

Study of Electrical Transport Properties in Thermally Evaporated Cu₂S Thin Films

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Abstract

Cu₂S was synthesized by vacuum evaporation under a pressure of 10⁻⁶ torr at an evaporation rate of 3 Å/sec. The electrical properties of Cu₂S thin film were studied using current-voltage characteristics. The conductivity was found to exhibit two distinct mechanisms within the applied field i.e., low voltage region and high voltage region. The most probable conduction mechanism prevailing in Cu₂S thin film is found to be Poole-Frenkel effect. Activation energy has been calculated for different thicknesses and is found that activation energy decreases with increase in film thickness. Dielectric constant of the film was measured using impedance analyzer. Plot of dielectric constant vs. frequency shows normal dielectric behavior of the film. Photocurrent of Cu₂S film increases with increase in film thickness.

Key Words: I-V Characteristics, C-V Characteristics, Dielectric Effect, Photocurrent

1. Introduction

Copper sulphide being an II-VI compound is an important material from the point of basic research, because it is known to exist in several crystallographic and stoichiometric forms. The principal advantage of II-VI compounds for terrestrial solar photovoltaic are low cost and ease of deposition of good quality films from these materials by a variety of growth methods. Due to their high photosensitivity, high absorption coefficient and direct band gap these II-VI semiconductors are of great interest in electronic and opto-electronic devices. Copper sulphide film exhibit several crystalline forms and variety of orientation with respect to substrate depending on deposition parameters.

Copper sulphide is found to exist in two forms at room temperature as ‘copper-rich’ and ‘copper-poor’.

Copper rich phases exist as chalcocite, djurlite, digenite and anilite [1] and copper poor phase are existing as co-vellite. It is well established that Cu₂S is the optimum proportion for obtaining high efficiency solar cells with high stability against oxidation and degradation. Due to the presence of copper vacancies in the film, Cu₂S exhibit P-type conductivity and act as electron acceptors that give rise to free holes. Cu_xS (x = 1 to 2) thin films have numerous technological applications such as sensors [2], as thermo electric and photoelectric converters [3], as field emission and photoluminescence [4], as solar controller and solar radiation absorber [5] as thermo-electric cooling material [6] and in micro-electromechanical-systems (MEMS) devices. Much attention has been focused on thin film CdS/Cu₂S hetero-junction due to their great promise as low cost solar power converter owing to their high efficiency up to 9.1% with improved stability [7]. Cu₂S thin films have been prepared by various techniques such as spray pyrolysis [8], modified

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chemical method [9], photochemical method [10], vacuum evaporation method [11], chemical bath deposition (CBD) and atomic layer deposition (ALD) method. Investigation on current-voltage characteristics seems to provide more adequate method for distinguishing between different mechanisms of charge transport in the material. The purpose of the present work was therefore to report the current-voltage characteristics and dielectric properties of Cu_2S thin film as a function of film thickness prepared by vacuum evaporation method.

2. Experimental

2.1 Film Formation and Characterization

Cu_2S thin films of different thicknesses were deposited onto properly cleaned glass substrate with the help of hind high vacuum coating unit at vacuum, 10^{-6} torr. Initially glass substrates were thoroughly washed with soap solution and then with distilled water. Next the glass substrates were kept in ultrasonic agitator for 45 min to remove the impurities present in the substrate. Pure (99.99%) copper sulphide powder was used as the source material and high purity aluminum electrode was vacuum evaporated onto cleaned glass substrate on which the films were deposited. Molybdenum boats were used as source heater and the glass substrate was mounted on the holder with heating arrangement. Thicknesses of the film were measured using quartz crystal monitor whereas temperature was measured with the help of fine wire chromel and alumel thermocouple. Substrate to source distance was optimized to be at 18.1 cm inside the vacuum chamber. Rotary drive was employed to maintain uniformity in film thickness. Constant rate of evaporation $3 \text{ \AA/sec}$ were maintained throughout the sample preparation.

