Transport Properties of $Ga_{0.45}In_{0.55}Sb$.

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The thermoelectric power and Hall measurement carried out on the as-prepared sample of $Ga_{0.45}$ $In_{0.55}$ Sb confirmed that the sample is n-type semiconductor. The existence of two minima at 3.5 and 4.8 Vcm^{-1} on the σ - E characteristic reflects the occurrence of the Gunn effect. A metallic like behavior of the sample is observed and interpreted. The effect of the magnetic field on the concentration of the electrons is apparent

1. Introduction

The compound semiconductors GaSb and InSb and the alloys based on these compounds are widely used in electronic and optical devices, such as microwave rectifiers, tunnel diodes, Hall effect devices, infrared photocells, optical filters, etc., so many researchers [1-7] paid special attention to these compounds. The band gap of the compound Ga_x In_{1-x} Sb found to be varied from 0.7 to 0.2 eV with changing the value of x [8]. Accordingly [9], GaSb and InSb alloys are elaborated in order to obtain Gunn oscillators with low threshold voltage. Nowadays, polycrystalline semiconducting alloys have received special attention because of their use in devices reduces the complications of ultrapurification and highly controlled conditions required for the growth of single crystals[10].

The present work aims to attempt to prepare the alloy $Ga_{0.45}$ $In_{0.55}$ Sb and to study some of its transport properties, namely electrical resistivity, Hall coefficient and mobility as a function of temperature and / or magnetic field.

2. Experimental Procedure

To prepare the nominal composition $Ga_{0.45}In_{0.55}Sb$, high- purity gallium, antimony and indium (each 99.999% pure) were used. The appropriate proportions of the elements, were weighted, mixed together and then sealed in a thick wall transparent quartz tube with internal diameter 8 mm under a vacuum of 10^{-5} mmHg. The ampoule was heated at a rate of 3 $Kmin^{-1}$ to 50 K above the liquidus temperature [11] and held for about 20 h with periodic shaking. Then, the ampoule was quenched in ice. The resultant cylindrical shape ingot with its length 4 cm was cut in equal pieces of thickness ~ 3 mm and then polished mechanically.

Special attention was paid to fine polishing so that, each of the samples has two parallel and optically flat surfaces satisfactory for the proposed measurements. The X-ray diffraction analysis revealed that the as-prepared composition is having a polycrystalline structure as shown in Fig. (1). The main phases coexist together are Ga, Sb, GaSb, GaIn and GaInSb. Using Scherrer equation:

$$r = \lambda / \Delta\theta \cos\theta$$

where λ is the wave length of the CuK_{α} X-ray radiation, $\Delta\theta$ is the half peak width of the corresponding phase and θ is the diffraction angle, the average grain size of the grains of the coexisted phases GaInSb, GaSb, Ga and Sb was found to be 2558, 2326, 1722 and 1940Å respectively. These results are confirmed by the surfaces morphology studies carried out by using the scanning electron microscope type Jeol JsM-5300(Japan). Since the average grain size was equal to about 2000 Å. The photo-micrograph shown in Fig. (2) indicates that most of the grain structure are complex. Each large grain contains spread small grains which are probably unlike phases. Also, precipitates are present at the grain boundaries of some grains.

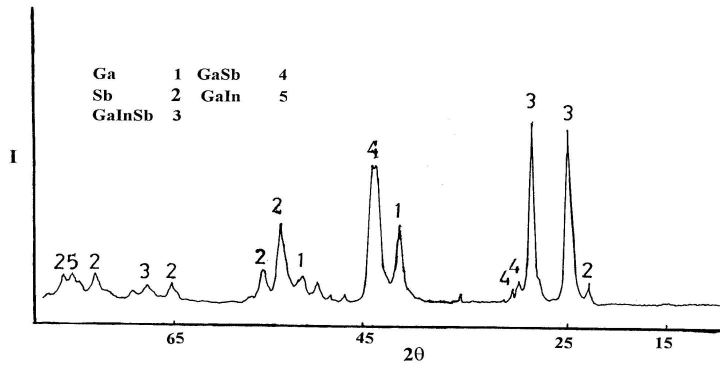

Fig. (1): X-ray diffractograms of $Ga_{0.45}In_{0.55}Sb$.

