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(Ph. D. Thesis)

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ABSTRACT

We have observed standing spin wave resonance in evaporated films of 81-19 permalloy and pure nickel. The permalloy measurements were made in a room temperature microwave transmission spectrometer at 14.5 Gc/sec. In this system we observed not only excitation of unpinned spin wave modes but also a previously unreported interference effect between the propagating electromagnetic and spin wave modes in the film. A 15.1 Gc/sec reflection spectrometer was used to measure the temperature dependence of the exchange stiffness of nickel. We have extended the temperature range of these measurements up to 540°K, which is in excess of 85% of the Curie temperature of nickel, $T_c = 627^\circ\text{K}$. Above $.8 T_c$ the exchange stiffness appears to vary directly as the magnetization.

I. INTRODUCTION

At present, there is no theoretical model which adequately describes the magnetic behavior of the ferromagnetic 3d transition metals. Usually one of two limiting cases is considered: either the Heisenberg model¹ in which the 3d electrons are localized at each atomic site, or the band model² where the electrons are itinerant and to first approximation are described by plane wave states. In finding the true description, which probably lies somewhere between these two extreme cases, it is important to obtain precise experimental data on model-sensitive phenomena.

One source of such data is the direct measurement of the dispersion relation for the low-lying elementary excitations of the magnetic system (spin waves). The Heisenberg model³ predicts the well-known quadratic dispersion relation, $\omega = \omega_0 + D k^2$, for long wavelength spin waves. Kittel and Herring⁴⁻⁶ have shown by general arguments that this result should be independent of the model in the long wavelength limit ($ka \ll 1$, where "a" is a lattice constant). This has been independently confirmed on a theoretical basis by Izuyama and Kubo^{7,8} for the itinerant model. For this reason, the measurement of the exchange stiffness, D , at a single temperature offers no fundamentally interesting physical information. However, the temperature dependence of D is model dependent. For localized spins, the exchange stiffness is reduced by spin wave-spin wave interactions, which lead to the low temperature expression⁹ $D(T) = D(0)(1-bT^{5/2})$. In the band picture, interaction of spin waves with single particle excitations gives a leading term proportional to T^2 in the renormalization of D , $D(T) = D(0)(1-cT^2)$.

The two experimental techniques used to measure θ are standing spin wave resonance in evaporated thin films¹⁰ and inelastic scattering of thermal neutrons.¹¹ The techniques are complementary in the range of k over which spin waves can be observed. The upper limit for resonance,¹² $k \approx .03 \text{ \AA}^{-1}$, is just the lower limit for neutrons.¹¹ Although the resonance experiments have much greater resolution (better than .1% for well resolved modes compared to 2-3% for neutron experiments), the properties of evaporated films can deviate markedly from those of pure bulk samples.¹³ Therefore, careful analysis of film quality is mandatory in interpreting resonance results.¹⁴

Because 81-19 permalloy is particularly well suited for evaporation of magnetic films,¹⁵ many of the standing spin wave experiments have been performed on this alloy. However, theoretical analysis is much simpler for pure metals than for alloys. For this reason, Phillips and Rosenberg^{14,16} have measured the exchange stiffness in thin films of Fe, Ni, and Co from 4° to 295°K. In all three cases, the observed variation in θ was much too large to be explained on the basis of a localized spin model. They found that the predictions of Izuyama and Kubo⁸ for the itinerant model gave the right order of magnitude for the observed effect.

We have used the standing spin wave resonance technique to study the temperature dependence of θ in pure nickel films over the extended temperature range 120° to 540°K. The upper limit is in excess of .85 T_c , where the Curie temperature of nickel, T_c , is 627°K. Below room temperature our results are in qualitative agreement with those obtained by Phillips and Rosenberg.¹⁶ Over the entire range we also find reasonable agreement

with the results obtained by Stringfellow¹⁷ using the less accurate method of small angle scattering of thermal neutrons. We have found that above $.8 T_c$ the exchange stiffness appears to vary directly as the magnetization. As discussed in Part III, this may indicate that at these elevated temperatures the effect of transverse spin correlation is negligible for the long wavelength ($k < .01 \text{ \AA}^{-1}$) spin waves which we excite.

We have also studied the room temperature resonance spectra of permalloy films in a microwave transmission spectrometer. In this system, in contrast to the standard reflection spectrometer,¹⁸ the exciting microwave field is not uniform in the film. We have confirmed that such a field will excite spin wave modes even in the absence of surface spin-pinning. This is important in light of recent experiments which indicate that surface pinning may be exceedingly small in high quality single crystal films.¹⁹ As mentioned above, the reliability of film measurements depends critically upon film quality. We have also observed an interference effect between the propagating electromagnetic and spin wave modes in the film. Analysis shows that this effect can give additional information about the variation of internal field in the film. Such information is useful in determining what parameters must be varied during evaporation to improve film quality.

II. MICROWAVE TRANSMISSION

A. Experiment

The samples studied by microwave transmission²⁰ were vacuum evaporated permalloy films (81% Ni, 19% Fe) ranging in thickness from 1000Å to 3000Å. This particular alloy composition is ideally suited for magnetic resonance studies of evaporated films because of its low anisotropy and zero magnetostriction.²¹

The films were deposited on unheated glass substrates at a pressure of approximately 3×10^{-6} Torr. The substrates were cut from microscope slides, cleaned with a liquid detergent, and rinsed in a distilled water bath. The final cleaning was done in the bell jar by positive ion bombardment. The source was an 81-19 permalloy rod, one end of which was heated by bombardment with electrostatically focused 4 KeV electrons. Best results were obtained for an evaporation rate of approximately 18Å/sec.

The film thickness and the evaporation rate were determined by measuring the frequency shift of a quartz oscillator placed near the substrate. The fractional uncertainty in the values obtained by this method is of the order of $\pm 10\%$. The problems inherent in this technique are discussed in Appendix I where the entire vacuum evaporation system is described in more detail.

Figure 1 is a schematic diagram of the transmission spectrometer used in this experiment. The films were mounted across a circular iris cut in the common side-wall of the two cavities, as shown in Fig. 2. The seal was made with conducting silver print to prevent power leakage

around the film. In order to compensate for the loading effect of the dielectric glass substrate, the resonance frequency of the second cavity was adjusted by means of a capacitive tuning screw.

The coupled cavity system has two normal modes of oscillation. Figure 3 shows the configuration of the microwave magnetic field, h , for the two modes. Placing a conducting film across the iris virtually eliminates any excitation of Mode 2. For Mode 1, the coupling was small enough that the cavities could be treated as independent resonant systems. This was experimentally verified by measuring both the reflected and transmitted power as a function of both the applied frequency and the resonant frequency of Cavity 2. The maximum power absorption by Cavity 1 occurred at 14.485 Gc/sec and was independent of the tuning of Cavity 2. The peak in the power transmitted from Cavity 2 varied in both frequency and intensity as a function of tuning but always exhibited an absolute maximum at 14.485 Gc/sec.

The fact that the resonant frequency of Cavity 1 was not pulled by tuning Cavity 2 implies that there was a negligible amount of power transmitted from Cavity 2 to Cavity 1. In this limit it is a good approximation to assume that the field transmitted through the film couples to an outgoing wave propagating in a medium with a characteristic impedance nearly equal to that of free space. This assumption is made in Section C, and the results predicted there are in good agreement with experimental spectra. Further experimental justification was obtained by removing the second cavity and coupling the transmitted power directly into a wave guide. In this case the approximation is certainly valid. Apart from a poorer signal-to-noise ratio, the spectrum was unchanged.

To obtain the spin wave spectrum of a given film, the klystron was locked to Cavity 1 by an AFC system, and Cavity 2 was tuned to give maximum power transmission. Straight dc detection was used to monitor both the power reflected from Cavity 1 and that transmitted from Cavity 2 as a function of the applied dc magnetic field.

The dc field, $\vec{H}$, was supplied by an electromagnet, as shown in Fig. 2. The magnet was rotatable about an axis parallel to the incoming wave guides. In this configuration, $\vec{H}$ was normal to the microwave magnetic field at the film, $\vec{h}$, for all magnet orientations. Spectra were recorded for a series of values of Φ , $0 \leq \Phi \leq \frac{\pi}{2}$, where Φ is defined as the angle between $\vec{H}$ and the film normal. The dc field was swept at a rate of 10 Oe/sec, and its absolute value was measured with a rotating coil gaussmeter.

Finally, we wished to compare our spectra with those obtained with a uniform exciting field. This was easily accomplished by interchanging the cavities and blanking off the coupling iris in the side-wall with a copper plug. The modified system is just a standard reflection spectrometer. Spectra were obtained with this system for the same range of Φ .

All experiments were performed at room temperature which was usually near 295°K.

B. Results

By exercising great care to avoid contamination during evaporation, we were able to prepare films comparable in quality to those produced by Nisenoff and Terhune²² using an "ultra-high" vacuum evaporator. In the transmission spectra of these samples, we observed not only excitation of unpinned spin wave modes but also an unexpected interference effect between the photon and spin wave modes in the film. In interpreting our experimental spectra we will find it helpful to compare our results with those predicted for an ideal film and those obtained in other experiments.

In the absence of magneto-crystalline anisotropy, the dispersion relation for a uniform film is given by²³

$$\left(\frac{\omega}{\gamma}\right)^2 = \left[|\vec{H}| \cos(\phi - \theta) - 4\pi |\vec{M}| \cos^2 \theta + \frac{\mathcal{D}}{\gamma} k^2 \right] \times \left[|\vec{H}| \cos(\phi - \theta) - 4\pi |\vec{M}| \cos^2 \theta + \frac{\mathcal{D}}{\gamma} k^2 \right] \quad (1)$$

where ϕ is defined as above, and θ is the angle between the magnetization, $\vec{M}$, and the film normal. In general the magnetization is not parallel to the applied field, and θ is a function of both $H = |\vec{H}|$ and ϕ . This can be seen by considering the magnetic energy density, E_M , in the system:²⁴

$$E_M = -\vec{H} \cdot \vec{M} + \frac{1}{2} (4\pi M^2 \cos^2 \theta) \quad (2)$$

The second term on the right arises from the demagnetizing field in the direction of the film normal. The equilibrium orientation of M , from $\partial E_M / \partial \theta = 0$, is determined by the relation

$$H \sin(\phi - \theta) + 2\pi M \sin 2\theta = 0 \quad (3)$$

The net result is that the magnetization changes direction as the dc field is swept, and the observed mode spacing is not quadratic in k . However, in the two limiting cases of $\phi = 0$ and $\phi = \pi/2$, the situation is greatly simplified.

At perpendicular field with $H > 4\pi M$, $\theta = \phi = 0$. Equation (1) reduces to the familiar resonance relation of the Kittel model:²⁵

$$H = \frac{\omega}{\gamma} + 4\pi M - \frac{\beta}{\gamma} k^2 \quad (4)$$

At parallel field, $\theta = \phi = \pi/2$, and the mode spacing has the same quadratic dependence on k :

$$H = 2\pi M \left\{ \left[\left(\frac{\omega}{2\pi\gamma M} \right)^2 + 1 \right]^{1/2} - 1 \right\} - \frac{\beta}{\gamma} k^2 \quad (5)$$

The possibility of exciting a mode depends critically on the boundary conditions on the transverse magnetization. If the surface spins are completely pinned, the resonance condition is given by $k = n\pi/L$ where n is an integer and L is the film thickness. Assuming the film normal is z directed, the transverse magnetization varies as $\sin(n\pi z/L)$, and the uniform ($n = 0$) mode cannot be excited. A uniform exciting field will only couple to modes with a net transverse magnetization, which corresponds to odd values of n in this case. The mode intensity decreases as $1/n^2$ and the spacing, $H_0 - H_n$, is proportional to n^2 for both $\phi = 0$ and $\phi = \pi/2$.²⁵ H_n is defined as the resonance field for the n th mode.

If the surface spins are completely free the transverse magnetization varies as $\cos n\pi z/L$, and the predicted mode spacing is the same as for the pinned case - however, a uniform field will couple only to the uniform mode. Finally, in the case that the surface spins are neither completely

pinned nor completely free, the transverse magnetization varies as

$$\alpha \sin kz + \beta \cos kz \quad (6)$$

The resonance condition is $k = \frac{n_{\text{eff}}\pi}{L}$, and the boundary conditions on the transverse magnetization, m , are usually expressed by

$$\left. \frac{\partial m}{\partial \eta} \right|_{0,L} + \lambda m|_{0,L} = 0 \quad (7)$$

where η is the outward directed unit normal and the film surfaces are at $z = 0$ and $z = L$. The ratio α/β and the effective mode number, n_{eff} , are determined from (6) and (7) as a function of the pinning parameter λ . The limits $\lambda \rightarrow 0$ and $\lambda \rightarrow \infty$ correspond to the unpinned and pinned cases respectively.

In a transmission spectrometer, such as the one described in the previous section, the exciting field is not uniform. In fact, in the limit $L \ll c/\sqrt{2\pi\sigma\omega}$, it is easily shown that

$$\vec{h}(z) \approx \vec{h}(0) \left\{ 1 - \left(1 - \frac{c}{4\pi\sigma L} \right) \frac{z}{L} \right\}$$

where σ is the electrical conductivity. We have assumed that a microwave magnetic field, $\vec{h}(0) = h\hat{x}$ is incident at the surface $z=0$ and that the field $\vec{h}(L)$ couples to a propagating wave. For a 2000Å permalloy film, $c/4\pi\sigma L$ is of the order of 10^{-3} and to a good approximation $\vec{h}(z) = \vec{h}(0)(1-z/L)$. This field will couple to all pinned modes ($n > 0$) with the intensity varying as $1/n^2$. In the unpinned case, the uniform mode as well as higher modes corresponding to odd values of n should be excited. The intensity will fall off as $1/n^4$, and it is probable that only a very few modes can be detected experimentally.

Spin wave spectra nearly always show some deviation from the behavior predicted above.¹³ The only consistently reproducible film property has been the variation of the effective pinning as a function of the angle ϕ . The perpendicular field spectrum is usually characteristic of strong pinning. However, the spacing and intensity of the low order modes often tend to vary as n and $1/n$, respectively. Also, modes associated with even values of n can often be detected.

As $\vec{H}$ is rotated away from the normal direction, the higher order modes diminish rapidly in intensity. Without exception, a critical angle²⁶ ($\phi = \phi_c$) is reached where only a single resonance is observed. This is characteristic of a uniform film in the absence of any pinning. For $\phi_c < \phi \leq \pi/2$, very weak higher order modes usually reappear. The spectrum in this range is characteristic of weak pinning, and the deviations from quadratic spacing at parallel field are much less pronounced than at perpendicular field.

Two models have been proposed to account for the observed behavior; the surface anisotropy model of Soohoo and the volume inhomogeneity (VI) model originally proposed by Portis²⁷ and extended by Hirota,²⁸ Wigen and Kooi,¹³ and others. The latter is based on the assumptions that the surface spins are free and that the internal field varies in the film due to a non-uniform magnetization. While both qualitatively predict the observed features of experimental spectra, the VI model has been more successful in quantitatively explaining mode positions and intensities.

Nisenoff and Terhune²² (NT) have reported the first observation of standing spin wave spectra in permalloy which showed no deviation from the predicted quadratic spacing at either parallel or perpendicular field.

They found that their experimental results could be explained very well by a simplified version of the VI model -- i.e. a uniform film with thin layers ($\sim 25\text{\AA}$) of reduced magnetization at each surface. We have obtained similar results and we feel that the model proposed by NT is also appropriate for our films.

The top traces in Figs. (4), (5) and (6) show spectra of a $1200\text{\AA}$ film in a single reflection cavity for $\phi = 0$, $\phi = \phi_c = 14^\circ$, and $\phi = \pi/2$. At $\phi = 0$, the intensity decreases slightly more rapidly than $1/n^2$, consistent with strong surface pinning. However, the weakly excited even modes indicate slight asymmetry in the boundary conditions. A critical angle, ϕ_c , is observed at which there is apparently no pinning. At $\phi = \pi/2$, the high field mode is identified as $n = 0$ and the two weakly excited modes as $n = 1$ and $n = 2$. Figure 7 shows a plot of $H_0 - H_n$ vs n^2 at both parallel and perpendicular field for this film. The apparently smaller exchange stiffness at perpendicular field was also observed by NT. They showed that this was not inconsistent with their model since extremely thin surface layers of reduced magnetization can lead to a larger effective film thickness in the perpendicular field orientation. For this reason we used the parallel field slope and the real thickness, L , to calculate $\tilde{A}$.

The values of $4\pi M$ and g ($g = \gamma\hbar/\mu_B$) can be calculated from the experimentally determined values of H_0 ($n = 0$) and simultaneous solution of Eqs. (4) and (5). The line shapes at perpendicular field were very nearly Lorentzian and the linewidths nearly constant for $n > 1$. In this case it is appropriate to define a spin wave lifetime $T_2 = 2/\gamma\Delta H$ where ΔH is the full width at half-maximum.

A summary of the experimentally determined parameters for the 1200Å film and a 2000Å film which exhibited virtually identical spectral features is given in Table I. The values of $4\pi M$ and g are in reasonably good agreement with bulk values, as they should be for high quality films of this alloy composition. Magnetostriction and crystalline anisotropy, which can cause uniform shifts in spectra,²⁹ are extremely small in 81-19 permalloy. The T_2 's at this frequency are also in good agreement with values determined from both standing spin wave and parallel pumping experiments.³⁰ The absolute values of δ' , although reasonable, should not be taken too seriously because of the uncertainty in L .

The reflection and transmission spectra of the 1200Å film, as measured in the transmission spectrometer, are shown in the two lower traces of Figs. 4, 5, and 6. There is no detectable change in the mode spacing or the linewidth from the single cavity result, but the qualitative change in mode intensities are of great interest. As expected the even modes are strongly excited and the intensity variation of all modes is slightly faster than $1/n^2$ for $n > 1$. At $\phi = \phi_c$ there is now clear evidence of an $n = 1$ mode. The identification is confirmed from Eqs. (1) and (3). Using $\phi = 14^\circ$ and the values of $4\pi M$ and g given above yields a spacing $H_0 - H_1 \approx 400$ Oe, which is just the observed spacing within experimental error. Thus we have direct evidence of excitation of unpinned modes in a transmission system.

