INTERNATIONAL JOURNAL OF
ADVANCED SCIENTIFIC RESEARCH AND INNOVATION

Print ISSN: 2785-9533 - Online ISSN: 2785-9541

VOLUME 6, ISSUE 2, 2023, 70 – 79

Growth and characterization of Ag_2Te single crystals

M. Fangary, M. Nassary
Physics Department, Faculty of science, south valley University

Abstract

A special design, based on the Bridgman technique, was used in our laboratory for preparing single crystals of Ag_2Te. By the combination of electrical and thermoelectrical measurements, electronic transport parameters of Ag_2Te single crystals have been studied. Through the transport experiment, the semiconducting properties of this compound could be revealed. The compound Ag_2Te, which has p-type conductivity, is studied throughout a wide temperature range extending from 163- 520 k.

Keywords:
Ag_2Te single crystal, Hall coefficient, thermoelectric power

Introduction

The semiconducting phases of the silver chalcogenides, Ag_2S, Ag_2Se and AgTe, Ag_2Te and Ag_5Te, historically have received less attention than other semiconductors with narrow band gaps and high carrier's mobilities and Kane's dispersion law. Perfectly stoichiometric material has negligible magneto resistance. But recent Silver telluride compounds that can be used in data storage devices [1], optical materials [2] and magnetic measurements [3]. Like numerous metal telluride (PbTe , Bi_2Te_3, $Cu_{1.75}Te$ …), Ag_2Te is of high interest as thermoelectric material, converting heat into electricity and vice versa [4] The optical properties of silver tellurides thin films was investigated by Apple [5]. Sharma [6] and Dhere and Goswami [7] have studied the structural properties of silver telluride thin films by scanning electron microscopy. Recently galvanomagnetic studies have also been carried out in silver telluride thin films [8-10]. P. Gnanadurai [9] have studied the influence of the doping concentration and magneto resistivity in silver telluride thin films. the electrical and thermoelectrical properties of p-type Ag_2Te were investigated by [11,12]. Now it is evident that more work is required in order to understand the transport properties of the Ag_2Te in the single crystalline form and to reveal the discrepancies concerning the physical properties. the goal of the current paper is to estimate some important physical parameters of this compound

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Experimental

In order to grow perfect Ag_2Te single crystal a modified Bridgman -stock Barger technique was used. the ampoule is charged with a required amount of material 6.16g of pure silver (Aldrich mark) 99.9999% representing 62.79% and 3.65 g of pure Tellurium (Aldrich mark) 99.9999% representing 37.21%.the appropriate amount was first sealed in silica ampoule at a pressure of 10^{-5} Torr. At the beginning of the growth run, the ampoule was held in the hot zone of the furnace at 1263 k to about 24 hours for melt homogenization. Then, the melt was shaken during heating several times to accelerate the diffusion of the constituents through each other. the mechanical system is always used to draw the charged ampoule from zone to another with the required rate.in our case , the charged ampoule is lowered gradually and slowly through temperature steps at rate of about 1.6mm/h. the temperature of the middle zone is 1233k corresponding to the crystallization temperature of Ag_2Te according to the phase diagram [13].The duration time for producing Ag_2Te as single crystal is about 11days.the produced ingot was identified by means of x-ray analysis to be Ag_2Te crystal . The results were in good agreement with the published values [14] recorded in the international center for diffraction data, ICDD standard. For studying the electrical conductivity and Hall effect, the sample was prepared in a rectangular shape with dimension $10.45x3.5x2.65mm^3$. silver paste contact was used as ohms' contact. The ohmic nature of the contacts was checked by recording the current -voltage characteristic. the conductivity and the Hall coefficient were measured by a compensation method in a special cryostat [15]. with a conventional D.C. type measuring system by using a tinsley (UJ33E Mark) potentiometer in a magnetic field of 0.5 tesla. the temperature range of investigation was extend from 163k up to 520 k. All measurements were carried out under vacuum condition of about 10^{-3} Torr. For studying thermoelectric power (TEP), an evacuated calorimeter (10^{-3}) was used to protect the sample from oxidation and water vapor condensation at high and low temperatures respectively. the calorimeter has two heaters. the outer heater (the external source) discharges its heat slowly to the specimen environment. The inner heater (connected to the lower end of the crystal) was made purposely to control the temperature and its gradient along the specimen. the TEP is calculated at different temperatures by dividing the magnitude of the thermo voltage difference across the crystal by the temperature difference between the hot and cold ends.

