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Thermoelectric, transport and microstructure properties of binary chalcogenide Ga_2Te_3 Crystals

R. H. Al-Orainy [1], A. T. Nagat [1], S. A. Hussein [2], A. A. Ebnalwaled [*3]

[1] *Physics Department, Sciences of Faculty of Girls, King Abdulaziz University – KSA*
[2] *Physics Department, Faculty of Sciences, South Valley University, Qena, 83523 Egypt*
[3] *Electronic & Nano Devices lab, Physics Department, Faculty of Sciences, South Valley University, Qena, 83523 Egypt*
kh_ebnalwaled@yahoo.com

Abstract:

High quality crystals of Ga_2Te_3 compound were prepared by modified Bridgman technique, the microstructure and transport properties were investigated. The dependence of electrical conductivity, Hall effect, Hall mobility and charge carriers concentration on temperatures was performed in the temperature range (294 K - 553 K). The temperature dependence of the ac conductivity and the frequency exponent, s is reasonably well interpreted in terms of the correlated barrier-hopping CBH model. The dependence of thermoelectric power in the temperature was studded and a lot of semiconducting parameters (effective mass, life time, diffusion length and diffusion coefficient) were calculated.

***Keywords*:**

Crystal Growth, Semiconductors, Transport Properties, Ga_2Te_3.

1. Introduction

Metal sesqui-chalcogenides, $M_2^{III}X_3^{VI}$ (where M = Ga, or In and X = S, Se or Te) crystallize in the zinc-blende or wurtzite structure, where one-third of the cation sites is vacant in order to satisfy the chemical valences. These vacancies noted as ''structural vacancies'' are inevitable to hold tetrahedral crystal structures. The orderings of the large amount of vacancies have received crystallo-chemical interest, and have been studied for a fairly long time. In earlier works on Ga_2S_3, Ga_2Se_3 and Ga_2Te_3, Hahn and Klinger [1] reported that Ga_2S_3, Ga_2Se_3 crystallizes in zinc-blende and wurtzite structures and Ga_2Te_3 crystallize only in the zinc-blende structure.

In recent years, a great deal of interest has been focused on semiconducting III–VI compounds. This interest has been driven by their possible device applications. The electrical and optical properties of Ga_2Te_3 are very important for different applications, since this material is a semiconductor whose properties are dominated by stoichiometric vacancies in one-third of the cation sites [2]. The bonding is tetrahedral, and the compound has very large concentration of vacancies. The presence of the gallium vacancies in the Ga_2Te_3 lattice makes it an interesting material for fabricating switching devices. Pearson [3] showed that Ga_2Te_3 has a cubic structure with 5.886 A° lattice constant. A greater portion of published data deal with structure, switching, and memory properties of Ga_2Te_3 crystals [4 – 9]. It is clear from a literature survey that there is a phase transition in Ga_2Te_3 [10, 11].

The lack of literature concerned with transport properties Ga_2Te_3 crystals led us to perform this paper in order to:

1- Prepare and characterize Ga_2Te_3 crystals (microstructure, dc electrical conductivity, Hall coefficient, carrier concentration, charge carrier mobility, ac conductivity and thermoelectric properties).

2- Estimate semiconducting parameters for Ga_2Te_3 crystals.

These investigations are essential for the understanding of the materials and consequently open up the new possibilities of practical applications.

2. Experimental

Crystals of Ga_2Te_3 were grown from a melt using a modified Bridgman technique. The silica glass ampoules were cleaned and then weighted with high-purity (6 N) elements. The ampoule is coated internally with a thin layer of pyrocarbon to prevent the produced ingot from adhesion with silca tube. The ampoule was evacuated to a pressure of less than10^{-6} Torr and sealed off. The mixture was slowly heated (1.5 ^{o}C min^{-1}) over a temperature range of 200 – 250 ^{o} C to minimize the risk of cracking the ampoule. The temperature was kept at 1065 ^{o} C for two days to homogenize the melt. The crucible was then lowered to the cold side at 300 ^{o}C at a speed of 1.6 mmh^{-1}. The growth method and the experimental apparatus have been discussed in details elsewhere [12].

