CHAPTER V

ELECTRO LUMINESCENCE

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5.1 INTRODUCTION :

Electroluminescence (EL) is a phenomenon of light emission by luminors due to the sole action of electric field (a.c.ord.c). Studies of voltage and frequency dependence of time averaged electroluminescent brightness, brightness waveforms are the important means to understand the mechanism of electroluminescence. In present chapter studies on voltage and frequency of the time averaged electroluminescent brightness are studied, so as to obtain the information about -

i) effect of activators on EL behavior,
ii) Vòltage dependence of EL brightness.
iii) frequency dependence of EL brightness,
iv) mechanism of electroluminescence,
v) kinetics involved in EL process.

5.2 THEORETICAL BACKGROUND :

5.2.1:Relation between brightness and applied voltage :

The electroluminescent brightness depends on the exciting voltage, A variety of mathematical forms proposed by various workers are presented here and there appears to be a considerable disagreement as regards the exact quantitative relationship.

Destriau (1) in (1936) observed that the time average of emission intensity of electroluminescent zinc sulphide phosphor increases exponentially with increasing amplitude of exciting a.c. field. The first relation proposed by him is

$B = a \exp.(-b/v)$ ------- (5.1).

Where B and V are brightness and voltage respectively, and 'a, and 'b, are constants independent of V, but depends on temperature, type of phosphor and fequency of applied voltage. The relation is amenable to simple theoretical interpretation(2) Later Distriau (3) changed this to $B = av^n \exp. (- b/v)$ --(5.2) Where n is a constant.

For single crystal of ZnS a relation

$$B = av^n \exp. (-b/ v) \qquad \text{....(5.3)}$$

is given by Alfrey and Taylor (4). This equation with n=1 is found to hold good for ZnS:Mn films (5). The equation(5.3) with n=0 reduces to

$$B = a \exp. (-b/ v) \text{ ----------} \qquad (5.4)$$

This equation was first employed by Alfrey and Taylar (4) and Zalm (6,7), Zalm and his co-workers (6,7) have reported that his equation is valid over seven decades of brightness for microcrystalline ZnS phospors.

Later on Thornton and others (8,9,10) have shown that the equation (5.4) holds only at low voltage but at high voltages the equation

$$B = a \exp. (- b/v) \qquad \text{.......(5.5)}$$

Works more satisfactorily

Lehmann (11) found the emission of individual particles to obey equation (5.5) and found that summation of the output of large number of particles leads to the normally observed equation (5.4).

A relation similar to equation (5.5)

$$B = a \exp. (-b / V + V_0) \qquad \text{.....(5.6)}$$

Was first proposed by Destriau and Ivey (12) and subsequently used by Morehead (13) with some success in ZnS and ZnS:Cu;Cl for microcrystalline ZnS activated with Cu, Howard et.al (14) and Lehmann proposed

$$B = aV \exp. \left(- b/V+Vo \right) \qquad \ldots\ldots (5.7)$$

Where b and Vo increases slowly with frequency. On the basis of exhaustion barrier theory Taylor (16) deduced the following relation.

$$B = Bo \exp. - \left(\frac{V_o}{V} \right)^{\frac{1}{2}} \qquad \ldots\ldots (5.8)$$

Where as on the basis of impact ionisation mechanism Magy (17) derived the following relation

$$B = a \exp. - b / (1 + CV)^2 \qquad \ldots\ldots (5.9)$$

Luyckz and Stokkink (18) reported a relation

$$B = a \exp. (-bv) \qquad \ldots\ldots (5.10)$$

for microcrystalline ZnS : Mn

Harman and Raybold (19) have suggested the powerlaw relation

$$B = a (V - Vo)^n$$

For Zns phosphors with $n = 3$. A similar expression with $Vo = 0$ and $n = 2.5$ was given by Diemer (20) for single crystals of ZnS and was found to be valid at high voltages over five decades of brightness.

Halsted and Kollar (21) used equation (5.11) with $Vo = o$ and $n = 7$ Zalm et al (22) used it with $n = 3.5$, Gaur and Ranade (23) used this equation with $Vo = o$ and $n = 6.5$ for CaS : Ag phosphors. Lawangar and Pawar (24) used equation (5.11) with $Vo = o$ for Cas : Bi : Pd. phosphors.

On the basis of Piper and Willisms theory Howard etal (14) obtained another relation as

$$B = a v^{3/2} \left(1 - \frac{3\, v^{\frac{1}{2}}}{C} \right) \exp. \left(- C/V^{\frac{1}{2}} \right) \qquad \ldots\ldots (5.12)$$

With a and C as constants

Vlasenka and Popkar (25) have found another empirical formula for film panels i.e.

$$B = g \exp. \left(\sqrt{v/h} \right) \qquad \ldots\ldots \qquad (5.13)$$

Where g and h are constant.

Chan and yu (26) have shown that EL emittance of film panels dependson various parameters such as composition and aging of panels and applied voltage. They have studied evaporated film panels of ZnS: Mn, Cu, Cl and have given an emparical formula.

$$B = A \exp. \left(- G / F + \sqrt{v} \right) \qquad \ldots\ldots\ldots (5.14)$$

Where A, G and F are functions of operating time and can be regarded as constant during any measurement of EL emittance.

Z. Porada (27) applied the EL model suggested by Alfrey and Taylor (28) to explain the observed EL emmission excited by high energy electrons in the strong electric field of the metal semiconductor junction and arrived at the equation.

$$B = B_o \exp. \left(A/f \right) \exp. \left(- \alpha/\sqrt{v} \right) \qquad \ldots. (5.15)$$

Where B_o is a constant independent of units in which B is expressed, A is a parameter dependant on the technological conditions under which samples are prepared and temperature, f is a frequency and α is a parameter dependent on the value of the critical intensity of the electric field causing ionisation of the luminescent centre with V as applied voltage.

5.2.2 Relation between brightness and applied frequency :

The EL brightness is found to be a function not only of the applied voltage but also of the frequency. Various relations between brightness and frequency given by different workers are given below.

Curie (29,30) has derived an expression on the assumption that there exists a bimolecular process of recombinations of electrons with recombination centres, and that the same number of excited centres are produced per cycle

The expression

$$B = B_0 \frac{n_0^2 \; \alpha}{(1 + \frac{n_0 \alpha}{2f})} \qquad \ldots\ldots(5.16)$$

Where α is the recombination coefficient, n_0 the initial concentration of free electrons in the conduction band, f the frequency of applied field and B_0 a proportionality constant

Thornton (8) considering the recombination rate as determined from the electrons released from traps and controlled by electric field obtained

$$B = n_0 f \left[1 - \exp.\ (- A/f) \right] \qquad \ldots\ldots (5.17)$$

Where n_0 is number of excited centres for each half cycle , assumed to be constant $A = \int_0^{2\pi} \exp\ (av \sin x)\ dx$ with $x = \omega t$ is a field dependent factor which increases with voltage .

Alfrey and Taylor (28) has given a relation

$$B = B_0\ P \exp.\ (- P / 4\ f)\ \exp.\ (C/\sqrt{V}) \qquad \ldots\ldots (5.18)$$

Where C is constant V being applied voltage and P the rate constant for thermal emptying of traps and is given by

$$P = S \exp.\ (- E/KT) \qquad \ldots\ldots\ldots\ldots(5.19)$$

Here S and E represents the frequency factor and trap depth respectively.

Zalm (31) showed that the light emission during each cycle of the applied voltage is not constant but is dependent on the frequency of the applied field. He gave a relation

$$B \propto \exp (- \text{constant} / \sqrt{V}) \qquad \ldots\ldots (5.20)$$

Where $V = F (f,t)$, f, being frequency and t , the time. Thus light emitted for a cycle is inversely proportional to f so that the total light emitted per unit time is independent of frequency which is contrary to the observation.

Zalm considerd that the actual voltage across the EL cell is governed by extra capacitance and resistance present in the experimental cell. Thus for a series of capacitor of value C and the resistance R of EL cell, the actual voltage across the cell may be given

$$WRC\, V_o / \sqrt{1+ \omega^2 R^2 C^2} \qquad \ldots\ldots\ldots (5.21)$$

Where Vo is amplitude of the applied voltage to the whole system of the capacitor and EL cell.

If $Rc \gg \frac{1}{\omega}$, the voltage across the cell and concequently, the luminous emittance is independent of frequency, while with $RC \ll \frac{1}{\omega}$, the emittance increases more linearly with the frequency. It is thus possible to describe any frequency dependence with a suitable distribution in RC values of the system.

Johnson, Piper and Willians (32) deduced the brightness frequency relation by considering both excitation and recombination process. Johnson etal found

$$B \propto 1 - a \left[1 - \exp.\left(1 - \tfrac{1}{2} f T_2 \right) \exp.\left(- \tfrac{1}{2} f T_1 \right) \right] \quad \ldots.(5.22)$$

Where a is a constant, T_1 is the relaxation time of the excited electrons returning to empty luminescence centres and T_2 is the time constant of creation of primary electrons.

Chan and Yu (26) have empirically suggested that the EL emittance (B) varies with frequency (f) of applied sinusoidal field as

$$B = B_s - B_m \exp\left(- f / f_c \right) \quad \ldots\ldots (5.23)$$

Where B_s B_m and f_c are constants (B_s , B_m). The equation indicates that B approches a saturation value B_s when $f \gg f_c$ and a linear relation between B and f when $f \ll f_c$.

5.2:3 : Brightness Waves :

The variations in the EL brightness with time during the applied a.c. voltages are called as 'Brightness Waves ". Besides it is a graphic indication of Phase relationship of light component (applied to y plates) and applied voltage (to x plate) on a double beam cathode ray oscilloscope.

The shape of the brightness waves depends on the field intensity, frequency and the direction in which the light out put is observed. It is also depends on the nature of activators, and the spectral characteristics of the detector (PMtube). The existance of brightness waves and the fact that its frequency is twice the frequency of applied voltage were first reported by Destriau (33,34) in year 1937.

In general, a brightness wave exhibits two ' Primary ' peaks during each cycle of the applied voltage. Some phosphors, however, show additional much smaller or minor peaks referred as ' secondary ' peaks. The existance of secondary peaks is related to the electrons, which recombines with luminescence centre after a delay resulting from trapping (2,35) . The prominance of primary peaks than the secondary peaks at the applied voltages suggests that in the phosphor deeper trap levels are less in number. Moreover, absence of secondary peak suggests that the electron traps are located close to the luminescence centres (3,37,24).

5.2.4 : Efficiency of electroluminescence :

The lumineous efficiency (lumen/watt) of electroluminescence of powdered crystal phosphor embeded in an insulating medium and excited by alternating fields, depends on the quality of the phosphor, the EL cell design and conditions of excitation.

Lehmann (38) has given an empirical relation for efficiency of an EL phosphor namely

$$\eta^2 = C^2 \left(B/V^4 \right) \qquad \ldots\ldots (5.24)$$

Where C is a constant. According to this equation efficiency increases so long as the brightness increases faster than the fourth power of the applied voltage. The efficiency is seen to be approximately independent of frequency of excitation but depend on the magnitude of the applied voltage.

