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The Effects of Crystallographic Orientation of Tl_2Se Crystals on Hall Mobility

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Abstract: ***New crystal growth method was used in the present investigation to grow the Tl_2Se crystals.Electrical conductivity has been measured in two crystallographic directions ($\sigma_{//}$ and $\sigma_{\perp}$) forTl_2Se crystals. In addition, our work was extendedto cover Hall Effect phenomenon in the same temperature range (225-500K). In this way, the Hall mobility behavior and the scattering mechanism of the charge carriers were discussed. The energy gap and the ionization energy ware estimated as an important physical constant for the first time for this semiconductor compound.***

Keywords: crystallographic orientation, Hall mobility, Tl_2Se

1. Introduction

In the last few years, much attention has been devoted to semiconductors of the $A^{III}B^{VI}$ group. These chalcogenide semiconductor compounds are of interest because investigations of their physical properties open up the possibility of applications in technology. The layer structure compounds are promising materials in applications. This is due to their unique isotropic semiconducting properties. Although Tl-Se crystals are unique semiconducting materials, they are the less studied compounds in the $A^{III}B^{VI}$ group. The characteristic of thallium chalcogenide Tl_2Se has been investigated by using x-ray photoelectron spectroscopy approach [1]. The work of TI_2Se compound was extended to deal with the crystalline form. The specific heat capacity of TI_2Se has been measured by applying a heat pulse technique in the temperature range 3-300K and differential scanning calorimetric in the temperature range 190 –640K [2]. Heat capacity and phase transitions in highly anisotropic $A^{III}B^{VI}$ - type semiconductors and their analogs at low temperatures were investigated [3]. TI_2Se was involved in that work. Their results were in a satisfactory agreement with other investigators [4].Stasova and Vainshtein reported that the structure of thallium selenide TI_2Se to consist of Tl tetrahedral united by layers of Se atoms [5]. The sequence of layers Tl and Se should be similar to the sequence, whichis found in the hexagonal structure of Tl_2Se. They found that the interatomic distances in the $T1_2Se$structure correspond to those of structure of TISe and TI_2S [6].Tl_2Se is a semiconductor with a layered structure of tetragonal symmetry with unit-cell dimensions a =0.852A, c =1.268 Å and space group p4/n. [7]. Anisotropic properties of semiconductors are associated with their layered structure with strong covalent bonding in the layers and weak Van Der Weal's bonding between the layer as in Bi_2Te_3 [8]. We concluded from the literature survey that there is lack of data of the structure and anisotropy of electrical conductivity. Therefore, we undertook such a work to elucidate this confusion. Accordingly, this workwill be published for the first time.

2. Experimental Technique

2.1 Crystal Growth

Preparation of Tl-Se needs unusual precautions for growing crystals. So a special growth technique was used as follows:a mixture of thallium and selenium in correct stoichiometric proportions was used to produce Tl_2Se (13.62 g of Tl (purity 99.9999 %) and 2.63 g of Se (purity 99.9999 %). The percentages of the charge elements are 83.808% Tl, and 16.192% Se. The chemicals were obtained from Aldrich. Theywere placed in a sealed evacuated tube, then heated up in the first zone of the furnace and held at 663K(corresponding to the crystallization pointof Tl_2Se compound[9]. We used for growth of the single crystals of Tl_2Sea new method, whichhas already been [10]. This technique is very simple but it enables us to obtain a highly good quality of crystals. This is because of the absence of the motor vibrations whichis not beneficial. Also the growth rate can be controlled easily. Conformation of the product crystal showed that it is single crystal with a tetragonal structure and the unit cell parameters obtained are the same as reported earlier[7]. This is done in Central Metallurgical Research Development Institute(CMRDI)Egypt.

2.2 Electrical conductivity and Hall Effect measurements

A sample of 6.2x2.2x2.1 mm dimensions was prepared directly by a gentle cleavage from the product version ingot.This is because the product crystal is of a layer type. The sample in this way has length of three times its width (to avoid Hall voltage drop as recommended by Insberg[11]. The sample was placed in an evacuated Pyrex cryostat. The cryostat end finger has an isolated electric heater (for high-temperature measurements) and can be immersed in a liquid nitrogen Dewar flask (for low-temperature measurements).

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A variac transformer was used for adjusting the required ambient temperature, which was measured by K-type thermocouple. If *ab* is the crystal layer plane and *c* is the direction perpendicular to both *a* and*b*, then the measuring condition can be summarized as follows: *a* is the current direction, *b* is the magnetic field direction and *c* is the developed Hall voltage. The magnetic field value was 5000g in this experiment. It was supplied from a PHEWE (Germany) electromagnet. In the present work D.C conductivity and Hall measurements were performed as the method recommended by ASTM (F67) [12].It must be mentioned that for electrical conductivity and Hall measurements the following points were considered:

- The silver conducting paste was employed as an Ohmic contact and its Ohmicnature was checked by recording the I-V characteristics in both forward and reverse bias directions.
- A potentiometer type (UJ33E-England) was used for measuring the Hall voltage.