Examination of the transmission spectra gives a surprising result. At resonance there is an alternating increase and decrease in the total power transmitted. This effect is not restricted to one field orientation. As the angle ϕ is varied, the high field mode, whether pinned or unpinned,

always corresponds to a decrease in transmitted power. The next mode is always observable and always corresponds to an increase in transmission. The alternation continues for all detectable modes.

The physical origin of this effect can be understood if one remembers that there are two modes of propagation in the film, a spin wave mode and a photon mode. The rotating magnetization associated with the spin wave mode induces eddy currents

$$\left(\vec{\nabla} \times \vec{J} = - \frac{\sigma}{c} \left[\frac{\partial \vec{H}}{\partial t} + 4\pi \frac{\partial \vec{M}}{\partial t} \right] \right) .$$

Eddy currents will couple to a propagating wave at the surface $z = L$. The phase of the radiating eddy currents depend directly on the phase of the transverse magnetization at $z = L$, which changes by 180° for each successive value of n . Thus the spin wave contribution to the transmitted power either add to or subtract from the contribution of the electromagnetic (photon) mode. From the perpendicular and critical angle results we see that modes of even symmetry about the center of the film always correspond to a decrease in transmitted power. This is confirmed in the detailed calculations of the next section. It will also be seen that observation of this interference effect can be used to obtain additional information about mode symmetries and boundary conditions which cannot be obtained from a single reflection cavity experiment.

The occurrence of this interference effect should only depend on mode symmetries and not on the specific eigenfunctions describing the spatial variation of the modes. This was experimentally confirmed with films which showed the usual deviation from quadratic spacing. These included ones we prepared and two that were kindly given to us by Wigen

and Kooi.³¹

In conclusion we wish to make a brief comment on film quality. Nisenoff and Terhune²² prepared their films in an oil free, vacuum pumped system at a pressure of 10^{-7} Torr. They suggested that the deviations from quadratic behavior usually observed are largely due to contamination from residual gases present during evaporation. They specifically listed backstreaming of oil in conventional oil-pumped systems as a possible source of contamination. Our results indicate that this may be the most critical factor in determining film quality in this composition permalloy. We took great pains to eliminate any backstreaming from either the mechanical fore-pump or the oil diffusion pump, and although our evaporation pressure was more than an order of magnitude higher than that of NT, we produced comparable films.

C. Transmission Theory

The qualitative behavior of the observed transmission spectra is easily understood from the physical arguments of the previous section. A detailed mathematical analysis requires simultaneous solution of Maxwell's equations and the spin equations of motion in the film. The problem is greatly simplified if the film is uniform or if the variation in the internal field is confined to thin surface layers and can be incorporated into the boundary conditions for the transverse magnetization. The films reported on here appear to fall into this category, and the following calculation will be restricted to this case. Furthermore, only the perpendicular field geometry will be considered. In the parallel field orientation, the appearance of a transverse demagnetizing field gives rise to elliptically polarized normal modes. The dispersion relation has three branches, and the algebra becomes extremely tedious.

Let us now consider a film of infinite extent in the x and y directions with planar surfaces at $z = 0$ and $z = L$. The applied dc magnetic field and the saturation magnetization are normal to the plane of the film: $\vec{H}_0 = H_0 \hat{z}$, $\vec{M}_0 = M_0 \hat{z}$. Denoting the microwave components of the field and the magnetization by lower case letters, the equation of motion for the magnetization, in the limit that $|\vec{h}| \ll H_0$ and $|\vec{m}| \ll M_0$, is

$$\pm i \frac{\partial m^\pm}{\partial t} = D \frac{\partial^2 m^\pm}{\partial z^2} - \omega_0 m^\pm \mp \frac{1 m^\pm}{T_2} + \gamma M_0 h^\pm \quad (8)$$

The rotating components $m^\pm$ and $h^\pm$ are defined in the usual way, and $\omega_0 = \gamma(H_0 - 4\pi M_0)$, where γ is the gyromagnetic ratio. A phenomenological Bloch damping term, $\partial m_{x,y} / \partial t = -m_{x,y} / T_2$, has been introduced. This

generates the observed Lorentzian line shape which is characteristic of relaxation broadening. In the present limit of $|\vec{m}| \ll M_0$ it is formally equivalent to a Landau-Lifshitz³² damping term which is proportional to $\vec{M}(\vec{M}\vec{H})$. In the notation of Landau and Lifshitz $D = \gamma(2A/M_0)$.

In describing the electromagnetic properties of the system, the displacement current (which is important in a ferromagnetic insulator) can be ignored. Eliminating the electric field from the Maxwell curl equations gives

$$\frac{c^2}{4\pi\sigma} \frac{\partial^2 h^\pm}{\partial z^2} = \frac{\partial h^\pm}{\partial t} + 4\pi \frac{\partial m^\pm}{\partial t} \quad (9)$$

Assuming $m^\pm$ and $h^\pm$ vary as $e^{i(\omega t - kz)}$ and substituting into Eqs. (8) and (9), one obtains the dispersion relation

$$\pm \omega = \omega_0 + D k^{\pm 2} \pm \frac{1}{T_2} + \frac{4\pi\gamma M_0}{1 - \frac{1}{2} \delta^2 k^{\pm 2}} \quad (10)$$

where $\delta = c/(2\pi\sigma\omega)^{1/2}$ is the classical skin depth and $k^\pm$ are the propagation vectors for the two senses of circular polarization.

For $\omega_0 > 0$, m^- is being driven far from resonance, and its contribution is negligible. The propagation of h^- is that of an electromagnetic wave of frequency ω in a medium of conductivity σ . This is easily verified by substitution of $-(\omega - i/T_2)$ for $(\omega - i/T_2)$ in the following analysis which will be confined to h^+ and m^+ . However, if the driving field at $z = 0$ is linearly polarized, the contribution from h^- must be incorporated in the final result.

Returning to Eq. (10) and considering only the right circularly polarized components, we see that there are two solutions of k^+ corresponding to the two modes of propagation in the system. Near the crossover of the spin wave and photon dispersion curves ($\omega \approx \omega_0$), there is a strong mixing, and both solutions have both spin wave and photon character:

$$k^2 = \left(-\frac{1}{\delta^2} + \frac{k_0^2}{2} \right) \left(1 \pm \left[1 - i \frac{A}{(2i/\delta^2 + k_0^2)^2} \right]^{1/2} \right)$$

$$A = \frac{8\pi\gamma M_0}{\theta \delta^2} \quad (11)$$

$$k_0^2 = \frac{\omega - \omega_0 - i/T_2}{\theta}$$

In the region of strong interaction, the mixing results in eddy current damping of the spin wave modes as discussed by Pincus.³³

Equation (11) can be greatly simplified if we restrict our calculation to the region for which $\omega - \omega_0 > 5 \times 10^{+9} \text{ sec}^{-1}$. In evaluating the terms in k^2 we will use the typical parameter values $L = 2 \times 10^{-5} \text{ cm}$, $\delta = 3 \times 10^{-4} \text{ cm}$, $4\pi\gamma M_0 = 2 \times 10^{+11} \text{ sec}^{-1}$, and $\theta = 5 \times 10^{-2} \text{ cm}^2 \text{ sec}^{-1}$. We then find

$$k_1^2 \approx k_0^2 \left(1 - i \frac{A}{k_0^4} \right) \quad (12)$$

for the spin-wave-like mode, and we find

$$k_2^2 \approx \frac{2i}{\delta^2} \left(1 - \frac{A\delta^2}{2k_0^2} \right) \quad (13)$$

for the photon-like mode. The restriction on $\omega - \omega_0$ limits the validity of these expressions to spin wave modes for which $n \geq 2$, where n is a

positive integer defined by the resonance condition $\text{Re}(k_1) \approx \text{Re}(k_0) = n\pi/L$. The predicted behavior for $n = 1$ should, however, be qualitatively correct.

We now define the susceptibility of mode α ($\alpha = 1, 2$) by the relation $m_\alpha^+ = \chi_\alpha h_\alpha^+$. From (10), (12), and (13) we have

$$\begin{aligned} \chi_1 &= \frac{1}{8\pi} \delta^2 k_0^2 \\ \chi_2 &= \frac{-\gamma M_0}{\rho k_0^2} \end{aligned} \quad (14)$$

At this point, we have sufficient information to explicitly evaluate microwave transmission through the film.

Because experiments are performed in resonant cavities, it is appropriate to consider the case of a linearly polarized incident field. A microwave magnetic field of frequency ω and amplitude h_0 is applied in the x direction at $z = 0$. If the film is terminated at $z = L$ by a medium with a characteristic impedance equal to that of free space, the boundary conditions on $h^\pm = h^\pm(z)e^{\pm i\omega t}$ are

$$\begin{aligned} h^\pm(0) &= h_0 \\ h^\pm(L) &= \lim_{z \rightarrow L^-} \left(-\frac{c}{4\pi\sigma} \frac{\partial h^\pm(z)}{\partial z} \right) \end{aligned} \quad (15)$$

Because there are two modes of propagation of h^+ in the film, the transmitted wave will be elliptically polarized. However, only the component $h_x = \frac{1}{2} (\text{Re}[h^+(L)e^{i\omega t}] + h^-(L)e^{-i\omega t})$ will couple to a propagating mode in the microwave system. In this case, the transmitted power is proportional to the quantity W defined by the following relation:

$$W = \frac{\{h^-(L) + \text{Re}[h^+(L)]\}^2 + \{\text{Im}[h^+(L)]\}^2}{4h_o^2} \quad (16)$$

In the absence of any wave spin excitation, Eq. (16) reduces to

$$W = W_o = \left(\frac{c}{4\pi\sigma L} \right)^2 \quad (17)$$

In order to determine the transmission at resonance, we let

$$k_1 = \frac{n\pi}{L} - i L/2n\pi DT_2 = \frac{n\pi}{L} - iK \text{ and assume standing wave solutions for } h^+ \text{ and } m^+.$$

$$\begin{aligned} h_1^+(z) &= h_o (\alpha \cos k_1 z + \beta \sin k_1 z) \\ h_2^+(z) &= h_o (\rho \cos k_2 z + \eta \sin k_2 z) \\ h^+(z) &= h_1^+(z) + h_2^+(z) \\ m^+(z) &= x_1 h_1^+(z) + x_2 h_2^+(z) \end{aligned} \quad (18)$$

Substitution of Eq. (18) into Eq. (15) gives

$$\frac{h^+(L)}{h_o} \approx \frac{c}{4\pi\sigma L} \left\{ 1 + [-1 + (-1)^n - i(-1)^n n\pi KL] \alpha - (-1)^n n\pi\beta \right\} \quad (19)$$

where the coefficients α and β are to be determined from the boundary conditions on the transverse magnetization.

The observed perpendicular field spectra indicate strong surface spin pinning so we choose the boundary conditions

$$m^+(0) = m^+(z) = 0 \quad (20)$$

The values of α and β are then given by

$$\begin{aligned}\alpha &\approx -\frac{x_2}{x_1} \\ \beta &\approx -\frac{1}{KL} \alpha\end{aligned}\tag{21}$$

As must be the case for pinned surface spins the spatial variation of the spin wave amplitudes h_1^+ and m_1^+ is dominated by the term proportional to $\sin k_1 z$. Finally, from Eqs. (16), (19), and (21) the fractional change in transmitted power at resonance is

$$w = \frac{W-W_0}{W_0} \approx (-1)^n \frac{16\pi\gamma M_0 L^2 T_2}{\delta^2 (n\pi)^2}\tag{22}$$

This expression, however, is not complete. At resonance, the spin waves are absorbing power, and the Q of both cavities is altered. The field applied at $z = 0$ is not constant, and the effect of its variation must be included.

The most direct way of solving this problem is analysis of the equivalent circuit of the transmission system,³⁴ as shown in Fig. 8. It is assumed that the film is symmetrically placed with respect to the two cavities and that the coupling of power through the film can be represented by a mutual inductance M. At resonance the equivalent resistance of each cavity is increased by an amount ΔR , and the mutual inductance changes by ΔM . The total transmitted power, P, is just $i_2^2 R_L$. Making the simplifying assumptions that $R_x = R_y = R$, $M_1 = M_2$, $\Delta R \ll R$, $\Delta M \ll M$, $L_1 = L_2$, the fractional change in transmitted power is given by

$$\frac{\Delta P}{P} \approx \frac{2\Delta I_2}{I_2} \approx \frac{2\Delta M}{M} - \frac{2\Delta R}{R} \frac{1}{(1+\beta_1)} \quad (23)$$

where the coupling coefficient, β_1 , is given by

$$\beta_1 = \frac{\omega^2 M_1^2}{Z_1 R}, \quad Z_1 = R_0 + i\omega L_1 \quad (24)$$

The quantity w given by Eq. (22) is just $2\Delta M/M$. Exact evaluation of the term proportional to ΔR is somewhat more difficult. One must know cavity Q 's, coupling coefficients, and filling factors. However, the qualitative behavior of this term can be determined. ΔR is proportional to ΔR_s , the change in the real part of the surface impedance per unit area of the film.

$$R_s \approx \text{Re}(-k_1 \beta) = \frac{16\pi\gamma M_0 L^2 T_2}{\delta^2 (\pi\pi)^2} \quad (25)$$

It was found that amplitudes of the resonances in the transmission spectra were described well by the relation

$$\frac{\Delta P}{P} = (-1)^n \frac{C_1}{n^2} - \frac{C_2}{n^2}; \quad C_1 = \frac{16\pi\gamma M_0 L^2 T_2}{\delta^2 \pi^2} \quad (26)$$

for $n \geq 2$. The values of T_2 calculated from the experimentally determined parameter C_1 are listed in the last column of Table I. The agreement with the T_2 's determined from the resonance linewidths is quite satisfactory, considering the uncertainty in the values of L and δ . Furthermore, the intensity of the modes in the reflection spectra, which vary

directly as ΔR_s , decreased slightly faster than n^{-2} for $n \approx 2$. This coupled with the fact that the even modes did not disappear completely in a single reflection cavity experiment means that the pinning was not complete nor was it identical at the two surfaces. However, these effects are small, and the predictions of the perfect surface pinning are in good overall agreement with our experimental results.

Recent experiments³⁴ indicate that for high quality epitaxial permalloy films the surface pinning is extremely weak, even in the perpendicular field orientation. In the absence of pinning, a uniform exciting field will couple only to the uniform ($k = 0$) mode. In a transmission spectrometer, the non-uniform exciting field will couple to modes for which $k \neq 0$. Because of the importance of film quality in making a reliable determination of the bulk exchange stiffness, we will extend the previous analysis to the case of unpinned surface spins.

The boundary conditions on the transverse magnetization are

$$\left. \frac{\partial m^+(z)}{\partial z} \right|_{z=0} = \left. \frac{\partial m^+(z)}{\partial z} \right|_{z=L} = 0 \quad (27)$$

From Eqs. (15) and (18)

$$\beta = \frac{1}{n\pi} \frac{x_2}{x_1}$$

$$\alpha = \frac{1}{KL} (1 - (-1)^n) \beta$$

The modes are predominantly cosine-like, and there is significant excitation only for the odd-numbered ones. The fractional change in the transmitted power at resonance is

$$\frac{2\Delta M}{M} = w = [1 - (-1)^n] \frac{32\pi\gamma M_0 L^2 T_2}{\delta^2 (n\pi)^4} \quad (28)$$

The power absorption also varies as n^{-4} :

$$\Delta R_s \propto \frac{1 - (-1)^n}{n^4} \quad (29)$$

For $|\Delta M/M| > |\Delta R/R(1+\beta_1)|$, excitation of the odd modes will cause an increase in transmitted power.

Our experimental data give a qualitative verification of this behavior. As described in the previous section, there is an increase in transmitted power for the $n = 1$ mode at the "critical angle." Although the mathematical analysis of this section is not valid except in the perpendicular field orientation for $n \neq 2$, the sign of $\Delta M/M$ depends only on the spatial variation and symmetry of the transverse magnetization. The surface spins are free at the "critical angle," and for $n = 1$, $m^+(z)$ will be dominated by a term proportional to $\cos \frac{\pi}{L} z$.

Although unpinned modes can be excited in a transmission spectrometer, both reflected and transmitted intensities fall off as n^{-4} . The mode intensities can be increased by pinning the spins at one surface. This can be achieved experimentally by evaporating an antiferromagnetic layer onto the exposed surface of the film, as reported by Kooi and coworkers.³⁵ In this case the resonance condition is $\text{Re}(k_1) = (n + 1/2) \pi/L$, where n is a positive integer.

We first examine the configuration in which the first surface is pinned and the second surface is free:

$$m^+(0) = 0 \quad \left. \frac{\partial m^+(z)}{\partial z} \right|_{z=L} = 0 \quad (30)$$

By straightforward algebra, we find

$$\frac{2\Delta M}{M} = w \approx -(-1)^n \frac{16\pi\gamma M_0 L^2 T_2}{8^2(n+1/2)^3 \pi^3} \quad (31)$$

while

$$\Delta R_s \propto \frac{1}{(n+1/2)^2} \quad (32)$$

The large power absorption will probably dominate the transmission spectrum and mask any alternation.

If the film is turned around,

$$\left. \frac{\partial m^+(z)}{\partial z} \right|_{z=0} = 0 \quad m^+(L) = 0 \quad (33)$$

and we find the same expression for w ,

$$\frac{2\Delta M}{M} = w \approx -(-1)^n \frac{16\pi\gamma M L^2 T_2}{8^2(n+1/2)^3 \pi^3} \quad (34)$$

The power absorption on the other hand is very much weaker,

$$\Delta R_s \propto \frac{1}{(n+1/2)^4} \quad (35)$$

Comparison of Eqs. (31), (32), (33), and (34), suggests that the maximum number of spin wave modes can be observed when the boundary conditions of Eq. (30) are satisfied. However, for a film with one surface pinned and one surface free, a uniform exciting field will couple to all modes

and the power absorption will fall off as $1/(n+1/2)^2$. The transmission technique has no advantage over a standard reflection experiment unless one is interested in looking specifically at the interference effect.

