CHAPTER VI

DIELECTRIC AND AC CONDUCTION PROPERTIES

6.1 Introduction

An important fundamental property of a semiconducting material is its dielectric permittivity. A knowledge of this parameter is necessary for many problems in semiconductor physics and in particular, it is essential for electric field simulated emission studies. Another important aspect for the study of the dielectric properties of materials is to understand certain fundamental physical properties of the system, like the presence of impurities, voids, structural defects, various polarisation and relaxation mechanisms, etc., [1-10].

The study of ac conductivity also yields information about the mechanism of conduction phenomena in the films [11, 12]. The chief advantage of ac measurements is that it permits investigation in the interior of the insulating materials. Secondly, the ac voltage bias being 0.05 V, the maximum field within the insulating material is kept to a minimum and there is little danger of more than one conduction process being active.

The study of ac conductivity (σ_{ac}) in disordered solids has attracted much attention in the last decade. One of the main reasons for this interest is the assumption, that conductivity is dominated by the deep defect levels lying in the mobility gap of these materials; hence a study of ac conductivity in these mate-

rials will throw light on the nature of these levels.

A close survey of the literature reveals that even though some work has been carried out on electrical [13,14] properties of Sb_2Se_3 thin films, their dielectric and ac conduction properties are seldom subjected to a systematic study.

This investigation is mainly intended to comprehend the basic dielectric and ac conduction properties of the material. A brief but relevant theory is given in the first part followed by the experimental details with a discussion on the results.

6.2 General Theory of Dielectrics

6.2.1 Complex dielectric constant (ϵ^*) and dielectric loss (tan δ)

The capacitance C (farads) of a parallel plate capacitor neglecting edge effects is given by

$$C = \epsilon' \epsilon_o A / d \qquad (6.1)$$

where A is the area of the electrodes (m^2), d is the thickness (m), ϵ' is the relative permittivity and is the permittivity of free space (8.85×10^{-12} Fm^{-1}).

When an alternating electric field E is applied to a dielectric, the displacement D varies periodically with time. However D lags behind in phase relative to the field E, so that for example if

$$E = E_o \cos wt \qquad (6.2)$$

We have

$$D = D_o \cos (wt - \delta) = D_1 \cos wt + D_2 \sin wt \qquad (6.3)$$

where δ is the phase angle and

$$D_1 = D_o \cos\delta \text{ and } D_2 = D_o \sin\delta \quad (6.4)$$

For most dielectrics, D_o is proportional to E_o, but the ratio D_o/E_o is generally frequency dependent. To describe this situation, two frequency dependent dielectric constants $\epsilon'(w)$ and $\epsilon''(w)$ are introduced which are given by

$$\epsilon'(w) = D_1 / E_o = (D_o/E_o) \cos\delta \quad (6.5)$$

$$\epsilon''(w) = D_2 / E_o = (D_o / E_o) \sin\delta \quad (6.6)$$

It is convenient to combine these two constants into a complex dielectric constant.

$$\epsilon^* = \epsilon' - j\,\epsilon'' \quad (6.7)$$

The relation between D and E, both expressed as complex quantities, is then simply

$$D = \epsilon^* E_o e^{iwt} \quad (6.8)$$

From equations (6.5) and (6.6)

$$\tan\delta = \epsilon''(w) / \epsilon'(w) \quad (6.9)$$

Since both ϵ' and ϵ'' are frequency dependent, the phase angle δ is also frequency dependent. The $\tan\delta$ is frequently called as the loss factor.

6.2.2 Polarisation

The storage capacity of a capacitor system is increased by inserting a dielectric material between the two plates and it is done by neutralizing some of the free charges at the electrodes which would otherwise contribute to the external field. This

phenomenon is known as dielectric polarisation. Thus polarisation P is related to the displacement D and electric field E by the relation in the vector form as,

$$\bar{D} = \epsilon_o \bar{E} + \bar{P} \quad \text{(for vacuum)} \tag{6.10}$$

However when a dielectric material of permittivity is inserted then this reduces to,

$$\bar{P} = \epsilon_o (\epsilon' - 1)\bar{E} \tag{6.11}$$

$$\text{also} \quad \bar{P} = N \bar{\mu}$$

where N is the number of dipoles and $\bar{\mu}$ the average dipole moment due to the charged particles, which is assumed to be proportional to the local electric field of strength E, that acts on the particle, then,

$$\bar{\mu} = \alpha \bar{E} \tag{6.12}$$

where α is the polarizability of the dipole. Combining equations (6.10), (6.11) and (6.12), we have,

$$\bar{P} = \epsilon_o (\epsilon' - 1) \bar{E} = N \alpha \bar{E} \tag{6.13}$$

which is the well known Clausius equation and links the macroscopically measured permittivity to three molecular parameters viz., N, α and E.

6.2.3. Relaxations

Relaxation is a process, which is applied to linear systems where a polarisation and an electric field are proportional to one another in equilibrium. The lag between field and polarization implies an irreversible degradation of free energy to heat.

The dielectric flux density D resulting from an applied alternating field E, would differ in phase from E. This is due to the inertia of the polarisation which, when the frequency becomes high enough, cannot follow the field variation giving rise to a 'relaxation' of the measured permittivity. This behaviour may be stated by an equation of the type,

$$\epsilon_r(w) = \epsilon_\infty + \int \alpha\ (t) \exp\ (jwt)\ dt \qquad (6.14)$$

in which ϵ_∞ is the value of permittivity at infinite frequency which is a constant and α (t) is a sort of decay factor accounting for the lagging of polarisation behind the applied field. Most of the relaxations can be described by means of Debye equations.

Debye proposed an exponential form for the decay factor,

$$\alpha\ (t) = \alpha\ (0)\ e^{-t/\tau_a} \qquad (6.15)$$

where τ_a is a relaxation time, characteristic of the dielectric which may be a function of the temperature but not of time. Substituting this in the above equation,

$$\epsilon_r(w) = \epsilon_\infty + \int \alpha(0) \exp^{(jw - 1/\tau)\ t}\ dt \qquad (6.16)$$

Cole and Cole (15) showed that the above equation (6.16) should be replaced by the more general expression,

$$\epsilon' - j\ \epsilon'' = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 - (jw\tau_a)^{1-\beta}} \qquad (6.17)$$

where τ_a is the mean relaxation time and β is a constant of

value between 0 and 1. If the results are plotted in an argand diagram, the points still lie on a circle, but its center is depressed below the ϵ' axis. If the radius of the circle is drawn to the points (ϵ_s , 0), the angle between the radius and the ϵ'axis is equal to $\beta\pi$ / 2. β may be expressed as a spreading factor of the actual relaxation time about the mean value τ_a. There is no molecular interpretation for this factor, but it is useful in analysing a complicated relaxation spectrum.