3. Results and Discussions

In general, two considerations were taken into account in the present work; firstly , it is preferable to study the conductivity rather than the conductance because thus ensures that the measurements are of a property of the material rather than of the contact. Secondly; several cuts front the product version were considered for measurement to ensure crystal homogeneity.

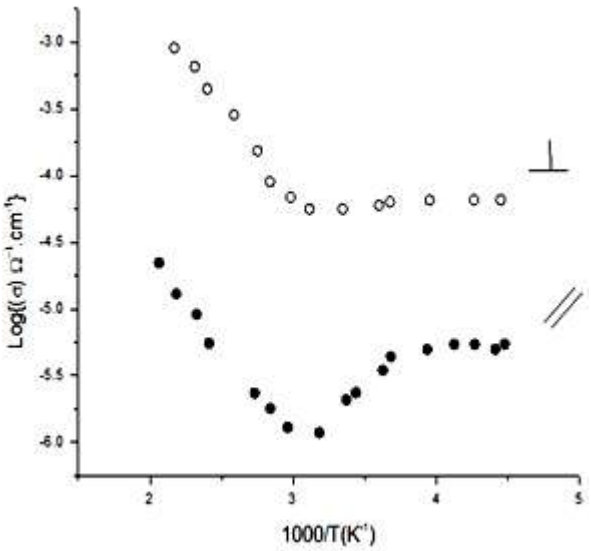


Figure 1: Variation of the electrical conductivity of Tl_2Se in two crystallographic directions

Fig (1) depicts the variation of $\sigma_{//}$and $\sigma_{\perp}$ with temperature.The influence of temperature on the electrical conductivity σ was investigated in a temperature range from 225 k to 500K. From the curve, we can notice the following:-

- It is observed that a increases slowly at the low temperatures (the extrinsic region) as a result of liberation of the ionized acceptors from the impurity level.
- Generally $\sigma_{\perp}$is always higher than $\sigma_{//}$for instance $\sigma_{\perp}$at room temperature equals 4.17 x 10^{-5} $\Omega^{-1}cm^{-1}$ while $\sigma_{//}$at the room temperature equals 2.51 x 10^{-6} Ω^{-1} cm^{-1}.
- The general mode of σ variation against temperature is typically as othersemiconductor' behaviors.
- The electric conductivity is highly anisotropic where values of that of $\sigma_{\perp}$exceeds the $\sigma_{//}$by almost one order of magnitude. Such variation can be attributed to the anisotropy of the carrier mobility.

Table 1: The following of the summarizes the different regions and their corresponding temperatures

Region	Corresponding temperature in the case of $\sigma_{//}$ (Solid circles)	Corresponding temperature in the case of $\sigma_{\perp}$ (circles)	Estimated value
Extrinsic	225 to 270 K	225 to 270 K	ΔE_a= 0. 11 eV
Transition	270 to 350 K	270 to 350K	
Intrinsic	350 to 500 K	350 to 500 K	ΔE_g = 0.55 eV

In the *intrinsic region* the following equation was applied:

$$\sigma = \sigma_o \exp(-\Delta E_g/2KT) \quad (1)$$

Where ΔE_g is the energy gap and σ_o is the pre-exponential factor.From the above equation we estimated the value of $\Delta E_g$and it was found to be 0.2 eVfor$\sigma_{//}$ and $\sigma_{\perp}$. The dependence in the low temperature range (extrinsic region) follows the relation: -

$$\sigma = A \exp(-\Delta E_g/2KT) \quad (2)$$

Where, *A* is a constant (the mobility varies much more slowly with temperature),$\Delta E_a$is the ionization energy and K is Boltzmann constant. From the above equation we estimated the value of ΔE_a it was found to be 0.11 eV for$\sigma_{//}$and$\sigma_{\perp}$. The results obtained for both $\Delta E_g$and ΔE_a are independent of direction and there is no effect of the impurities on $\Delta E_g$and ΔE_a in both directions The same behavior was observed previously in an already [13].For gaining much understanding about the anisotropy of σ values.