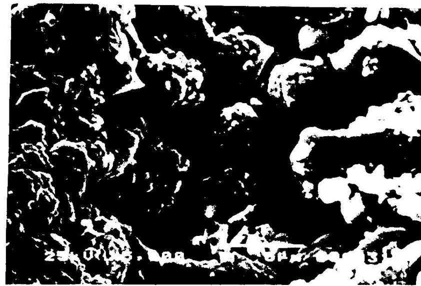

Fig. (2): SEM Photomicrographs of $Ga_{0.45}In_{0.55}Sb$.

D.C electrical conductivity and Hall measurements were carried out using the standard Van-Der Pauw method [12, 13] within the temperature range 175-390K. An Al 745 Ax type voltage/current source was used for constant current supply, and the voltage was measured using a Keithly 177 microvoltmeter. The temperature of the assembly was monitored using chromel–alumel thermocouple. For Hall measurements, an electromagnet (Magnet type M77 Oxford) with magnetic field of about 1.2T was used. The thermoelectric power was measured using a conventional method with copper electrodes and pressure contact. Also, the current – voltage characteristics was measured by using a conventional pressure contact sample holder.

3. Results

The current density (J)–electric field (E) characteristic of the as–quenched bulk sample of the composition $Ga_{0.45}In_{0.55}Sb$, was as shown in Fig. (3-a). As shown in Fig. (3-a), up to ~ 3.2 Vcm^{-1}, the characteristic seems linear verifying ohmic conduction. This is followed by two other non-linear parts limited in the range 3.2 – 3.6 Vcm^{-1} and 3.6 – 4.8 Vcm^{-1}, both are verifying power dependence of J on E. Further increase of E was followed by an abrupt increase of J which is followed by an evidence of the possibility of attaining switch- ON process. The double logarithmic relation between J and E (though is not here) seemed consisting of three main linear parts with the slopes equal respectively to 1.02 that is very near to unity verifying ohmic conduction at the lower range of E, 0.34 that is far below unity verifying field inhibition of conductivity (σ) and 0.48 indicating a strong inhibition of conductivity. Furthermore, the σ - E plot shown in Fig. (3-b) has a zero coeffecient below ~ 3.2 Vcm^{-1} and two successive negative differential conductivity regions with their minima appearing respectively at ~ 3.5 Vcm^{-1} and ~ 4.8 Vcm^{-1}. Such irregularity in the σ - E plot can reflect a complexity in the band structure of the prepared material and in turn can reflect Gunn effect.

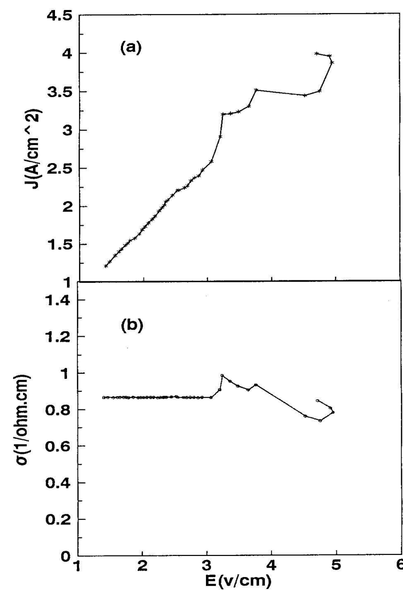

Fig.3:(a) J vs E for as-prepared sample.
(b) σ vs E for as-prepared sample.