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Results and discussion

1- Temperature dependence of electrical conductivity and Hall effect for Ag_2Te: -

The temperature dependence of electrical conductivity of Ag_2Te single crystal samples was studied, over temperature range from 163k to 520k. Fig [1] shows the variation of the electrical conductivity σ with temperature. In the region from 163k to 248k showed unchangeable of electrical conductivity with temperature, i.e. the conductivity remains almost temperature independent. After this range the compound takes the general behavior of the semiconductor materials and the curve can be subdivided into three regions: the first region (248k-403k) represent the extrinsic region. In this region the electrical conductivity increases slowly with temperature and this is due to the fact that the carrier concentration, in this region, is determined by the number of ionized acceptors liberated from the impurity level. In this range the following formula describes the relation between σ and T :-

$$\sigma = \sigma_0 \exp(-\Delta E_a/2K_BT)$$

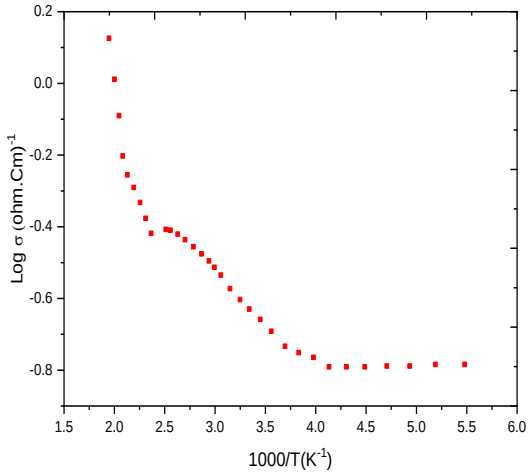

Fig (1): -Temperature dependence of the electrical conductivity for Ag_2Te single crystal.

Where σ_0 is a pre-exponential factor and K_B is the Boltzmann constant. From the above relation we calculated the impurity ionization energy ΔE_a. It was 0.11 ev. the second region from 403k to 433k, the electrical conductivity decreases with increasing temperature, which it caused by the phase transition from the

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monoclinic α-Ag_2Te to the FCC β- Ag_2Te; which the cubic phase exhibits metallic character. The third region from 438k-520k represents the intrinsic region. In this region the conductivity σ increases with increasing temperature. The excitation of the carriers from the valence band to the conduction band is responsible for this rise of the conductivity where the temperature is high enough. The following equation is used to determine the width of the energy gap: -

$$\sigma = \sigma_0 \exp(-\Delta E_g/2K_BT)$$

The value of the energy gap ΔE_g is 0.9 ev as calculated from the last equation. the room temperature conductivity of this crystal is 0.249 $\Omega^{-1}.cm^{-1}$.

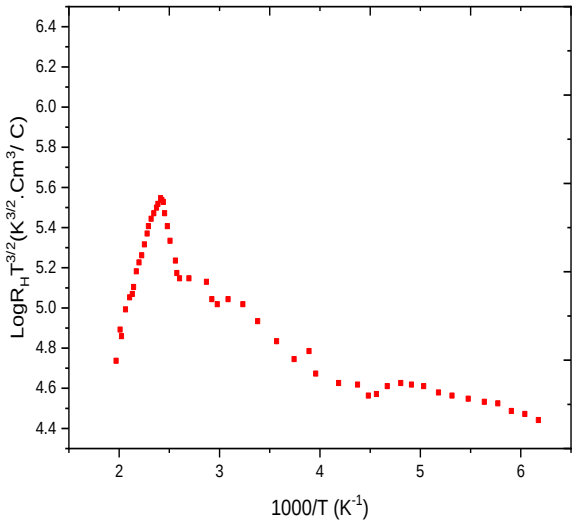

Fig (2): -Relation between $R_HT^{3/2}$ and $10^3/T$ for Ag_2Te Single crystal.