We have shown here that the interference between a photon mode and a spin wave mode can lead to either an increase or a decrease in the microwave transparency of a thin film. From the exact nature of the transmission spectrum, one can obtain information about the symmetry of and boundary conditions for the spin wave modes in the film. While this type of information is helpful in understanding film properties, it offers nothing new to our understanding of metallic ferromagnetism. We feel that the outstanding feature of transmission spectrometry is its potential usefulness in measuring the exchange stiffness in high quality single crystal samples in which there is little or no surface spin pinning. It should be pointed out, however, that the existence of a non-uniform exciting field is not an exclusive property of the transmission system. The origin of the field attenuation in the film is the impedance mismatch at the surface $z = L$. Excitation of unpinned modes should be observed in the reflection spectrum of any film which is terminated by a non-conducting medium.

III. TEMPERATURE DEPENDENCE OF THE EXCHANGE STIFFNESS OF NICKEL

A. Experiment

Nickel films ranging in thickness from 1600 Å to 2800 Å were deposited on heated glass substrates at a pressure near 3×10^{-6} Torr. The source was a 99.999% pure, vertically oriented nickel rod 3/16" in diameter. Evaporation took place from the top of the rod, which was heated by electron bombardment. The melt was confined to a small region at the top by placing the bottom of the rod in thermal contact with a water cooled copper block. The substrates, which were held at a temperature of 290°C, were shielded from the source by a manually operated shutter. The shutter was removed when a uniform evaporation rate (~ 16 Å/sec) was attained. The thickness of a given film was determined within ± 50 Å with a multiple beam interferometer.

Resonance spectra were obtained at 15.1 Gc/sec over the temperature range 77°K to 600°K. A standard field modulation and lock-in detection scheme was used. The resulting experimental traces gave the derivative of the power absorption with respect to the applied dc field which was swept at 2 Oe/sec and monitored by a rotating coil gaussmeter. Careful calibration of the gaussmeter enabled determination of the center of well resolved lines within ± 2 Oe.

Observation of spectra over such a wide temperature range required the use of two separate resonant cavities. For measurements above room temperature (295°K), films were placed on an end wall of a cylindrical cavity resonant in a TE_{111} mode at 15.10 Gc/sec, as is shown in Fig. 9. The coupling iris in the top of the cavity removes the infinite degeneracy of this mode. The end wall was separated from the rest of the cavity by

.010", and the entire system was evacuated to a pressure of $\sim 2 \times 10^{-5}$ Torr. Microwave continuity was maintained at the break by a radial choke joint. A direct current resistance heater was used to raise the temperature of the end wall and sample while the water cooled cavity remained at room temperature.

In order to eliminate temperature drift, a simple servo control system was used to regulate the heater current. A thermistor, which comprised one arm of an ac bridge, was placed in direct contact with the heating coils. The bridge was driven at 86 cps and the bridge output fed into a lock-in detector. The lock-in output drove a dc motor coupled to a helipot which in turn varied the heater current. With this system (shown schematically in Fig. 10) the sample temperature was stable to better than $\pm 1^\circ\text{K}$ over periods as long as 2 hrs.

The low temperature cavity was identical with the one just described except in two respects. The water cooling jacket was replaced by a brass can which was immersed in a liquid nitrogen (or liquid helium) bath. Also, this system was sufficiently stable that the temperature control unit was not needed. Maximum temperature drifts did not exceed $\pm 1.5^\circ\text{K}$ over the entire range of 77°K to 295°K . For both cavities a copper-constantan thermocouple was used to monitor film temperature.

Heating the films well above room temperature in the cavity presented significant experimental problems. Irreversible changes, both physical and chemical, can take place. We found that annealing a film for 30 min at 300°C in the bell jar immediately after evaporation was helpful. Annealed films showed less deviation from the predicted quadratic mode spacing than unannealed films. Also, the spectra of

annealed films were quite reproducible between 295°K and 465°K. This behavior was not always observed for unannealed films. However, drastic changes in the room temperature spectra of films which had been heated well above 465°K were observed independent of past annealing. These changes included broadening of resonance lines and a decrease in the exchange stiffness and were attributed to oxidation. Although the cavity was evacuated, the design required that the substrate be cemented to the cavity wall. We believe that the apparent chemical changes in the film were caused by reactions with outgassing components from the cement.

The upper limit of the range of reproducibility was extended well above 500°K by evaporating a 2000Å layer of gold over a given film. The gold was added after the film had been removed from the evaporation system and its room temperature resonance spectrum recorded. Exposure to air always causes some surface oxidation, and the function of the gold was to prevent further oxidation. The evaporation of the gold onto a "dirty" surface probably prevented the formation of an alloy at the Ni-Au interface. This was confirmed from the room temperature spectrum which showed no detectable change after the addition of the protective gold layer.

B. Results and Discussion

In all Ni films which we prepared, only a single resonance was detected with the applied dc magnetic field parallel to the plane of the film. As discussed in Part II, a spectrum consisting of a single line is characteristic of extremely weak surface spin-pinning and is commonly observed in the parallel field orientation. For this reason, the exchange stiffness was determined only from the perpendicular field spectrum which consisted of several well resolved modes. However, the resonance spectra of the Ni films, in contrast to the permalloy samples described above, always deviated from the behavior predicted for an ideal film²⁵ in two respects: the apparent room-temperature magnetization of the films determined from resonance was from 10% to 40% smaller than the known bulk value; also, the low order modes in the perpendicular field spectrum always showed some deviation from the predicted quadratic spacing.

The apparent reduction in magnetization has been observed in previous studies of Ni films by both Nose²⁹ and Phillips and Rosenberg.¹⁶ Nose has suggested that the effect is probably due largely to a strain-induced anisotropy field rather than an actual reduction in magnetization. In a material such as nickel, where magneto-elastic effects are not negligible, the respective perpendicular and parallel field resonance equations should be modified in the following way:³⁴

$$\left(\frac{\omega}{\gamma}\right)^2 = \left(H - 4\pi M - \frac{3\lambda\sigma}{M} - \frac{3\lambda\sigma_u}{M} + \frac{D}{\gamma} k^2 \right) \times$$

$$\left(H - 4\pi M - \frac{3\lambda\sigma}{M} + \frac{D}{\gamma} k^2 \right) \quad (36)$$

$$\left(\frac{\omega}{\gamma}\right)^2 = \left(H + 4\pi M + \frac{3\lambda\sigma}{M} + \frac{3\lambda\sigma_u}{M} \cos^2 \theta + \frac{\delta'}{\gamma} k^2 \right) \times \quad (37)$$

$$\left(H + \frac{3\lambda\sigma_u}{M} \cos 2\theta + \frac{\delta'}{\gamma} k^2 \right)$$

where λ is the saturation magnetostriction constant, σ is the isotropic stress in the plane of the film, and σ_u is a uniaxial stress at an angle θ with respect to the magnetization. Nose points out that the uniaxial stress is usually small in evaporated films in which case (36) and (37) reduce to (4) and (5) with M replaced by

$$M_{\text{eff}} = M + \frac{3\lambda\sigma}{4\pi M} \quad (38)$$

Simply stated, an isotropic stress in the plane of the film causes a uniaxial anisotropy in the direction of the film normal. The associated anisotropy field uniformly shifts all modes leaving the mode spacing unaffected.

If the film is not uniformly strained, then the magneto-elastic coupling will contribute to the non-uniformity in the internal field. We feel that this effect, in addition to slight film contamination, was responsible for the deviation of the low order modes. When the variation in the internal field is small, a simple perturbation calculation¹⁶ shows that the dispersion relation measured from the quadratically spaced higher order modes accurately gives the exchange stiffness of the material.

The films selected for the study of the temperature dependence of the exchange stiffness were those which showed the smallest deviation from quadratic spacing and an apparent magnetization most nearly equal to the

bulk value. The direct effect of strain on the exchange stiffness is not known, and we felt that samples with the largest apparent magnetization would most accurately reflect properties of strain free bulk material. The results for two samples (which we shall refer to as Film A and Film B) will be presented here.

The room-temperature, perpendicular field resonance spectrum and corresponding plot of the resonant field vs n^2 are shown in Figs. 11 and 12 for Film A ($L = 2660\text{\AA}$) and in Figs. 13 and 14 for Film B ($L = 2150\text{\AA}$). The measured values of the exchange stiffness are compared with values determined from previous thin film^{16,29} and neutron scattering^{11,17} experiments in Table II. The agreement is satisfactory. The fractional uncertainty in neutron measurements is at least 2%-3% due to resolution problems inherent in the experimental technique. Variation in film results is to be expected even if all samples are of reasonably high quality. Standing spin wave resonance is sensitive to small changes in the exchange stiffness, but determination of the absolute value depends on a knowledge of the film thickness, which is usually known only within $\pm 1\%$. Also, the effective thickness for resonance, as we observed in permalloy, can differ from real thickness.

The features of the resonance spectra of our two films are similar. Each shows only a slight deviation from quadratic spacing for the low order modes. For $n > 3$, where the modes are quadratically spaced, the intensity decreases somewhat more rapidly than $1/n^2$. In each case the highest order mode which is well resolved corresponds to $n = 7$. In Film A an extremely weak mode identified as $n = 9$ is detected but is not well resolved. Its

position indicates that there is no drastic deviation from quadratic spacing. In both films a weakly excited $n = 6$ mode is observed, indicative of a slight asymmetry in the internal field.

The rapid intensity decrease of the high order modes, although undesirable from a resonance point of view, was a consistently observed property of high quality films. We prepared films in which modes as high as $n = 11$ were well resolved; however, the deviation from quadratic spacing was much more drastic in these samples. We conclude from this that the mode pinning in nickel is due principally to variations in the internal field rather than intrinsic surface effects.

The line shapes at room temperature were nearly lorentzian and the linewidths nearly constant as a function of n . In Film B there was a slight (less than 10%) but noticeable broadening for $n = 7$. This was undoubtedly due to surface irregularities which become increasingly more effective in scattering spin waves as the wavelength decreases.³⁵ The peak to peak linewidths were ~ 265 Oe in A and ~ 350 Oe in B. From Rodbell's³⁶ observation of ferromagnetic resonance in thin single crystal nickel whiskers, we would expect a value of ~ 190 Oe at 15 Gc/sec. However, evaporated films are in general polycrystallines and any randomness in orientation of the crystallites will broaden the lines because of the magneto-crystalline anisotropy.

This assumption appeared to be borne out by the temperature dependence of the linewidths. Below room temperature the linewidths increased rapidly and the line shapes became more nearly gaussian. Above room temperature the lines narrowed slowly in each case reaching an absolute minimum near 450°K (~ 230 Oe in Film A and ~ 240 Oe in Film B). In addition it

was found that the rate of change of linewidth below room temperature scaled very nearly as dK_1/dT , where K_1 is the first anisotropy constant of nickel. It should also be pointed out, however, that non-uniform strain in the plane of the film will also contribute to inhomogeneous broadening and have a qualitatively similar temperature dependence.

The combination of line broadening and the weak intensity of the higher order modes restricted our measurement of the exchange stiffness to temperatures above 120°K. Below 120°K modes 5 and 7 were poorly resolved and below 100°K were unresolved.

The high temperature limit of resolution was ~540°K in both films. Above 450°K the lines broadened, but somewhat more slowly than at low temperatures. Between 450° and 540° the relative increase was within 10% of that observed by Rodbell,³⁶ and the line shapes remained very nearly lorentzian. In this region the exchange stiffness was decreasing rapidly, and the corresponding decrease in mode spacing was the determining factor in "washing out" modes 5 and 7. In fact, the resolution was poor above 500°K. Between 500°K and 540°K mode positions, intensities, and linewidths had to be determined by making a least squares fit of a spectrum of lorentzian lines to the experimental data.

Although there was clear evidence of spin wave excitation up to nearly 580°K, computer analysis was not helpful above 540°K. The overall signal to noise ratio was falling off both with M and $1/\Delta H$ (ΔH being the peak to peak linewidth). The positions of the high order modes, as determined by the least squares fit, were extremely sensitive to the exact values of experimental points read from the strip chart recording.

Apparent line centers would shift by as much as 20-30 Oe if the experimental values fed into the computer were varied within the range of experimental error.

In an attempt to obtain some idea of the actual Curie temperature, spectra for Film A were taken up to 606°K. Above 550°K, where the effects of crystalline anisotropy and magnetostriction should be negligible, we analyzed both parallel and perpendicular field spectra to determine $4\pi M$ and g . We found that if we assumed a Curie temperature of $T_c = 612^\circ K = .976 T_{cNi}$, that $M(T/T_c)$ scaled exactly with the magnetization of bulk nickel in the presence of an applied field, $M(T/T_{cNi})$, as measured by Weiss and Forrer.³⁷ This can be seen in Fig. 15. The g value in this region was 2.19 which is in fairly good agreement with the bulk value of 2.22.³⁶ We did not have sufficient high temperature data to unambiguously establish $M(T)$ for Film B. However, it appeared that $T_c \approx 602^\circ K = .96 T_{Ni}$, as can also be seen in Fig. 15.

As we mentioned in the previous section, some care must be exercised in interpreting film results, particularly when the samples are heated well above room temperature. We have used the reproducibility of the room temperature spectrum as a measure of the reliability of high temperature results. For this reason the sample was returned to room temperature periodically. Even with the protective gold coating slight changes were observed when the samples had been heated up to ~540°K. In both films, there were slight (less than 200)e) but uniform shifts in the spectra. In Film A, a broadening of approximately 40 Oe in the room temperature linewidths was observed. These effects could easily arise from slight change in the internal strain in the film. Because relative mode intensities and the exchange stiffness were unchanged, we believe

that the results up to 540°K are reliable.

After Film A had been heated to 606°K the changes in the room temperature spectra were considerably more profound. Specifically, the linewidth increased to nearly 450 Oe. The broadening caused the same resolution problems encountered at both high and low temperatures. The resultant uncertainty in the determination of the exchange stiffness was $\pm 3\%$, and within these limits $\mathcal{D}$ was unchanged. This fact by itself is not convincing proof that the film has not been contaminated at high temperatures, but the direct measurement of the magnetization above 550°K tends to confirm this assumption. The temperature dependence of M would certainly not scale with that of a pure bulk sample if oxidation of the film were increasing at high temperatures.

In Film B, which had only been taken to 584°K, the room temperature linewidth showed a slight decrease, the higher order modes became relatively weaker, and the exchange stiffness remained unchanged within $\pm 1\%$. This behavior is characteristic of annealing. It would seem therefore that in both samples the principal effect of raising the temperature was to change the state of internal strain, although the reason for the qualitative difference in results is not clear.

One other factor should be considered in analyzing the temperature dependence of the exchange stiffness. Phillips¹⁴ has pointed out that if the strength of surface pinning changes as a function of temperature, the effective film thickness will also change. Since the mode spacing is proportional to $1/L^2$, a spurious term will be introduced into the measured variation of $\mathcal{D}$. However, any significant change in pinning conditions would cause a large change in relative mode intensities.

Within the limits of resolution, the relative mode intensities in our films remained constant below 540°K.

Plots of D vs T are given in Figs. 16 and 17 for Films A and B respectively. Below room temperature our data are in good qualitative agreement with that of Phillips and Rosenberg¹⁶ (PR). PR, using a much thicker film ($L \sim 5000\text{\AA}$), were able to resolve modes up to $n = 15$ and measure the exchange stiffness down to 15°K. Between 70°K and 295°K, they found that the renormalization of D was very well described by $D = D_0 (1 - aT^{3/2})$. We know of no theoretical justification for a $T^{3/2}$ variation in this region, and in fact it is probably not appropriate to try to fit D to a single term expansion in a power of the absolute temperature over such wide temperature range. However, to compare the functional dependence of D in our films with that observed by PR we have attempted to fit our low temperature data to such a single term expansion. We find that the most reasonable fit is in fact $D(T) \propto 1 - aT^{3/2}$. Plots of D vs $T^{3/2}$ are given in Figs. 18 and 19 for the two films.

We do find a quantitative discrepancy with PR. From 120°K to 295°K we observe a reduction in D of about 13% in Film A and 14% in Film B compared with a reduction of approximately 17% observed by PR. Although our resolution at low temperatures was substantially less than theirs, the observed difference is too large to be explained by experimental error. It is possible that their film may have a lower Curie temperature than ours, a fact which would lead to a more rapid reduction in D over the given temperature range. However, we do not at present have sufficient information about their film, to make such a judgment with any degree of certainty. In any case, the order of magnitude of the

magnitude of the reduction of σ is much too large to be explained on the basis of a Heisenberg model which predicts a change of ~1% over this range.

Over the entire temperature range we find reasonable agreement with the results of Stringfellow,¹⁷ who observed small angle scattering (SAS) of thermal neutrons from nickel powder. From 4°K to 560°K, he finds $\sigma \propto \sigma_0 \left(1 - b \left(\frac{T}{T_c}\right)^{2.6 \pm .7}\right)$. We definitely do not find a single term expansion which reasonably fits our data over our temperature range, but the percentage reduction between 120°K and 540°K is very close to that observed by Stringfellow. In Film A we find $\sigma(540^\circ\text{K}) \sim .52 \times \sigma(120^\circ\text{K})$. Using the best fit curve of the neutron data, i.e. $\sigma \propto 1 - b(T/T_c)^{2.6}$, gives $\sigma(540^\circ\text{K}) \sim .56 \sigma(120^\circ\text{K})$. From 120°K to room temperature, the best fit gives a reduction of 9% compared to our value of 13%.

Although the difference in results can almost be accounted for by the experimental uncertainty in the neutron data, it would appear that σ is varying slightly more slowly in the bulk sample. This is not inconsistent with the fact that the Curie temperature in Film A is ~2-1/2% less than in bulk. Also, in Film B where there is an apparent 4% reduction in the Curie temperature, we find $\sigma(540^\circ\text{K}) \sim .49 \sigma(120^\circ\text{K})$. The neutron measurements would appear to favor our film results over those of PR.