6.2.4 Temperature Coefficient of Capacitance

Dielectrics are used at all temperatures and the temperature variation of capacitance is hence crucial. An important parameter used to indicate the variation of capacitance with temperature is the temperature coefficient of capacitance (TCC) which is defined by

$$TCC = \Gamma_C = 1 / C\ (dC/dT)_p \qquad (6.18)$$

where T is the temperature and p the pressure (constant). Γ_C is usually expressed in parts per million (ppm) per degree Celsius.

6.2.5 Frequency dependent behaviour of dielectric constant and dielectric loss in thin films

The dielectric constant ϵ' and the loss factor tanδ are not absolute constants for an actual dielectric. Both these constants depend on many factors, namely the temperature, the frequency, the magnitude of the alternating voltage, the humidity of air, etc. The frequency dependence of tan δ in thin films is a very important factor. Usually the dielectric constant decreases

with increasing frequency. At very low frequencies, the dipoles are oriented by the electric field without time lag. In this case the orientation of the dipoles is not accompanied by any loss of energy and hence tanδ is very low. At very high frequencies the dipoles are not able to follow the variations of the field, whereas the normal agitation is still present. The loss factor of thin film dielectric shows a general behaviour of increasing loss at very high frequencies with a wide minimum in the intermediate range, as a result of superposition of various loss mechanisms.

The loss minimum observed in many cases [16-21] was explained by Goswami and Goswami [22]. The capacitor element was assumed to comprise (i) an inherent capacity element (C) unaffected by frequency and temperature, (ii) a discrete resistance element (R) due to the dielectric film in parallel with C and (iii) a series resistance (r) due to lead lengths etc. It was assumed that while R would be affected by temperature due to the exponential factor in the relation

$$R = R_o \exp(\quad E/kT) \tag{6.19}$$

the lead resistance r would be more or less of constant value. In general R would be >> r. A mathematical analysis of the equivalent circuit lead to several conclusions.

The series capacitance and the loss factor (tan δ) were obtained as

$$C_s = \frac{1 + w^2 R^2 C^2}{w^2 R^2 C} = (1 + D^2) C \quad (6.20)$$

and $\tan\delta = 1/wRC + r/wR^2C + wrC = D(1+r/R) + wrC$ (6.21)

where $D = 1/wRC$.

The loss minimum would be observed at a frequency given by

$$w_{min} = \sqrt{1/rRC^2} \quad (6.22)$$

When $R^2C >> r$ or $r/R << 1$ is true for all cases equation (6.21) reduces to

$$\tan\delta = 1/wRC + wrC = D + RC \quad (6.23)$$

when w is small $1/wRC >> wrC$, then

$$\tan\delta = 1/wRC = D \quad (6.24)$$

when w is large $1/wRC$ is generally $<< wrC$ and hence

$$\tan\delta = wrC \quad (6.25)$$

Thus equations (6.24) and (6.25) predict a condition when the loss factor will be inversely proportional or directly proportional to w.

It is observed from equation (6.22) that w_{min} is determined by $(1/rRC^2)^{\frac{1}{2}}$. Since R would considerably increase with the lowering of temperature, r remaining practically unchanged, w_{min} will also be lower at lower temperatures. Thus a shift of w_{min} would be to a lower frequency at lower temperature and vice versa. In a similar way, the variation of $\tan\delta$ with temperature could be explained.

The effect of temperature on capacitance was obtained from equation (6.20) which could be rewritten as

$$C_s = 1 / \; w^2R^2C + C = (1 + D^2)C \qquad (6.26)$$

If however $1 / w^2 R^2 C << C$ or $D^2 << 1$, then

$$C_s = C \qquad (6.27)$$

This condition can be realised in a capacitor system either by increasing R, ie., by lowering the temperature or by raising it. A suitable combination of these factors along with an appropriate value of C will lead to the frequency independent capacitance.

6.3 AC conduction

The ac conductivity of amorphous materials gives an idea about the conduction mechanism in such materials. In a parallel plate capacitor, if C_o is it's capacitance value in air and ϵ_r is the permittivity of the medium that filled the condenser, then, neglecting the fringing effects, the capacitance can be expressed as,

$$C = \epsilon_r C_o \qquad (6.28)$$

when an alternating e.m.f. V with an angular frequency $(2\pi f)$ is applied across this condenser, the alternating current is given by,

$$i = jw\epsilon_r C_o V \qquad (6.29)$$

Substituting the values of C_o as $C_o = \epsilon_o A/d$, where A is the common area of the plates, d the separation between them and the angular frequency, equation (6.29) becomes

$$i = jw \; (\epsilon' - \epsilon'') \; (A/d)\epsilon_o V \qquad (6.30)$$

By definition the field strength is $E = V/d$ where V is the voltage applied and the current density is $J = i/A$.

Therefore,

$$J = jw\epsilon_o\epsilon' E + \epsilon_o \epsilon'' E \tag{6.31}$$

The conductivity is,

$$J / E = jw\epsilon_o\epsilon' + w\epsilon_o \epsilon'' \tag{6.32}$$

In a dielectric, the imaginary part is accounted for the capacitance which involves the real part ϵ' of the relative permittivity. Thus the dielectric conductivity is defined as the real part of the ratio J/E, that is,

$$\sigma = w\epsilon_o \epsilon'' \tag{6.33}$$

The dielectric conductivity σ, represents the sum of all the loss mechanisms in the material and is a measure of the performance of the dielectric as an insulator. For a parallel equivalent circuit,

$$\tan\delta = 1 / wC_pR_p \tag{6.34}$$

where C_p is the parallel equivalent capacitance and R_p is the parallel ac resistance. Therefore the equivalent parallel conductance G_p will be

$$G_p = 1 / R_p = wC_p \tan\delta \tag{6.35}$$

The frequency dependence of conductivity of different materials has been widely investigated by many researchers [23-27] because they can give information about the electronic structure of these materials. In all these cases the conductivity was found to be a non-decreasing function of frequency. Such a behaviour is suggestive of hopping conduction [28], either by electrons or ions. This theory is in line with Anderson's theory [29], according to which, the localised states generally exist in

disordered systems, and with Mott's [30] model of the pseudogap in amorphous solids.

Pollack and Geballe [31] have given a detailed interpretation of the frequency dependence of the electrical conduction in amorphous solids. The ac conductivity should increase with frequency of the applied field and saturate at frequencies high enough to be comparable to the natural frequency of hopping between the centers. It has been pointed out that it is necessary to average conductivity over all hopping distances and activation energies for all pairs of occupied and empty centers [32]. This averaging process yields, for single or multiple hops the ac conductivity $\sigma(w)$ in amorphous solids depends on the angular frequency w obeying the relation

$$\sigma(w) \;\alpha\; w^n \tag{6.36}$$

where n decreases from 1.0 for single hops and 0.5 for multiple hops. This relation is in reasonable agreement with the observations [24,33]. The increase of n at low temperatures is due to the predominance of single hops. This indicates that the frequency dependence of ac conductivity saturates at very high frequencies.