The simple calculations of luminescence efficiency η_0 , in the terms of brightness units (arbitrary), as a function of applied voltage and frequency are possible using Lehmann's (38) relation, viz

$$\eta_0 = \frac{B}{W} = \frac{B/f}{av^2 \left[K_o'' + b \left(B/f \right)^{1/2} \right]} \qquad \ldots (5.25)$$

Where K_o, a and b are constants. The values of a and b are to be taken from the work of Schwertz et al (39) while K_o'' is taken to be unity.

5.3 : RESULT AND DISCUSSION:

EL studies are carried out in the voltage range from 0 to 900 volts (r.m.s.) and frequency range from 500 Hz to 9 KHz.

5.3.1 : The voltage dependence of EL brightness :

The plots between EL brightness (in arb units) and applied voltages at different frequencies for typical sample are shown in fig (5.1). From these it suggests that there is no linear relation between applied voltage and EL brightness. It is also observed that the threshold voltage (the voltage above which EL brightness start) decreases with increase in frequency and it veries between (370 V r.m.s. and 615 V rms) Table(5.1) However the variation of threshold voltage with activator concentration is not symmetric except for the sample in which

Threshold voltage may be higher for sample KD_1, & KD_4 (Cas : Dy).

The nature of possible relationship existing between brightness and applied voltage is examined by plotting. (i) $\log\left(\frac{B}{V}\right)$ Vs $1/V$, (ii) $\log\left(\frac{B}{V}\right)$ Vs $1/\sqrt{V}$, (iii) log B Vs V (iv) log B Vs $1/V$, (v) log B Vs $1/\sqrt{V}$ It is observed that none of these plots (fig. 5.2 - 5.6) show a definite linearity, indicating that the observed brightness voltage dependence is not exponential. However the plots between log B and log V fig. (5.7) show a linear relation with slight deviation at higher and vary low voltages, some typical plots log B Vs log V are shown in fig. (5.8 - 5.11). From these graphs the plots are closely linear showing a bending towards the voltage axis at high voltage. This indicates that saturation of brightness occurs at high voltage. How ever for 1 KH_z log B - log V relation may be holding good among phosphors studied within the limitted range of voltage.

The linearity between log B and log V suggests that the brightness and voltage follow the power low relation of the form (23)

$$B = av^n$$

a and n being constant to exist over the observed region of linearity. The relation is similar to equation (5.11) with $V_\sigma = 0$ similar, relation is observed by Gour (40) for Cas:Ag phosphors Lawangar et al (24) for CaS: Bi : Pd phosphors, and by Patil M.G. (41) for Cas:Ag:Dy phosphors. The values of n are calculated from the slopes of the (log B - log V) plots and are listed in table (5.2).

The bending of plots at high voltages is also observed by Patil M.G.(41). It might be due to one or more reason amongst (i) the loss of voltage in conducting electrodes, there by lowering the field strength at high voltages (ii) the trap can not be emptied as fast as they are filled at high frequencies and give rise a space charge scattering and hence an increase in relation less transitions at higher fields (iii) the distribution of applied a. c. voltage to the microcrystalline powder form phosphors. How ever second reasoning given above seems to be more probable.

5.3.2 Frequency dependence of EL brightness :

A brightness is also found to be a function of frequency of applied voltage. Typical graphs between log B and log f are shown in fig. (5.12 - 7.17) the result obtained can be summarised as (i) At low frequency the brightness increases almost lenearly (ii) The frequency range over which the linear relationship hold depends on the exciting voltage. At higher voltages brightness increases more rapidly than at lower voltages. (iii) At moderately higher frequencies the brightness increases slowly and finally it saturates. (How ever the sample KD 31 fig (5.17) does not behave as described above, brightness decreases for higher frequencies).

The above said behaviour can be understood on the basis of the equation given by Curie (30) Viz.

$$B = B_0 \frac{n_0^2 \cdot \alpha}{1+ (n_0 \alpha / 2f)} \qquad \ldots(5.26)$$

Where α is the time constant, n_0 being initial concentration of free electrons in the conduction band f is the frequency of applied alternating field and B_0 is propertionality constant.

At lower frequencies, the life time of the excited centres is short compared to the peridicity of applied voltage and factor α becomes large compared to f. Under these conditions the equation (5.26) takes the form $B = B_0 n_0^2 f$ which explains the observed linear behavior.

At moderately high frequencies the term $n_0 \alpha / 2f$ becomes much smaller than unity and equation (5.26) reducess to $B = B_0 n_0^2 \alpha$ The expression is independent of frequency and point the saturation of E.L. brightness.

5.3.3 Relative efficiency :

Fig. 5.18 (a) depicts the variation of brightness (B) with power (W = V r.m.s. X device current) supplied to EL cell. It may be be seen that the brightness increases with supply of energy and then saturates. This is expected since there is an increase in radiationless transitions at high voltages.

The ratio B/w terms as relative efficiency (in arb units) is plotted as function of W and it shows that with increase in energy the efficiency increases, goes through maximum and then decreases fig. 5.18 (b). This finding is Similar as the findings of Kulkarni, Ambardekar (42) Bhand (43).

5.3.4 : Brightness waves :

Fig. (5.19) shows typical brightness wave pattern obtained by varying either the voltage or the frequency. It may be seen that the brightness waves exibits two peaks per cycle of the applied voltage . According to Waymouth and Bitter (44) the light-emission from single grain is concentrated in certain small spots and these spots emit brightness waves. On this basis the occurance of brightness waves in the present phosphor can be interpreted by assuming spots themselves to be minute rectifying junctions (8). These junctions can be oriented in any direction. The excitation will occur in first half cycle and in other during next half cycle giving two peaks in one cycle.
It may be observed from fig.(5.19) that the observed peaks are not of equal amplitude. This type of asymmetry was observed by Zalm et al and by Lawangar et al (24) and they concluded that this lack of symmetry may be due to the emission of light at

the side facing to the cathode. The observed asymmetry may also be due to the self - absorption of light or due to scattering of light by inter adjescent layers of powder or by individual particles. Thus there seems to be no single reason for the observed asymmetry.

The brightness waves do not show the secondary peaks. This indicates that the electron traps are located close to luminescent centre.

5.3.5: Effect of activators on electroluminescence behavior :

It may be seen from the fig. (5.1 - 5.5) that for various samples with different concentrations of activators (Dy:Tb) the nature of voltage and frequency dependance of brightness is almost similar. The activators have only the effect of modifying the relative intensities in unsymmetric way.

5.3.6 : Mechanism of electroluminescence :

To observe. EL emission comparatively high fields are required for all samples studied the threshold voltage is found to be grater than 370 V rms. This requirement of an intense electric field for EL emission along with the existance of power low relationship between brightness and voltage suggests that the probable mechanism of EL in these phosphors is likely to be the direct field ionisation of either Valence Band electrons or impurity centres with the transfor of electrons in to the conduction band takes place by a quantum mechanical tunneling process (40,45).

5.3.7 : Kinetics involved in EL process :

In present investigations,the existance of power law relationship between brightness and voltage indicates the bimolecular process of recombination of electrons with luminescence centres (23) Moreever, variation of brightness with frequency as per equation (5.16) given by D.Curie also exhibits bimolecular process.

5.4: SUMMARY :-

The important features of this chapter can be summarised as follows.

1) The EL brightness (B) and voltage V follow power law relation over a wide range of voltage.

2) At lower frequencies the brightness increases with frequency almost linearly. At higher frequencies brightness saturates.

3) The activators (Dy : Tb) have no significant effect on the EL behavior of phosphors but they modify the relative intensities only.

4) The probable machanism of EL in these phosphors is likely to be the direct field ionisation of either velence band electron or impurity centre. Where the transfer of electron into conduction band takes place by quantum mechanical tunneling process.

5) Kinetics involved in EL process is likely to be bimolecular.

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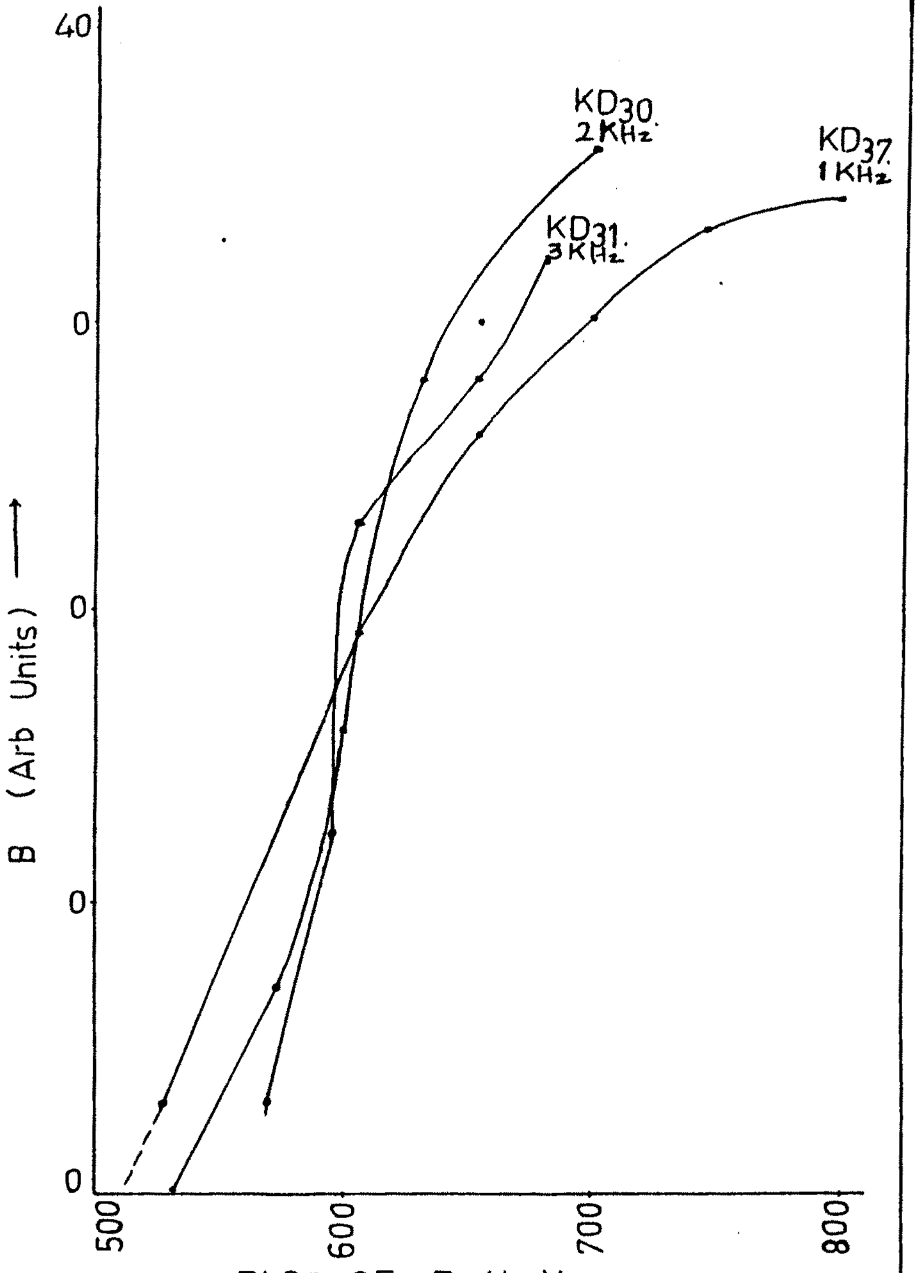
-x-x-x-x-x-x-