Hall effect measurements are very important. Thus, the present investigation is extended to cover this unique phenomenon. In the same temperature range (163-520 k) the variation of the hall coefficient R_H against temperature was examined as seen in Fig .2. The positive sign of the hall coefficient indicates p-type conduction. The curve is divided mainly into three different regions: - The first region in the temperature range from 163k-393k represents the extrinsic region. From the slope of this part, the activation energy is calculated to be E_a=0.14ev. The second region in the temperature range from 393k to 433k represents the transition region. In the transition region the behavior is governed mainly by the concentration of the charge carriers and their mobility. Above 433k, the third region at high temperature represents the intrinsic region. It obvious that R_H varies rapidly with temperature in a linear fashion.

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From the slope of this part, the energy gap is calculated to be E_g=0.93ev. This value is in a good agreement with the conductivity measurements. A combination of the Hall measurements and the electrical conductivity data was used to study the temperature dependence of the mobility of the charge carriers in the two directions parallel and perpendicular to the layers plane.

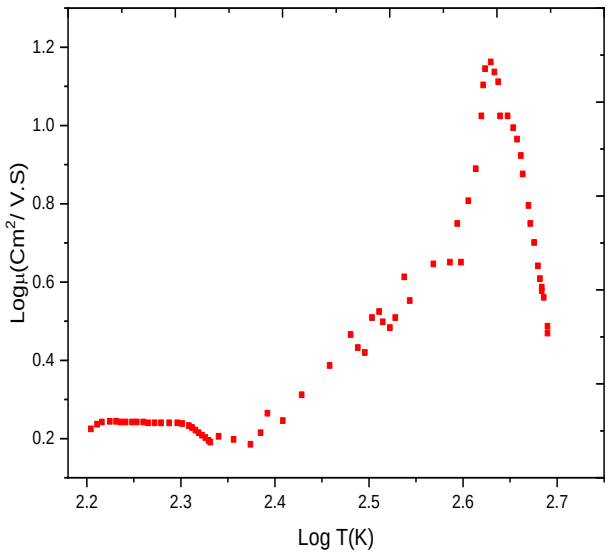

Fig (3): -The behavior of Hall mobility as a function of temperature for Ag_2Te single Crystal.

Fig (3). Indicated the relation between the carrier mobility μ_H and T. The relation $\mu_H \alpha\ T^n$ governs the variation of the Hall mobility against temperature. This curve distinguishes three regions. In the first region at low temperature from 163k to 248k the mobility seems to be fairly constant with temperature. the second region in the temperature range from 248k to 433k the μ_H increases with temperature obeying the law $\mu_H \alpha\ T^{4.36}$. This behavior agrees with semiconductors behavior. such behavior is characteristic of a scattering mechanism of the charge carriers on ionized impurities. In the third region in temperature range from 438k to 520k the hall mobility μ_H decreases with temperature by relation $\mu_H \alpha\ T^{-8.55}$.

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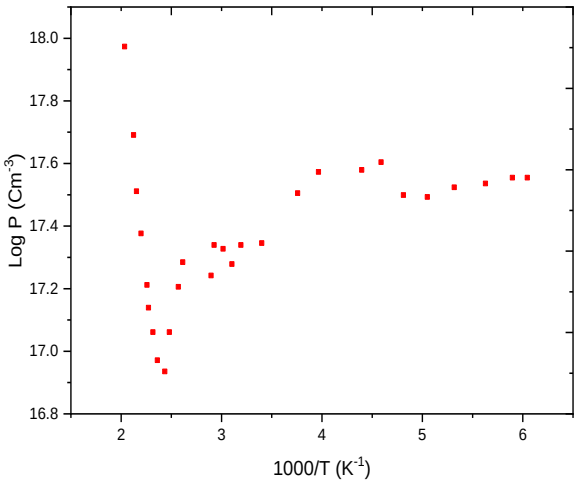

Fig (4): - Variation of hole concentration with temperature for Ag_2Te Single crystal

Fig (4) represents the dependence of charge carrier concentration on temperature .The charge carrier concentration was calculated from hall data by using the relation ($p=1/R_H$ e) where, p is the hole concentration , e is the electron charge .From this figure we notice that the concentration of currieries in the extrinsic region increases slowly with increasing temperature , while it increases rapidly with temperature in the intrinsic region .The energy gap calculated from the slope of this curve in the high temperature range is found to be E_g =0.98ev.The hale concentration at room temperature is equal to $p=2.33x10^{17}$cm.