However, there are a few cases where a square law dependence is observed [23,34]. This is explained on the basis of two center hopping as

$$\sigma' \;\alpha\; w^2 \tag{6.37}$$

i.e., the conductivity increases as the square of the frequency.

The dielectric films which have high densities of localized levels in the forbidden gap also exhibit the hopping mechanism. The presence of compensated donors and acceptors in the crystalline dielectric also associates the hopping process with them. These properties could be expected in the case of amorphous substances. In these substances, electronic transitions between trapping levels suggest a more fundamental hopping mechanism which gives the Mott $T^{-1/4}$ law [35]. Jonscher has theoretically shown that the change in the character of transitions of the carriers is related to the slope of the $\log\sigma$ versus frequency curve.

6.4 Measurements

The dielectric and ac conduction studies on Sb_2Se_3 films have been carried out by forming metal-semiconductor-metal (MSM) structures. Aluminium films were used as metal electrodes. The series capacitance (C_s) and the dissipation factor (D) of the respective films in the frequency range 5 Hz to 10 MHz at various temperatures (296 - 456 K) were measured using Hewlett Packard digital low frequency impedance analyser (4192A). A precaliberated copper-constantan thermocouple was used to measure the temperature.

Since the deposited films at T_s = 303 and 493 K contain many defects such as voids, grain boundaries, pinholes, etc., they were subjected to annealing treatment at about 490 K in a vacuum

of 1.3 Pa for repeated one hour cycles. The measurement of dielectric constant and other related parameters of Sb_2Se_3 thin films was carried out on well stabilized capacitors.

The equivalent parallel capacitance (C_p) and series capacitance (C_s) are connected by the relation

$$C_p = C_s / (1 + D^2) \quad (6.38)$$

and $$C_s = C_p (1 + Q^2) / Q^2 \quad (6.39)$$

where D is the dissipation factor or loss factor (tanδ) and Q is the quality factor.

The r.m.s voltage applied across the thin film capacitors throughout the study is 50 mV. The dielectric constant has been calculated from the measured value of capacitance, capacitor area (A) and the dielectric film thickness (d) using the relation

$$\epsilon' = Cd / \epsilon_o A \quad (6.40)$$

The loss factor tanδ is given by

$$\epsilon'' = \epsilon' \tan\delta \quad (6.41)$$

The ac conductance (G_p) has been evaluated from the measured values of series capacitance and dissipation factor using the relation

$$G_p = 2\pi f \left(\frac{C_s}{1+D^2} \right) . \frac{\epsilon''}{\epsilon'} = wC_p \tan\delta \quad (6.42)$$

6.5 Results and discussion

6.5.1 Effect of substrate temperature and thickness on dielectric constant

The thickness dependence of dielectric constant of Sb_2Se_3 films deposited at T_S = 303 and 493 K before annealing are shown in Fig. 6.1. The dielectric constant of the films deposited at T_S = 303 K are found to increase with thickness in the lower thickness range and attains a constant value above 200 nm. This can very well be attributed to the reduction of a number of defects like voids, discontinuities, stacking faults, etc., as the thickness increases [36]. The large increase in dielectric constant of the films deposited at T_S = 493 K is due to the fact that at higher substrate temperature; defects like interstitials are recovered considerably. The unusual oscillatory behaviour of dielectric constant of polycrystalline films with thickness above 200 nm can be due to random variation in grain size as observed from SEM analysis (Fig. 3.5 (a) and 3.5(b)).

6.5.2 Effect of frequency and temperature on dielectric constant

The variation of dielectric constant (ϵ') and loss factor ($\tan\delta$) with frequency (5 Hz - 10 MHz) at various temperatures for annealed Sb_2Se_3 films deposited at T_S = 303 K of typical thickness 125 nm are shown in Figs. 6.2 and 6.3. The large increase in dielectric constant with decrease of frequency (Fig. 6.2) can be explained on the basis of charge carriers being blocked at the electrodes. The space charge layer existing in the dielectric