TABLE 5.1 : VALUES OF THRESHOLD VOLTAGES FOR DIFERENT SAMPLES AT DIFFERENT FREQUENCIES (at 300^{o}k)

Sample No.	Frequency in H_z						
	100	500	1 K	3 K	5 K	7 K	9 K
KD 1	700	572	530	465	445	415	315
KD 4	635	560	530	475	445	435	424
KD 29	-	550	509	500	530	550	595
KD 30	520	515	510	500	495	485	475
KD 31	595	572	551	495	465	455	445
KD 37	550	530	490	450	425	400	390
KD 39	560	530	485	445	425	400	370
KD 40	615	583	550	465	445	425	415

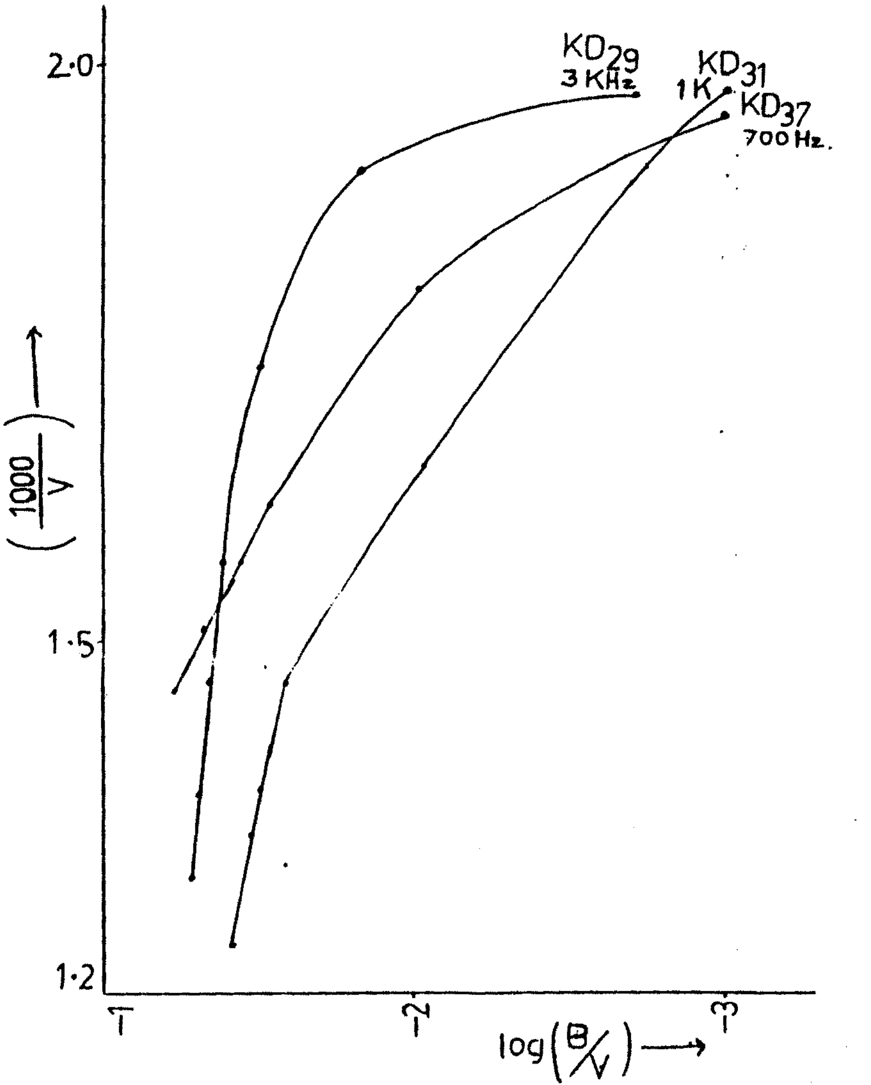
TABLE 5.2 : VALUES OF 'n, FOR DIFFERENT SAMPLES FOR DIFFERENT FREQUENCY :

Sample No.	'n, value for frequency				
	1 KH_z	3 KH_z	5 KH_z	7 KH_z	9 KH_z
KD 1	7.00	3.45	3.5	3.2	-
KD 4	7.00	3.45	4.6	4	-
KD 29	7.1	7.1	6	8 K	-
KD 30	9.6	-	-	-	-
KD 31	6.3	3.1	3.4	2.8	2.1
KD 37	2.7	3.1	2.3	2	-
KD 39	8.6	4	2.8	3.4	3.2
KD 40	8.6	3.8	2	3.8	3.4



PLOT OF B Vs V

FIG. NO.: 5·1.



PLOT OF log(B/V) Vs (1000/V) → FIG. NO: 5·2

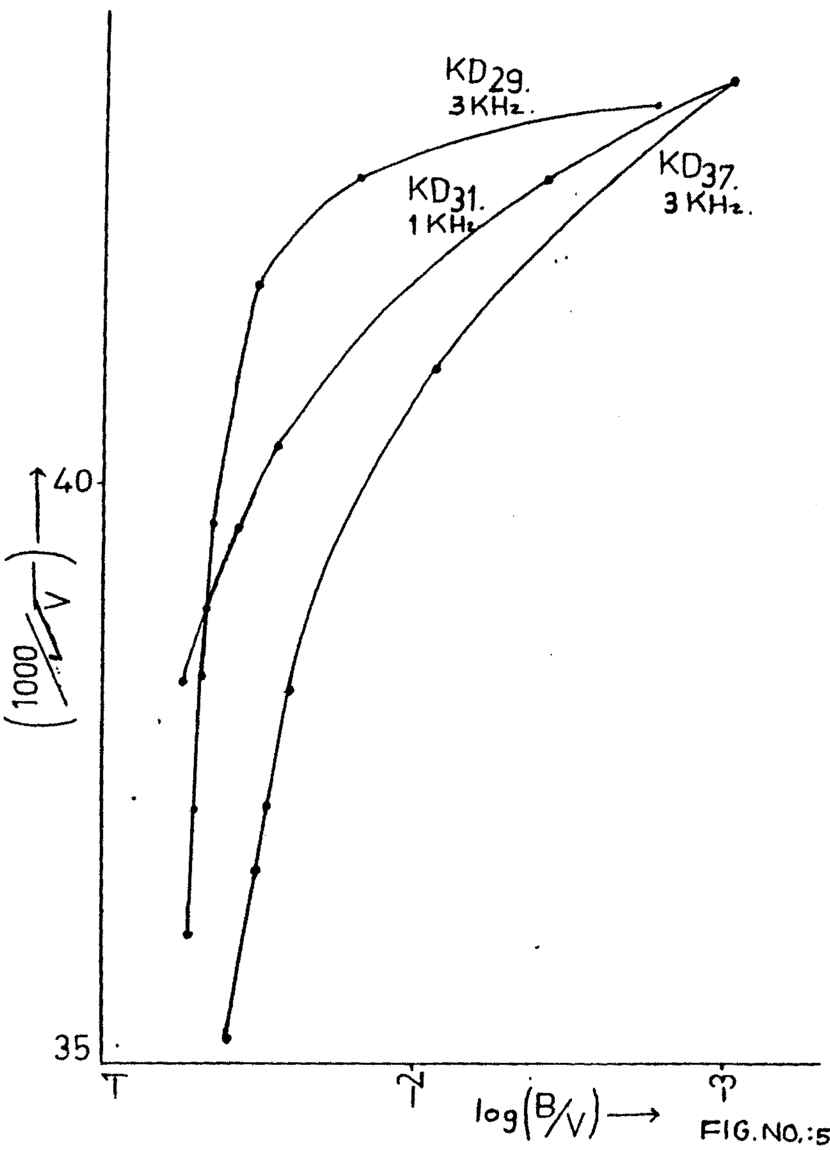
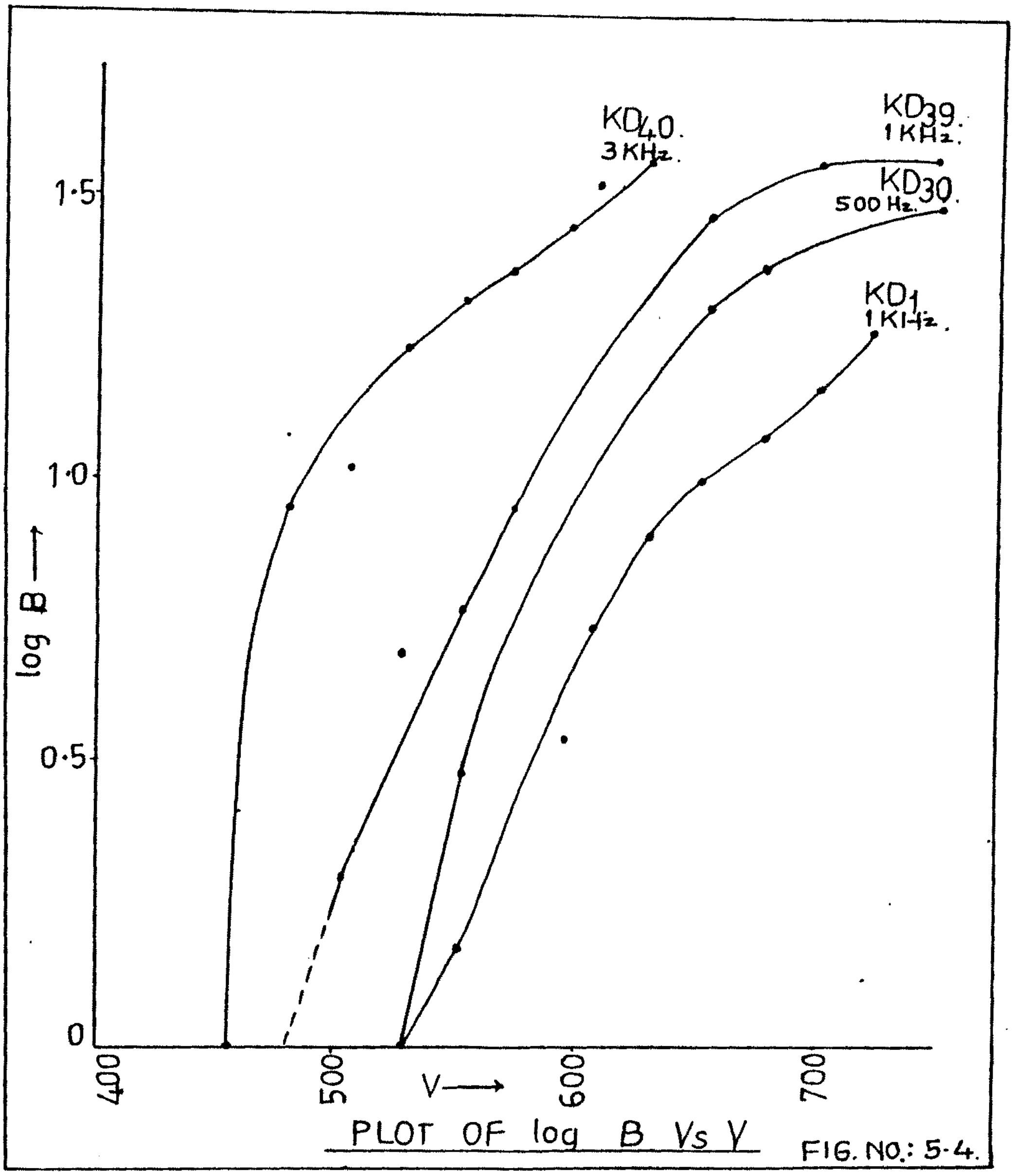