It is interesting to note that we find the greatest discrepancy with Stringfellow below room temperature, where his experimental technique is least sensitive (except near T_c where critical fluctuations and lifetime broadening of spin waves cause considerable resolution problems). Small angle scattering involves the observation of neutrons scattered near

the forward direction. At a critical angle, θ_c , with respect to the forward direction the neutron intensity drops off sharply, and analysis shows that θ_c is proportional to $1/\bar{D}$. The origin of this lies in the fact that long wavelength spin waves and neutrons both have free particle dispersion relations and that the effective mass for spin waves, $\mu = \hbar/(2\bar{D})$, is much smaller than the neutron mass, M . Simultaneous solution of the energy and momentum conservation equations gives $\theta_c \approx \mu/M = 1/2 \bar{D}M$. The result is independent of incident neutron wavelength so that this experiment has the great advantage of being able to use a thermally white neutron beam.

The difficulty at low temperatures arises from the fact that $\theta_c \lesssim 2^\circ$ in nickel. At such small angles the unscattered neutron beam provides a strong background and interferes with resolution. Also, the spin wave dispersion relation, $\omega = \omega_0 + \bar{D}k^2$, is not truly that of a free particle. The gap frequency, ω_0 , must be corrected for, and small angle scattering affords no means of directly determining ω_0 . This causes particular problems below room temperature where the anisotropy (and therefore ω_0) increases rapidly.

The range of k for which the exchange stiffness is determined by this method depends on the cutoff angle and the spread in k_n , the wave-vector of the incident neutrons. The cutoff wave vector, $k_c \approx k_n \theta_c$, is roughly $.04\text{\AA}^{-1}$ at room temperature. We see that the small angle scattering technique is complementary in range of k to our thin film resonance experiments for which $k < .01\text{\AA}^{-1}$.

A great deal more information about the spin wave dispersion relation can be obtained from inelastic neutron scattering by using the "constant Q"

technique developed by Brockhouse.³⁸ This method has the distinct advantage of being able to measure the temperature dependence of the energy of a spinwave of a given wave vector k . The need for a high intensity monochromatic neutron beam presents experimental problems; however, Nathans and coworkers¹¹ at Brookhaven have recently extended the range of k for which spin wave energies can be measured by this technique to $.03\text{\AA}^{-1} \leq k \leq .5\text{\AA}^{-1}$. The Brookhaven group is currently studying the temperature dependence of $\omega(k)$ in a single crystal of Ni,³⁹ and we shall find it very interesting to compare their results with ours and those of Stringfellow, particularly for values of k near $.03\text{\AA}^{-1}$.

The most interesting physical property which we have observed was the variation of the exchange stiffness with respect to the magnetization. Figure 20 is a plot of $\bar{D}(T/T_c)$ vs $M(T/T_c)$ for Film A. The value of T_c has been taken to be 612°K , and $M(T/T_c)$ has been appropriately scaled from the data of Weiss and Forrer.³⁷ On the basis of our direct measurement of the high temperature magnetization as shown in Fig. 15, we definitely feel such a scaling is reasonable.

At low temperatures $\bar{D}$ is decreasing more rapidly than M . This can be expressed mathematically by the relation $d\bar{D}/dM > \bar{D}/M$. As the temperature is increased the ratio $(d\bar{D}/dM)/(\bar{D}/M)$ gradually decreases and appears to approach a constant value of unity for $T > .8T_c$. Above $.8T_c$ the resolution is decreasing, and there is necessarily some uncertainty in the value of $(d\bar{D}/dM)/(\bar{D}/M)$. However, it appears that the best fit to the experimental data in this region is in fact $d\bar{D}/dM = \bar{D}/M$. In other words, the exchange stiffness is varying directly as the magnetization, and the physical implications of this

would appear to be significant.

The magnetization, M , is just proportional to $\langle S_z \rangle$, the average z -component of the spin at each atomic site. Then for $d\mathcal{O}/dM = \mathcal{O}/M$, we have $\mathcal{O} \propto M \propto \langle S_z \rangle$, which implies that transverse spin components do not contribute to the renormalization of $\mathcal{O}$. It would appear initially that this effect could be understood on the basis of a simple physical argument.

The long wavelength standing spin waves which we excite are being driven in a system where the spins are thermally excited. For a sufficiently high density of short wavelength thermal excitations one might expect the contribution of transverse spin components to the renormalization of $\mathcal{O}$ to average out. That the situation is not quite so simple can be seen from the work of Horowitz and Mattis⁴⁰ who have theoretically treated energy renormalization for a system where the moments are localized but the exchange is long range.

They find that if they define a thermal screening length

$$R_0 = \left[\frac{\hbar^2 \pi^2}{8\mu kT} \right]^{1/2}, \quad \mu = \frac{\hbar}{2\mathcal{O}} \quad (39)$$

the effective long range exchange $J_T(R)$ scales directly with the magnetization for $R > R_0$:

$$J_T(R) \approx J(R) \frac{M(T)}{M(0)}$$

From the above definition of R_0 , we see that $R_0 \sim \frac{\lambda_T}{4}$ where λ_T is the wavelength of a thermal magnon. In effect, the thermal excitations screen out transverse spin components for excitations of wavelength $\lambda > \lambda_T$. In essence, this is what we have suggested with our physical

argument.

The calculation of the point at which the thermal screening is complete and $\mathcal{O} \propto M$ should be observed is somewhat more complicated. The energy of long wavelength spin waves is given by⁴⁰

$$\begin{aligned} \hbar\omega &= \sum_{\mathbf{R}_1} J_T(\mathbf{R}_1)(1 - \cos \mathbf{k}_1 \cdot \mathbf{R}_1) \\ &\approx k^2 \left\{ \sum_{\mathbf{R}_1 < \mathbf{R}_0} J_T(\mathbf{R}_1) \mathbf{R}_1^2 + \sum_{\mathbf{R}_1 > \mathbf{R}_0} \mathbf{R}_1^2 J(\mathbf{R}_1) \frac{M(T)}{M(0)} \right\} \quad (40) \end{aligned}$$

where $\mathbf{R}_1$ is a lattice vector. For nickel at $T = 500^\circ\text{K}$, $\mathbf{R}_0 \sim 3\text{\AA}$ and the effective exchange coupling scales with the magnetization except for nearest neighbor (and perhaps next nearest neighbor) interactions. Since we are summing over the volume, the term proportional to $M(T)$ is weighted by a factor of the order of $\mathbf{R}_E^5 - \mathbf{R}_0^5 / \mathbf{R}_0^5$, where $\mathbf{R}_E$ is the range of the exchange. Although $J_T(\mathbf{R})(\mathbf{R} < \mathbf{R}_0)$ and $J(\mathbf{R})$ are not known exactly, it is not unreasonable to expect that the second term on the right in (40) will be dominant at and above 500°K where we have observed $\mathcal{O}(T) \propto M(T)$. It should be emphasized that this argument does not apply to thermal renormalization. The rapidly varying short wavelength excitations are much more sensitive to short range interactions. In fact, the above expansion is not valid unless $k\mathbf{R}_E \ll 1$.

From these arguments we would draw the following conclusions. The effectiveness of screening due to thermal excitations depends not only on the density of excitations but also on the range of the exchange. The exchange stiffness will not scale with M until the mean thermal wavelength is much smaller than the range of the exchange. This not only

explains why the temperature at which we observe $\mathcal{O} \propto M$ is so high but would also imply that the effect might not occur at all in systems where the exchange interaction is of short range. Also, equation (40) suggests that the effect will occur uniformly for all spin-waves in the wave vector range $k \ll 1/R_E$. Finally we arrive at the not too surprising conclusion that in nickel $R_E \gg 3\text{\AA}$.

A question arises as to the applicability of this model to nickel. We have seen that the reduction in $\mathcal{O}$ at low temperatures is much too large to be explained by a simple Heisenberg model. However, results of paramagnetic resonance experiments above the Curie temperature⁴¹ cannot be explained by a simple band model. As we mentioned in the introduction, the true picture probably lies somewhere between the two limiting cases, and it is not clear that the model we have considered here is totally inappropriate for nickel. In fact, we find that the existence of and temperature at which the $\mathcal{O} \propto M$ effect is observed is plausible in terms of the theory of Horowitz and Mattis.

Unfortunately the results from Film B above $.8 T_c$ are not sufficiently precise to confirm this behavior. The fact that we have directly measured the high temperature magnetization at only two points raises a further question as to the reliability of the value $T_c = 602^\circ$. For this reason we have compared $\mathcal{O}(T/T_c)$ with $M(T/T_c)$ for values of T_c from 590°K to 627°K . In this range the assumption that $d\mathcal{O}/dM = \mathcal{O}/M$ is consistent with the results above $.8T_c$ within the experimental error. At $T_c = 590^\circ$ the value of $(d\mathcal{O}/dM)/(\mathcal{O}/M)$ immediately below $.8 T_c$ appears to be slightly less than in Film A, and at $T_c = 627^\circ$ it is

somewhat larger. For $T_c \sim 600^\circ\text{K}$ the agreement with Film A is excellent from $.8T_c$ to the lowest temperature values. This can be seen in Fig. 21 where $(d\sigma/dM)/(\sigma/M)$ is plotted vs T/T_c for both films. That the agreement is good for $T_c(B) \approx 600^\circ\text{K}$ is not unexpected. As mentioned above, the slightly more rapid fall off of $\sigma(T)$ in Film B is consistent with a lower Curie temperature.

The results from Film A if not absolutely conclusive are strongly suggestive that above $.8T_c$ σ varies directly as M . The results from Film B are in such close agreement with those of A below $.8T_c$ that it seems probable the behavior above $.8T_c$ is similar. In both films the physical properties which we could determine (e.g. σ , $\Delta H(450^\circ)$, g) were in reasonably good agreement with bulk values, but the reduced Curie temperatures shows that the samples were not pure. While this fact indicates that the quantitative temperature dependence of σ is probably not particularly reliable for pure bulk material, it would not appear to reduce the significance of the high temperature results.

Our understanding of the observed $\sigma(T) \propto M(T)$ effect is predicated neither on the fact that the sample is nickel nor on the fact that it is absolutely pure. If our arguments are valid, then this effect could presumably be observed in other materials.

The only previously reported measurement of exchange stiffness in films above room temperature is that of Nose²⁹ who studied two Cu-Ni samples. One was 22% Cu with a Curie temperature of 130°C and the other 15% Cu with $T_c = 210^\circ\text{C}$. In each case spin waves could not be resolved within 30° of T_c . The magnetization of the films was measured independently. In both films Nose noticed that σ was decreasing more rapidly

than M . However the upper limit of resolution in the 22% Cu film was less than $.8T_c$ and in the 15% Cu sample was $\sim .85T_c$. In neither case was extensive data taken near the high temperature limit nor a careful study of $(d\sigma/dM)/(\sigma/M)$ made. Nose's results, while not encouraging are not necessarily inconsistent with ours. Furthermore, since there have been no quantitative theoretical predictions as to when the $\sigma \propto M$ effect should occur, it is possible that in other systems the onset would not appear at the same value of T/T_c which we have observed. And in fact, if the range of exchange is smaller in Cu-Ni than in Ni, we would expect the effect to occur at a higher value of T/T_c .

Finally it should be pointed out that assumed absence of transverse spin correlation is just the random phase approximation (RPA). We wish to emphasize that we are not suggesting that the RPA is valid to describe the thermodynamic properties of the system. We do feel though that it is valid over the limited region of the magnetic excitation spectrum which we observe.

IV. CONCLUSIONS

We have found in our study of the temperature dependence of the exchange stiffness of nickel that above $.8 T_c$ $\bar{\alpha}$ appears to scale directly with M . This would seem to indicate that at these elevated temperatures transverse spin correlations play a negligible role in the energy renormalization of the long wavelength excitations which we observe. Our understanding of this phenomenon is based principally on physical arguments rather than a rigorous theoretical prediction. However, we see no reason why this effect should not be observable in other ferromagnetic systems where the exchange is of sufficiently long range.

High temperature measurements of the magnetization by resonance indicated that the Curie temperature of these films was less than the bulk value. The precise quantitative variation of $\bar{\alpha}$ with respect to T is therefore not to be taken as a reliable bulk measurement. However, the fractional change in $\bar{\alpha}$ between 120 K and 295 K was an order of magnitude too large to be explained on the basis of a Heisenberg model, even taking into account the reduced critical temperature.

Detailed studies of the intensity variation of standing spin wave modes in several films led us to conclude that mode pinning in nickel is primarily a volume inhomogeneity effect, as suggested previously by Nose.²⁹ It is therefore likely that "bulk quality" films will exhibit little or no surface spin-pinning. Although unpinned modes cannot be excited by a uniform microwave field, we have found that a non-uniform exciting field will couple to such modes.

A non-uniform field can be obtained in a transmission spectrometer

which we used to study room temperature standing spin wave resonance spectra in 81-19 permalloy. We have observed an interference effect between the propagating electromagnetic and photon modes in the film. Analysis of the observed effect gives additional information about the internal field in the film.

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Table I.

L	$4\pi M$	g	$\mathcal{D}$	$2/\gamma\Delta H$	T_2 (transmission)
$\sim 1200\text{\AA}$	$10.8 \times 10^3 \text{ gauss}$	2.12	$.046 \text{ cm}^2 \text{ sec}^{-1}$	$1.8 \times 10^{-9} \text{ sec}$	$1.4 \times 10^{-9} \text{ sec}$
$\sim 2000\text{\AA}$	$10.4 \times 10^3 \text{ gauss}$	2.09	$.042 \text{ cm}^2 \text{ sec}^{-1}$	$1.2 \times 10^{-9} \text{ sec}$	$.8 \times 10^{-9} \text{ sec}$

Table II.

	$\rho_{295^\circ K} (\text{cm}^2 \text{sec}^{-1})$
Inelastic neutron scattering:	
Nathans et al. ¹¹	.0568
Stringfellow ¹⁷	.0612
Spin Wave Resonance	
Nose ²⁹	.0538
Phillips ¹⁶	.0573
Holzer	
Film A	.0565
Film B	.0559

Fig. 1 Microwave transmission spectrometer

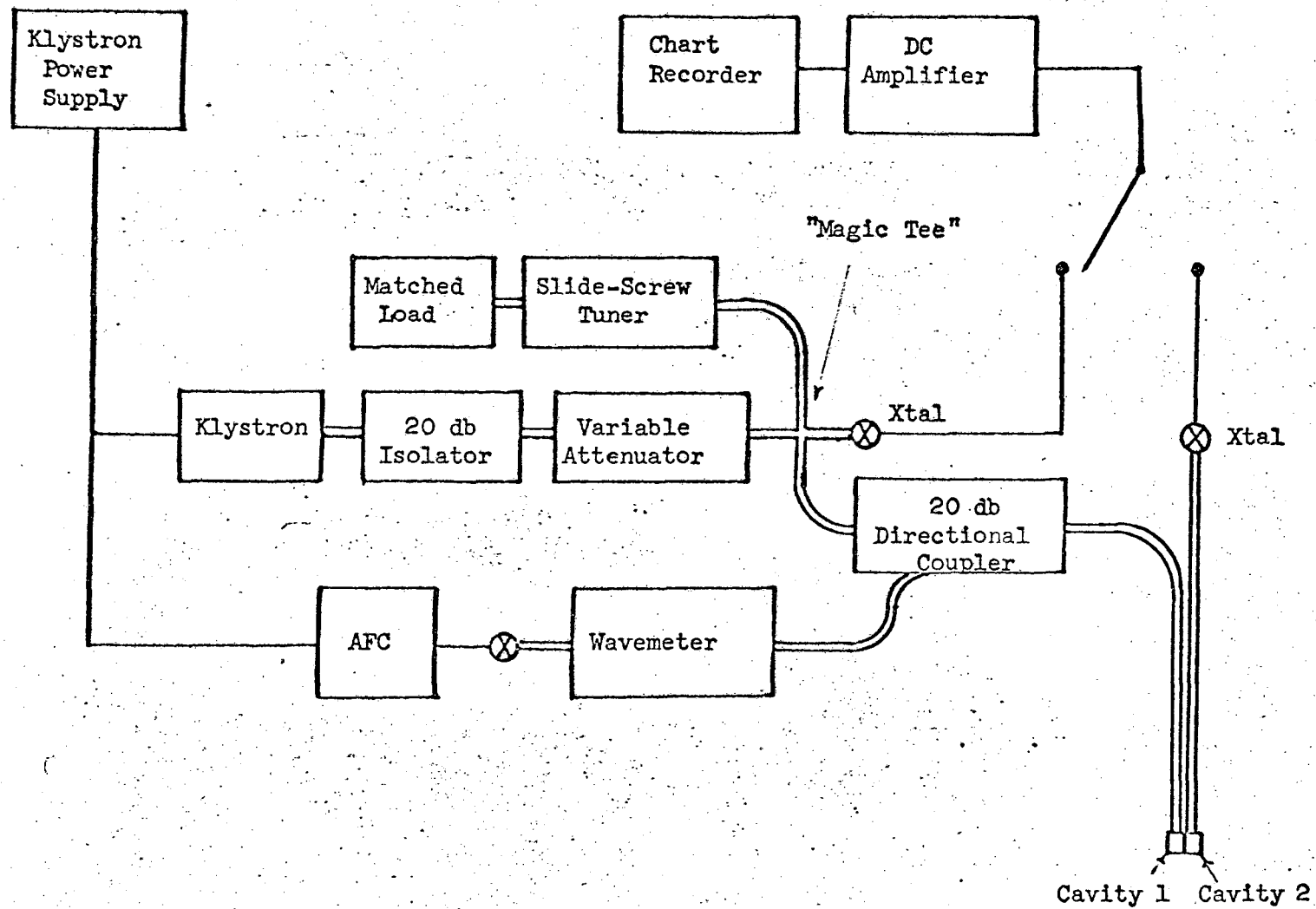


Fig. 2. Cross section of two cavity system showing sample placement and microwave magnetic field configuration. Cavity 2 is cut along a current node to allow sample insertion and removal.

