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High Temperature Electrical Characterization of Single Crystal of N-Type CDSE Optimally Annealed in a molten CD

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Abstract: In this paper high quality single crystal of n-type CDSE is grown from the vapor phase by the piper polich method. X-ray laue photographs from different directions are taken. DC galvanomagnetic properties are measured and analyzed in the temperature range 75 to 450 K in an undoped single crystal of n-type CDSE, which was optimally annealed in high purity molten CD at 850 ^{0}C for a period of ~ 400 h. α- the polaron coupling constant is estimated from the measured Hall mobility (μ^H) data at low and intermediate temperature by identifying a temperature region, wherein only the polar optical phonon scattering operates in the conduction band. The deformation potential E_b in the conduction band of CDSE is obtained from the measured μ_H data in the temperature region wherein the deformation potential also operates effectively. It is shown that a change in the microscopic mobility through photo excitation indicate a change in scattering.

Keywords- CDSE, Methodology

I. INTRODUCTION

Sample Preparation

Single crystals of n-type CDSE used in the present investigation were grown from the vapour phase by the Piper-Polich method [21].The crystalline nature of the boule was verified with the help of X-ray Laue photographs which were taken along different directions. The boule was found to be oriented along the c-axis as determined from the photographs exhibiting well defined sixfold symmetry patterns. From the boule ,sample of rectangular bar geometry of size about 7.0 X 2.0 X 1.0 mm^3 were cut perpendicular to the c-axis by using a microslice precision annular saw. The samples containing the largest- size single-crystal grains were selected for further experiment. These undoped as-grown samples ,being highly resistive ,were annealed in high purity molten CD at 850^oC for different annealing durations ranging from about 100 to 500 h in order to remove the compensating impurities and the native charge defects [22,23] .To anneal a sample, it was kept in an evacuated quartz ampoule along with the annealing metal whose weight was about ten times the sample weight .The mobility of the charge carries in the sample annealed for≈ 400 h was found to be maximum , and it was found to deteriorate in the under-annealed and the over-annealed samples.

Before providing ohmic contacts to the samples, they were roughened with the fine grained abrasive powder (3µm silicon carbide).After chemomechanical polishing with a solution of alcohol, and distilled water, the samples were etched [24] in a solution obtained by dissolving $KMnO_4$ in concentrated H_2SO_4, washed thoroughly in double-distilled and de ionized water several times and finally dried.

This etching left no surface deposits. Low-resistivity ohmic contacts on etched samples were made by diffusing high- purity indium at 250^0C in an inert atmosphere of high-purity argon. The voltage contacts measured about 0.6mm in size, whereas the current contacts were spread over the entire cross-sections of the samples. The ohmic nature of the contacts was verified for operating voltages by observing the linearity of the current –voltage characteristics throughout the temperature range of interest.

II. D.C. GALVANOMAGNETIC MEASUREMENTS

The low-field dc conductivity σ and the Hall coefficient R_H were measured in the temperature range 77 to 440 K by employing the five-probe technique [25] (i.e., the six-probe technique wherein one voltage probe is taken common). The experimental samples were affixed to a copper strip which had electrical insulation underneath, and then this assembly was placed in a double-walled metal vacuum cryostat [26].The samples were first cooled down to 77 K by thermal conduction through the copper strip which was kept immersed in liquid nitrogen. After taking measurements at 77 K, the temperature was raised by means of a mini-heater wound around the extended part of the copper strip. A precliberated copper-constantan thermocouple in direct contact with the sample surface was used to measure the ambient temperature T. During each measurement, we kept temperature fluctuations below 0.5 K with the help of a temperature controller. The Hall coefficient R_H was found to be independent of magnetic field up to 0.6 T which was the highest magnetic field available for us.Hall voltages were measured to an accuracy of ≈1.0% a Keithley dc nanovoltmetre and were foud to be proportional to the applied magnetic field and sample current. During each Hall measurement, the directions of the sample current and the magnetic field were reversed, and the observed Hall voltages were averaged in order to avoid thermoelectric and thermomagnetic effects. Taking in to account the errors involved in the measurement of the Hall voltage, magnetic field, and sample current, the error in the measurement of RH was found to be ≈2.0%. The dc conductivity σ was measured to an accuracy of about 1.5% and so the overall error in the estimation of Hall mobility µH was found to be ≈3.0%.