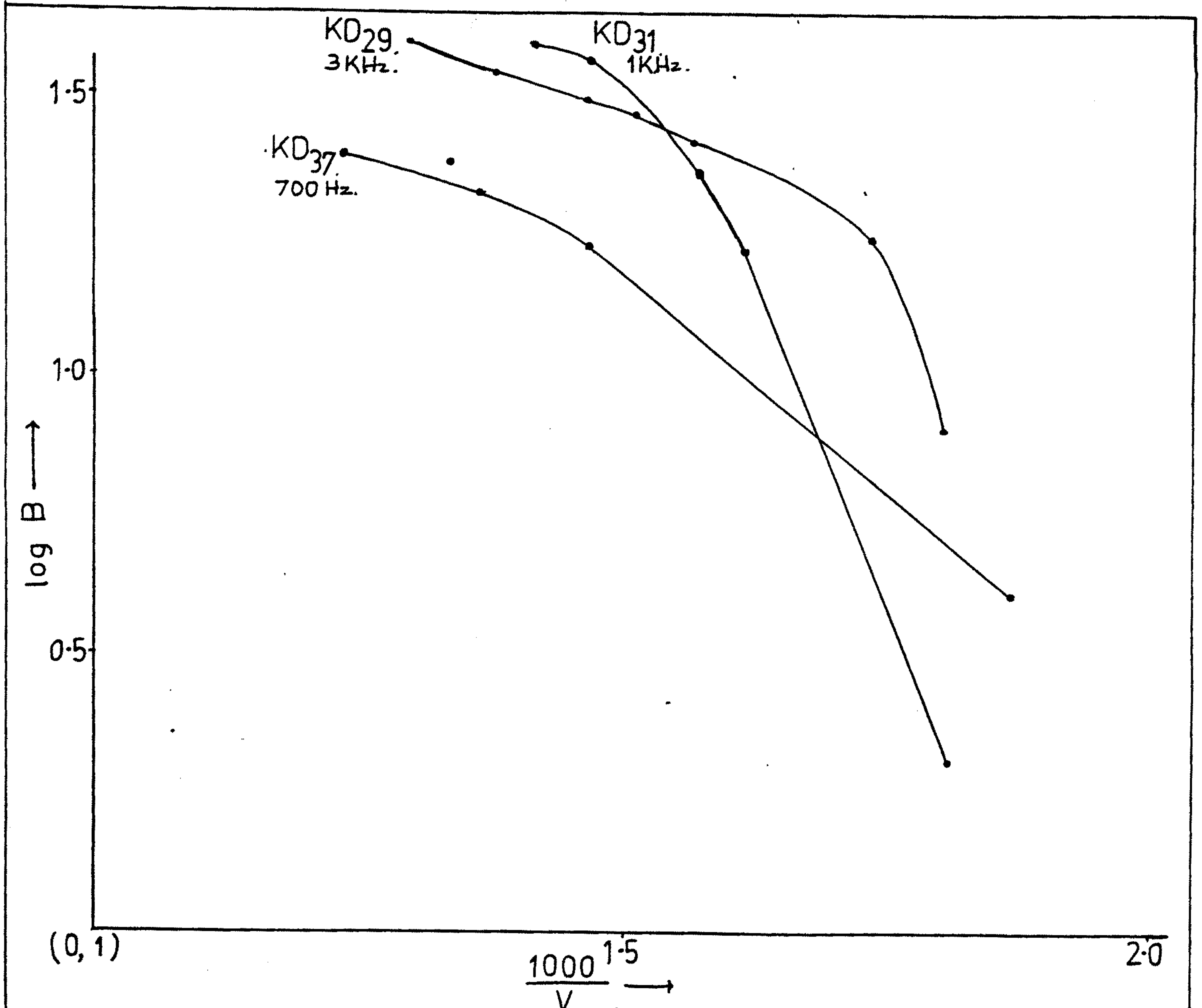
FIG. NO.: 5-3.

PLOT OF $\log(B/V)$ Vs $\left(\frac{1000}{\sqrt{V}}\right)$



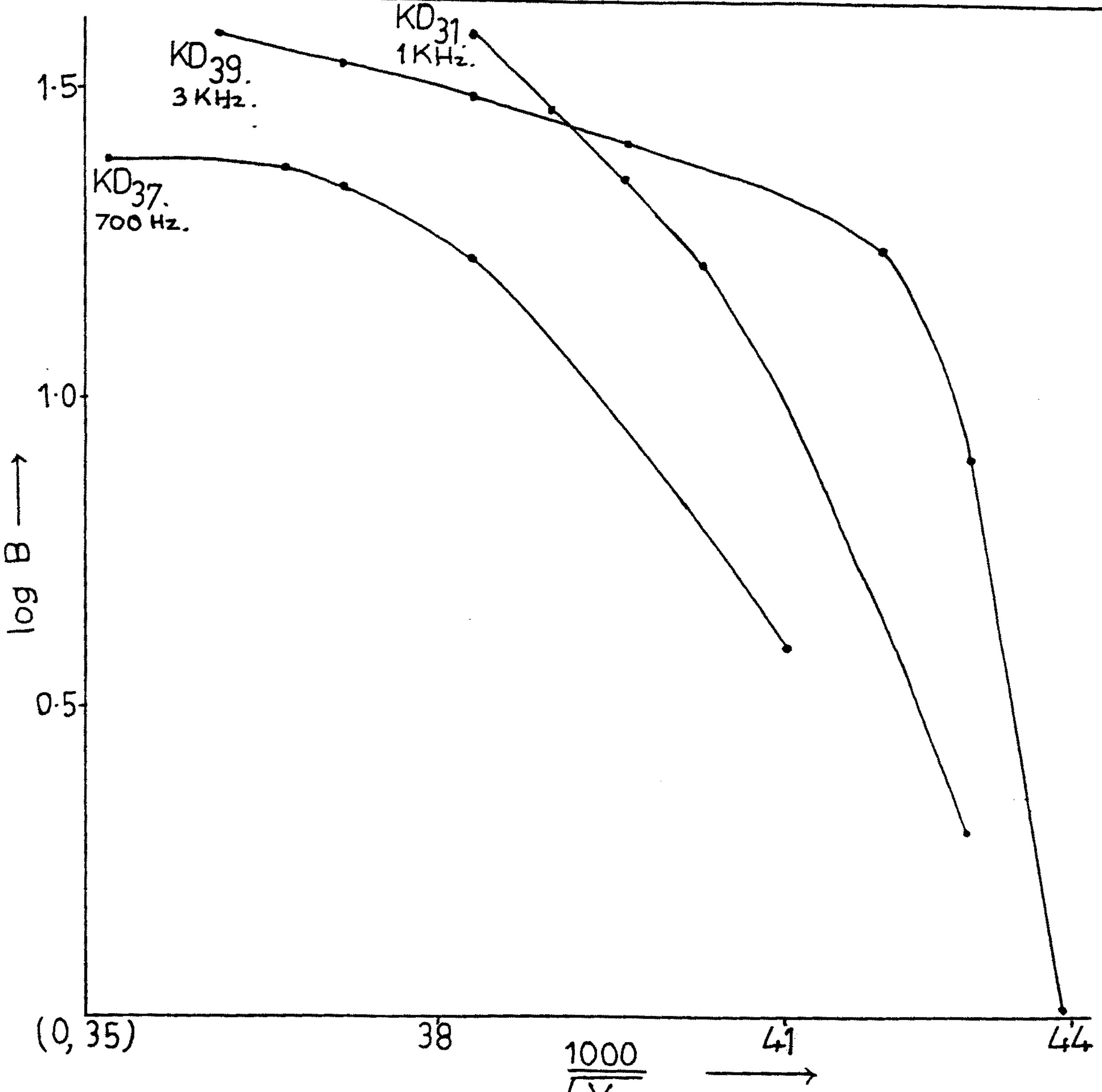
PLOT OF log B Vs V

FIG. NO.: 5·4.



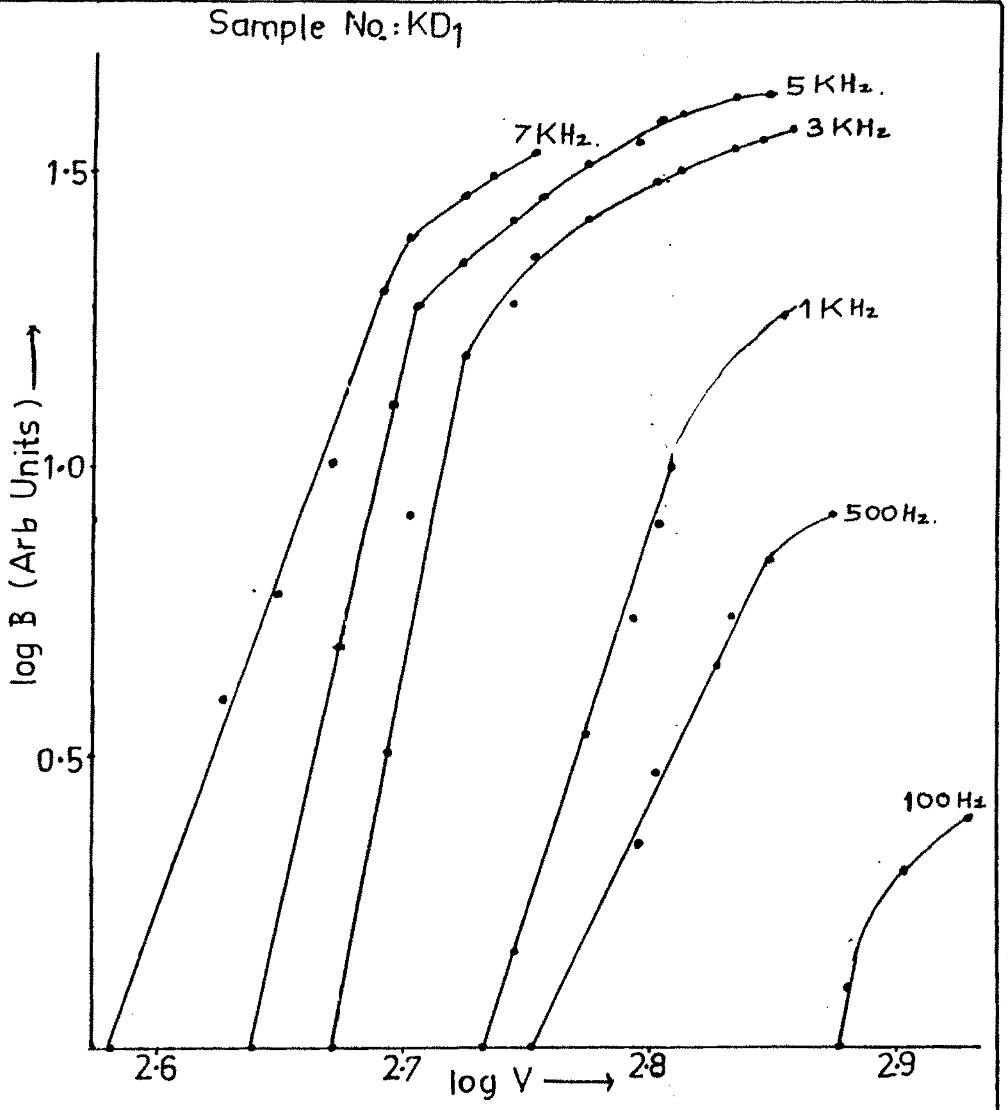
PLOT OF log B Vs $(1000/V)$ ⟶

FIG. NO.: 5.5.