Current-voltage characteristics of the Cu_2S thin films were studied by employing digital micro ammeter in series with the capacitor and the voltage source. All studies were performed under vacuum of about 680 torr. Current-voltage behavior was studied in the temperature range 300–400 K and the temperature during measurements was maintained using variable voltage transformer.

A copper constantan thermocouple was used for the temperature measurements. A potential of 20 volts was applied using DC regulated power supply in series with the digital voltmeter. Capacitance-voltage measurements were made at room temperature using LCR meter (LCR 819, G. W. Ltd., Taiwan). Capacitance dependence of bias was studied by applying reverse bias (0–2 V) in the frequency range 30 KHz to 100 KHz, where the internal bias of the LCR meter was switched OFF and the external bias was switched ON. The most commonly used technique for measuring resistivity is the four point probe method. The arrangement consists of PID controlled oven (model-PID-200 scientific equipment and services, Roorkee, India) in combination with low current source (model-LCS-01) and digital micro voltmeter (model-DMV-001). Photo current was measured by taking silver paste as a contact electrode at 1 cm separation, which was applied on the film surface. A halogen lamp was used for white light and the intensity of light was measured in mW/cm^2 by placing a surya-amp at the position of the sample. The samples are kept in measurement chamber and photocurrent was measured using micro ammeter.

3. Results and Discussion

3.1 I-V Measurements

Different types of mechanism namely tunneling, impurity conduction, space charge limited conduction, Schottky emission and Poole-Frenkel conduction take place in semiconductor films. Information about the conduction mechanism prevailing in the Cu_2S thin film is obtained from the current-voltage characteristics of Cu_2S thin films. Figure 1 shows the current-voltage characteristics of Cu_2S thin films of $1000 \text{ \AA}$ thickness at different temperatures. It is observed from the entire graph that current voltage curve has two distinct regions, low voltage region and high voltage region. At low voltage region ($I \propto V$), indicating that the current is controlled by thermally generating carriers and leads to ohmic behavior [12,13]. Ohmic behavior exists as long as free carrier density continues to be in thermal equilibrium.

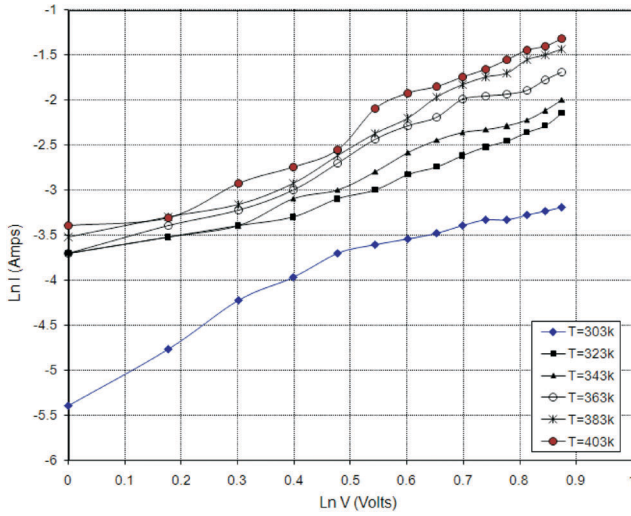


Figure 1. Variation of current with voltage measured at different temperature for Cu₂S film of 1000 Å thickness.

Conduction in the high voltage region does not seem to be ohmic, where the slope value is greater than 1, and this occurs when the carrier density becomes greater than the thermal equilibrium [14] in which square law region ($I \propto V^n$) is obtained. Hence it is inferred that the conduction mechanism in these films may be either Schottky or Poole-Frenkel. In the schottky mechanism electrons are transported by thermionic emission across metal-semiconductor interface i.e., when the potential barrier is sufficiently thick the current flowing through the semiconductor is limited, whereas in the poole-frenkel effect electrons are thermally emitted from traps to conduction band. As a result the columbic potential barrier is lowered by external field, thereby increasing the probability of electron that results in increase in flow of current through the semiconductor.