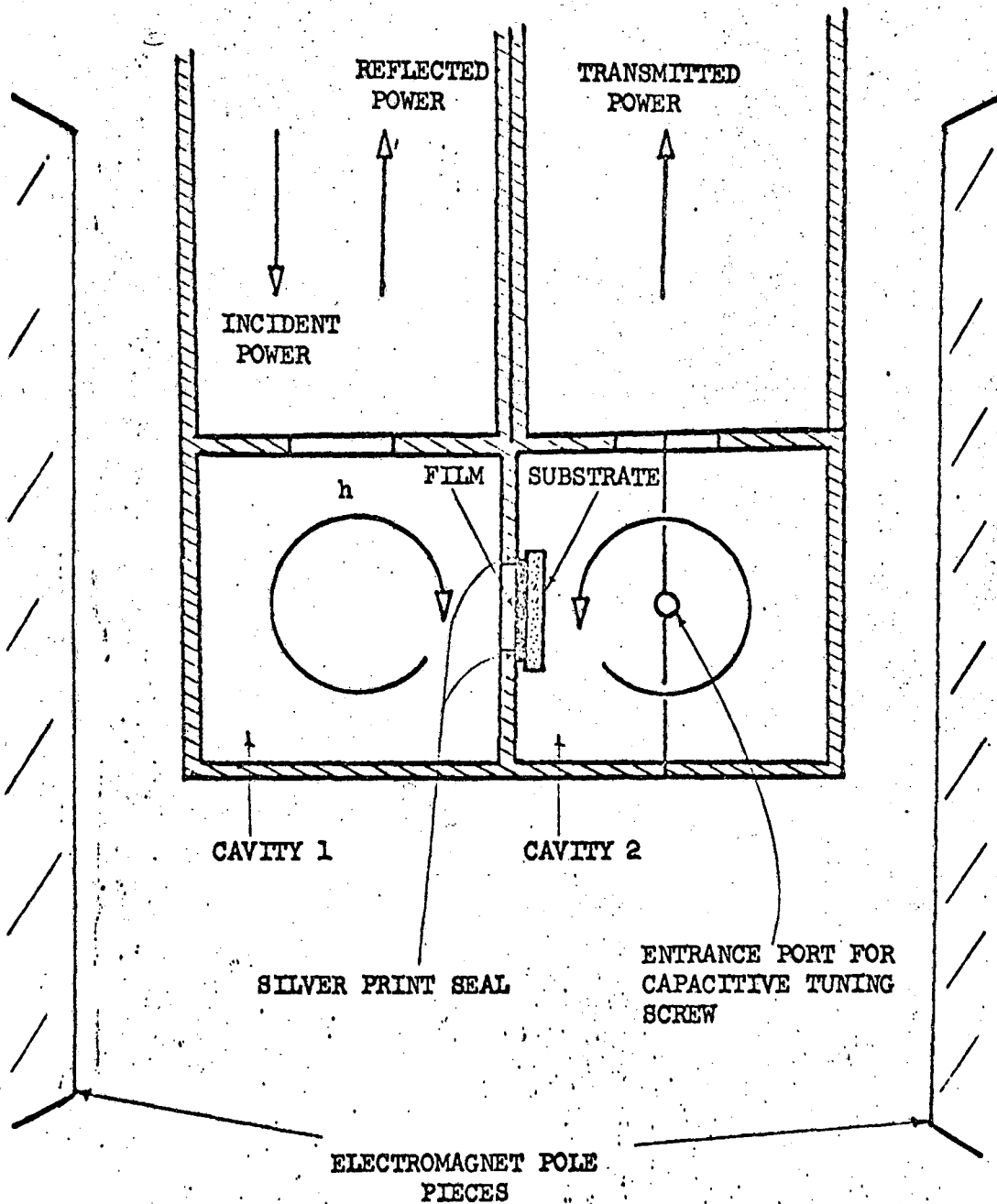
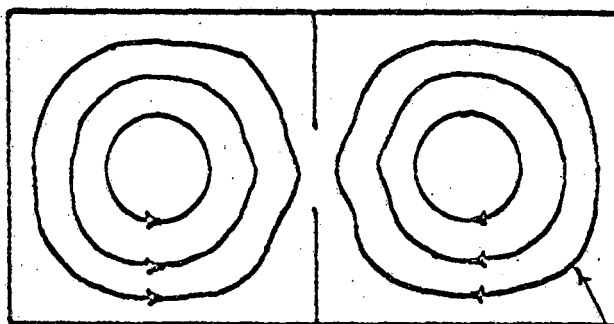
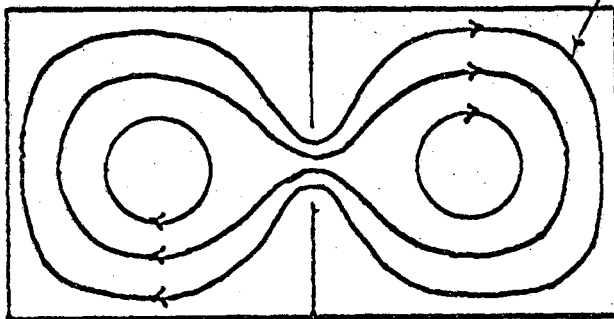


Fig. 3 Normal modes of coupled two-cavity system



Mode 1



Mode 2

h

Fig. 4 Perpendicular field resonance spectra for 1200Å⁰ film. The top trace was taken with the sample in a single reflection cavity and the lower traces with it in the transmission system. The high field mode is identified as $n = 1$.

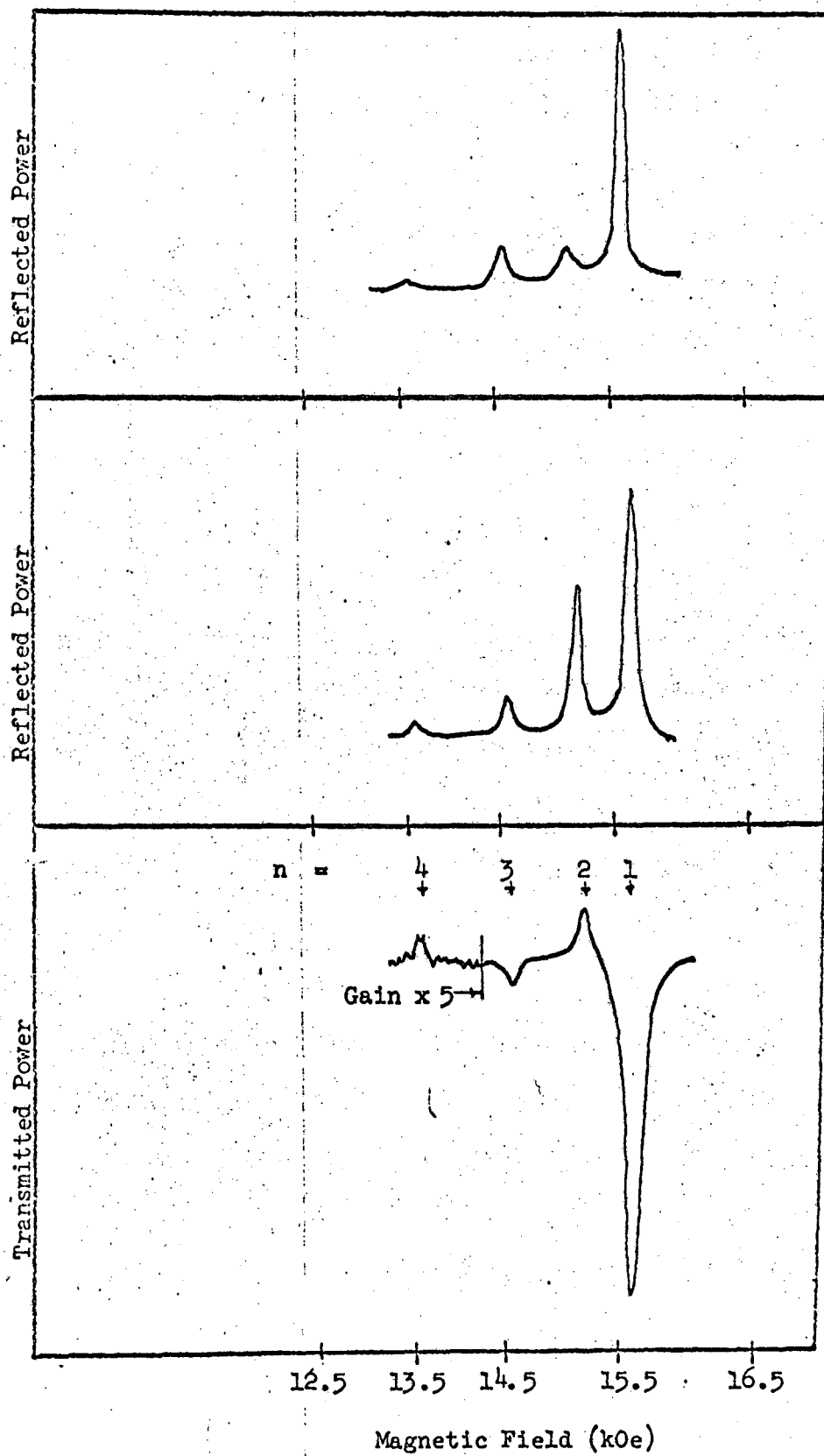


Fig. 5 Critical angle ($\phi_c = 14^\circ$) resonance spectra for 1200Å film. With the sample in a single cavity (top trace) only the $n = 0$ mode is detected. In the transmission system (lower traces) a second mode identified as $n = 1$ is detected.

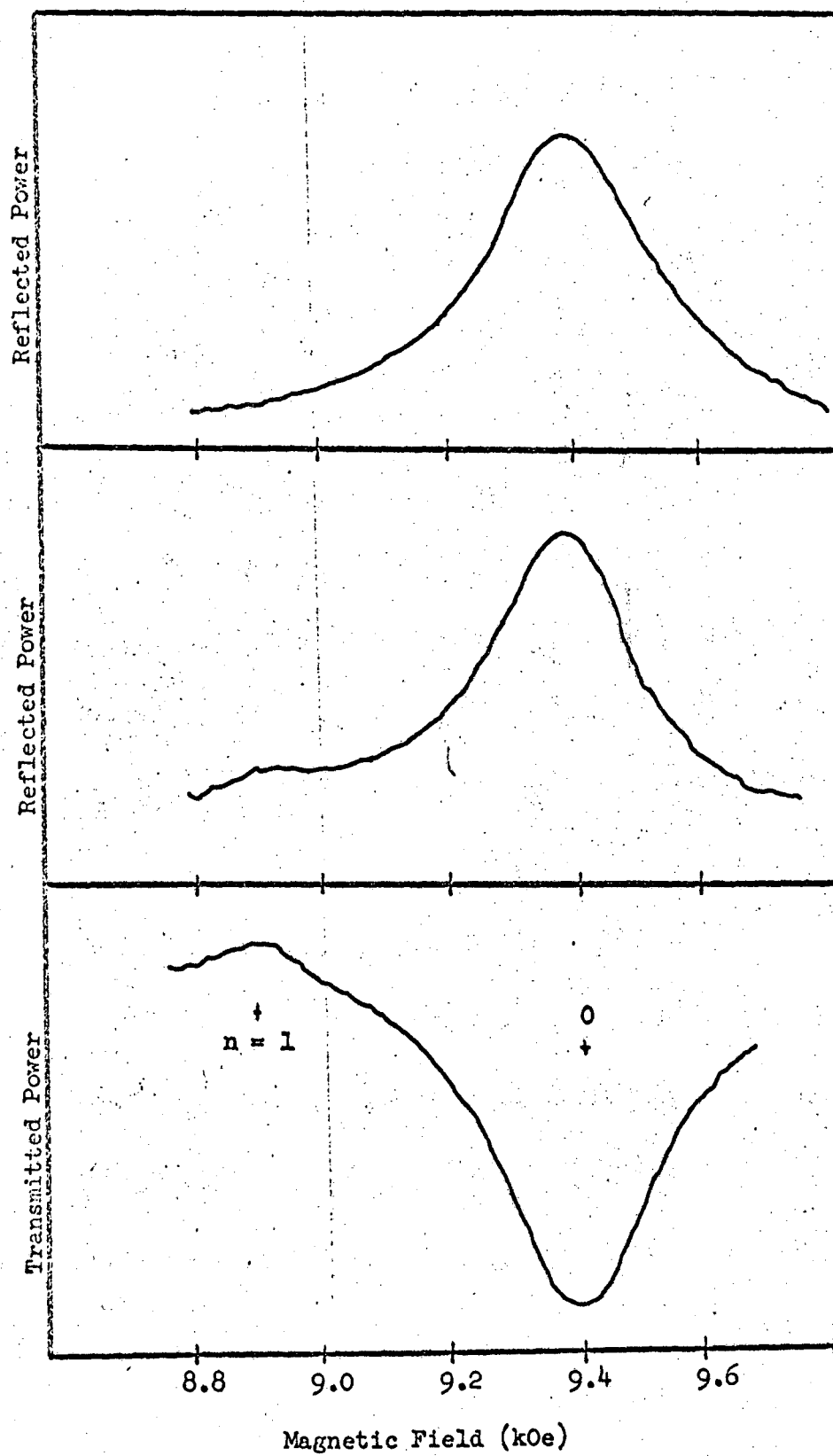


Fig. 6 Parallel field resonance spectra for $1200\text{\AA}$ film. The top trace was taken with the sample in a single reflection cavity and the lower traces with it in the transmission system. The high field mode is identified as $n = 0$. In the transmission spectrum mode $n = 2$ is not resolved.

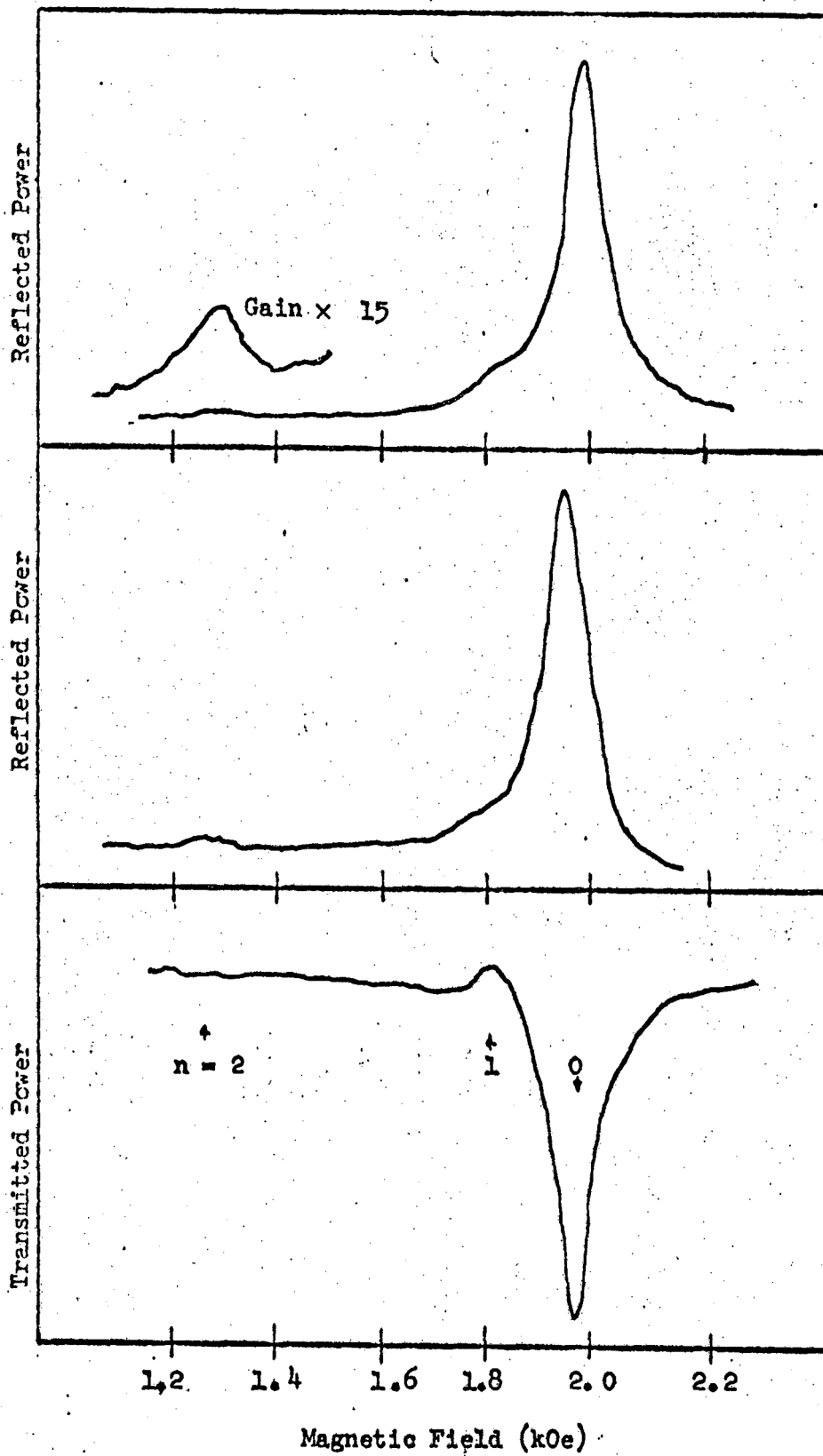


Fig. 7 $H_0 - H_n$ is plotted against n^2 . The value of H_0 at perpendicular field was determined by extrapolation. The smaller slope at perpendicular field indicates a larger effective film thickness.