PLOT OF log B Vs $(1000/\sqrt{V})$ ⟶

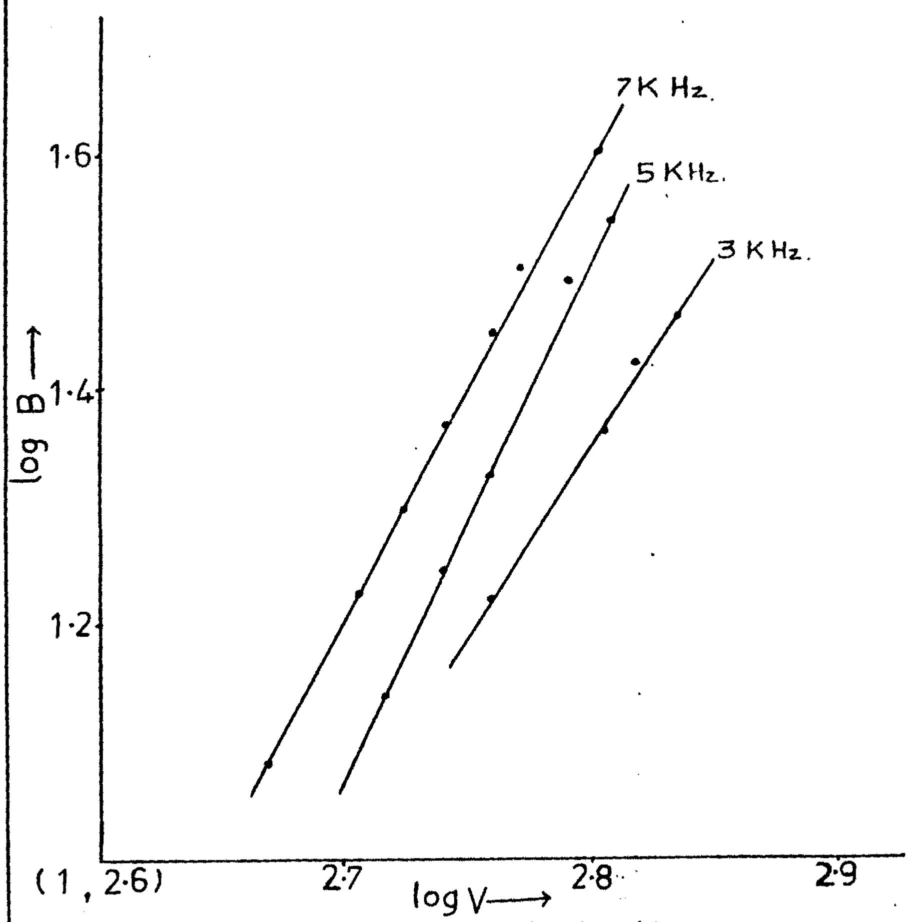
FIG. NO.: 5·6.



PLOT OF log B Vs log V

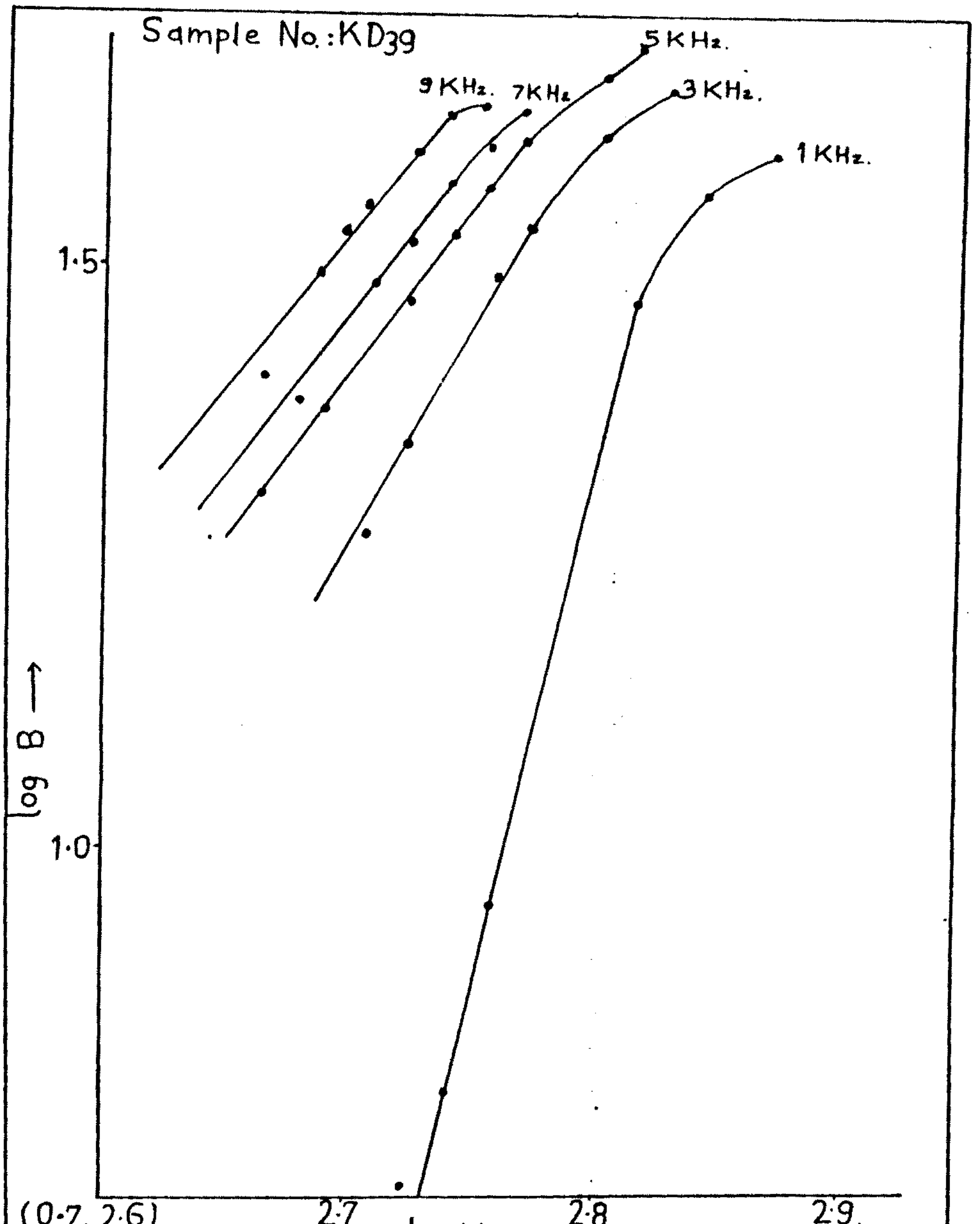
FIG.NO.: 5·7.