Within the whole considered range $125 \leq T \leq 435K$, the observed thermo-electric power (TEP) is negative confirming the domination of electron contribution to conduction. The S–1/T plot is consisting of three parts with different slopes as shown in Fig.(4). So, the temperature dependence of seebeck coefficient (S) can be described by

$$S = (-k/e) [(E_s/kT) + A] \qquad (1)$$

where k is the Boltzmann's constant, e is the electronic charge, E_s is the TEP activation energy which represents in that case the Fermi energy, since the TEP possesses minus sign[14,15] and A is a dimensionless parameter concerns the carriers scattering mechanisms[16] and assumed to be a measure of the kinetic energy transported by carriers[17]. E_s increased with increasing the range of T and possessed respectively the values 8.46×10^{-4}, 4.13×10^{-3} and 4.17×10^{-2} eV for the lowest, the intermediate and the highest range of T. This indicates more thermal activation of electrons with increasing T. Meanwhile, the relatively small value obtained for E_s with regard to the smallest value 0.2 eV obtained for the band gap[8] confirm mixed conduction and the results of TEP can be described in terms of the two carrier Model. One as donor and the other as acceptor are coexisted together in the composition. However, it is clear that, the contribution of holes is completely compensated with that of electrons. This is why the sign of TEP is negative over the whole range of T. This can be confirmed in terms of the apparently high value obtained for the concentration of negative charge carriers which exceed those known for nondegenerate semiconductors as will be stated later.

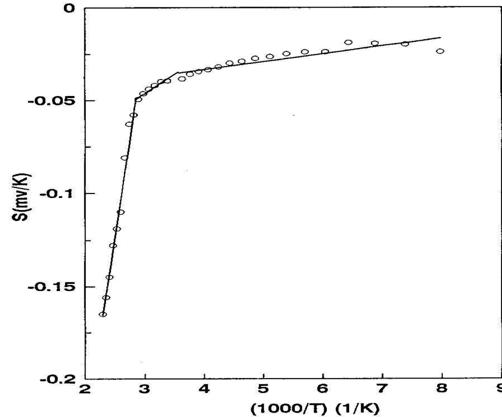

Fig.4 :Dependence of Seebek coefficient on temperature for the as-prepared.

The normal dc electrical resistivity ρ increases asymptotically with increasing T in the range 183 ≤ T ≤ 360 K leading to a metallic behavior conductivity. Such metallic behavior of conductivity is observed in pure isomolar composition $Ga_{0.45}In_{0.55}Sb$ with electron concentration from $6x10^{23}$ to $57x10^{23}$ m^{-3} in the interval 80 to 500 K [18].

However, the change of ρ with T becomes weaker with increasing the range of the latter. As shown in Fig.(5), the temperature dependence of ρ can be described by the following power equation

$$\rho = c\,T^{-n} \qquad (2)$$

Non of the calculated values for the power n of eqn.(2) which were equal to ~ 0.2, 0.1 and 0.0037 is coincided with the values 10 or unity characterizing the temperature dependence of metals respectively below and above the Deby temperature. Moreover, the absolute values obtained for ρ for this material are about two orders of magnitude higher with respect to those known for metals and metallic alloys.

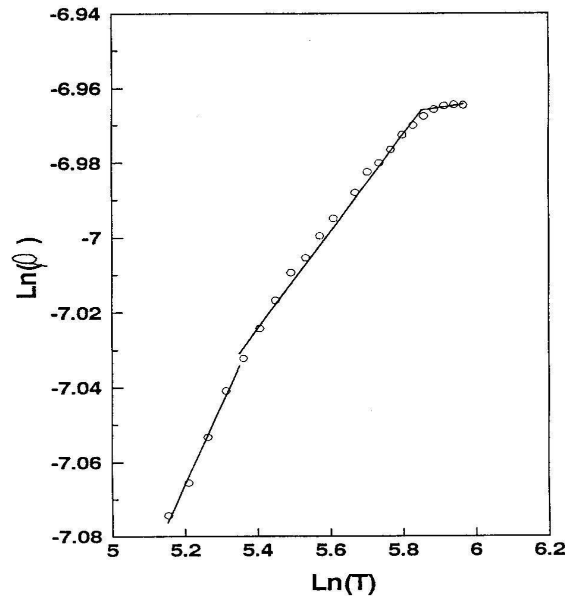

Fig.5 : Ln(ρ) vs Ln(T) for the as-prepared sample.