Temperature Dependence of thermoelectric power of Ag_2Te

The thermoelectric power TEP measurements were performed with the direction of temperature gradient parallel to the layer plane in a wide temperature range (172-430k).

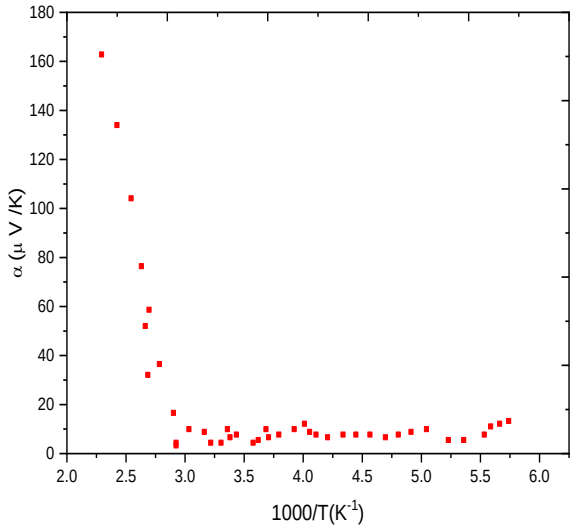

Fig (5): - Relation between α and 10^3 / T for Ag_2Te single crystal.

Fig (5): - Illustrates the general behavior of the variation of TEP with temperature. From this curve ,the magnitude (seebeck coefficient) α is constant below 340k (except for the anomaly at 298k) Above 340k , α increases sharply with increasing the temperature till reaching the maximum value 170.59μv/k corresponding to 430k .The rise of α is attributed to the thermal activation of the charge carriers in this rang .It is evident from these measurement that α has appositive sign in the whole temperature range of investigation .This is in agreement with results obtained from hall data .the behavior of thermoelectric power with temperature in the intrinsic region can be described the equation given by lauc[16]:-

$$(1)\propto = -\frac{K}{e}\left[\frac{b-1}{b+1}\left(\frac{\Delta E\, g}{2KT}+2\right)+\frac{1}{2}Ln\left(\frac{Mn*}{Mp*}\right)3/2\right]$$

Where b is the ratio of electron and hole mobility, ΔEg is the energy gap and (Mn*,Mp*) are the effective mass of electrons and holes, respectively. The relationship shows that a plot of α in the intrinsic range as a function of the reciprocal of absolute temperature in straight line shown in fig (5). The slope of the linear part is used to estimate the ratio of the electron to hole mobility. Taking ΔEg=0.9ev, from the previously obtained data ,the ratio b=(μ_n/ μ_p) is found to be 4.8. Hence by using the value of μ_p =3.64 cm^2/V.Sec, the electron

mobility can be deduced and its value is found to be 17.56cm^2/V.Sec .Another important parameter can be deduced with the aid of the obtained values of μ_n and μ_p using the Einstein relation ,from which the diffusion coefficient for both carriers(holes and electrons) can be evaluated to be 0.094 and 0.455 cm^2/Sec respectively. From the intersection of the curve, the ratio between the effective masses of the electrons and holes can be estimated to be ($m^*_n/m^*_p = 3.12x10^{-3}$) assuming that this ratio does not vary with temperature.
In the impurity region the following equation can be applied [17]: -
In impurity region the following equation can be applied:

$$\alpha = k/e[2-Ln(ph^3/2(2\pi m^*_p KT)^{3/2}] \text{ ---------(2)}$$

where, p is the hole concentration. The effective mass of holes m_p^* is calculated from the relation between thermoelectric power and Ln T. The relation is illustrated in fig (6) .