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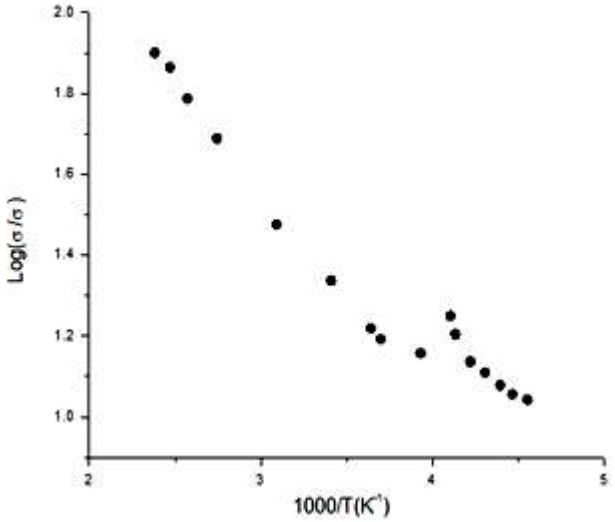


Figure 2: Anisotropy of the electrical conductivity of Tl_2Se in two crystallographic directions**.**

Fig.(2) is depicted .It illustrates the variation of the anisotropic factor $\sigma\perp/\ \sigma_{//}$ against temperature in the same investigated temperature range (i.e. 225 –500K). The, anisotropy law, as evident from the curve, has a general mode of variation similar to the variation of the electrical conductivity. In a previous work on GaTe [13]it was observed that the anisotropic ratio $\sigma\perp/\ \sigma_{//}$ is temperature independent. In 1972;Schmid and Mooserhave observed that the conductivity anisotropy in GaSe of the form[14]:

$$\sigma_{//} / \sigma\perp = A\ (-\Delta E_g/2KT) \qquad (3)$$

Where A depends on the ratio $(m^*_{//}/\ m^*\perp)$ where m is the effective mass of the electrons.From the above equation we calculated the energy gap value and was found to be equals 0.55 eV .Tl_2Se as seen on Fig (3) behaves likes the last formula observed by SchmidMooser[14].

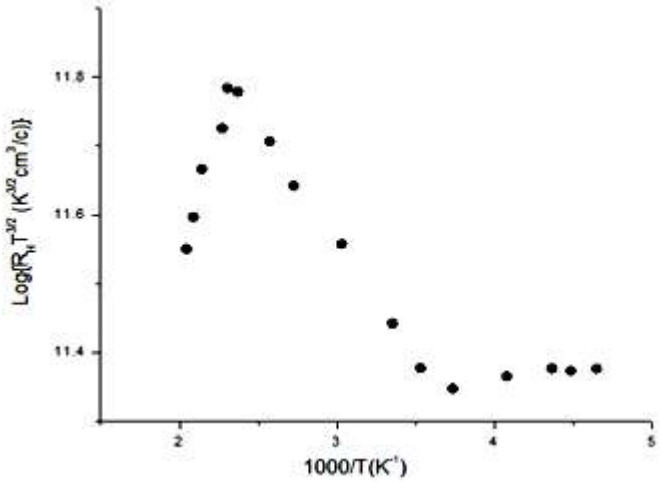


Figure 3: Influence of temperature on the Hall coefficient of Tl_2Se

As for the importance of Hall Effect work in the field of solid state and especially semiconductors, the present investigation is extended to cover this unique phenomenon.

In the same temperature range, the variation of the Hall coefficient against temperature was carried outas illustrated in Fig (4). This was donebased on the next relation:-

$$R_H T^{3/2}\ \alpha \exp\ (-\Delta E_g/2KT) \qquad (4)$$

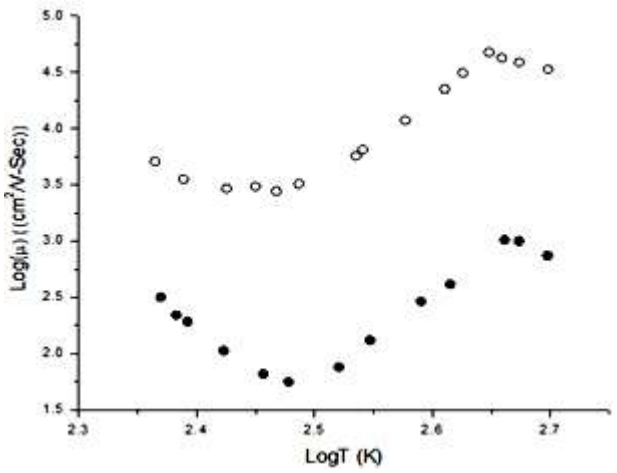


Figure 4: Variation of the Hall mobility of Tl_2Se in two crystallographic directions

Fig (4) represented the relation between $R_HT^{3/2}$ and $10^3/T$, which alsocan be divided into three different parts in the corresponding temperatures, appeared in Table (1). These values are in a good agreement with those obtained from the electrical conductivity measurements.It must be noticed that Hall coefficient is an isotropic against temperature in the both crystallographic directions because it is a scalar quantity.