Nevertheless, the similarity between the behavior observed for the present composition and that of metals where for both ρ increases with increasing T, besides, the relatively high values obtained for the charge carriers concentration as will be emphasized latter can prove that this composition can be considered as a degenerate system (the case for the heavy doped semiconductors). Meanwhile, the decrease in the value of the power n with transition from one range to the successive higher range of T, may reveal that lefting the degeneracy becomes possible with elevating T, and that the contribution of some of the charge carriers to degeneracy of the system and to the different process of scattering either ceases or becomes weaker or less thermally activated at elevated temperatures. This is a matter approaches the composition to be looked like a weakly doped or non-degenerate. However, the semimetallic behavior which is observed can be attributed to excess Sb with the metallic behavior as Bi which is separated as an elemental phase as it has been confirmed by XRD analysis.

Furthermore, considering the Matthiessen's rule, the extrapolation to T = 0 K gave the value 3.09×10^{-4} Ω/cm for the residual resistivity which is relatively high compared with those known for metals and metallic alloys.

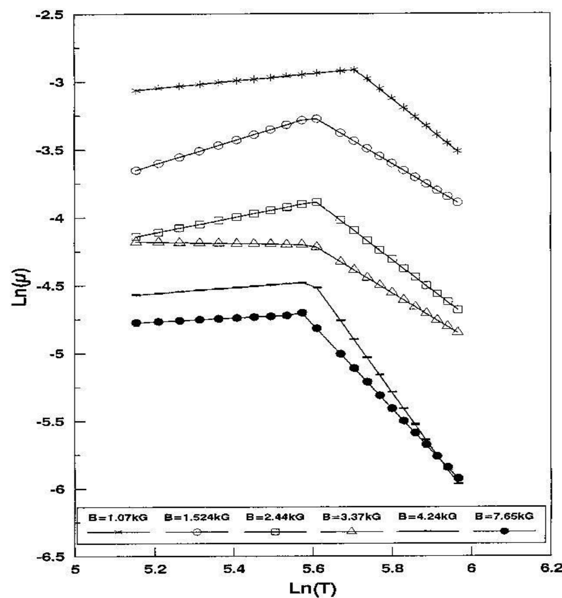

Fig.6 : Ln(μ) vs Ln(T) for as - prepared sample for different magnetic field.

As shown in Fig.(6), at fixed values of the magnetic field B in the range 1.07 to 7.65 kG, the temperature dependence of the carriers mobility can be described by a power equation as follows

$$\mu = C\,T^{\pm L} \qquad (3)$$

where C is a constant. Up to a certain temperature T_r, the power L possessed a plus sign while in the range above T_r , L possessed minus sign for all values considered for B, except at B = 3.37 kG, L possessed minus sign over the whole range considered for T. This indicates that μ can be thermally activated or inhibited with T, a matter depend on the range of the latter.

Table (1): Variation of the power L and the transition temperature T_r with the magnetic field intensity. The subscripts 1 and 2 denote the lowest and the successive higher range of T, respectively.