III. EXPERIMENTAL RESULT AND DISCUSSION

The observed temperature variation of Hall coefficient R_H (log R_H versus 1/T) for the sample at a typical magnetic field of 0.4 T and in the temperature range 75 to 450 K is shown in Fig. 1. Then it is observed that R_H decreases with increase of temperature in the entire temperature range-initially at a faster rate (freeze –

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out region) and thereafter at a slower rate, finally tending to be almost temperature-

independent (exhaustion region). Making use of the following charge-neutrality equation [27] for a non degenerate and compensative semiconductor,

$$\frac{n_c(n_c+N_a)}{N_{d-}N_a-n_c}=gN_c\exp(-\frac{E_D}{k_BT}) \qquad (1)$$

we have fitted this equation with the experimental data of R_H of Fig. 1 at various temperatures to estimate the donor thermal ionization energy E_D and the concentrations N_a, N_d of ionized acceptors and donors, respectively. In (1), g=1/2 is the donor level degeneracy factor, k_B is the Boltzmann constant, the difference (N_d-N_a) equals the free-electron concentration n_C at exhaustion, and N_C is effective density of states in the conduction band,

$$N_c=\frac{2(2\pi m^*k_BT)^{3/2}}{h^3}$$

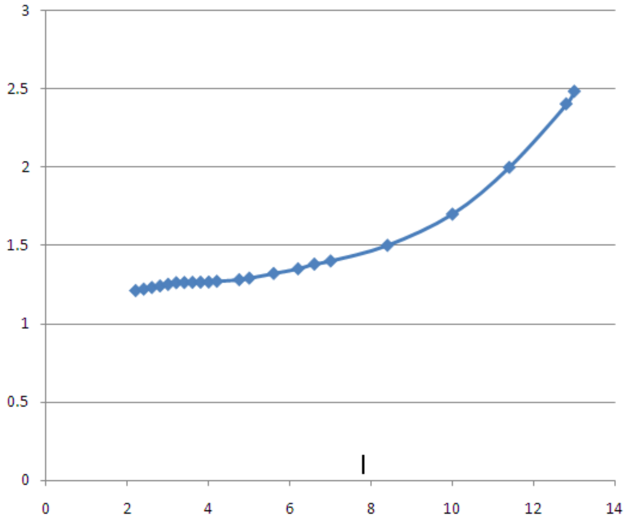

Fig.1. Temperature (K) dependence of the observed Hall coefficient R_H in a single crystal of n-type CDSE, optimally annealed in molten CD

Where m* (=0.13 m_O[28], m_O being the fre- electron mass) is the effective mass of free electrons and h the Planks constant. The values of E_D, N_a and N_d have been found to be 17.2 meV, 1.64×10^{15} cm^{-3} and $6.72\times10^{15}cm^{-3}$, respectively. The low compensation ratio 0.24 (i.e,N_a/N_d) for the sample shows that its annealing has been done more or less optimally . For an extrinsic semiconductor , the value of E_D is an indication of the purity of the sample .For sample having a total concentration of impurities well below the degeneracy level, the value of E_D approaches that given by the hydrogenic model [29], i.e,

$$E_D\rightarrow E_H=13.6\frac{m^*}{m_0}k_s^{-2} \qquad (3)$$

In case of CDSE, the hydrogenic value of E_D is 16.0 meV , taking K_s =10.2 [30]. This confirms the good quality of the sample.