The conduction mechanisms in these films are mainly governed by the grain boundary defect states and the defect states which effectively act as trapping or recombination centers play an important role in the determination of conduction mechanism in these films. Current-voltage relation according to Poole-Frenkel or Schottky has the form [15].

$$I = I_0 \frac{\exp(e\beta E^{1/2})}{K_B T} \quad (1)$$

where E is the electric field, β is the field lowering co-efficient, I is the applied current, K_B is the Boltzmann constant. The constant β is given by

$$\beta = \sqrt{\frac{e^2}{\alpha \pi \epsilon_0 \epsilon_r}} \quad (2)$$

where e is the charge of the electron, ϵ_r is the relative dielectric constant, ϵ_0 is the permittivity of free space, α is constant equal to 1 or 4 for Poole-Frenkel or Schottky effect respectively. In order to investigate the conduction mechanism prevailing in Cu₂S thin film, the samples are subjected to high electric field between $\ln I$ and $E^{1/2}$ and the values of field lowering coefficient (β_{exp}) were calculated using the slope value in equation (1). Figure 2 shows the variation of $\ln$ current with square root of applied field for Cu₂S thin film of 1000 Å thickness. Table 1 shows the calculated Poole-Frenkel co-efficient value of 1000 Å thickness and the values found to be in the range $1.07 \times 10^{-5} \text{ (mV)}^{1/2}$ to $1.99 \times 10^{-5} \text{ (mV)}^{1/2}$ for the temperature (303–403 K). The calculated values (β_{exp}) are compared with theoretical value (β_{theory}) and evidently it is found that the experimental value of ' β ' lie very near to the theoretical value of Poole-Frenkel co-efficient (β_{pf}) that was calculated using equation (2). Thus the conduction mechanism in Cu₂S thin film is most probably be the Poole-Frenkel

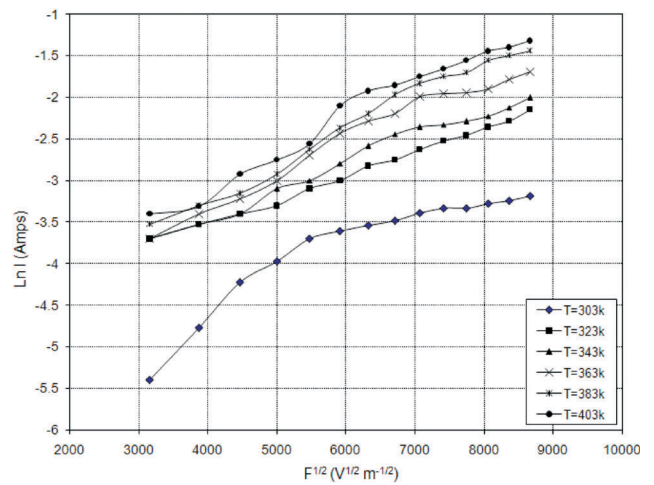


Figure 2. Variation of current vs. square root of applied field measured at different temperature for Cu₂S film of 1000 Å thickness.

Table 1. Experimental and theoretical values of β for Cu_2S film

T (K)	Experimental field lowering coefficient $\beta \times 10^{-5} (\text{mV})^{1/2}$	Theoretical field lowering coefficient $\beta \times 10^{-5} (\text{mV})^{1/2}$		ϵ_r from capacitance measure	ϵ_r from β_{pf} measure
		Poole Frenkel (a = 1)	Richardson Schottky (a = 4)		
303	1.07	1.51	0.75	40	50
323	1.78				
343	1.35				
363	1.79				
403	1.99				

effect based on the results obtained. It is observed that the curve exhibit linear current field characteristics at higher temperatures and within the ambient temperature range (323–343 K) the Poole-Frenkel co-efficient decreases as the temperature increases from (30 to 100 °C).