B (kG)	1.07	1.524	2.44	3.37	4.24	7.65
L_1	0.295	0.872	0.570	-0.060	0.220	0.180
L_2	-2.470	-1.910	-2.230	-3.220	-4.070	-2.930
T_r (K)	303	263	263	263	263	235

It is obvious from Table (1) that, at both ranges below and above T_r , L fluctuated unsequentially with changing the strength of the applied magnetic field. This may reveal the multi-phase structure of the prepared composition, which may reveal also in turn the possibility of contribution of different kinds of scattering and domination of a certain kind of scattering is a matter depends on the intensity of the applied magnetic field. The most interesting observation is that, except at B = 1.07 and 7.65 kG, T_r possessed a unified value 263 K which may have a special structure significance. For the sake of reminiscence, in case where scattering is controlled by the impurities the typical value for L is 3/2. However, the discrepancy from this value is large because the temperature of measurement is relatively exceeding that limit at which contribution of impurity scattering can be considered alone, and so, lattice scattering can not be neglected. On the other hand, the higher range of T would be controlled by the $T^{-3/2}$ law of the mobility which is not typically the case as shown in Table (1). The deviation from the $T^{-3/2}$ dependence which is observed in the present work

may be owed to the alloy multi-phase structure [19]. The lnμ vs 1/ T relations were kinked linear as shown in Fig.(7) suggesting the following formula to describe the temperature dependence of mobility

$$\mu = C \exp. (\pm E_{\mu} / kT) \quad (4)$$

where C is a constant has the significance of the value of the mobility at infinite temperature since the sign of the exponent is minus at the high range of T at all values considered for B. E_{μ} is the activation energy of mobility which is of two – fold significance, where at the ranges below and above T_r increasing T inhibits and promotes the process of scattering with the charge carriers (electrons), respectively.

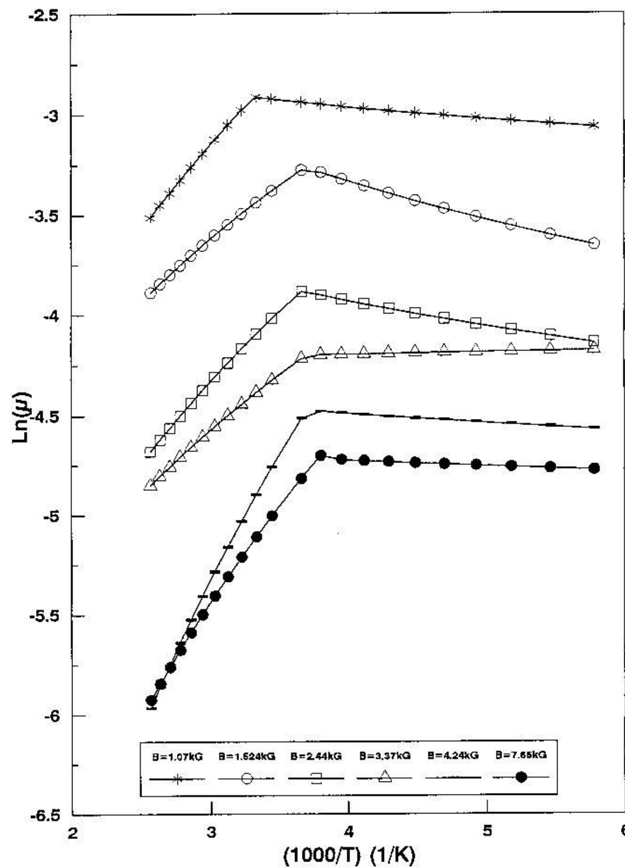

Fig.7 : Ln(μ) vs 1/T for as-prepared sample at different magnetic field.

Table (2): Values of E_μ with different magnetic field intensity.

B (KG)	1.07	1.524	2.44	3.37	4.24	7.65
$E_{\mu 1}$	-1.067	-0.10	-0.12	0.004	-0.045	-0.008
$E_{\mu 2}$	0.860	0.55	0.71	0.880	1.410	1.220

As it is seen in Table (2), neither $E_{\mu 1}$ nor $E_{\mu 2}$ changed sequentially with the magnetic field strength which is a matter can be explained on the same former bases. The minus sign for μ as marked on the coordinate confirms that the charge carriers are mainly electrons as demonstrated by the TEP measurements.