The platelet was cleaved with rectangular shape for the electrical conductivity and Hall effect measurements; the sample dimension was adjusted to be 9 X 3 X 2.4 mm^3 by gentle cleavage. For the purpose of thermoelectric measurements the length was adjusted to be 5 mm, while the crystal cross-section was 8 mm in diameter. Both electrical and Hall effect measurements were carried out in an evacuated Pyrex cryostat designed for this purpose [13]. For studying the thermoelectric power an evacuated calorimeter was used to protect the sample from oxidation and water vapor condensation at high and law temperature respectively. Details about the apparatus and method of measurements are obtained elsewhere [14, 15].

The silver conducting paste contacts were soldered on the Ga_2Te_3 to carry out the electrical conductivity, Hall effect and thermoelectric measurements. The ohmic nature of the contacts was verified by recording the current – voltage characteristics for forward and reverse directions. From the current – voltage characteristics, the ohmicity coefficient has the nearest value to 1.

3. Results & Discussions

3.1 Microstructur properties of Ga_2Te_3 crystals

Figure 1 depicts the X-ray diffractogram for the prepared Ga_2Te_3 crystals. The spectra confirm the crystalline nature of the prepared crystals. XRD measurements showed agreement with Cross-Ref PDF No. 00-035-1490.

Using known equations [16, 17] we can determine accurate lattice parameter *a* for the obtained crystals, we determine it to be 5.91, while it equal 5.89 for the standard Ga_2Te_3 crystals.

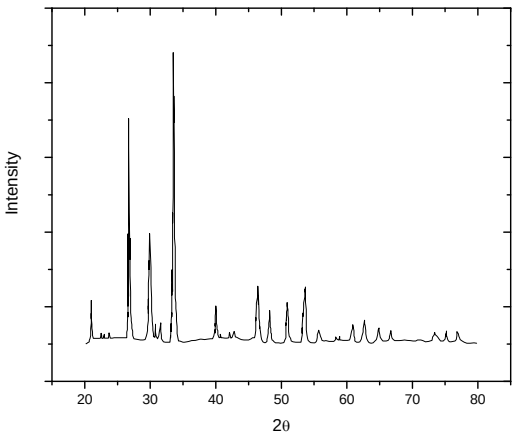

Fig. 1: - X – ray diffraction for Ga_2Te_3 crystals.

Crystallite size (*D*) of the obtained Ga_2Te_3 crystals was calculated to be 46.52 nm from the Debye–Scherrer's formula from the full width at half-maximum (FWHM) of the peaks expressed in radians [18, 19].

3.1 Electrical Properties for Ga_2Te_3 crystals

The present electrical conductivity work was carried out in a temperature range extending from 294 K up to 533 K. Fig. 2 shows the electrical conductivity σ *vs.* 10^3/T for Ga_2Te_3 crystals. As shown, in the investigated temperature range, the logarithm of the conductivity showed a linear dependence on the temperatures with two modes of conduction in addition to the transition region that appeared between them. From the relationships between the conductivity and the temperature, the energy gap is

deduced to be 0.9 eV while the ionization energy is 0.3 eV, The increase of σ in the intrinsic part (above 368 K) is regarded as a result of excitation of the carriers from the valence band to the conduction band. However in the extrinsic part (below 263 K), σ increment is regarded as a result of ionization of impurity atoms.

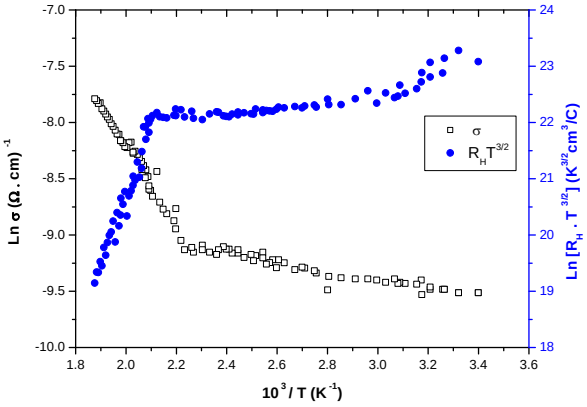

Fig. 2: Temperature dependence of electrical conductivity and Ln. R_H. $T^{3/2}$ vs. 10^3 /T for Ga_2Te_3 crystals.