Sample No.: KD_4

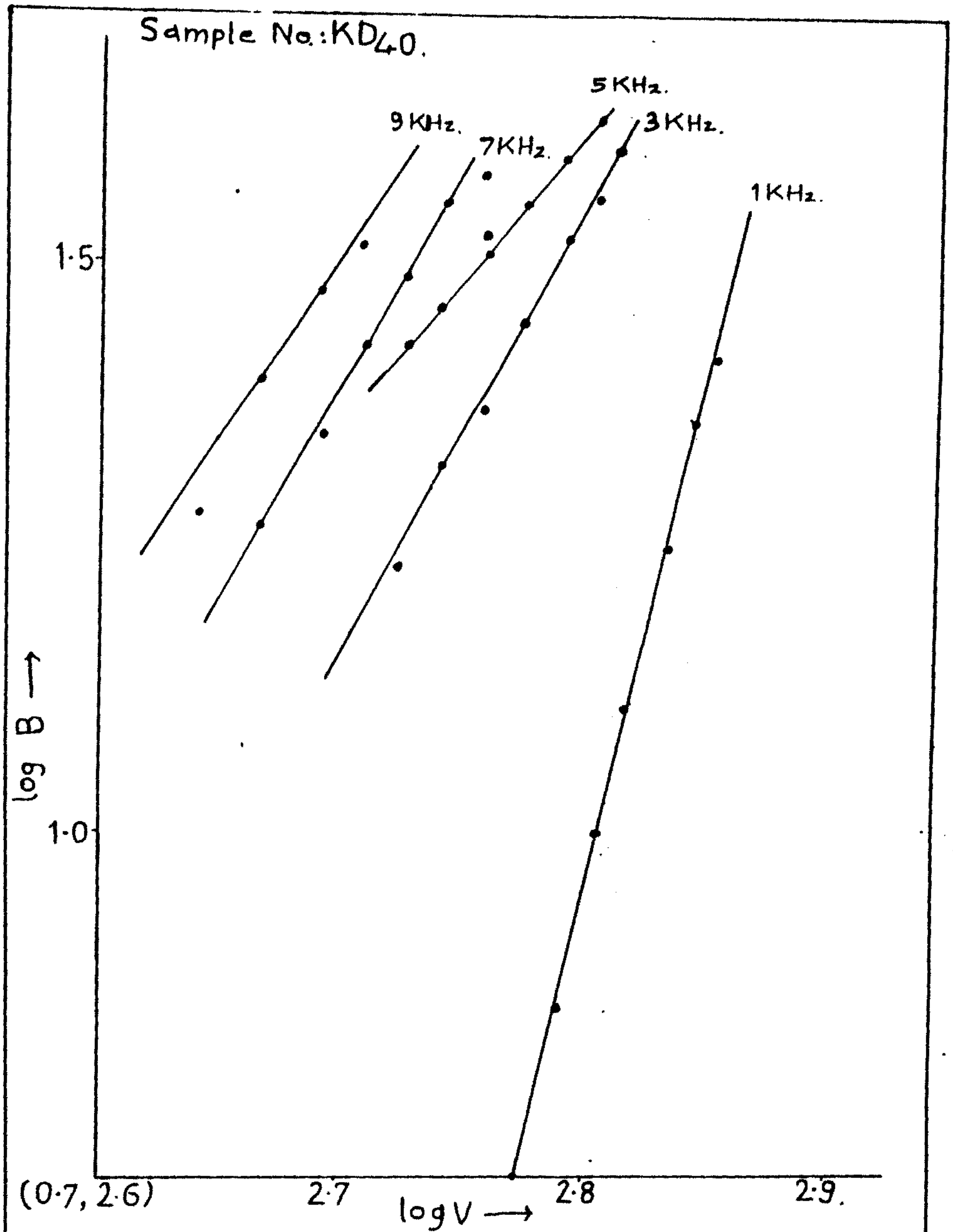


PLOT OF log B Vs log V

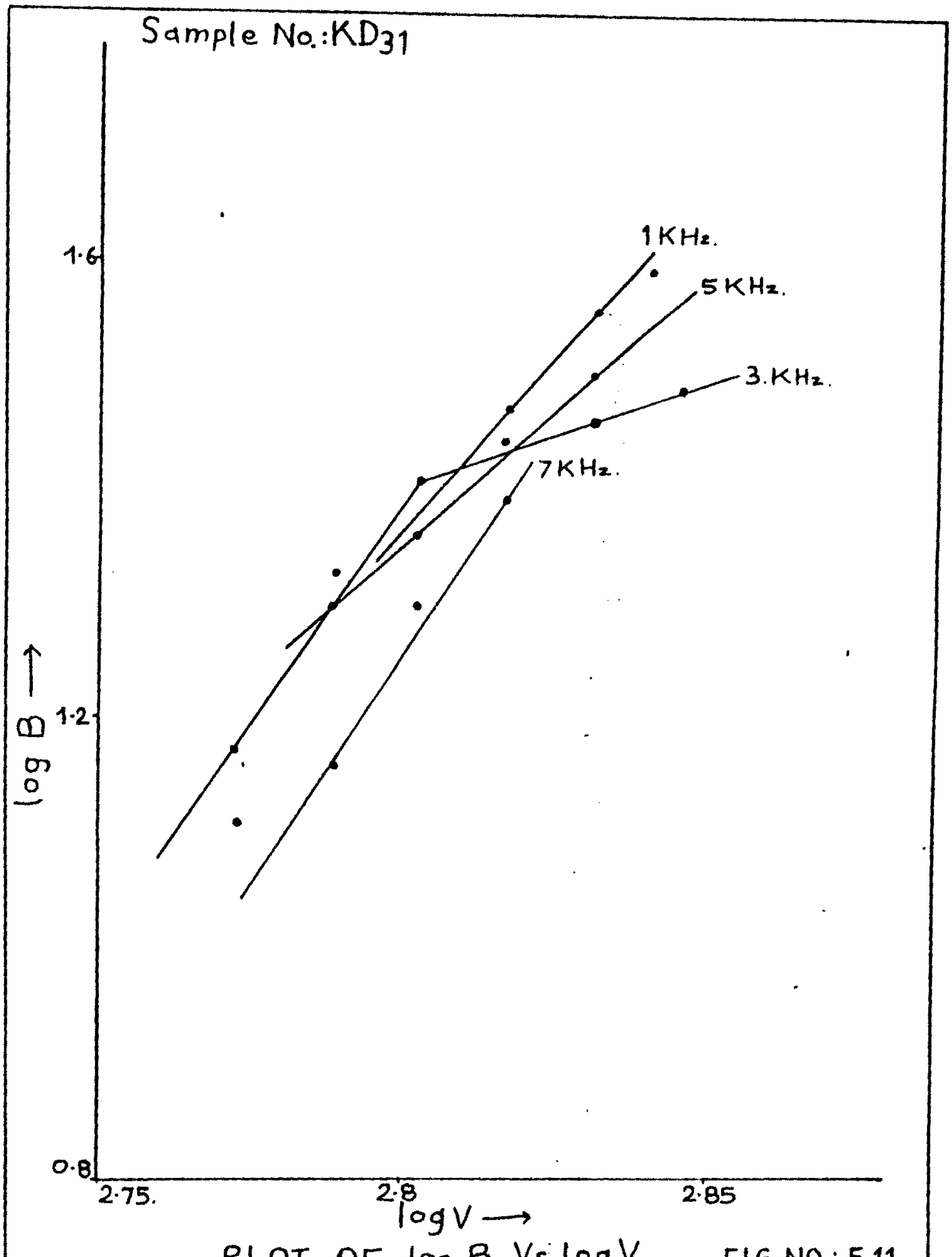
FIG. NO.: 5·8.



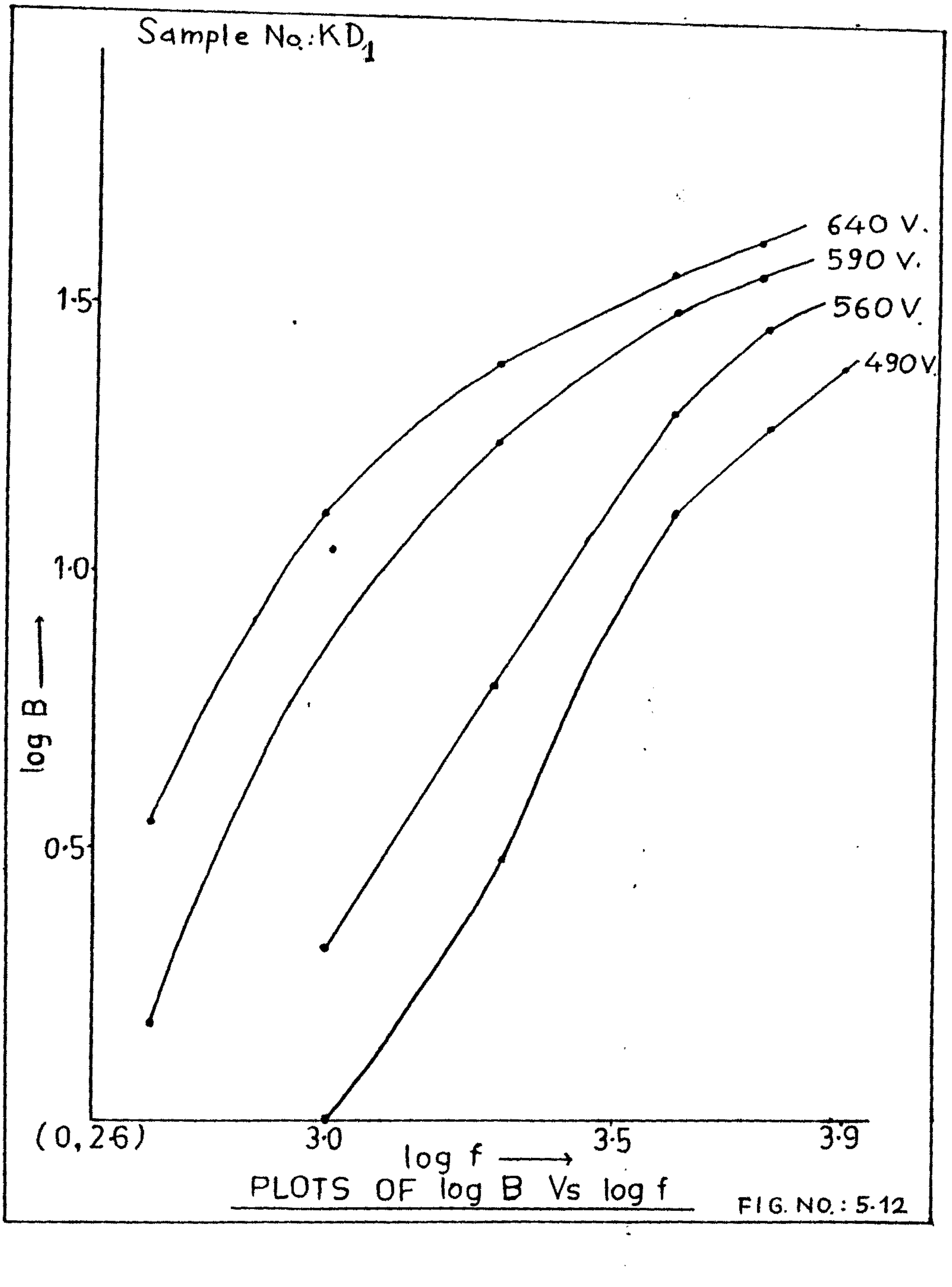
PLOT OF log B Vs log V FIG. NO.: 5·9.



PLOT OF log B Vs log V FIG. NO.: 5·10.

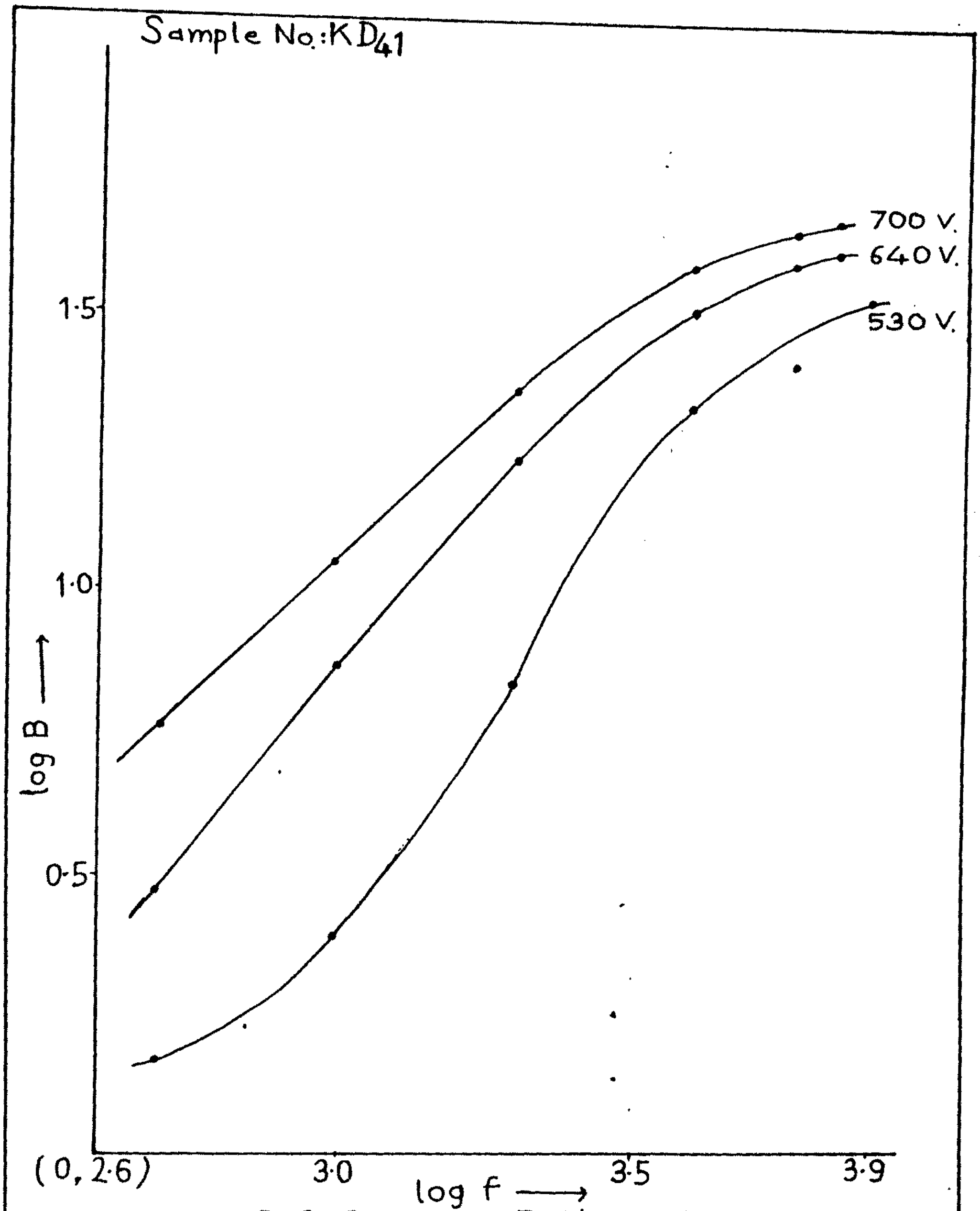


PLOT OF log B Vs log V FIG. NO: 5·11.