Typical examples for the dependence of μ on B at different fixed temperatures are those as plotted on Fig.(8). The monotonic decrease of μ with B at different fixed temperatures shown in Fig.(8) reflects a power dependence as follows[20];

$$\mu = C\, B^{-x} \tag{5}$$

Accordingly [21], the power x is equal to unity and the constant $C = E_y\, \sigma / J_x$, since E_y is the electric field along the y-direction, J_x is the current density along the x-direction and σ is the electrical conductivity.

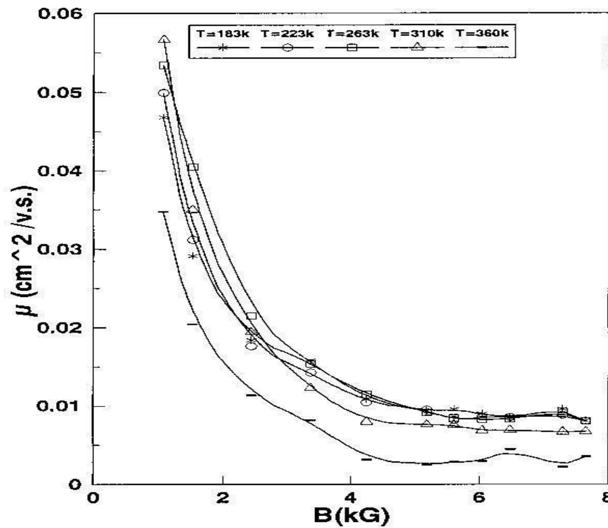

Fig.8 :The relation between the mobility and magnetic field for as-prepared sample.

Table (3): Variation of the power x with T. The subscript 1&2 denote the low and successive higher ranges of B.

T (K)	183	223	263	310	360
x_1	-1.170	-1.160	-1.026	-1.227	-1.460
x_2	-0.740	-0.900	--------	--------	0.340
B_r (KG)	2.440	2.440	--------	--------	5.180

In Table (3), the existence of two values for x indicates that the μ - B dependence possesses two ranges separated at a certain value of B denoted as B_r which possessed a unified value 2.44 kG at T = 183 and 223 K and 5.18 kG at T = 360 K, respectively. The power x_1 possessed value almost equal to unity at T equals 263 K. Otherwise, x_1 exceeds the value unity. On the other hand, at 360 K, x_2 possessed plus sign. Meanwhile, the dependence of μ on B becomes weaker in the range above B_r since $|x_2| < |x_1|$ and possessed values less than unity.

As for TEP and charge carriers mobility, the Hall coefficient R_H possessed minus sign (as denoted on the ordinate) at all the considered values of B and within the whole considered range of T which confirms the domination of contribution of electrons. Plots on Fig. (9) indicate that, the temperature dependence of R_H can be described by the following equation

$$R_H = C \exp. [E_{RH}/ KT] \qquad (6)$$

where C is a constant represents the value of R_H at infinite temperature (the pre – exponential). The plus and minus sign of the exponent has the significance that R_H increases and decreases respectively with increasing the range of T. The values of E_{RH} and its variation with B are as recorded in Table (4).

Table (4): Variation of E_{RH} with B. The subscript 1and 2 denote the lower and successive higher range of T, respectively.

B (KG)	1.07	2.44	4.24	5.60	7.65
E_{RH1} (ev)	-0.106	-0.103	-0.084	0.017	-0.071
E_{RH2} (ev)	0.084	0.870	1.268	1.392	0.785