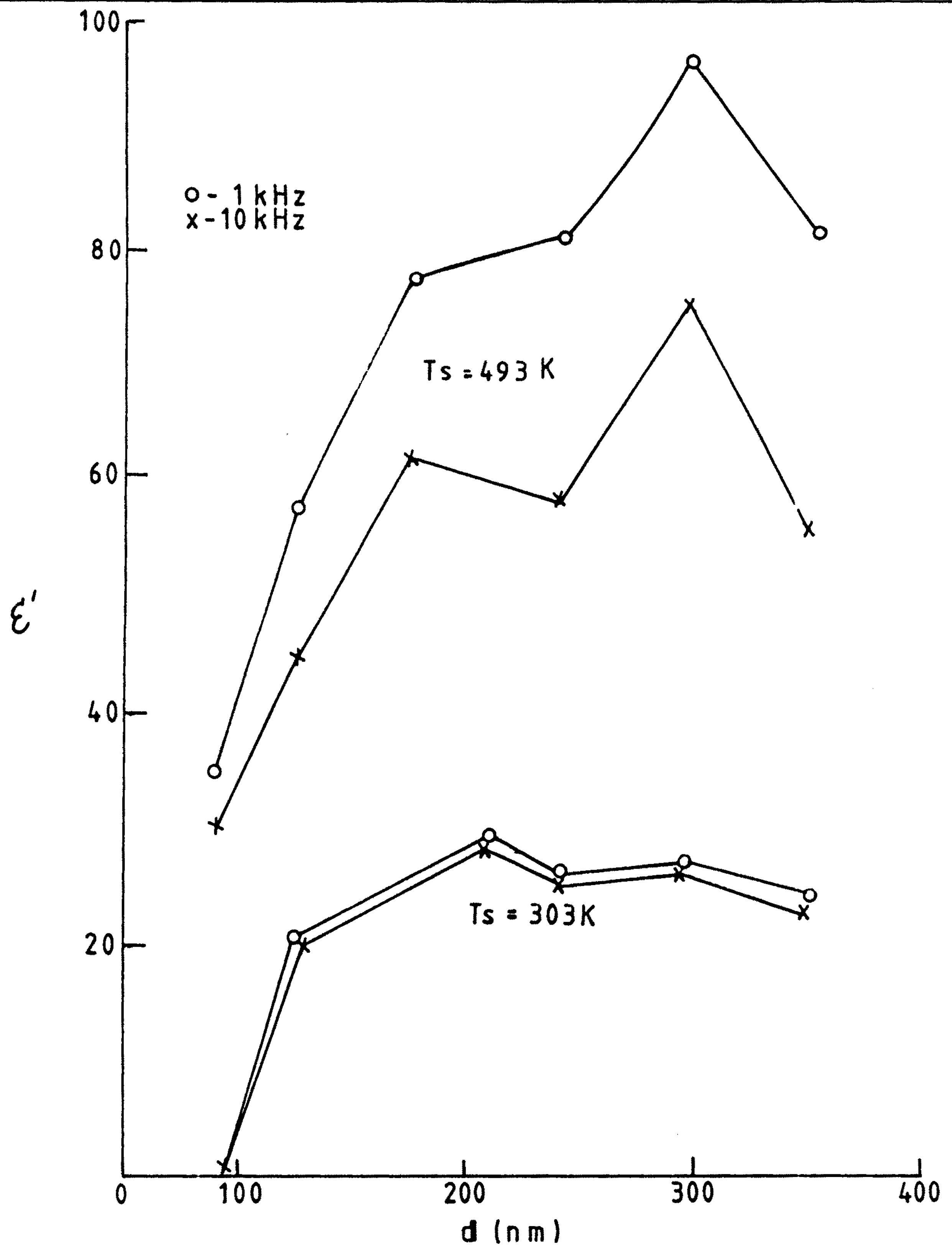


Fig 6.1 THICKNESS DEPENDENCE OF DIELECTRIC CONSTANT ε' OF $Sb_2 Se_3$ THIN FILMS

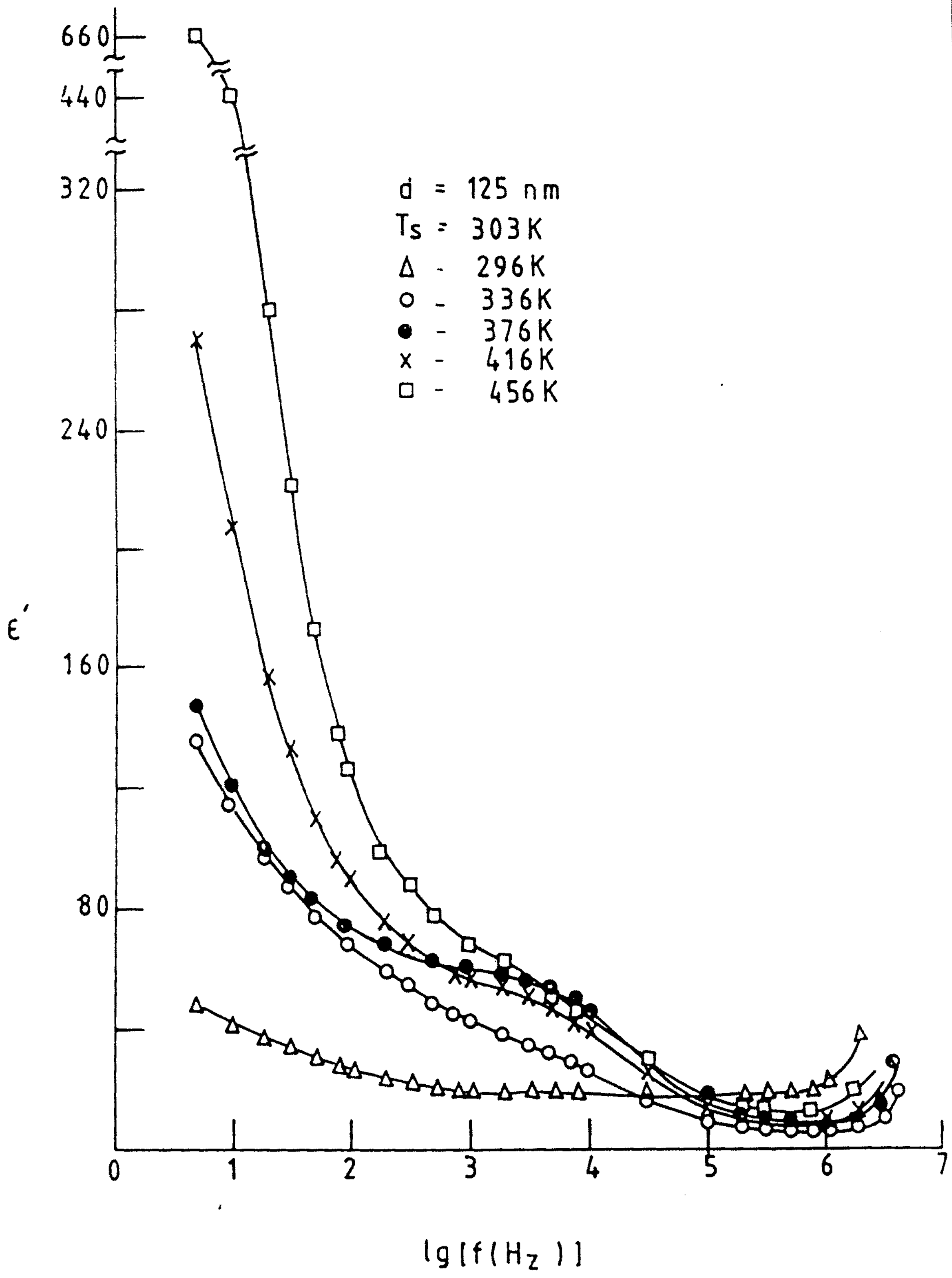


Fig 6.2 DEPENDENCE OF DIELECTRIC CONSTANT ε′ ON FREQUENCY (Lg f) AT DIFFERENT TEMPERATURES

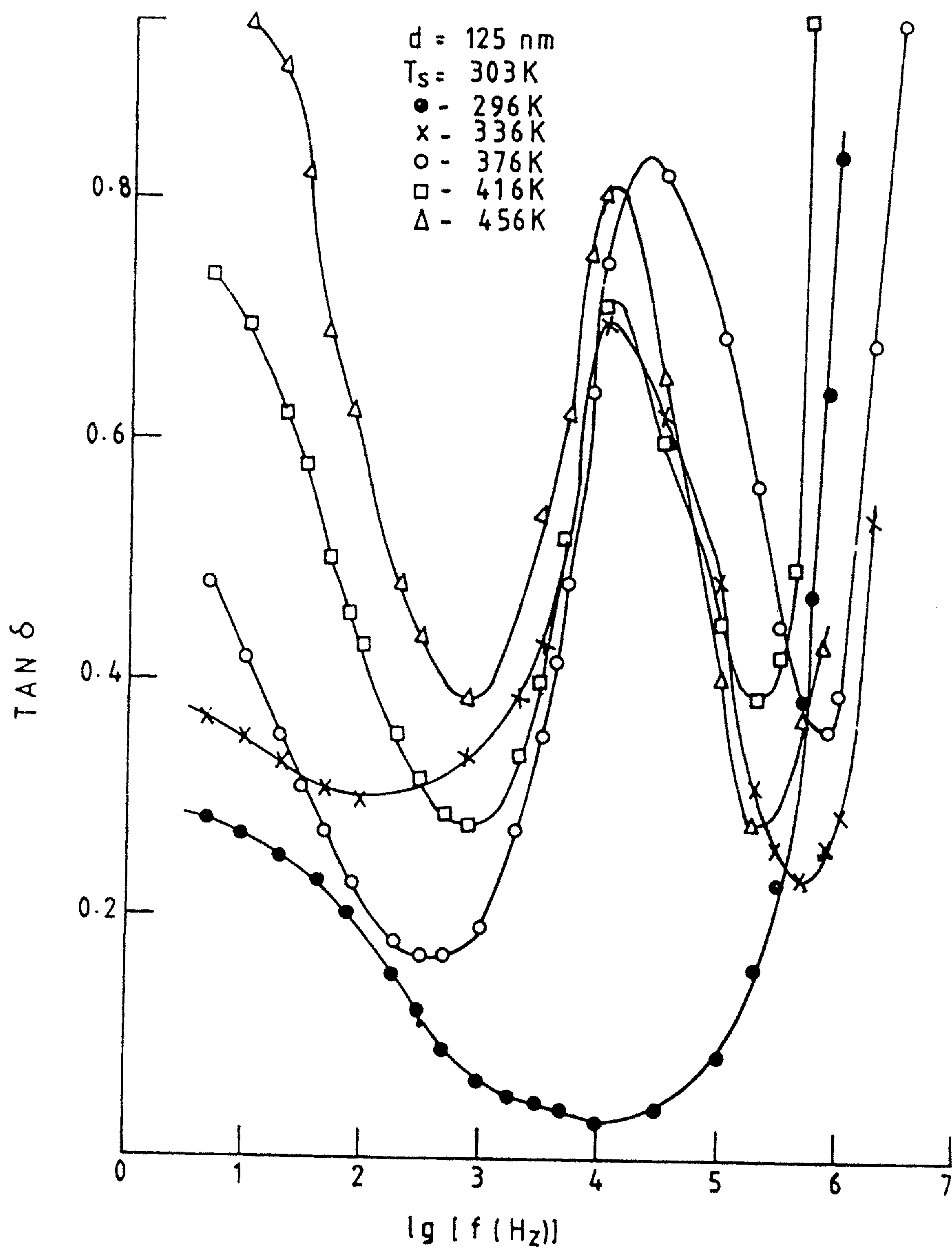


Fig 6.3 DEPENDENCE OF TAN δ ON FREQUENCY (Lg f) AT DIFFERENT TEMPERATURES

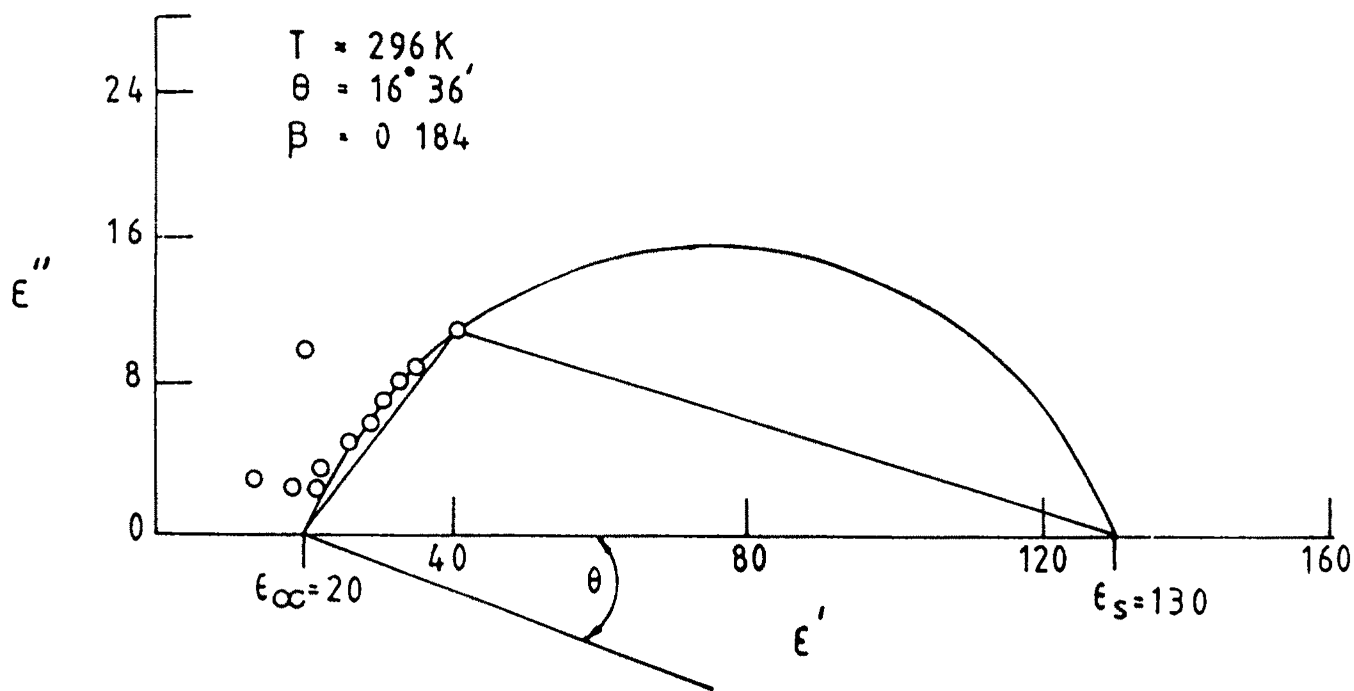
film should lead to a substantial increase in capacitance due to interfacial polarisation [37]. The striking loss peak minima and maxima observed in Fig. 6.3 is in accordance with the Debye's theory of dipole orientation [38]. The decrease in loss in the low frequency region may be ascribed to the relaxation of pairs of point-defects in the dielectric which causes a $\tan\delta_{min}$, that shifted to the high frequency region with the increase of temperature. Similar loss peaks are reported by earlier workers [39-41] and have been attributed to a dielectric relaxation phenomenon arising from dipolar reorientation. It is interesting to note that the magnitude of $\tan\delta_{max}$ increases and also shifts to the high frequency region with increase of temperature.