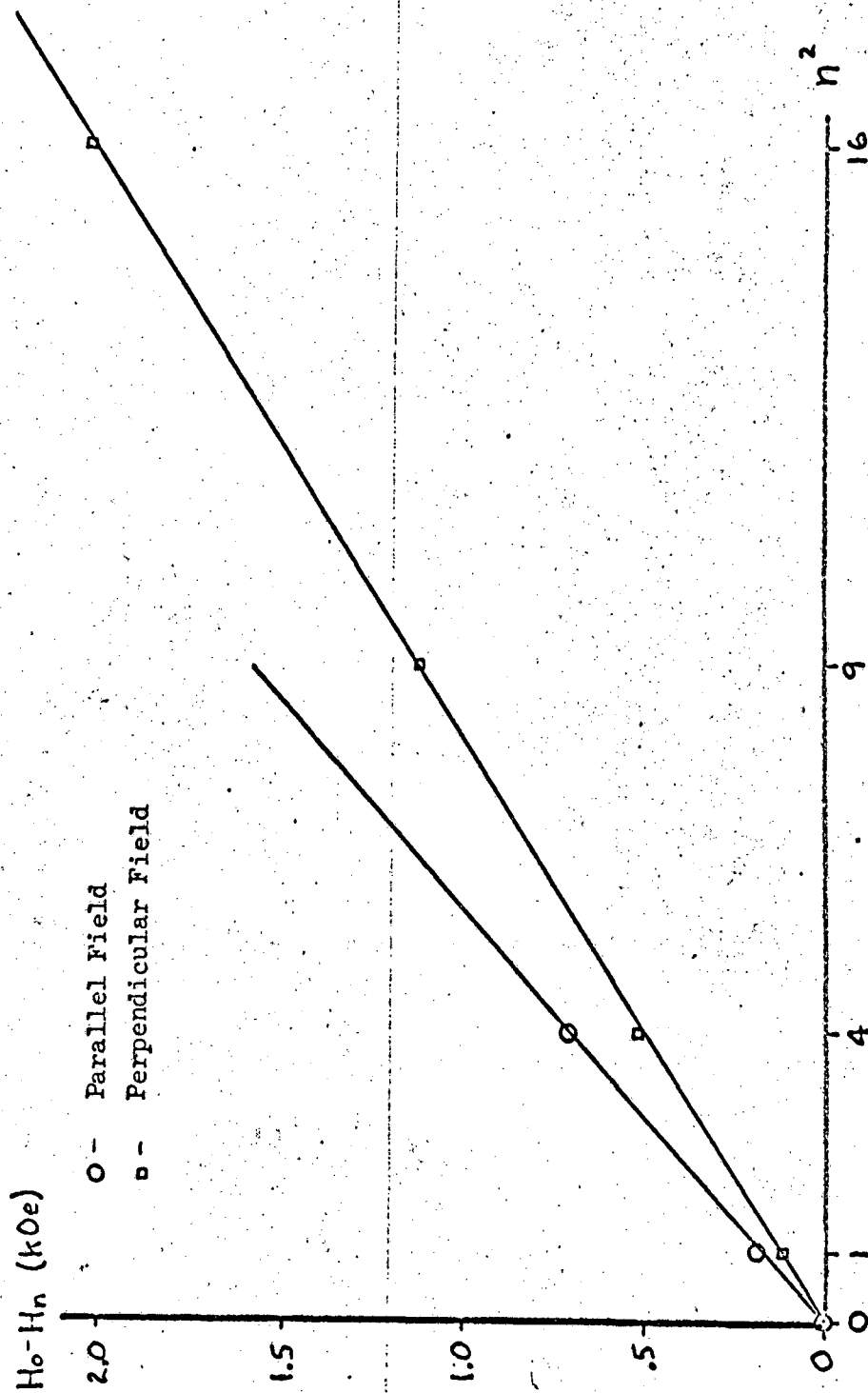


Fig. 8 Equivalent circuit for the two cavity transmission system. The coupling of power through the film is represented by the mutual inductance M .

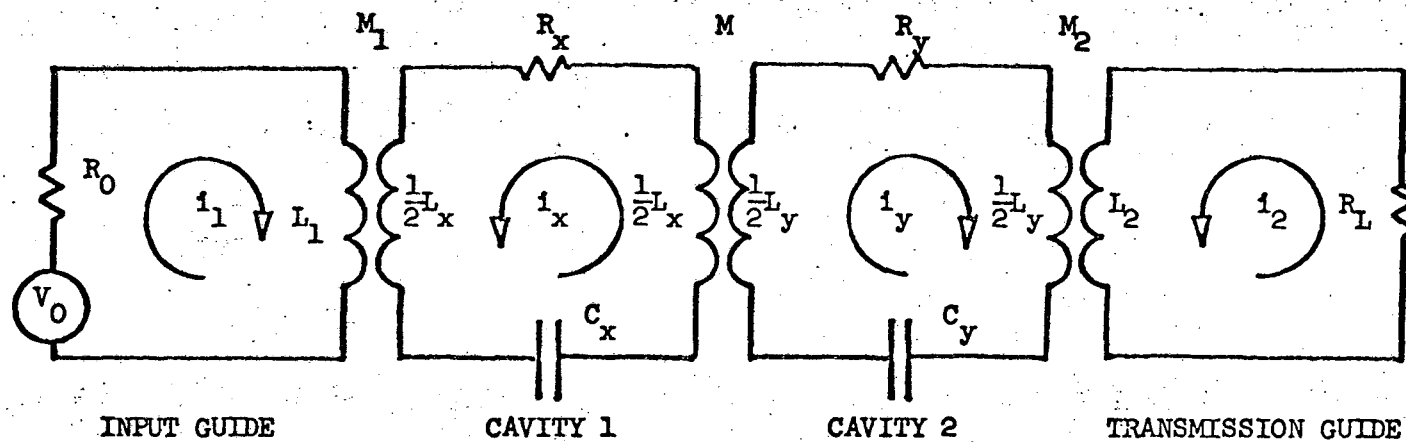


Fig. 9 Cross-section of the high temperature cavity.

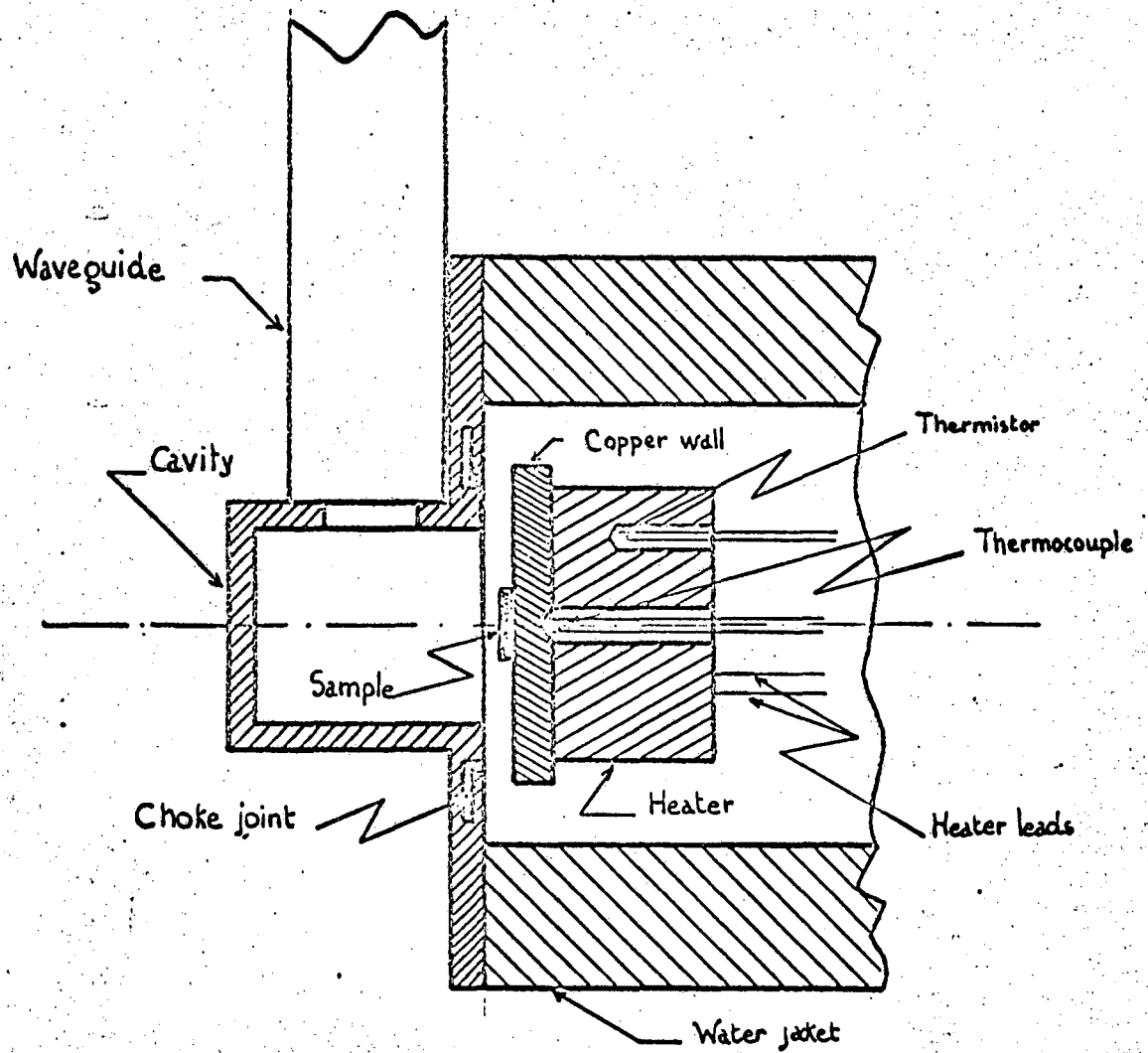


Fig. 10 Schematic diagram of temperature control system.

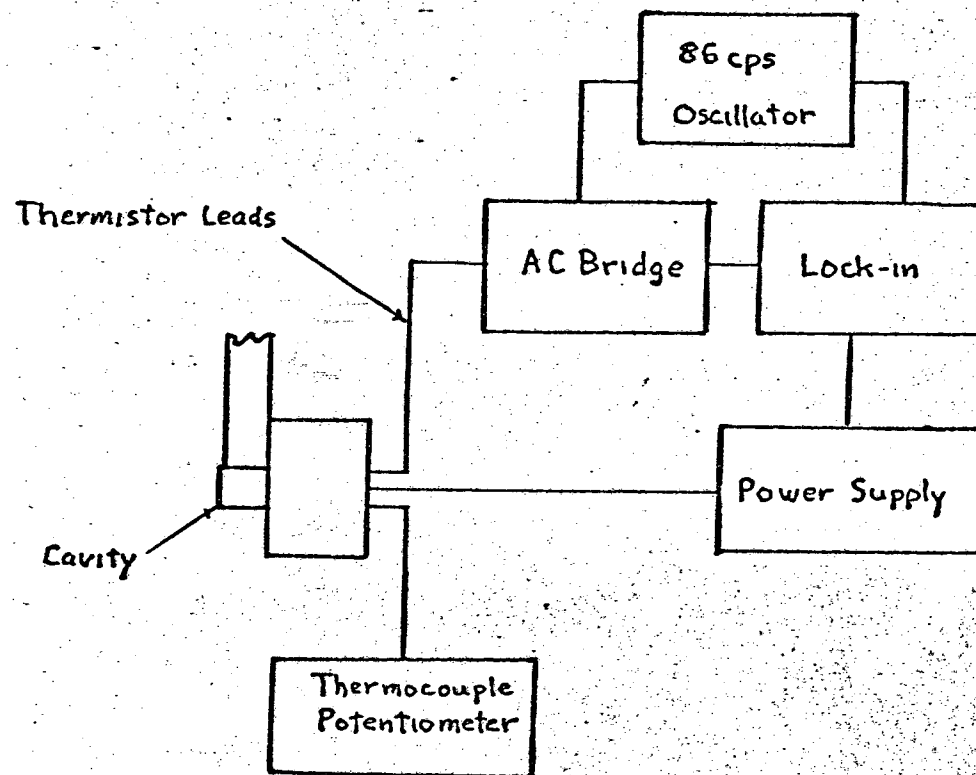


Fig. 11 Experimental trace of the derivative of power absorption, dP/dH , as a function of the applied field, H , for Film A at 295°K .

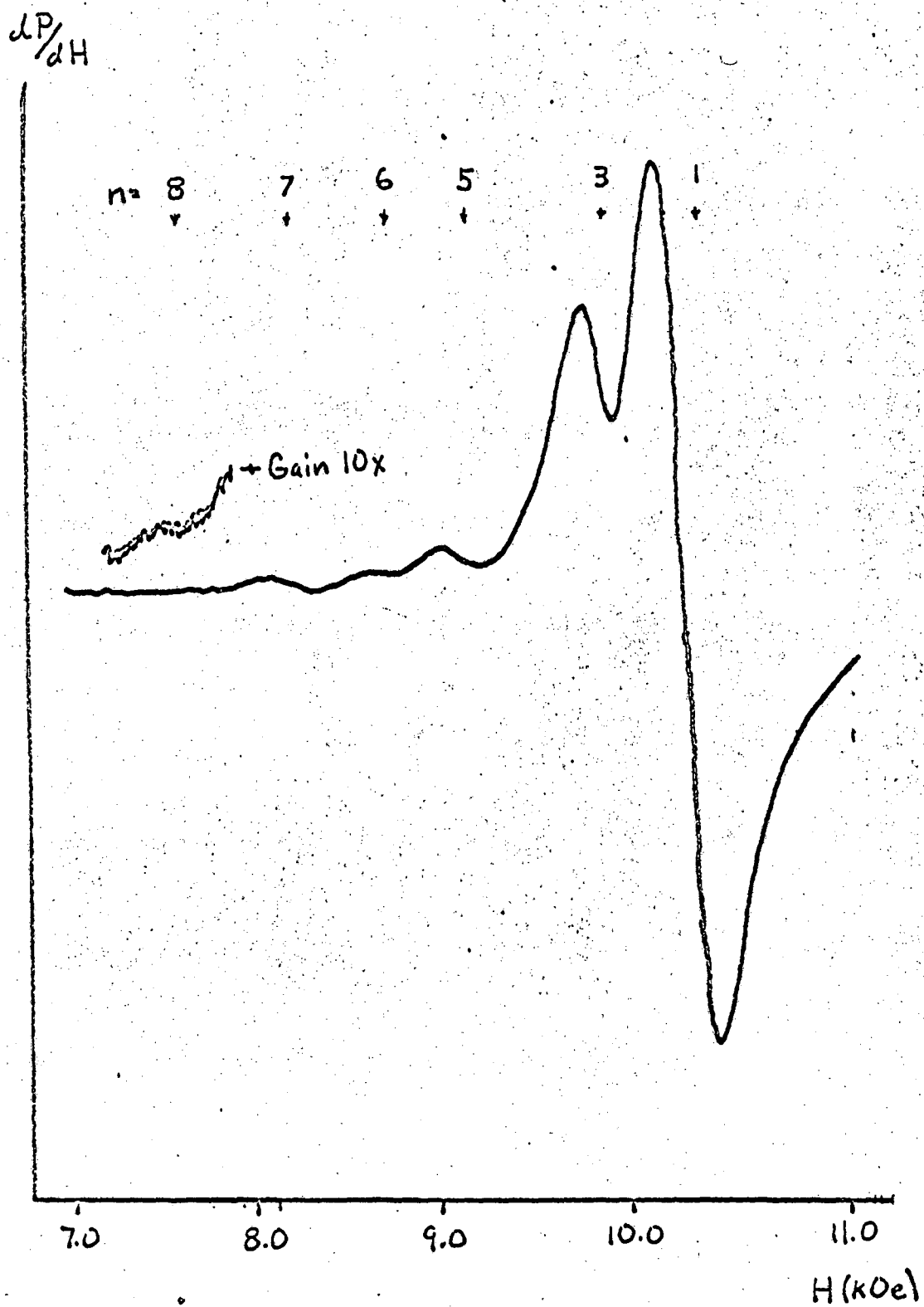


Fig. 12 Experimental trace of the derivative of power absorption, dP/dH , as a function of the applied field, H , for Film B at 295°K.

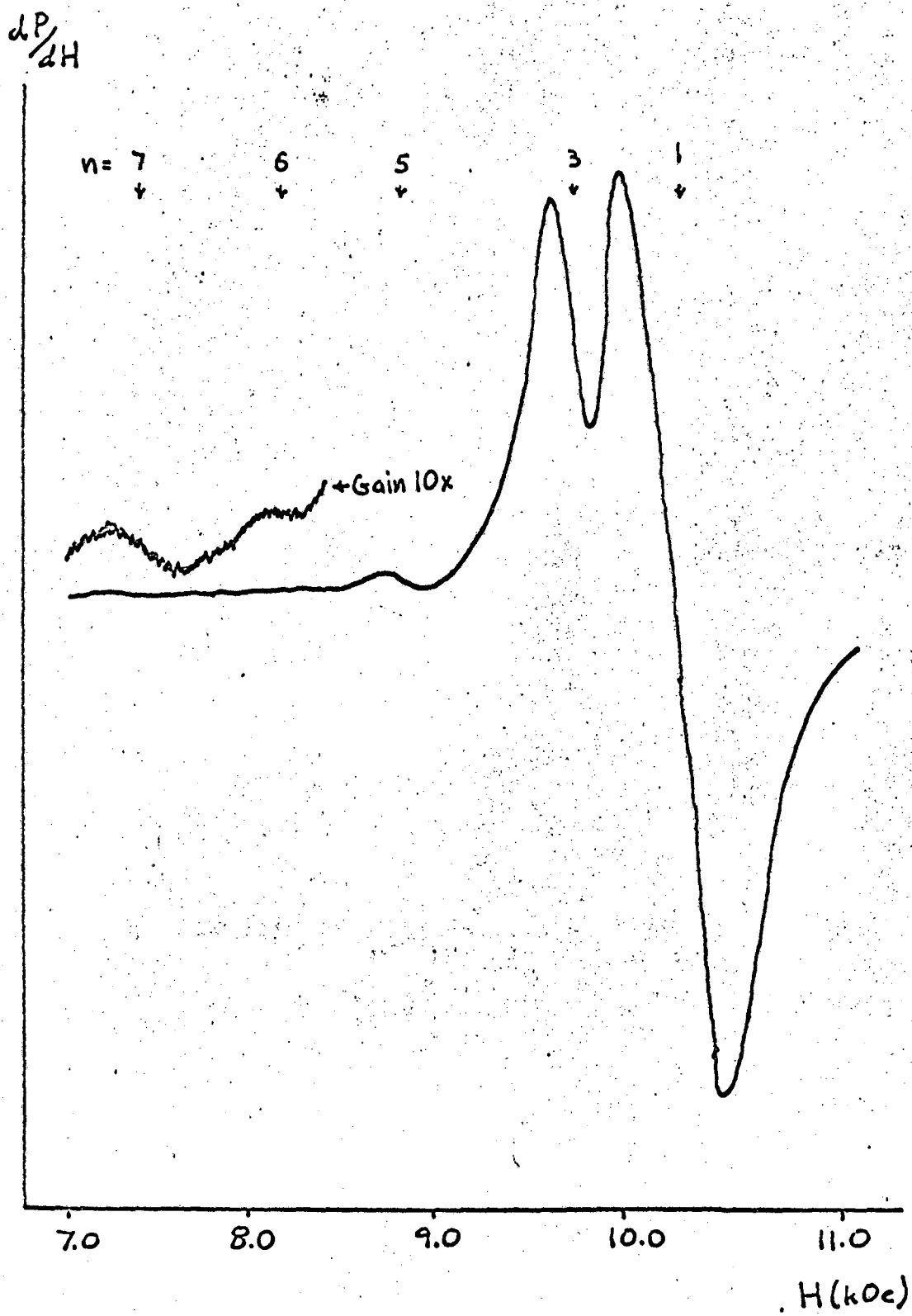


Fig. 13 Plot of resonance field, H , vs. n^2 for spectrum of Film A as given in Fig. 11.