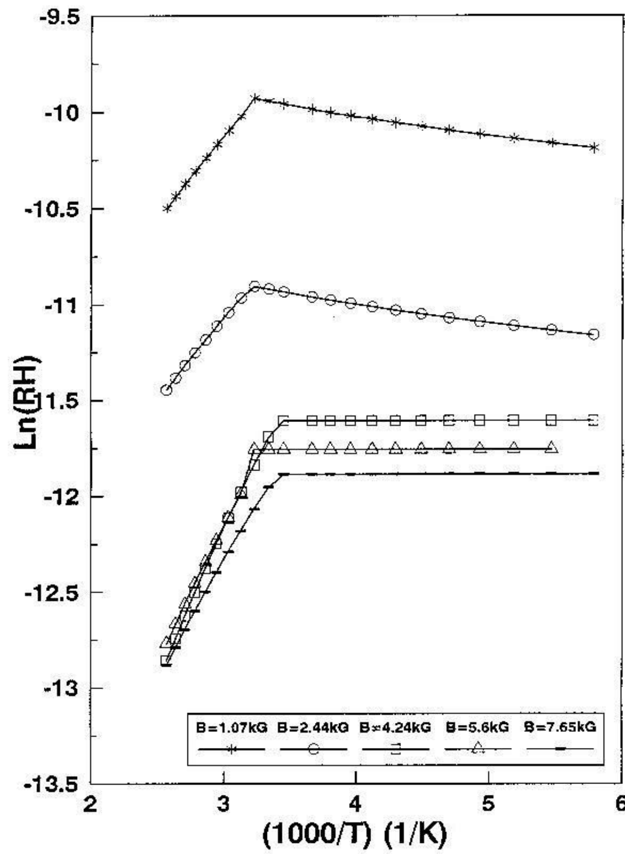

Fig.9 : Ln(RH) vs (1000/T) for as-prepared sample for different magnetic field.

It is obvious in Table (4) that, the sign of E_{RH1} is plus only at 5.6 kG where – R_H increased with increasing T. Otherwise, E_{RH1} possessed minus sign indicating a decrease in R_H with increasing T. In contrast E_{RH2} possessed plus sign at all values of B confirming an increase in – R_H with increasing T. Meanwhile, $|E_{RH2}|$ is about one order of magnitude higher with respect to $|E_{RH1}|$ confirming relatively strong thermal activation of – R_H as T exceeds T_r. In contrast to E_{RH1}, E_{RH2} is strongly dependent on B (within about one order of magnitude).

Accordingly [18, 22,23], the increasing in R_H with T which is observed on other $A^{III} B^{V}$ compounds too is explained by the influence of an additional conduction band characterized by the higher density of states and small mobility of charge carriers. This contradicts with results on Fig. (6 & 7), since |μ| increases with increasing T. However, it agrees well with results on Fig. (10), since the charge carriers concentration increases significantly with increasing T.

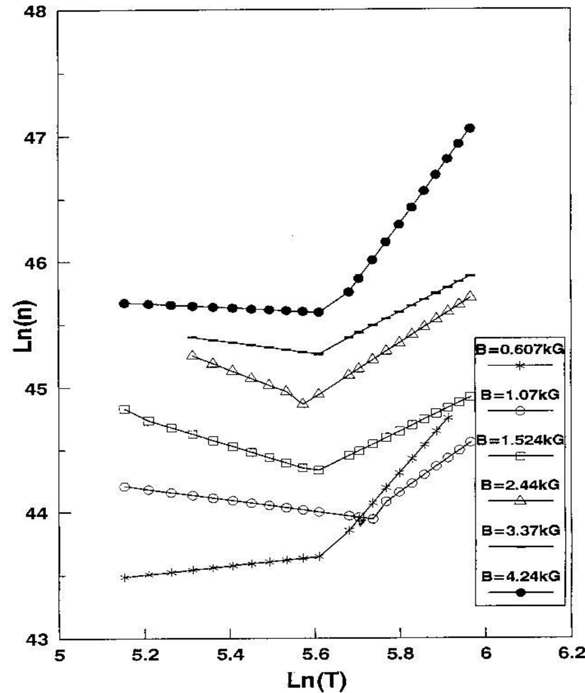

Fig.10 : Ln(n) vs Ln(T) for as-prepared sample.