A condition for impurity conduction is the presence of donors, acceptors, structural traps and there are so many of these that dominate the conduction processes. When the traps are in abundance and a trap having a columbic type barrier would experience Poole-Frenkel effect at higher fields and acts as insulator. Further to confirm Poole-Frenkel mechanism in Cu_2S thin film the dielectric constant ϵ_r that has been determined using equation (2) and is listed in Table 1 along with the high frequency values of ϵ_r obtained from equation (9). It is clear from Table 1 that the values of ϵ_r obtained from capacitance measurement are in close agreement with the

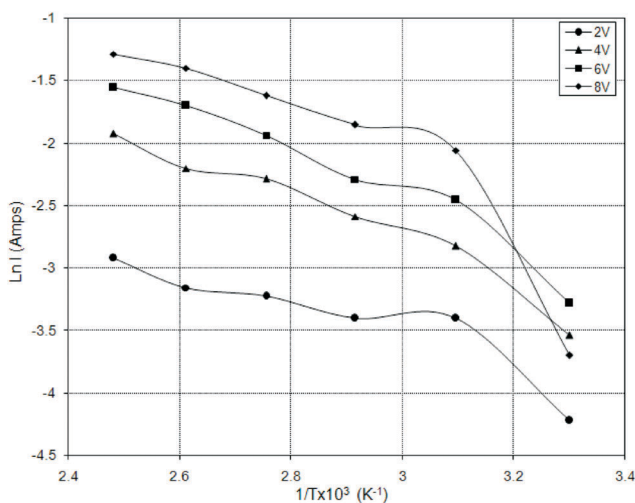
values obtained from β_{pf} in Cu_2S thin film.

Trap depth (E_t) for different voltages and film thickness of Cu_2S thin film are calculated from the plot of log current versus inverse of absolute temperature (Figure 3) and using the slope value in equation (3) the calculated values are found to be in the range (0.2 to 0.09) eV for Cu_2S thin film.

$$I = I_0 \frac{\exp(-E_t)}{K_B T} \quad (3)$$

where K_B is the Boltzmann constant, T is the absolute temperature. From Table 2 it is evident that trap depth decreases with increase in applied voltage, indicating that the trap depth has been lowered in the presence of applied field [16]. The reason is that conduction band is lowered and hence the probability of electron [17] escaping from the trap increases and thereby reduces trap depth. Moreover, trap depth decreases with increase in film thickness which is due to decrease in inter-grain boundary defect states and thereby reduces trap depth [18].

Figure 4 shows the variation of trap depth with applied voltage for Cu_2S thin film of different thickness

**Figure 3.** Variation of log current with inverse of absolute temperature for Cu_2S film.**Table 2.** Variation of trap depth with applied voltage

Bias applied (volts)	Trap depth E_t (eV) for different thicknesses	
	1000 Å	7000 Å
2	0.197	0.148
4	0.118	0.112
6	0.112	0.098
ΔE_0	0.30	0.15

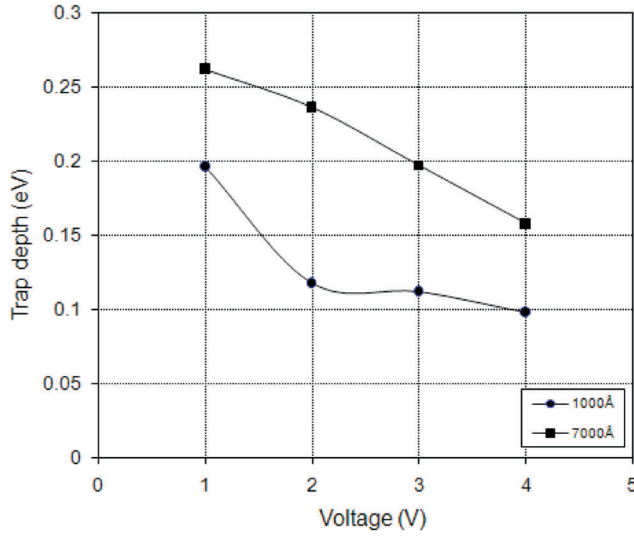


Figure 4. Variation of trap depth with voltage for different thickness of Cu₂S film.

and the intercept at $V = 0$ yields trap depth at zero field. The calculated value shows that at higher bias region the values almost remain constant, i.e., under increased bias the distance of the trap around the Fermi level to the top of valence band remain nearly constant.