The transition region (338 – 392 K) is characterized by a slight increase of σ as the temperature increases. This is due to the dominant charge carrier concentration effect in this range. At room temperature, σ has the value 7.38 × 10^{-5} Ω^{-1} cm^{-1}.

The Hall effect measurements were performed in the same temperature range. The sign of R_H indicates that Ga_2Te_3 behaves as a p – type semiconductor. The relationship between Ln (R_H . $T^{3/2}$) and 10^3/T was plotted in Fig. 2. From the figure we can notice that in the extrinsic region R_H decreased slowly with temperature but in the intrinsic region there is a sharp decrease.

Simultaneous measurements of the electrical conductivity and Hall effect permit us to investigate the influence of temperature on Hall mobility. This is typically presented in Fig. 3. The relationship between μ and T in the extrinsic region of temperature justifies the following relationship μ α T^{-4}. Such dependence leads to the assumption that the scattering mechanism is due to the ionized impurities in this range. At high temperature (intrinsic region) μ and T follow the relation μ α T^{-16}; this relation is abnormal if compared with other semiconductors [12 – 15, 20]. This high exponent leads to the assumption that the classical theories of semiconductors cannot be used for understanding the real scattering mechanism of the charge carriers in this temperature range. Hence more efforts are recommended for understanding the definite reason for this behavior. The room temperature Hall mobility is 182.13 cm^2 / V. s.

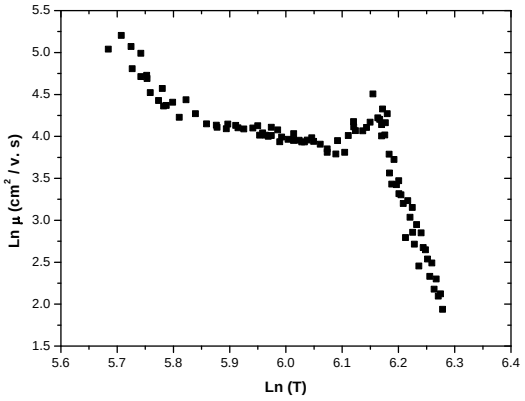

Fig. 3: - Hall mobility as a function of temperature for Ga_2Te_3 crystals

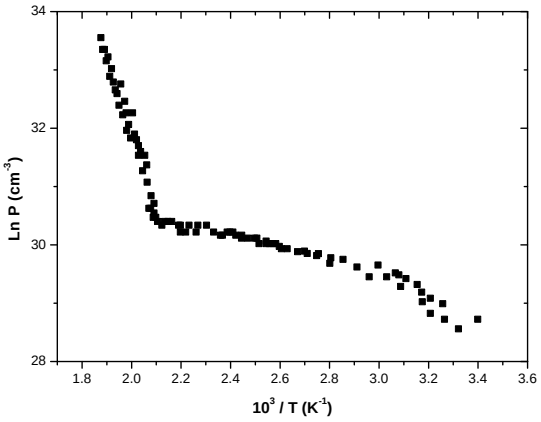

Fig. 4: - Variation of the carrier density versus reciprocal temperature.

Fig. 4 depicts the results of charge carrier density *vs.* reciprocal temperature in the same range as conductivity. There are three regions of this curve; we calculated the energy gap width from the slope of Fig. 6 in the intrinsic region. It is found to be 1.1 eV close to that deduced from other work [11]. The room temperature hole concentration is 2.53 x 10^{12} cm^{-3}.

3.3 ac Conductivity for Ga_2Te_3 Crystals

In order to make a good view about conduction mechanism of Ga_2Te_3, we extend our work to study the temperature and frequency dependence of ac conductivity.