The linear double logarithmic plot shown in Fig. (10) between the concentration of the charge carriers n and the temperature of measurement T suggested the following power relation

$$n = C\, T^{\pm m} \qquad (7)$$

where C is a constant represents the value of n at zero temperature. Values of the power m at the fields, which are denoted on the Fig. (10), are tabulated in Table (5).

Table (5): Variation of m with B at the lower and the successive higher ranges of T, m_1 and m_2, respectively.

B (KG)	0.607	1.070	1.524	2.440	3.370	4.240
m_1	0.600	-0.460	-0.670	-0.440	-0.200	-0.210
m_2	9.380	2.940	2.940	3.640	3.120	3.720

It is obvious from Table (5) that, except at B = 0.607 kG where n increased with increasing T over the whole considered range of temperature as shown in Fig. (10) the lower range of T seemed characterized by a decrease of the value of n with increasing T and so, the sign of m_1 was minus. In the successive higher range of T, n increased with increasing T and so, the power m_2 possessed plus sign. The temperature T_r which when exceeded the power m inverses its sign depends on B as shown in the same Table (5). However, neither m_1 nor m_2 changed sequentially with B.

As shown in Fig. (11), the concentration of the charge carriers increases with increasing B up to a certain value depends on T. Then it decreases with further increase in B. The turn down value of B decreases with increasing T and was equal to 5.18, 5.6 kG at T equals 360 and 340 K, respectively and 6.04 kG at both 310 and 240 K. Besides, such turn down field could not be attained at T = 183 K in the n–B isotherm.

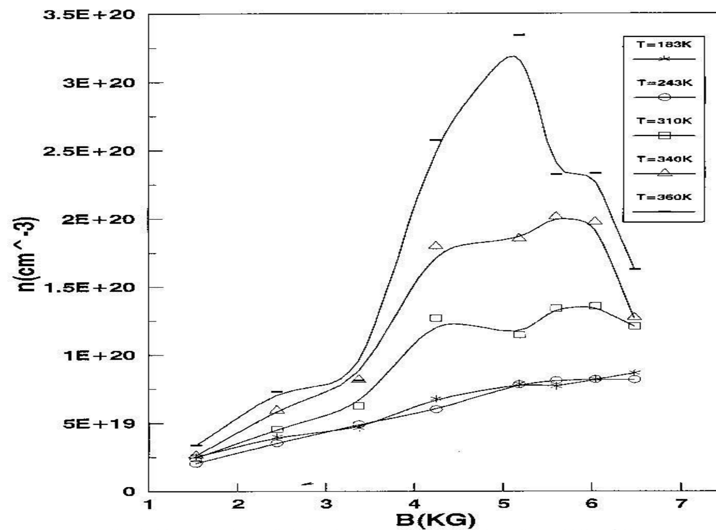

Fig.11 :The relation between carrier concentration and magnetic field for as-prepared sample.

Conclusions:

The irregular σ-E plot with the appearance of negative differential conductivity regions can reflect a complexity in the band structure and thence Gunn effect. The negative sign observed over the whole range of T proves that the material is n-type semiconductor as proved by the thermoelectric power. However, the valence exchange between the elements constituting the material results in formation of donor and acceptor subbands and the partial compensation of electrons by holes results in small values of the calculated thermoelectric power activation. The increase in resistivity with increasing T combined with the high values obtained for the charge carriers concentration may confirm that the prepared material behaves like n–type degenerate semiconductor. The thermoelectric power might be explained in terms of the two carriers model. The ρ-T characteristics showed a semimetallic–like behaviour, which can be attributed to the excess of Sb as an elemental phase. The variation of the mobility with temperature showed a unique transition temperature T_r = 263 K for all the applied magnetic fields except for B = 1.07 kG. On the other hand, the deviation from $T^{-3/2}$ dependence may be owed to the alloy multi-phase structure. The sign of the Hall coefficient R_H is always negative confirming the results of the thermoelectric power.

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