3.2 C-V Measurements

Figure 5 shows the frequency dispersion of capacitance of Cu₂S films for two different voltages. From the plot it is observed that low frequency has high capacitance value and this may be attributed to the significant polarization of charge carriers that give rise to ionic con-

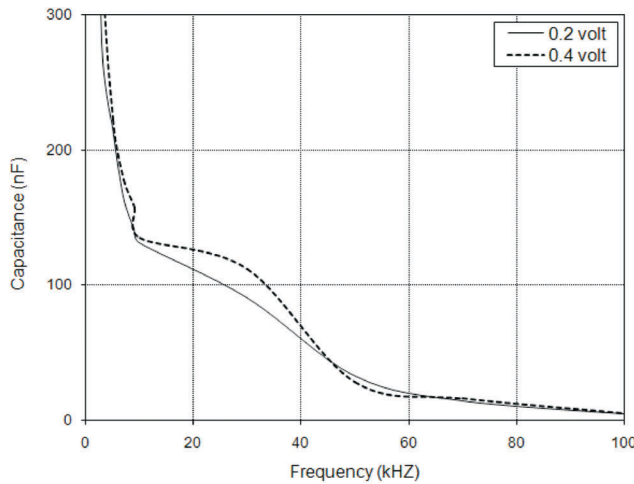


Figure 5. Frequency dispersion of capacitance in dark for Cu₂S film of 3000 Å thickness measured at different voltages.

duction. The dipoles cannot orient themselves at higher frequencies and hence the capacitance decreases [19]. Moreover at higher frequencies the frequency dispersion of capacitance was almost constant.

The number of states can be calculated from the measured capacitance at low and high frequencies, where the capacitance at lower frequency is associated with depletion layer and interface states, whereas capacitance at higher frequency is associated with depletion layer only. The total number of interface states can be calculated using the relation.

$$N_{IS} = \frac{(C_{LF} - C_{HF})}{q} \quad (4)$$

where N_{IS} is the total number of interfaces states, C_{LF} and C_{HF} are the capacitance at lower and higher frequencies respectively and q is the charge of an electron. Thus the total number of interface states (N_{IS}) calculated at low (70 kHz) and high frequencies (100 kHz) using equation (4) was found to be 1.8×10^{17} for 1000 Å thickness.

The ionized donor density (N_D) was calculated from the slope of $1/C^2$ vs. bias voltage curve (Figure 6) known as (Mott-Schottky plot) [20,21] at V_B close to zero and using the slope value in equation (5).

$$\frac{1}{C^2} = \frac{2(V_D + V)}{q\epsilon_0\epsilon_r A^2 N_D} \quad (5)$$

where A is the area of the electrode, ϵ_0 the permittivity

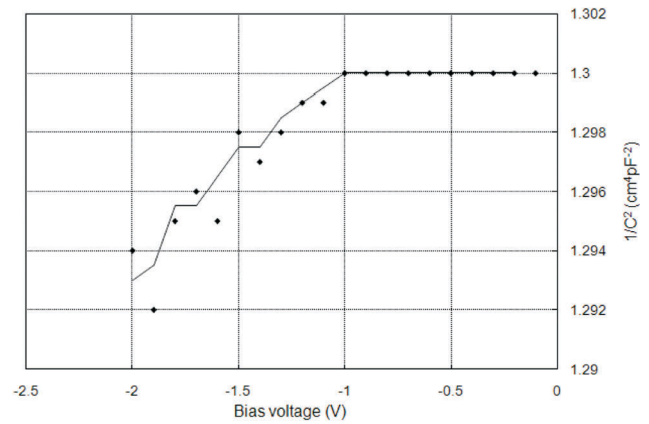


Figure 6. Variation of capacitance vs. voltage for Cu₂S film of 1000 Å thickness measured at constant frequency.

of free space, ϵ the dielectric constant of the material. Thus donor density (N_d) calculated from the above equation is found to be $5.57 \times 10^{16}/\text{cm}^3$. A negative voltage shifts in the frequency indicates the presence of positive charge carriers in the film [22]. The extrapolated value of voltage at $1/C^2 = 0$ gives the value of flat band potential as 2.4 V for 1000 Å thickness [17].