The Hall mobility was examined in two crystallographic directions. This is done because the electrical conductivity and Hall coefficients are available. Illustration of this combination(Hall mobility) ispresented inFig (4) where we can observe the following: -The general mode of $\mu_{//}$and $\mu\perp$ have the same variations.

-The Hall mobility μis an anisotropic quantity
-Values of $\mu\perp$are much higher than ofthose $\mu_{//}$. For instance at room temperature $\mu_{//}$=17.78 (cm^2/v.sec).Where it is for $\mu\perp$ at same temperature equals 500 (cm^2/v.sec). The general behavior of μagainst T can be divided into three parts:

At T < 270K, (the low temperature part) which corresponds to the extrinsic conduction. The middle region (270 <T < 240 K) and, the high temperatures part (T < 420 K) which is the intrinsic conductivity part.The expected negative temperatures coefficient of the mobility (in the high temperatures) appears only above 420K. This behavior was observed during investigation of GaTe[17] crystal. The observed behavior of the charge carrier's mobility needs much work for having an accurate explanation for understanding the real scattering mechanism of the charge carriers in this crystal. In the present time we consider the following power laws$\boldsymbol{\mu_{//}\alpha\ T^{2.5}}$ (in the low temperatures) and $\boldsymbol{\mu_{//}\alpha\ T^{-3.4}}$ (in the high temperatures).

The first power law means that the scattering of the carriers is attributed to impurities. While the second one suggests that the scattering mechanism, in this temperature range, is due to the polar optical mode which results from the polar interaction between charge carriers and optical phonons.The experimental results of the variation of the carrier density versus reciprocal temperature are depicted in Fig(6).

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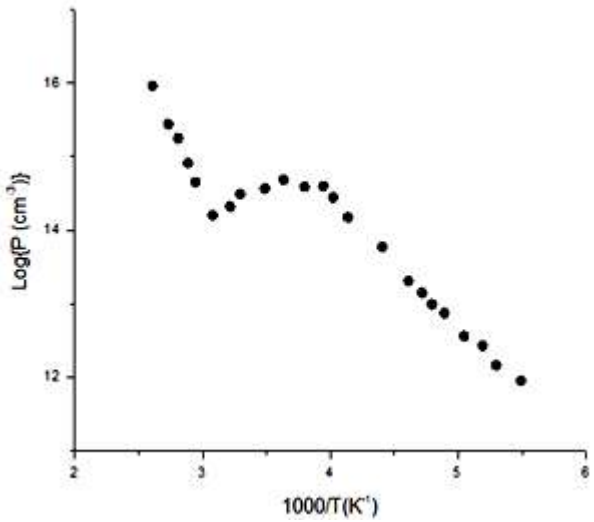


Figure 5: Influence of temperature on the charge carrier concentration of Tl_2Se

At low temperatures (below270K) the carrier concentration is determined by the number of ionized acceptors. This is noticed evidently because the slope of the variation of the carriers is quite slow. In this range we computed the depth of the acceptor level as 0.11 eV. At high temperatures the crystal is exhibiting an intrinsic behavior then the expected value for the intrinsic concentration could be given by:-

$$p_i = 2(2\pi k / h^2)^{3/2}(m_n^* m_p^*)^{3/4} T^{3/2} \exp(E_g / 2kT) \quad (5)$$

Where m_n^* , m_p^* are the effective masses of both electron and hole respectively . Utilization from this formula leads to calculation of the energy gap width of TI_2Se .It was found to be 0.55 eV . Both ΔEg and ΔEa values are in a good agreement with those obtained from the electrical conductivity and Hall measurements. Finally the charge carrier's concentration, at room temperature, equals to 12 x 10^{13} cm^{-3} in TI_2Se crystal.

4. Conclusion

1) The anisotropy of the electrical conductivity was studied beside the Hall Effectphenomenon was investigated in a temperature range extending from 225 up to 500K.
2) The dependence of anisotropic factor on temperature was checked and found to be similar to dependence of electrical conductivity on temperature.
3) The anisotropy of Hall mobility was also studied in the same temperature range.
4) It is proven that the Hall coefficient is a scalar quantity not a vector one i.e. it does not depend on the crystallographic directions.
5) The scattering mechanism of the charge carriers was discussed.
6) The energy gap was found to be = 0.55 eV;And the ionization energy = 0.11 eV.
7) $Tl_2Se$is a promising semiconductor with a layered structure of tetragonal symmetry.
8) In the present work, many physical parameters were estimated.

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