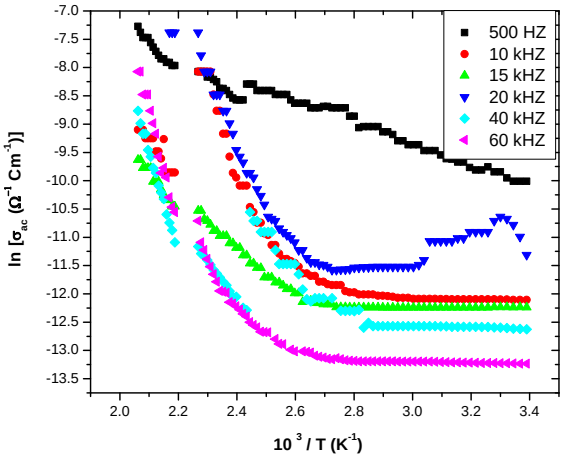

Fig. 5: Frequency dependence of σ_{ac} (ω) for Ga_2Te_3 crystals.

Fig. 5 shows the frequency dependence of ac conductivity (1kHz to 10 kHz) of Ga_2Te_3 crystals at different temperatures. The sample exhibits a frequency – independent conductivity at low frequencies, but at high frequencies σ_{ac} decreases with increasing frequency. The switch over from the frequency – independent region to frequency – dependent region is the signature of conductivity relaxation [21]. For semiconductors systems the ac conductivity follows the power law behavior $\sigma_{ac}(\omega) = A\omega^s$ [22], where A is temperature dependent constant and s is the frequency exponent and ω is angular frequency. As indicated from Fig. 5, s is slightly less than unity and decreases with increasing temperature, which indicates that the conduction in this region is due to correlated barrier hopping (CBH) [23].

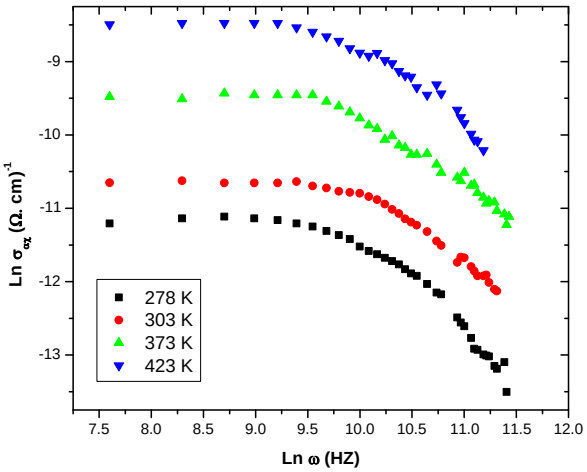

Fig. 6: - Temperature dependence of σ_{ac} for Ga_2Te_3 crystals

Fig. 6 shows the ac conductivity as a function of the reciprocal temperature (294 - 500 K) at different frequencies for Ga_2Te_3 crystals. It is clear from the figure that (σ_{ac}) increases with increasing the temperature. This suggested that the ac conductivity is a thermally activated process from different localized states in the gap or its tails [24]. The activation energy of conduction ΔE (ω) is calculated at different frequencies. From the frequency dependence of ac activation energy for the investigated sample we can obtain that ΔE (ω) decreases with increasing frequency. Such a decrease can be attributed to that, the increase of the applied field enhances the charge carrier's jumps between the localized states, and consequently the activation energy ΔE (ω) decreases with increasing frequency [25]. The decrease of ΔE (ω) with frequency confirms that the hopping conduction is the dominant conduction mechanism for Ga_2Te_3 crystals [24 - 26].

3.4 Thermoelectric properties for Ga_2Te_3 crystals

To gain a more definite idea about the semiconductivity and the behavior of Ga_2Te_3 crystals we measured the thermoelectric power when the temperature gradient was

produced in a direction perpendicular to the c-axis by the differential method in a wide temperature range from 239 up to 410 K. Fig. 7 shows the relationship between α and T for Ga_2Te_3 crystals. From the figure, we can find that the sign of α is positive indicating that Ga_2Te_3 is p-type semiconductor. The value of α is found to be constant with increasing temperature in the range from 239 up to 288 K. From 288 to 331K, α is increased with temperature. This is attributed to the thermal activation of the charge carriers in this range. Up to 331 K, the value of α decreases as the temperature increases. This is a result of the compensation process, which occurs in this temperature range. The value of α at room temperature is (65 μVK^{-1}).

