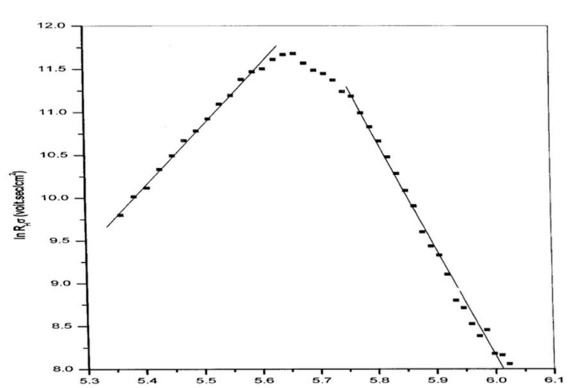

Fig (6): Behavior of μ as a function of temperature for $Tl_2GaInTe_4$

4- Conclusion

The preparation of our crystal sample of $Tl_2GaInTe_4$ was done by using a modified Bridgman technique for crystal growth from melt. The electrical conductivity and Hall coefficient is reported for the first time in the present work. Measurements of the electrical conductivity and Hall coefficient were performed over a temperature range 213-568 K.

All measurements were performed under vacuum in aspecial cryostat designed for this purpose. The measured Hall coefficient indicates p-type conductivity for our sample with a hole concentration of 5.369×10^{12} cm^{-3} at room temperature. The conductivity and Hall mobility at 300 K were 0.08033 Ω^{-1} cm^{-1} and 93500 cm^2/C respectively. The width of the band gap was estimated to be 0.88 eV. The position of the acceptor level was determined to be 0.32 eV. The diffusion coefficient relaxation time and the diffusion length of holes were also evaluated. This work provides a good picture of the actual physical behavior of the prepared quaternary $Tl_2GaInTe_4$ crystal, which leads to better application in many modern fields

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