6.5.3 Cole-Cole plots

Fig. 6.4 shows Cole-Cole plots [15] ϵ'' versus ϵ' for Sb_2Se_3 film of typical thickness 125 nm at two different temperatures. According to Cole-Davidson, the points lie on an arc of a circle which tend to become a semicircle that indicates the presence of a single relaxation time as in the simple Debye model. For such materials Cole and Cole have modified the Debye theory to obtain the more general expression

$$\epsilon'' - \epsilon' = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + (jw\tau_a)^{1-\beta}} \qquad (6.43)$$

where τ_a is the mean relaxation time, β the spreading factor of the actual relaxation time about the mean value τ_a, ϵ_s and ϵ_∞ are the static and high frequency dielectric constants respec-



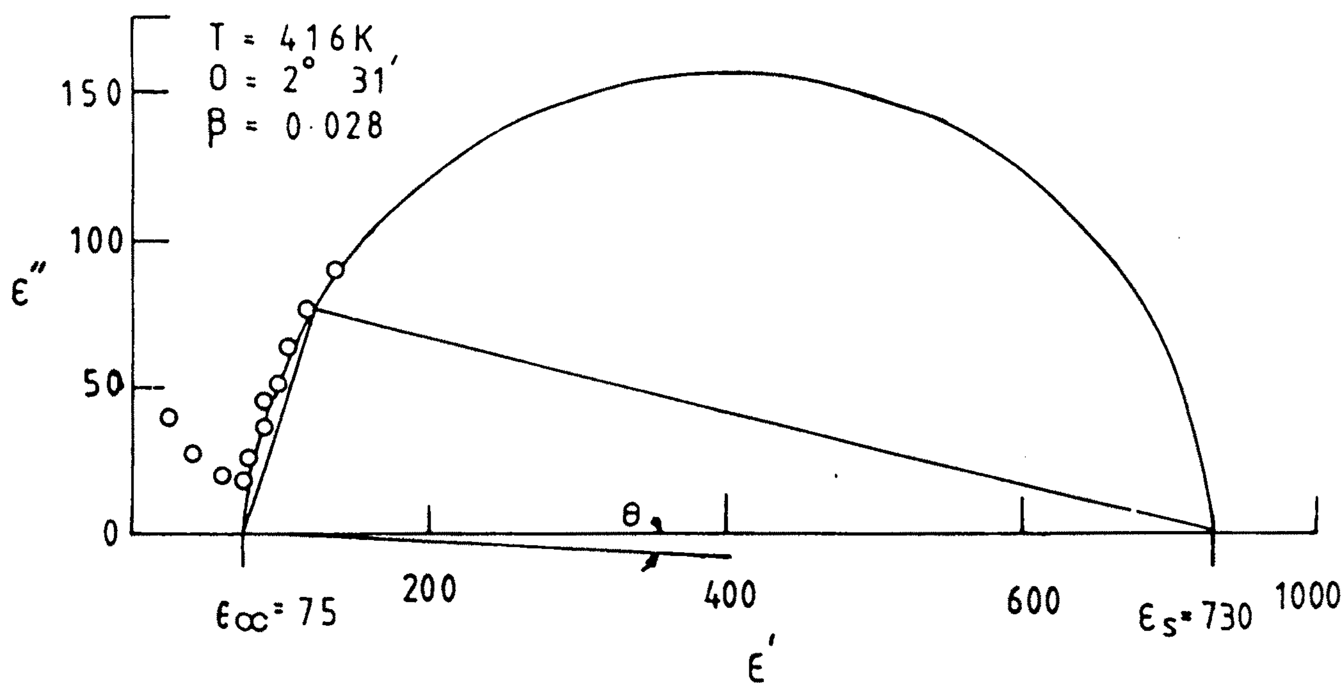


Fig 6.4 COLE -COLE PLOTS OF TWO RELAXATION PEAKS
(T_s = 296 K AND T_s = 416 K)

tively. The Cole-Cole plot of ϵ'' versus ϵ' is a semicircle with its centre below the ϵ' axis. The spreading factor β, ϵ_s and ϵ_∞ determined from the Cole-Cole plot are given in Table 6.1. It reveals that the spreading factor is found to be temperature dependent and its value tends to zero at higher temperatures. This is in accordance with the reports of the earlier investigators [42,43]. Knowing β, τ_a can be determined using the relation [22],

$$\frac{v}{u} = (\tau_a)^{1-\beta} \qquad (6.44)$$

where 'v' is the distance on the Cole-Cole diagram between ϵ_s and the experimental point, 'u' is the distance between that point and ϵ_∞ and w is the angular frequency. The values of β and τ_a obtained for two different temperatures are also presented in Table 6.1.

TABLE 6.1

Experimental values of β and τ_a

T (K)	ϵ_∞	ϵ_s	Spreading Factor β	Mean Relaxation Time τ_a (s)
296	20	130	0.184	5.6×10^{-2}
416	175	730	0.028	2.6×10^{-2}

6.5.4 Effect of temperature on capacitance

The temperature dependence of the capacitance of Sb_2Se_3 films for various constant frequencies is shown in Fig. 6.5. The experimental curves exhibit a gradual rise in the capacitance