Barrier height (ϕ_B) of the material can be calculated using the relation

$$\phi_B = V_i + V_n + \left(\frac{kT}{q} \right) - \Delta\phi \quad (6)$$

where V_i is the voltage intercept, and V_n is the depth of the Fermi level below conduction band. Barrier height thus calculated is seen to be 0.300 eV for 1000 Å thickness and is in good agreement with earlier reports of Cu_2S film [2]. It is also observed that barrier height decreases with increase in film thickness and the values found to be 0.100 eV for 7000 Å thickness. Thus it is concluded that higher thickness Cu_2S film have low barrier height and vice-versa.

3.3 Resistivity Measurements

According to the modern quantum electronic theory, electrical conduction in metals is due to electrons, while electrical resistivity results from the scattering of electrons by the lattice, therefore resistivity is the measure of metal lattice displacement from perfect regularity [23]. Figure 7 represents the variation of resistivity with inverse of absolute temperature for various currents (30–90 μA) of 7000 Å thickness. It is observed that resistivity initially decreases with temperature and attains a minimum value called as transition temperature and further increases with temperature. The transition temperature is estimated within (383–443 K) for 7000 Å thickness. These observations show that the material behaves as semi conducting nature at lower temperature and metallic above transition temperature [9,24] which is in good agreement with the earlier reports reported for Cu_2S thin film.

Figure 8 shows the variation of resistivity with tem-

perature of Cu_2S sample for different thickness at constant current ($I = 50 \mu\text{A}$). The resistivity of the film is seen to increase with increase in film thickness and this increase in resistivity is due to surface of the film, which is covered with circular islands with lot of empty spaces between them producing number of grain boundaries and hence results in increase in resistivity [25]. The grain size of the film is determined by using the Scherrer formula [2]

$$d = \frac{0.9\lambda}{\beta \cos \theta} \quad (7)$$

where λ is the wavelength used, β is the angular line

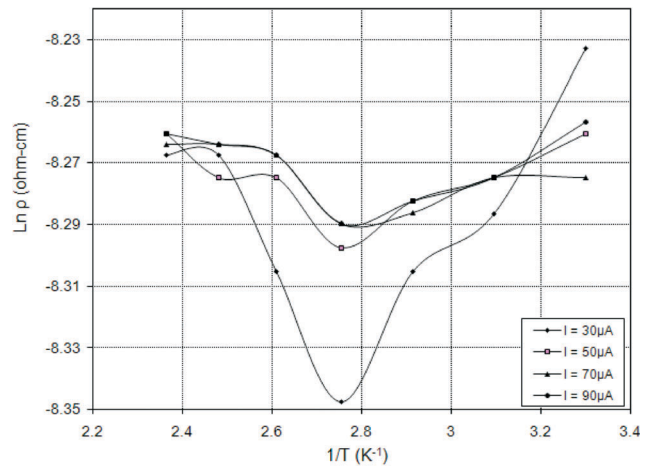


Figure 7. Variation of resistivity vs. inverse of temperature for Cu_2S film of 7000 Å thickness.

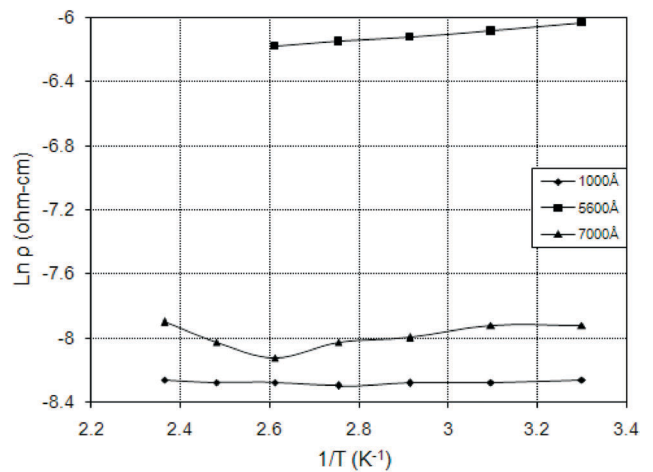


Figure 8. Variation of resistivity vs. inverse of temperature for Cu_2S of different thickness.