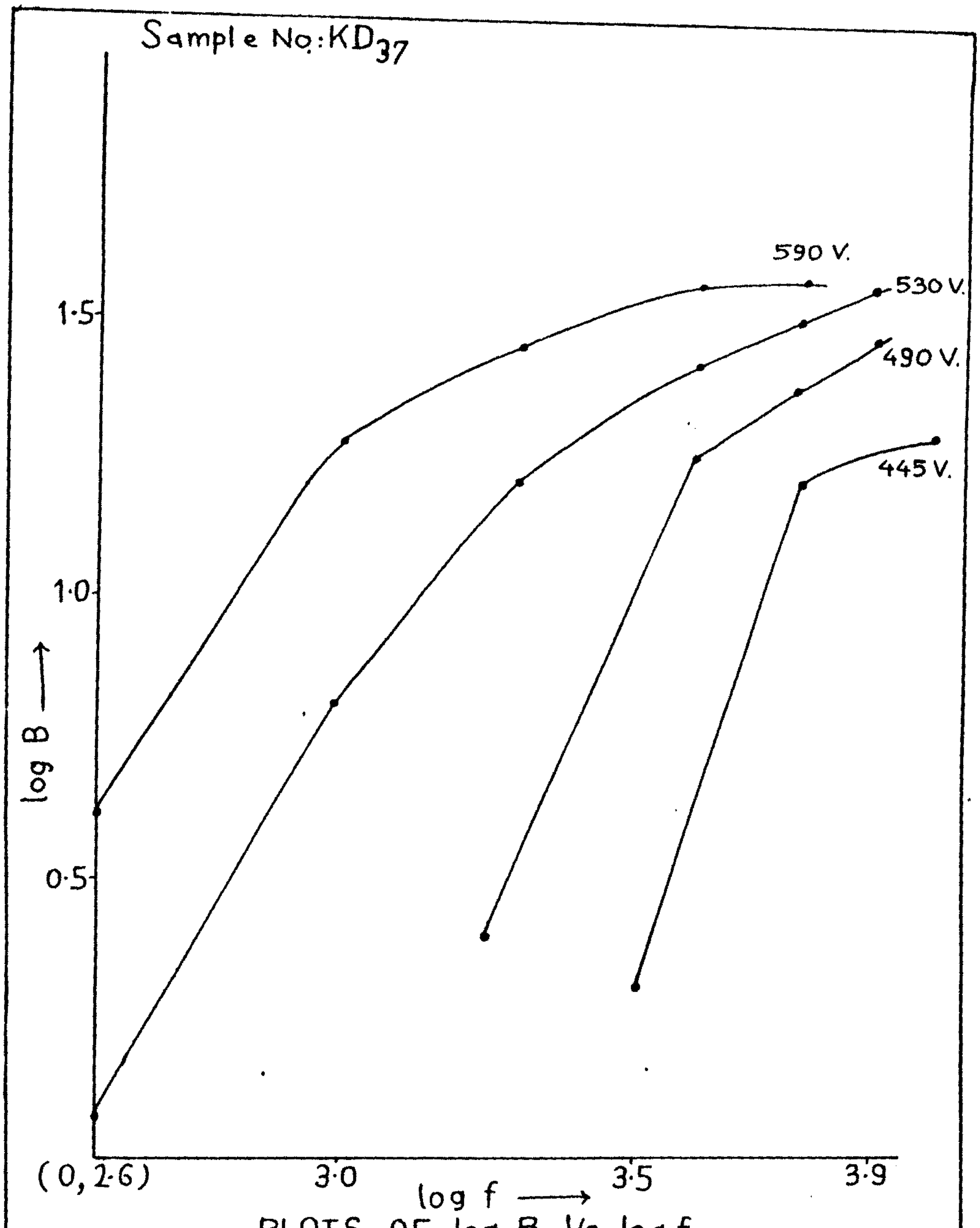
PLOTS OF log B Vs log f

FIG. NO.: 5·12



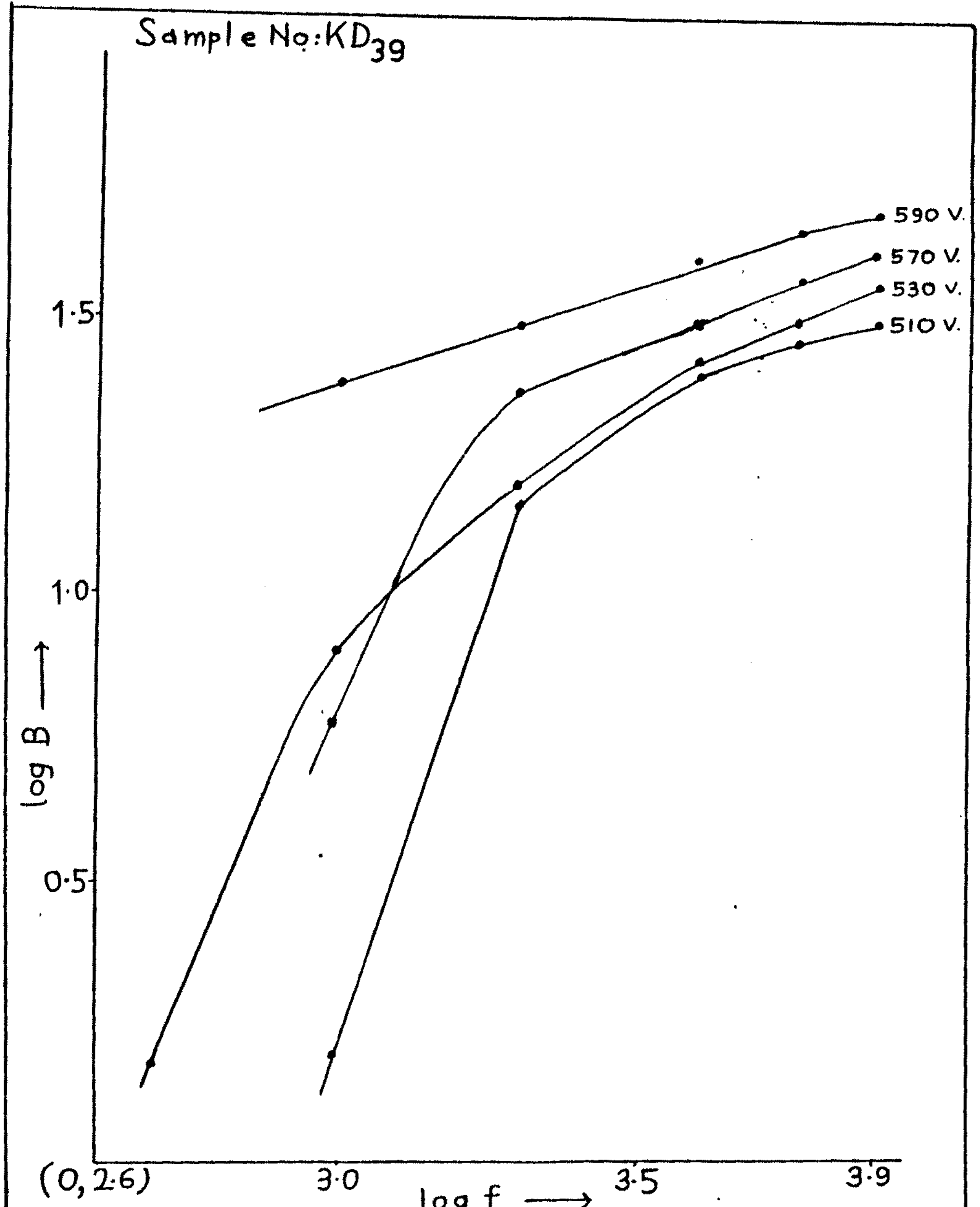
PLOTS OF log B Vs log f

FIG. NO.: 5-13.



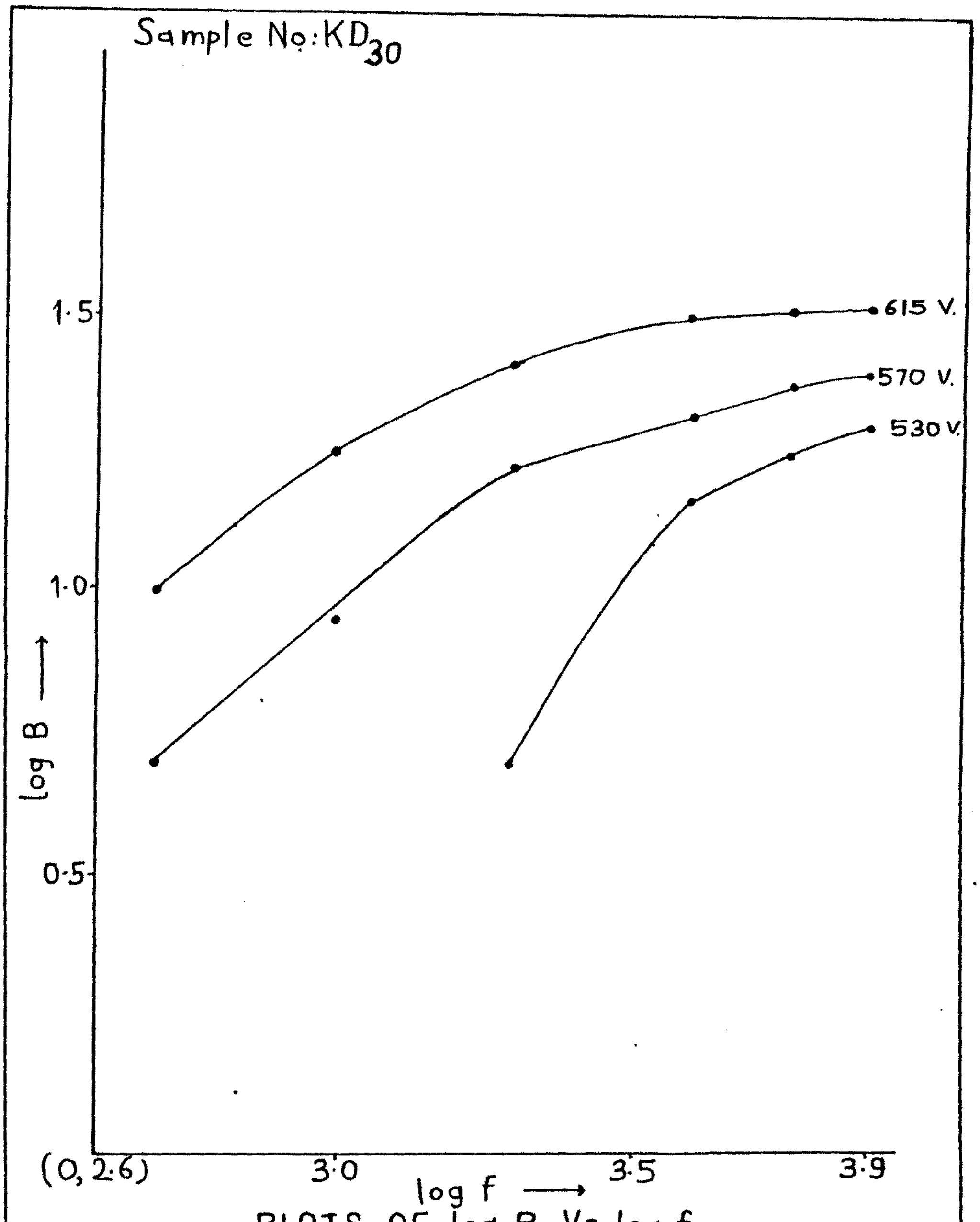
PLOTS OF log B Vs log f

FIG NO.: 5·14.