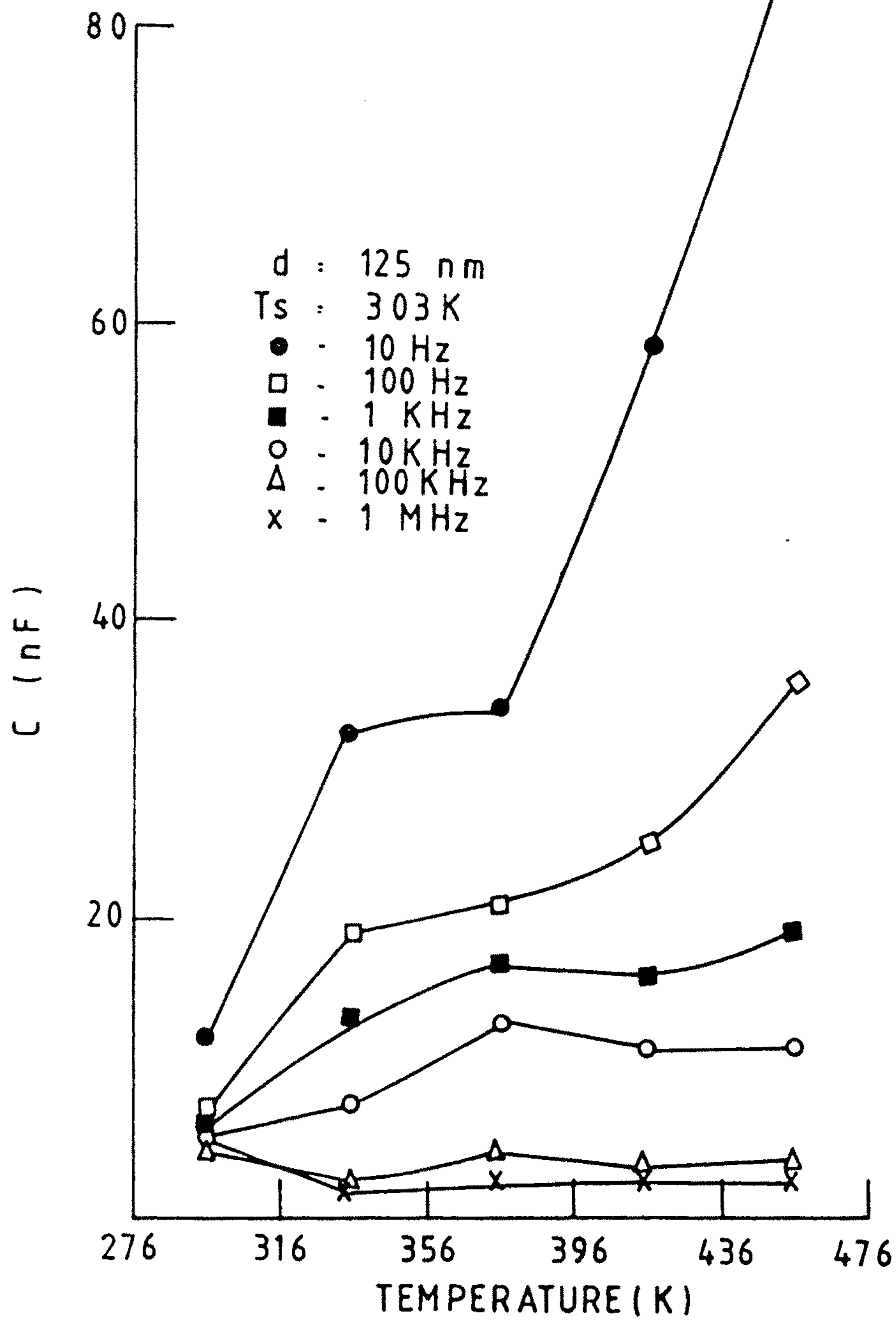


Fig 6.5 DEPENDENCE OF CAPACITANCE ON TEMPERATURES FOR VARIOUS FREQUENCIES

with temperature upto a certain value after which a steep increase is seen in the capacitance at low frequencies. A similar type of increase in capacitance with temperature beyond certain temperature has been observed by Parakh and Garg [44] in bulk Sb_2Se_3. The large increase in capacitance beyond certain temperature at low frequency may be due to ionic motion in the form of dipolar orientation. Decrease in capacitance with frequency is in agreement with Goswami Model [22]. The temperature coefficient of capacitance of a capacitor is determined from the relation as

$$TCC = 1/C \ (dC/dT)$$

and has been found to be 4690 ppm / K at 1 kHz and its value decreases gradually for higher frequencies.

6.5.5 AC conduction

The variation of ac conductance on frequency at different temperatures for Sb_2Se_3 films has been shown in a doubly logarithmic plot (Fig. 6.6). The curves exhibit two dispersive regions, one below 100kHz and the other above 100 kHz at all temperatures. In both the regions, ac conductance is proportional to w^n where the value of n depends on temperature and frequency. In the low frequency region, the value of n has been found to decrease from 0.8 to 0.4 as the temperature increase from 296 to 456 K. The differing contributions to conductance from the carrier movement (carrier movements between and within defect wells) at different temperature results in the decrease of n with