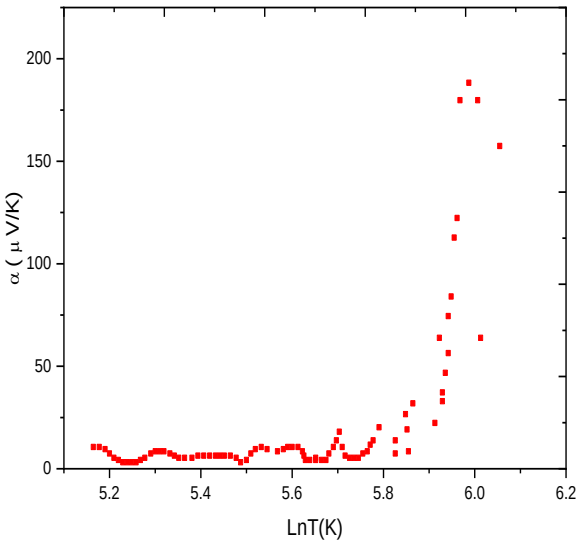

Fig (6) Relation between thermoelectric power and Ln T for Ag_2Te Single crystal

This curve yields a straight line in the impurity region. Calculation of the effective mass of the holes from the intersection of the curve yields the value $m_p^* = 7x10^{-34}$kg . Combining these values with the above-mentioned results for the ratio m_n^*/m_p^* , one obtains an effective mass of the electrons $m_n^* = 2.19x10^-$

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^{36}kg.The results indicate that the electron mobility is much higher than the hole mobility .this is acceptable since the hole effective mass is much greater than that of electrons .the calculated values of the effective masses for both minority and majority carriers can be used for the determination of the relaxation time for both current carriers .Its value for holes comes to be 1.59x 10^{-21}sec ,whereas for electrons 2.4x10^{-23} sec . Also from the diffusion length $L=\sqrt{D_T}$ the values of L_p and L_n are calculated and they are found to be 1.22x10^{-11}cm and 3.3x10^{-12}cm for holes and electrons , respectively .

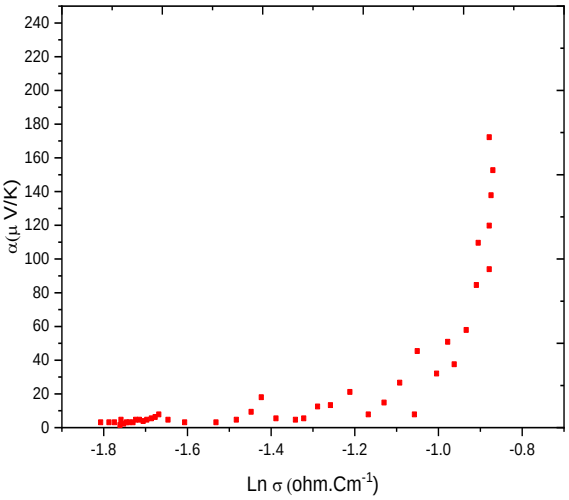

Fig (7): - Variation of thermoelectric power with electrical conductivity for Ag_2Te single crystal.

Fig (7) shows the dependence of α on the natural logarithm of electrical conductivity according to [18]: -

$$\propto = \frac{k}{e}\left[A + Ln\frac{2(2\pi m*pKT)3/2e\mu}{(2\pi h)3}\right] - \frac{k}{e}Ln\sigma \quad \textbf{(3)}$$

It seems that the magnitude α is constant as the electrical conductivity increase, in the lower conductivity region, up to temperature corresponds 340k. After which α increases gradually reaching a maximum value at temperature equal 430k.

Conclusions

In Summary, Ag_2Te single crystals have been successfully prepared by Bridgman technique. Electronic transport parameters of Ag_2Te single crystals have been studied by the combination of electrical and thermo electrical measurements. The compound Ag_2Te, which has p-type conductivity, is studied throughout a wide temperature range extending from 163- 520 k.

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