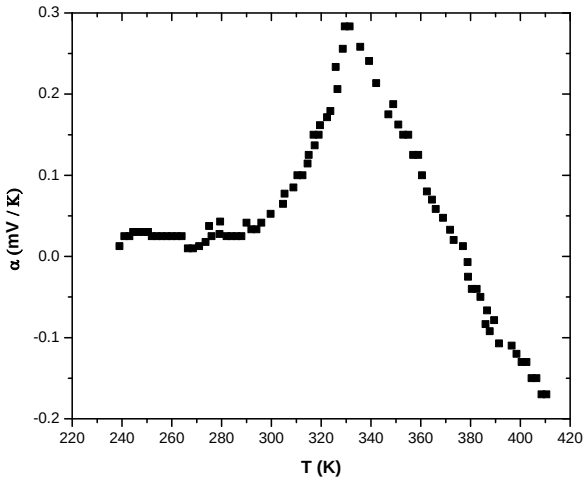

Fig. 7:- Temperature dependence of thermoelectric power for Ga_2Te_3 crystals.

Some major semiconductor parameters for Ga_2Te_3 crystals, such as the electron-to-hall mobility ratio, relaxation time of majority and minority carriers, effective mass of holes and electrons, diffusion coefficient and diffusion length can be estimated by using the formulas suggested in Refs. [25, 27]. The electron-to-hall mobility ratio was found to be 1.1 so μ_n = 200.3 cm^2 / V. s. The values of the relaxation time were calculated to be τ_p = 4.04 x 10^{-16} s and τ_n = 3.84 x 10^{-16} s, for holes and electrons, respectively. The diffusion constants for electrons and holes were calculated to be D_p = 4.7 cm^2 s^{-1} and D_n = 5.16 cm^2 s^{-1}. The diffusion length for holes and electrons respectively is L_p = 4.3 x 10^{-8} cm and L_n = 4.5 x 10^{-8} cm. finally the effective mass for holes and electrons respectively was found to be m_p^* = 1.7 x 10^{-37} kg and m_n^* = 1.3 x 10^{-37} kg .

Conclusion

- In the present work Ga_2Te_3 crystals were grown by modified Bridgman technique.
- The electrical conductivity at room temperature has the value 7.38 × 10^{-5} Ω^{-1} cm^{-1}. The energy gap is deduced to be 0.9 eV while the ionization energy is 0.3 eV.
- The sign of R_H and α indicates that Ga_2Te_3 behaves as a p – type semiconductor.
- The scattering mechanism in low temperature range is due to the impurities.
- The conduction mechanism was analyzed using the thermionic emission and variable range hopping of charged carrier's theories.
- The values of the relaxation time were calculated to be τ_p = 4.04 x 10^{-16} s and τ_n = 3.84 x 10^{-16} s, for holes and electrons, respectively. The diffusion constants for electrons and holes were calculated to be D_p = 4.7 cm^2 s^{-1} and D_n = 5.16 cm^2 s^{-1}. The diffusion length for holes and electrons respectively is L_p = 4.3 x 10^{-8} cm and L_n = 4.5 x 10^{-8} cm. finally the effective mass for holes and electrons respectively was found to be m_p^* = 1.7 x 10^{-37} kg and m_n^* = 1.3 x 10^{-37} kg .

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BIOGRAPHIES

A. A. Ebnalwaled was born in Qena, Egypt, in 1978. He received the B.Sc. degree in Physics (2000), the M.Sc. degree in experimental solid state physics (2003) and the Ph.D. degree in Materials science from the Faculty of Science, South Valley University. In 2013, he was promoted to Associate Professor of Materials science and Nanotechnology. He is currently the head of physics department, faculty of science, South Valley University.

He has been established Electronics and Nano Devices lab at the faculty of Science, South Valley University in Qena.

His research interests lie in the areas nanomaterials preparation, characterization and its uses in electronics, energy and biological applications.

He has many international publications and books in the field of materials science and nanotechnology. He has organized and served on scientific committees of several international conferences.