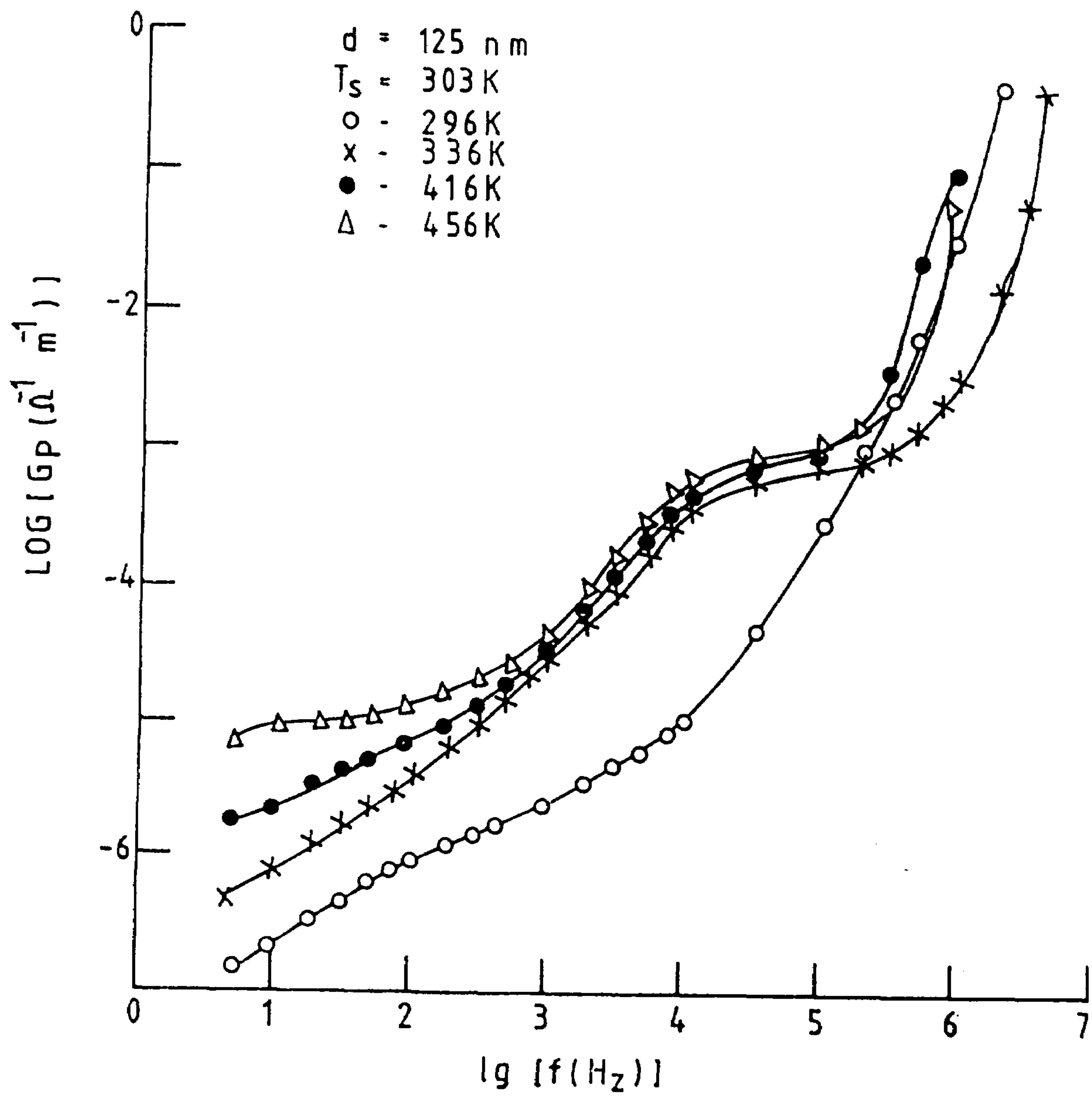


Fig 6.6 VARIATION OF AC CONDUCTANCE (Lg G_p) WITH FREQUENCY (Lg f) AT DIFFERENT TEMPERATURES

increasing temperature. The $\approx w^1$ ' dependence at low frequency is very similar to the behaviour predicted and observed by Pollak and Geballe [31] for hopping between a random distribution of sites in impurity conduction at low frequencies in Sb_2Se_3. They have explained that if hopping takes place between the random distribution of localised states then the value of n lies between 0.5 to 1.0, the lower value of n occurs for multiple hops while the higher occurs for single hops. Also a more detailed explanation has been given by Jonscher [45] for this behaviour on the basis of easy and difficult hops of the carrier between possible sites. Similar type of hopping conduction has been observed in many materials [23, 26,47,48]. At high frequencies the value of n is found to vary from 2.2 to 2.7 and suggests a square law dependence on frequency at all temperatures studied. The square law dependence of ac conductance at higher frequencies has also been explained by Argall and Jonscher [23] based on two center hopping. Though the high frequency (> 100 kHz) dependence of conductance is accompanied by the lead resistance and electrode effects, in the present study the effect of electrodes are substantially minimised by the use of thick aluminium deposits (300 nm).

6.5.6 Effect of temperature on ac conductance

The temperature dependence of ac conductance is shown in Fig. 6.7 for Sb_2Se_3 film. From the slope of the plot lgGp versus 1/T the activation energy is calculated and is found to

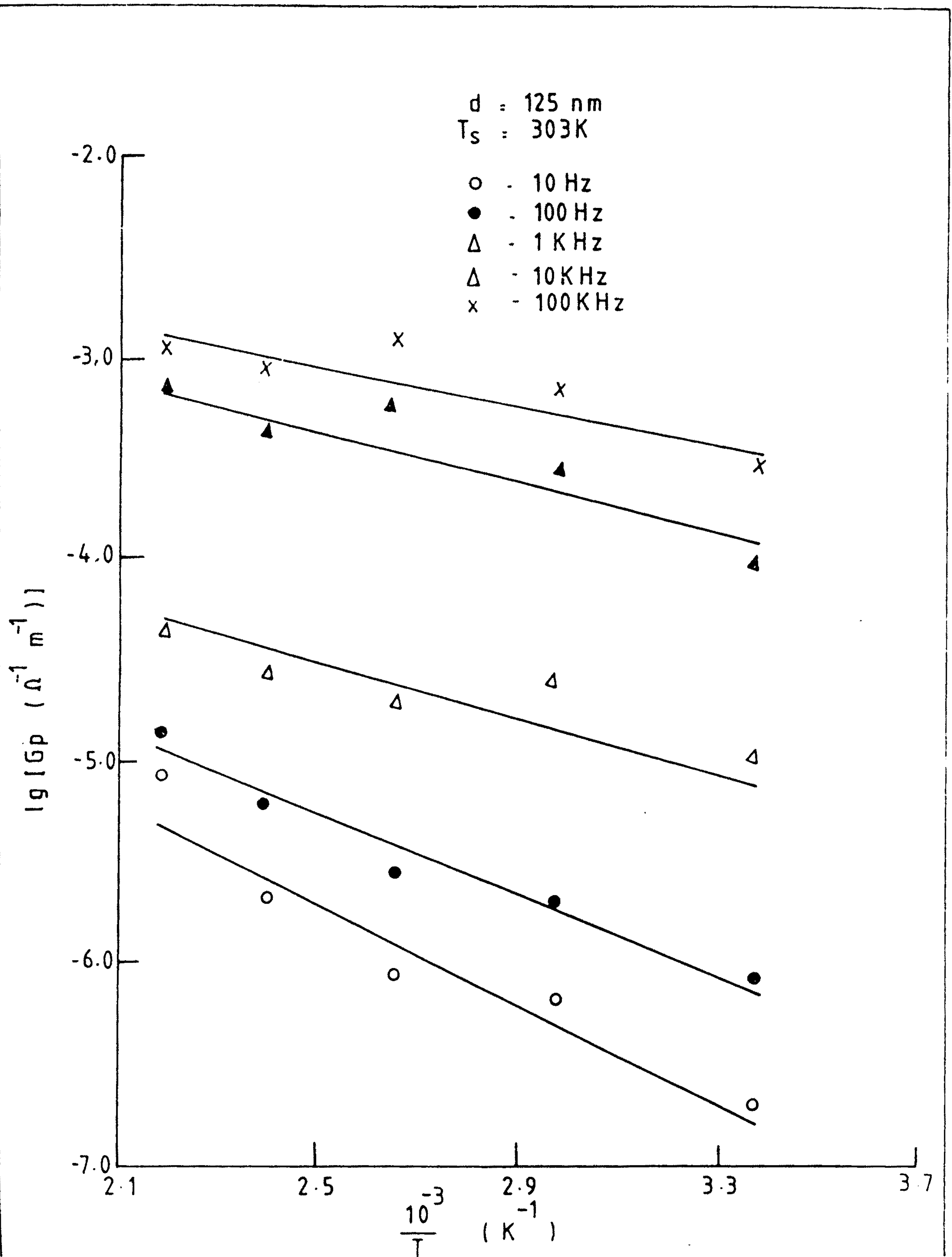


Fig 6.7 VARIATION OF AC CONDUCTANCE (Lg G_P) WITH TEMPERATURES AT DIFFERENT FREQUENCIES

decrease from 0.25 to 0.13 eV as the frequency increases from 10 Hz to 100 kHz. This is because of the low energy required for the hopping of charge carriers at higher frequencies. The observed activation energy represents the energy required for the motion of holes in the valence band due to excitation of electrons into acceptor level and to hopping of electrons between localised defects because of its p-nature which has been confirmed by the thermoelectric studies. The p-type nature of bulk Sb_2Se_3 has also been observed by Chakraborty et al [49].

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