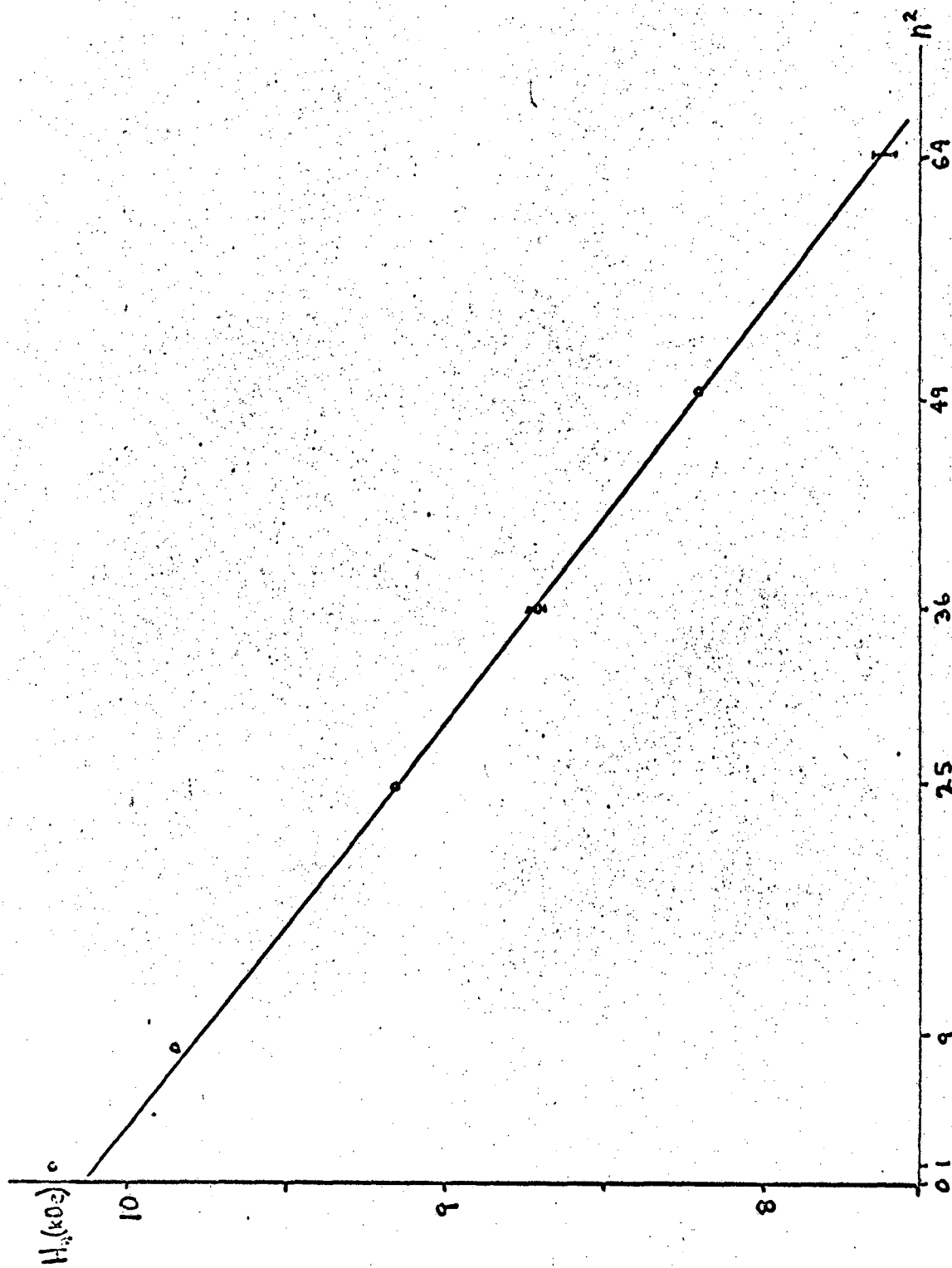


Fig. 14 Plot of resonance field, H_n vs. n^2 for spectrum of Film B as given in Fig. 12.

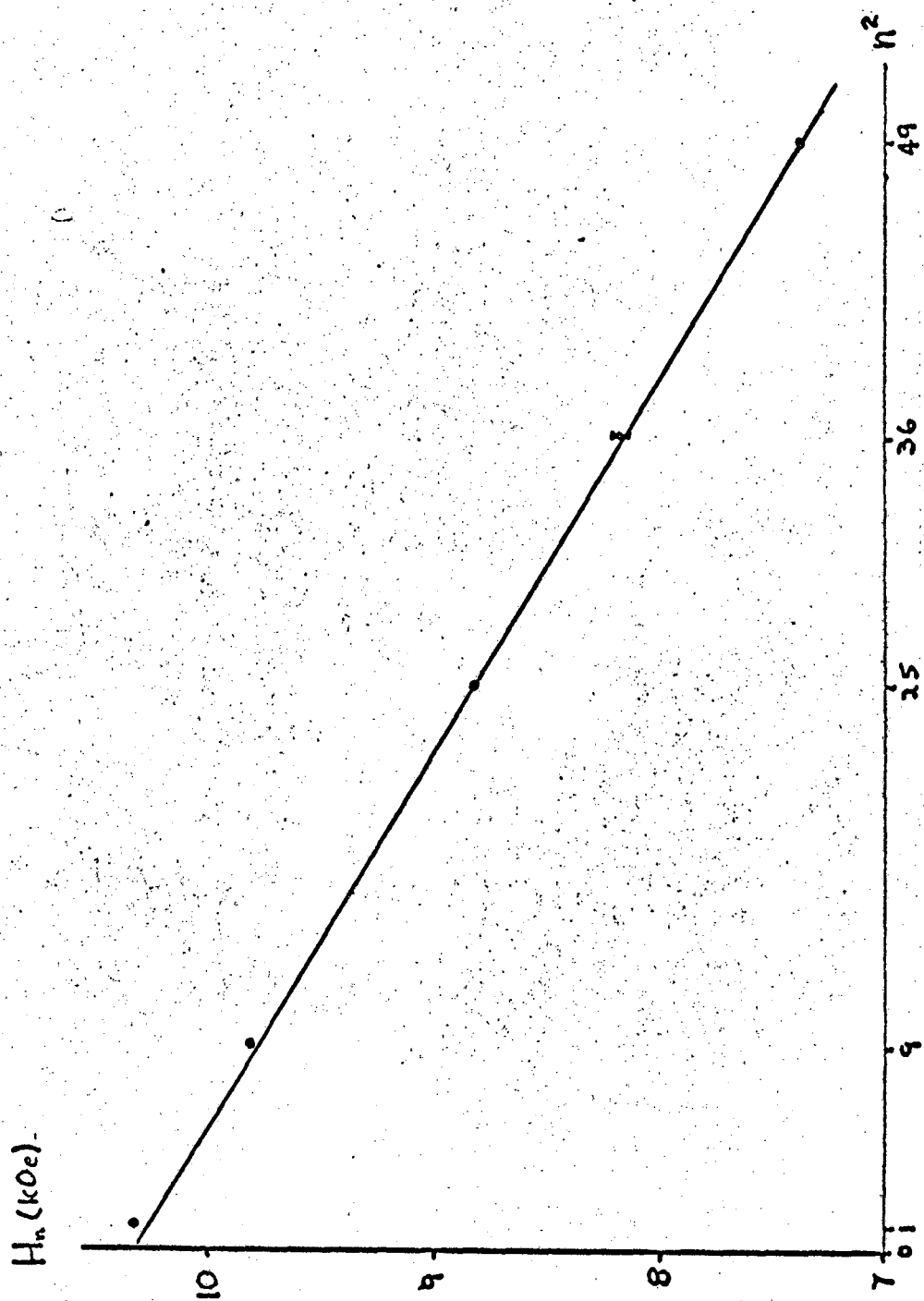


Fig. 15 Plot of specific magnetization, $\sigma(T)$, in a 3.5 kOe field as determined by Weiss and Forrer.³⁷ Values obtained by resonance are compared with $\sigma(T/.976)$ for Film A and $\sigma(T/.96)$ for Film B.

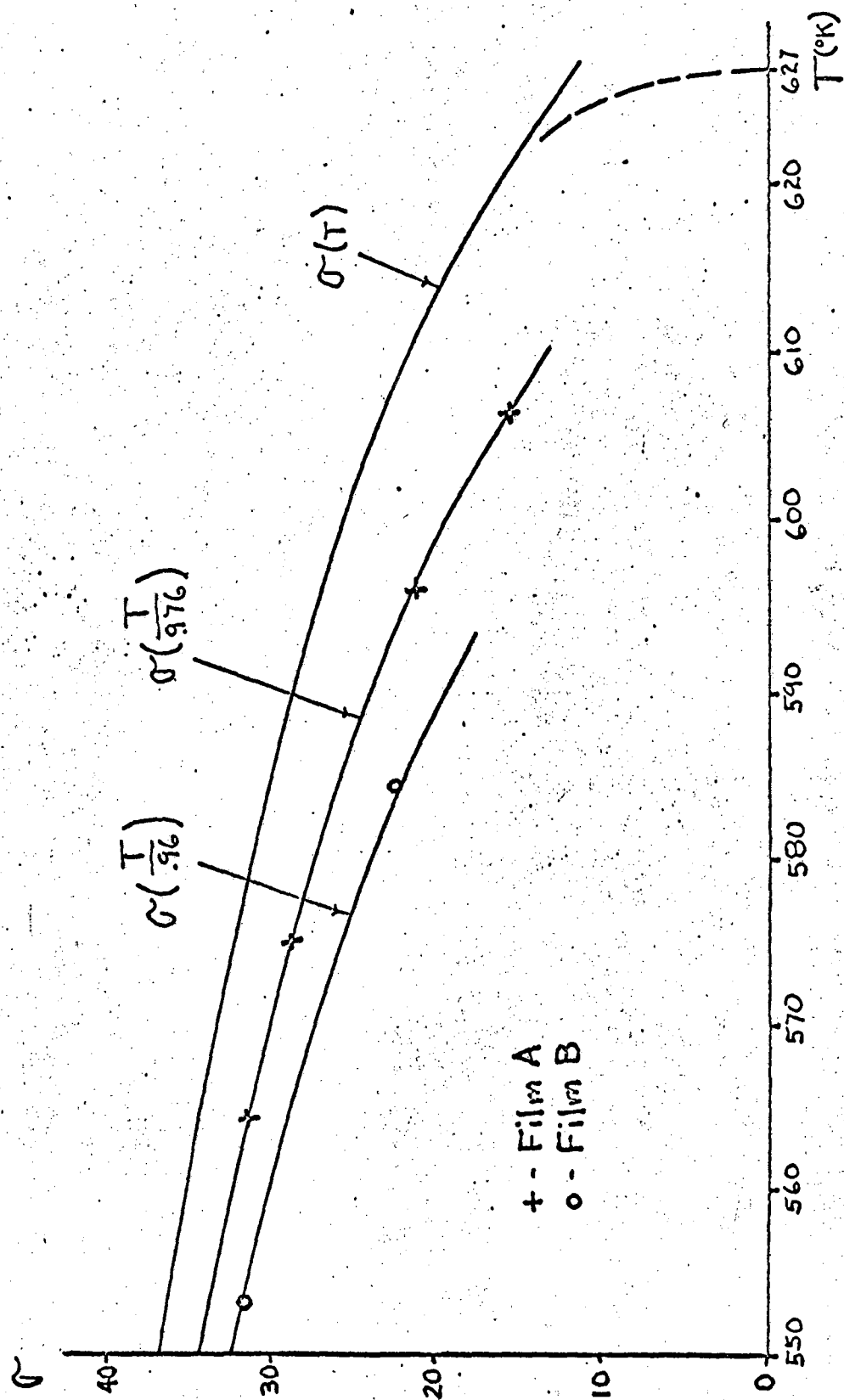


Fig. 16 $H_5 - H_7 = \frac{24\pi^2}{\gamma_L^2} D$ is plotted vs. T for Film A.

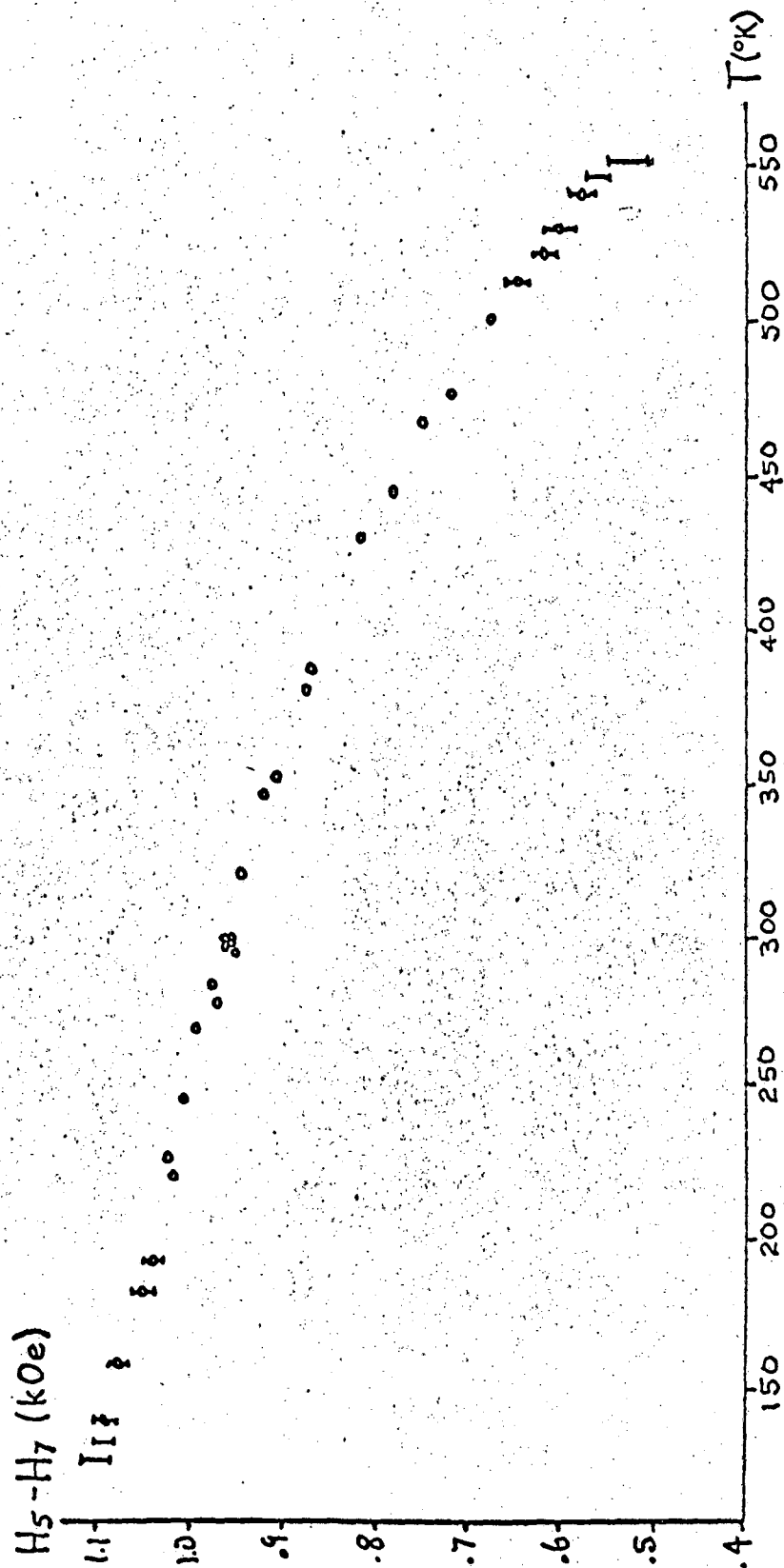


Fig. 17 $H_5 - H_7 = \frac{24\pi^2}{\gamma_L^2} \mathcal{D}$ is plotted vs. T for Film B.