To estimate the polaron coupling constant α from the measured data of Hall mobility μ^H (Fig. 2), we have to ascertain if there is any temperature region where only the polar optical phonon scattering operates in the conduction band and the other scattering mechanisms, like ionized impurity scattering and acoustic phonon scattering, are ineffective. If there is such a region, the experimental values of Hall mobility μ^H can be fitted with the theoretically computed values of Hall mobility μ^H_{OPS} for the polar optical phonon scattering by using a unique value of α. Such a fitting is possible only when scattering mechanism other than optical phonon scattering are effective in this temperature region. For the Hall mobility μ^H_{OPS}, the following Howarth and Sondheimer formula [31] for nondegenerate semiconductors has been employed:

$$\mu^H_{OPS}=\frac{8e}{6\sqrt{\pi}\alpha w_1m^*}Z^{-1/2}\Psi(Z)(\exp Z-1)r_H \qquad (4)$$

Where Z, called the reduced reciprocal temperature, is the ratio of Debye temperature θ_D ($\hbar w_1/k_B$=313 K [32]) and temperature T, Ψ (Z) is a function which has been evaluated by Howarth and Sondhimer [31] and later generalized by Ehrenreich [33] to include the carrier screening effects, r_H is the Hall scattering factor (i.e. Hall mobility to conductivity mobility ratio), and w_1 is the angular frequency of longitudinal optical phonons. As our material is polar, the effective mass m* of free electrons for the purpose of estimation of μ^H_{OPS} has been replaced by the polaron effective mass m^*_p which is higher than m* in accordance with [34]

$$m^*\rightarrow m^*_p=m^*(1+\frac{\alpha}{6}) \qquad (5)$$

The Hall scattering factor r_H, being characteristic of the scattering mechanism present, is given by [35]

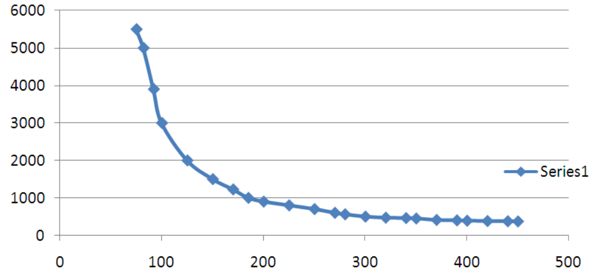

Fig.2. Temperature (K) variation of observed Hall mobilitiy μ^H (Cm^2 /v-Sec.) for α=0.51 in a single crystal of CDSE, optimally annealed in molten CD

$$r_H=\frac{\langle K'^2\tau^2\rangle\langle K'^2\rangle}{[K'^2\tau]^2}=\frac{\Gamma(5/2+2s)\Gamma(5/2)}{\Gamma(5/2+s)^2} \qquad (6)$$

Where K' is the wave number of free electrons, τ the free-electron relaxation time limited by the scattering mechanism present, angular brackets stand for the Maxwell averaging and s, called the scattering index for the scattering mechanism, is the exponent of free-electron energy E in the equation

$$\tau \sim E^s. \qquad (7)$$

In the temperature region where only the polar optical phonon scattering operates effectively, the scattering index s and the Hall scattering factor r_H take the values 0 and 1, respectively as according to the following equation for the free-electron relaxation time τ_{OPS} limited by the polar optical phonon scattering [28, 31],

$$\tau = \tau_{OPS} = \frac{\hbar^{3/2} 4\pi\varepsilon_0 (\exp Z - 1)}{e^2 (2m^* w_1)^{1/2} (1/n_0^2 - 1/K_s)} \qquad (8)$$

Where n_0, being the refractive index of the material at frequencies, equals the squar root of the dielectric constant $K\infty$ at optical frequencies for which the absorption is negligible .Equation (8) assumes that $T \ll \theta_D$ where $E \ll \hbar w_1$.

It has been found that a good fitting between μ^H and μ^H_{OPS} exist in the temperature region 200 K to 260 K for a unique value of α=0.51.The theoretically calculated values of μ^H_{OPS} using (4) with α=0.51 have shown in Fig. 2.