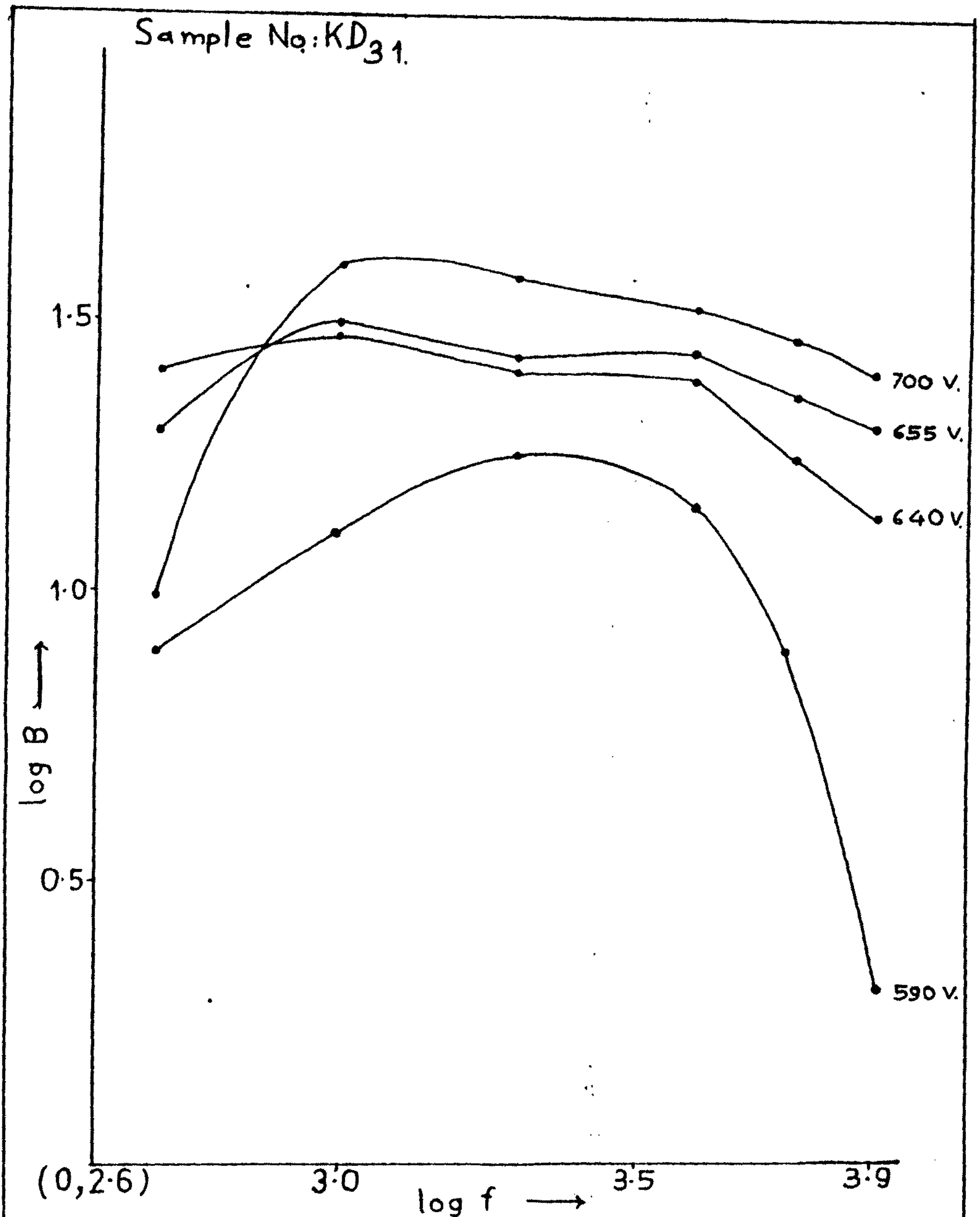
PLOTS OF log B Vs log f.

FIG. NO.: 5·15.



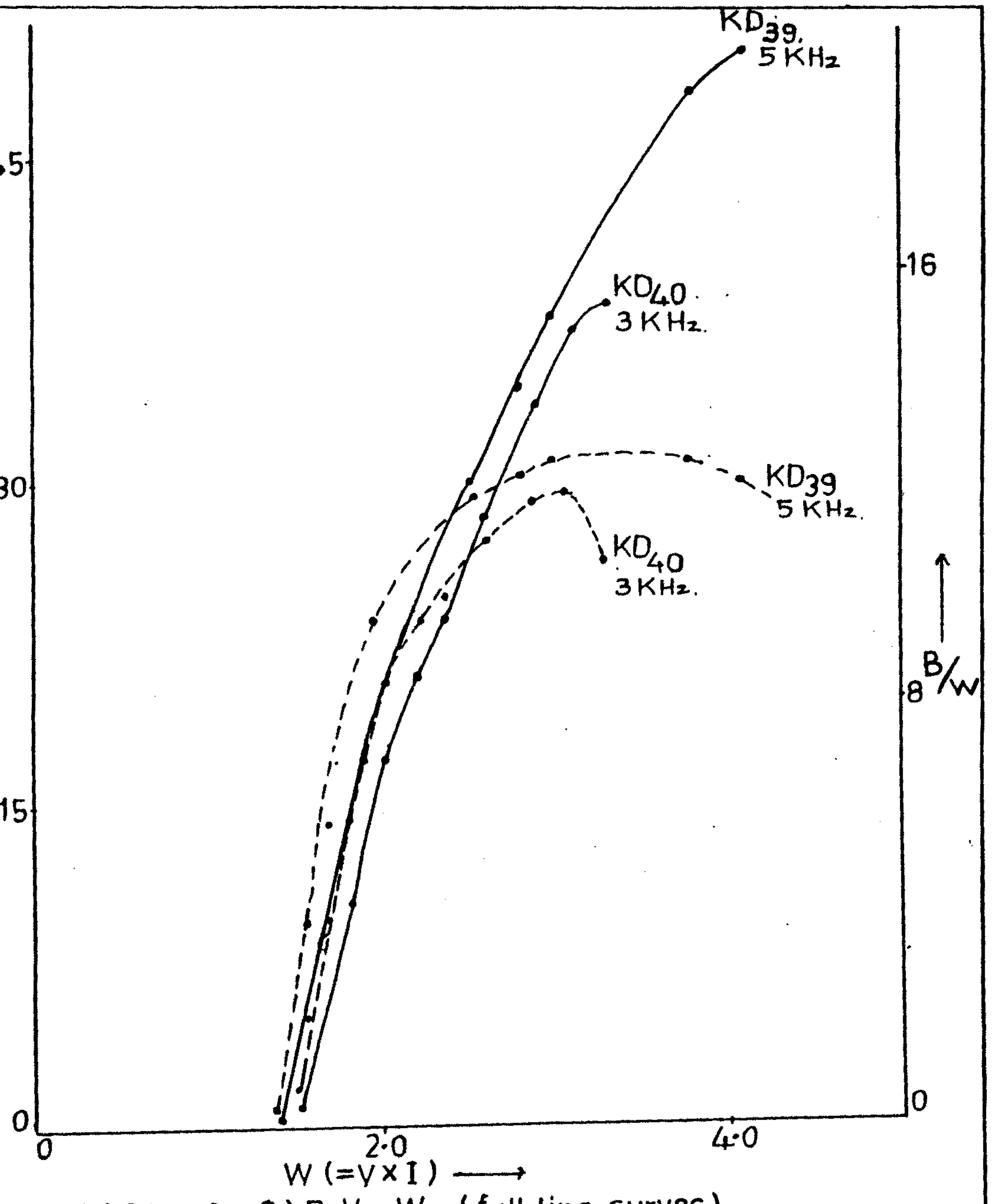
PLOTS OF log B Vs log f

FIG. NO. : 5·16.

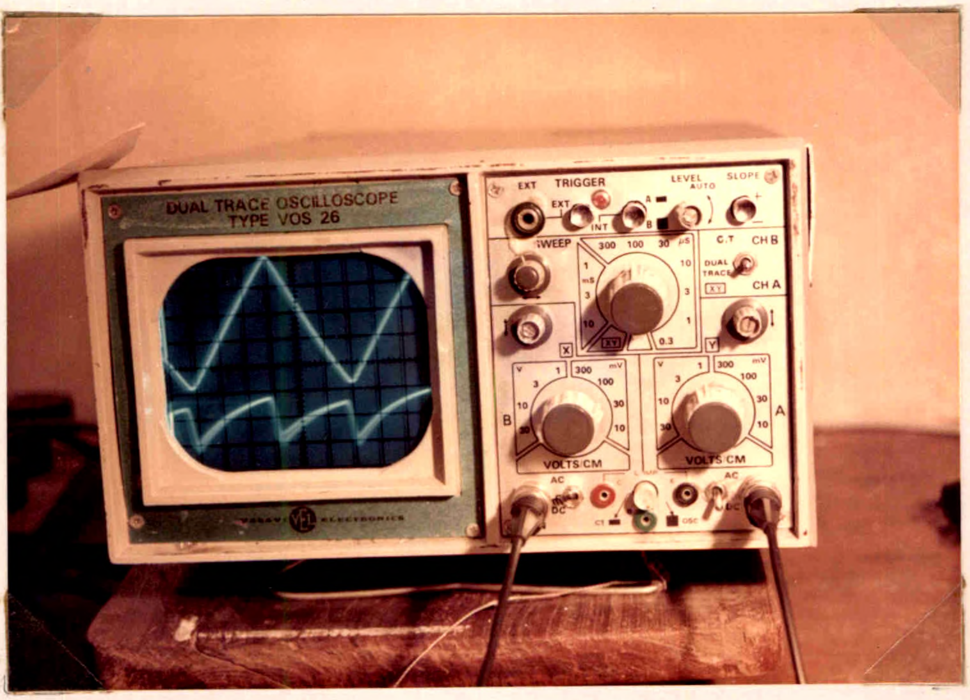


PLOTS OF log B Vs log f

FIG. No.: 5·17.



PLOTS OF a) B Vs W (full line curves)
b) B/W Vs W (dotted line curves) FIG. NO.: 5·18



BRIGHTNESS WAVE FORM

Frequency 2·2 KHz (Sample. No.: KD_{30})

Fig No.: 5·19.