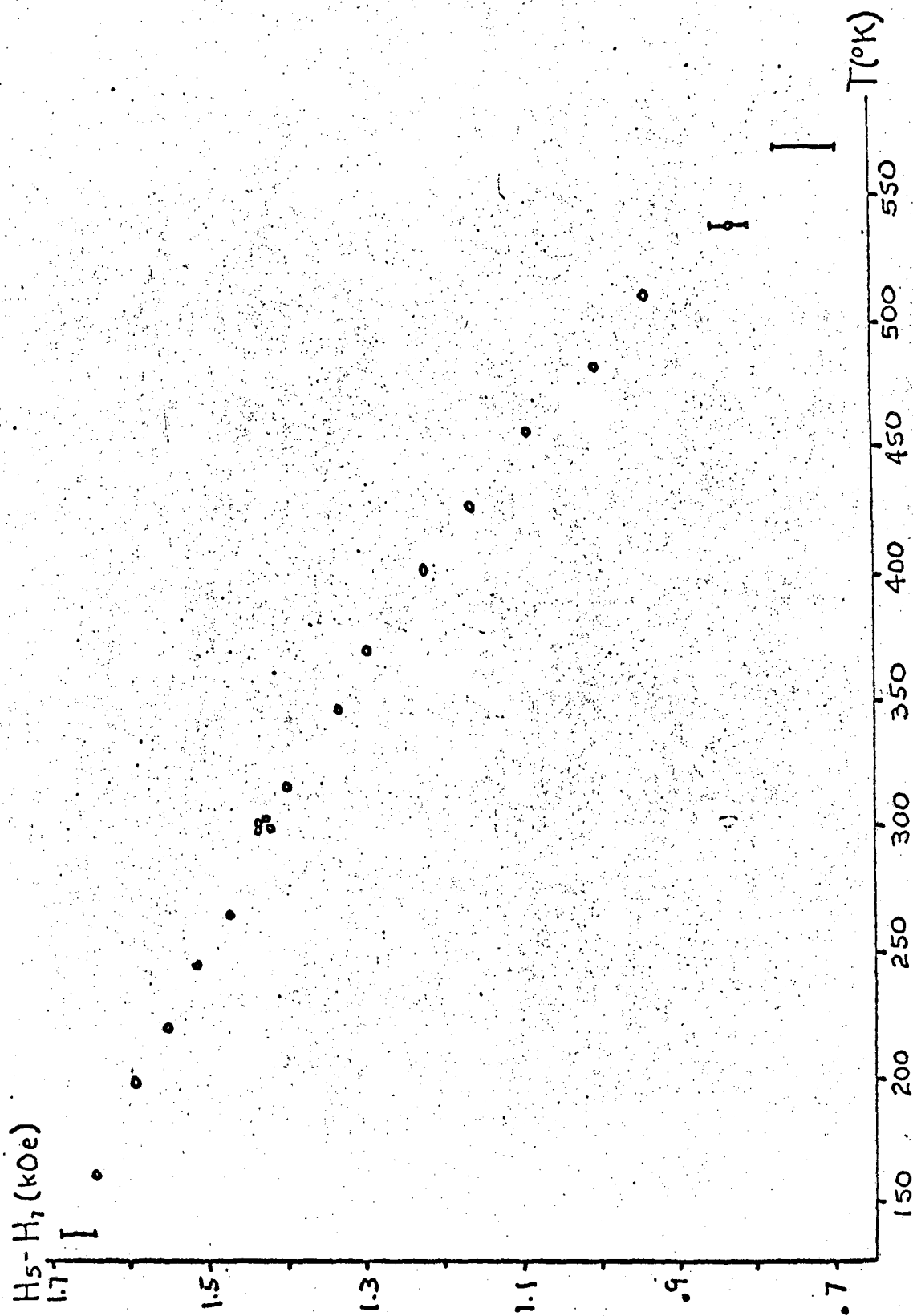


Fig. 18 $H_5 - H_7 = \frac{24\pi^2}{\gamma_L^2} D$ is plotted vs. $T^{3/2}$ ($T < 300^\circ K$)
for Film A.

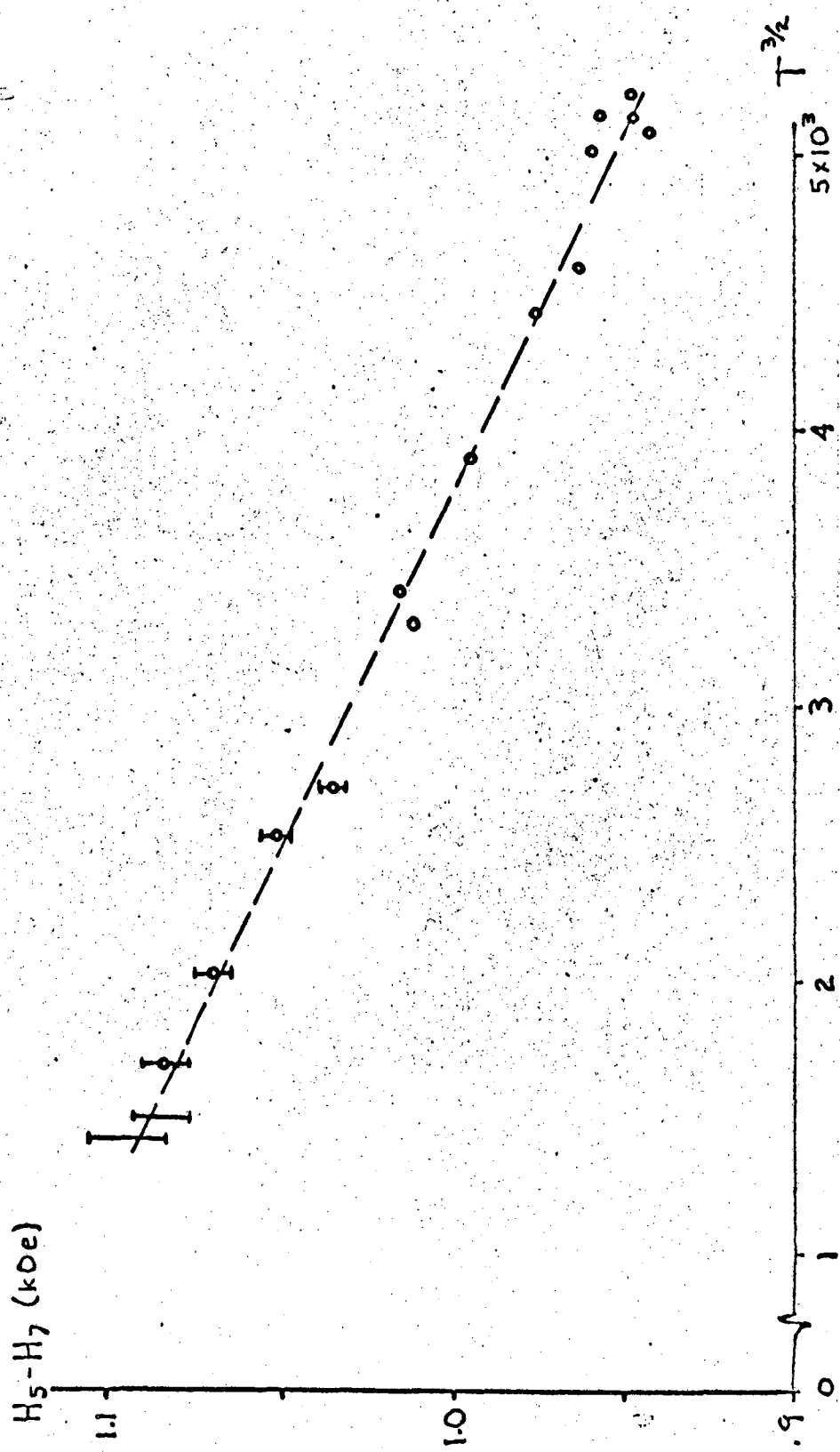


Fig. 19 $H_5 - H_7 = \frac{24\pi^2}{\gamma_L^2} D$ is plotted vs. $T^{3/2}$ ($T < 300^\circ K$)
for Film B.

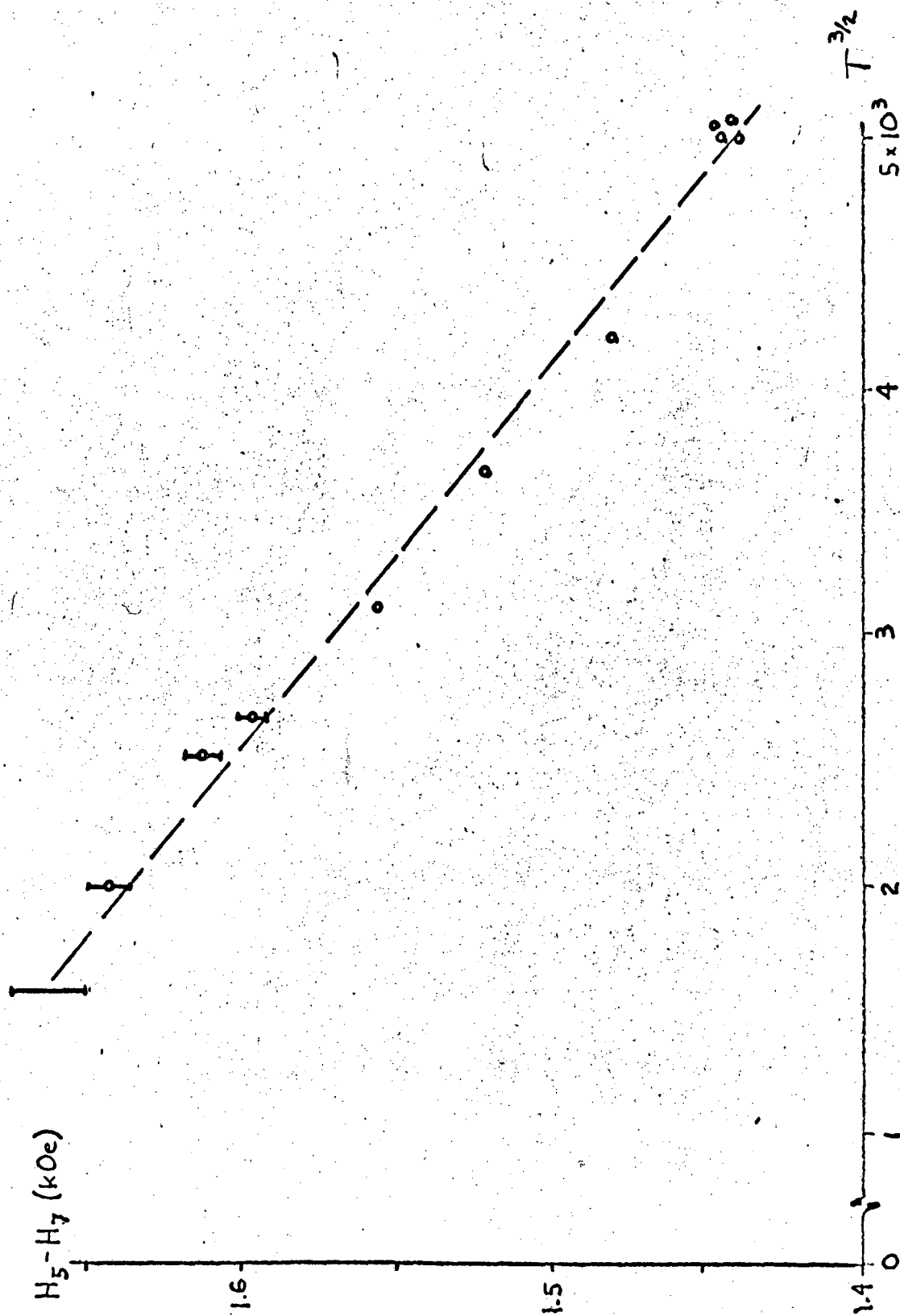


Fig. 20 The variation of $\mathcal{D}$ with respect to M . M is normalized to $M(T=0)$ and has been scaled to $T_c = 612^\circ\text{K}$. $\mathcal{D}$ is given in terms of H_5 - H_7 .

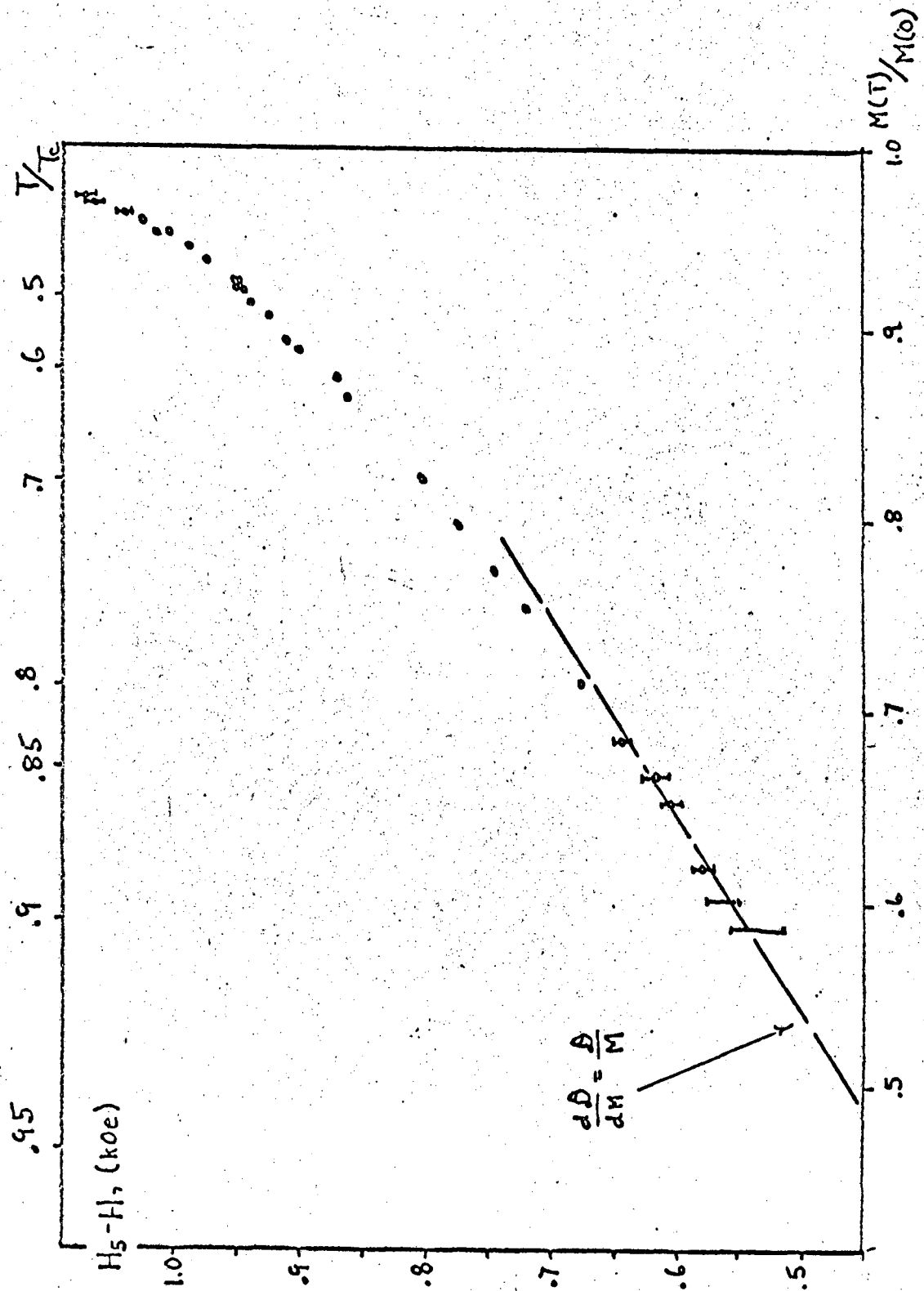
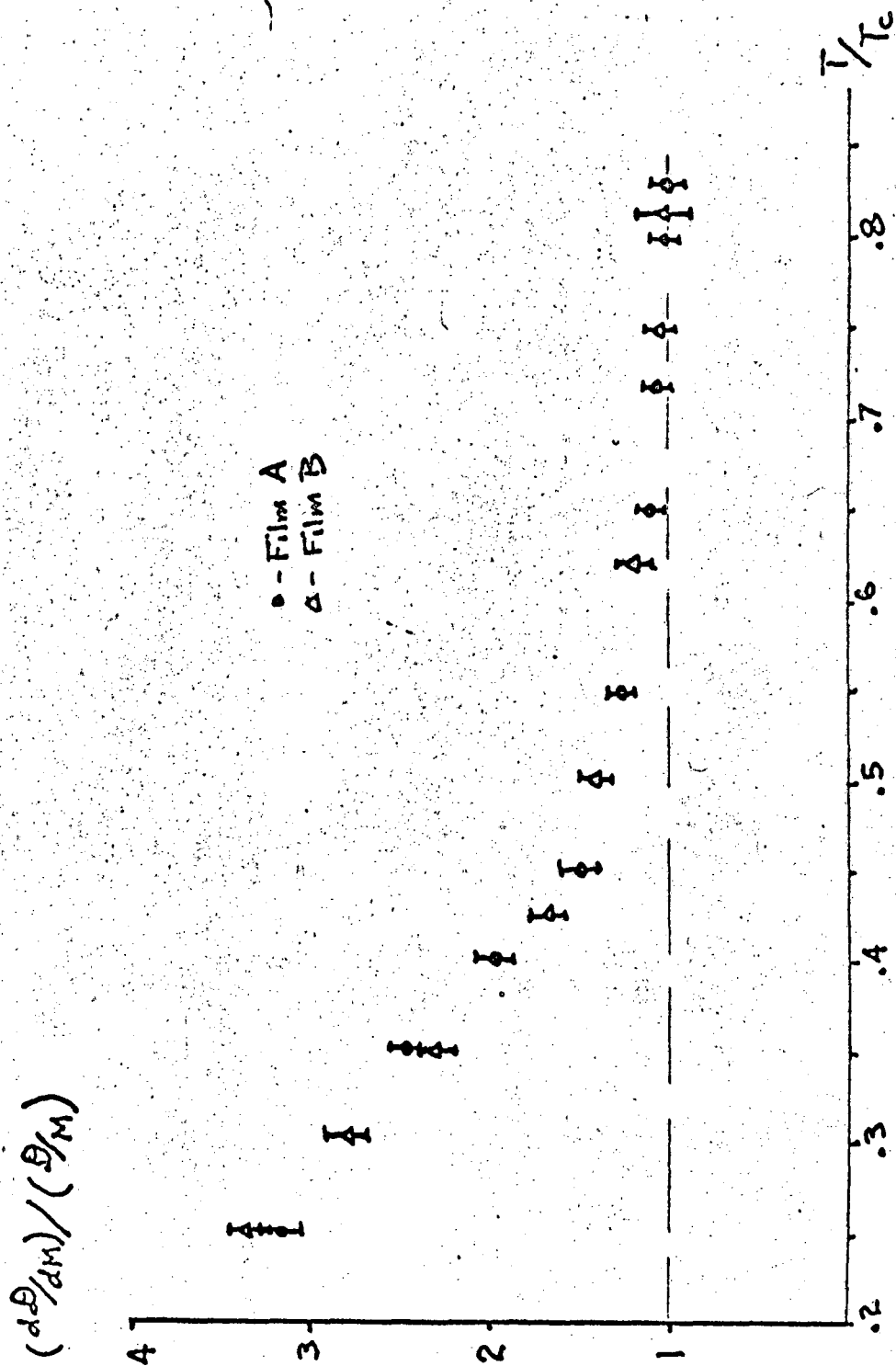


Fig. 21 $(dD/dM)/(D/M)$ vs. T/T_c . The values of T_c are 612°K for Film A and 600°K for Film B.



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