width at half maximum, θ is the Bragg's angle and the values are reported in Table 3. From Table 3 it is evident that the grain size decreases with increase in film thickness, which is due to the presence of new grains over older ones. Again it is seen from the graph that the resistivity decreases for the film coated at higher thickness, which is due to increase in grain size of the film at higher thickness and have opposite effect [26] (Table 3). The reason for the sudden decrease in resistivity of the film is that these islands continue to grow into bigger grains showing overgrowth on the film surface.

Activation energy (E_0) for different thicknesses of Cu₂S sample are calculated from Figure 8 and using the slope value in equation (8) the calculated values are entered in Table 4.

$$\rho = \rho_0 \frac{\exp(-E_0)}{K_B T} \quad (8)$$

Figure 9 shows the variation of log resistivity versus inverse of absolute temperature curve for 5600 Å film thickness at constant current ($I = 50 \mu\text{A}$). It is inferred from Table 4 that activation energy decreases with increases in film thickness and this may be due to the presence of defect states in the film.

3.4 Dielectric Measurements

The Poole-Frenkel effect is often observed in dielectrics and in amorphous materials due to the presence of large number of defect states in the energy gap of the

Table 3. Variation of grain size with film thickness

Thickness (Å)	Grain size (nm)
1000	207.59
3500	138.40
5000	19.77
7000	103.79

Table 4. Variation of activation energy with film thickness

Thickness (Å)	Activation energy E_0 (eV)
1000	0.595
5000	0.525
7000	0.450

material. The dielectric constant (ϵ_r) of the material in the frequency range (10–100 Hz) was calculated using the formula.

$$\epsilon_r = \frac{cd}{A\epsilon_0} \quad (9)$$

where C is the capacitance of the film, A is the area of the electrode, d is the thickness of the film and ϵ_0 is the electrical constant. Variation of dielectric constant as a function of frequency for different thickness is shown in Figure 10. The larger dielectric constant value of Cu₂S thin film is due to the fact that the particles act as dipoles under the influence of electric field. As the par-

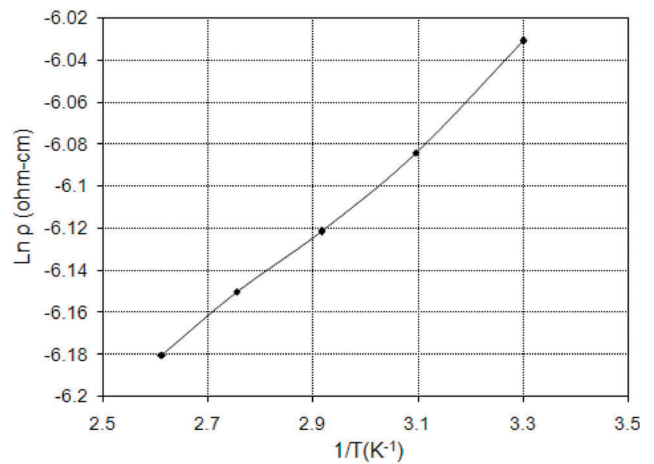


Figure 9. Variation of log resistivity vs. inverse of temperature for Cu₂S thin film.