The value of polaron coupling constant α as estimated from [28]

$$\alpha = \frac{e^2}{\hbar}\left(\frac{m^*}{2\hbar w_1}\right)\left(\frac{1}{K_\infty} - \frac{1}{K_s}\right) \qquad (9)$$

Involves an uncertainty due to uncertainties in the values of static and the optical dielectric constants K_s, K_∞ and their temperature variations. The method employed in this work for the estimation of α is free from this uncertainty. The temperature variation of the static dielectric constant K_s is expressed by [36]

$$K_{S(T)} = K_{s(0)}(1 + \lambda T) \qquad (10)$$

Where $K_{s(0)}$ =7.34 is the static dielectric constant extrapolated linearly to 0 K and $\alpha = 1.3\times10^{-3}$ [37] is constant for the material. The temperature variation of the optical dielectric constant K_∞ is shown from the fact given by Rode [38] that the ratio K_s/K_∞ is temperature independent. The temperature variation of K_∞ can be obtained by using (10) and the ratio K_s/K_∞ at room temperature [39] (K_s/K_∞=10.2/6.1).

At higher temperatures, the acoustic phonon scattering via the piezoelectric field is unimportant, and that via the deformation potential contributes significantly. The procedure adopted here for the estimation of α could have been employed at higher temperatures also, but the contribution of deformation potential scattering in the high-temperature region may be sufficient to change the effective Hall mobility of the charge carriers and, hence, to give a lower estimation of the polaron coupling constant α. Taking the already estimated value α (i.e., 0.51 which may be slightly underestimated due to the temperature variation of the two dielectric constants K_s and K_∞),we have computed the Hall mobility values μ^H_{OPS} for the polar optical phonon scattering in the high-temperature region also. The Hall mobility contributions μ^H_{DPS} due to the deformation potential scattering have been obtained by using Mattheisen's rule,

$$\frac{1}{\mu_H} = \frac{1}{\mu^H_{OPS}} + \frac{1}{\mu^H_{DPS}} \qquad (11)$$

and the deformation potential E_b in the conduction band has been calculated by employing the following formula due to Bardeen and Shockley [40] for this type of scattering for the long-wavelength acoustic modes:

$$\mu^H_{DPS} = \frac{2(2\pi)^{1/2} e\hbar^4 C_1 r_H}{3E_b^2 (m^*)^{5/2} (k_B T)^{3/2}} \qquad (12)$$

In this equation the elastic stiffness constant $C_1 = 8.39\times10^4$ MPa [30] equals the product of the density d of the material and the squar of the average longitudinal acoustic wave velocity v_1. The Hall scattering factor r_H for the deformation potential scattering is 1.18 due to the following energy dependence of the free-electron relaxation time τ_{DPS} limited by this scattering:

$$\tau_{DPS} = \frac{\pi\hbar^4 C_1}{\sqrt{2}E_b^2}(m^*)^{-3/2}(k_B T)^{-3/2}\left(\frac{E}{k_B T}\right)^{-1/2} \qquad (13)$$

Where the scattering index s is -1/2. The calculated value of E_b comes out to be about 10.9 eV which may be compared with values 3.7 and 11.5 eV as quoted by Finlayson at al. [18] and Rode [19], respectively.

Other approaches for the computation of the deformation potential E_b include the analyses of (i) the pressure dependence of electrical transport properties [41], (ii) the absorption of infrared signals [42], (iii) the acoustic absorption in the presence of magnetic field [43], and (iv) the phonon-drag thermoelectreic power [44]. But the accuracies of these approaches are not known, and the values of Eb given by such analyses do not agree with each other.

IV. CONCLUSION

It is observed that because of the proximity of the Hall mobilities μ^H and μ^H_{OPS}, the estimated values of E_b is subject to a large error. However, we have estimated E_b from a number of observations taken in a temperature interval of 50 K. From the fluctuations in the estimated values of E_b, an error of about 12% has been found.

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