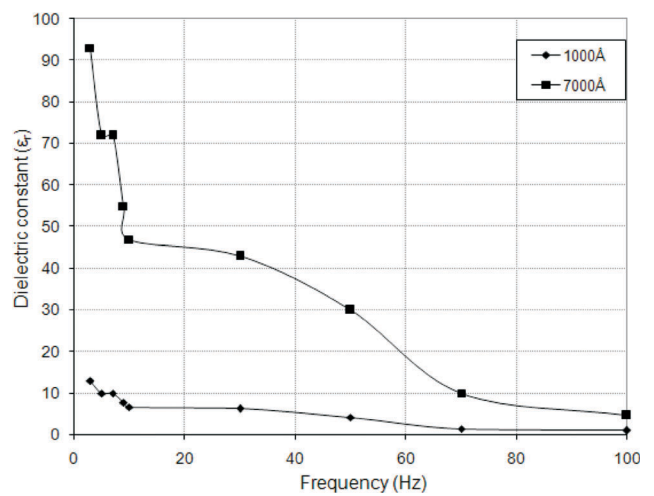


Figure 10. Variation of dielectric constant vs. frequency for Cu₂S of different thickness.

ticle size is in nanometer range, the number of particles per unit volume is very large and hence the dipole moment per unit volume increases, so the dielectric constant increases. Again it can be seen from the figure that dielectric constant increases with increase in thickness which is due to the decrease in size of the particle at higher thickness (Table 3). Due to this reason the number of particle per unit volume increases and hence the dielectric constant increases [27]. Dielectric losses are generally reflected in resistivity measurements ie, materials with high resistivity exhibits low dielectric loss and vice-versa [28]. Therefore it is concluded that low thickness Cu_2S film exhibiting high resistivity has low dielectric loss when compared to higher thickness film.

3.5 Photocurrent Measurements

When the film is illuminated by light, additional photo excited carriers are generated in the films. As a result, part of thermally excited carrier together with large number of photo-generated carriers neutralizes some fraction of localized charges in the depletion regions in the grain boundary potential barriers. Photo-conductivity signal versus intensity of incident light is described by the relation [29]

$$I_{pc} \propto (I_L)^\alpha \quad (10)$$

where I_{pc} is photo current, I_L is the incident photon flux and α is the empirical constant. The variations of photo current with applied voltage for different film thickness are shown in Figure 11. The conductivity is found to increase exponentially with applied voltage [18,30] as well as film thickness. It is observed from the graph that the film exhibit linear dependence of photocurrent with applied voltage and this behavior indicates that the film is free from trap.

4. Conclusion

The conduction mechanism in evaporated Cu_2S thin films was analyzed from the current-voltage characteristics. The possible conduction mechanism in Cu_2S films

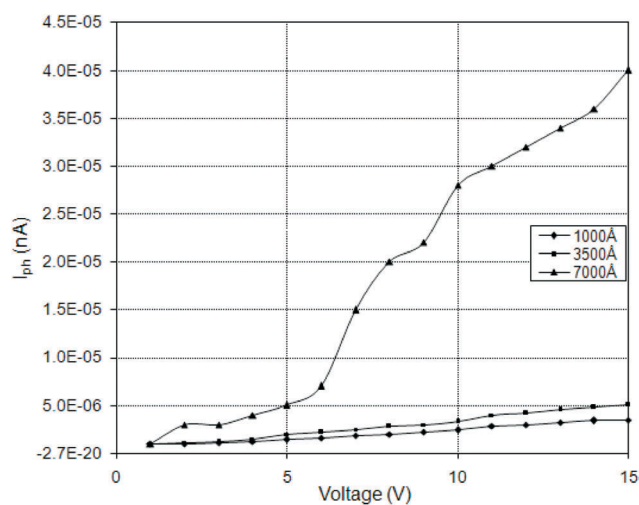


Figure 11. Variation of photo current vs. voltage for Cu_2S film of different thickness.

is found to be Poole-Frenkel type. The ionized donor density, flat band potential, barrier height are calculated from the capacitance voltage measurements. Resistivity studies revealed that low temperature Cu_2S thin films exhibit high conductance while high temperature film has high resistance. Higher thickness Cu_2S films with low resistivity and low barrier height has superior conducting behavior than low thickness Cu_2S films. The Cu_2S thin films exhibit high dielectric value and this dielectric increases with increase in film thickness.

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