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MICROWAVE RESONANCE IN THIN FERROMAGNETIC FILMS (1)

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Résumé. — Des couches minces de permalloy ont été étudiées par les techniques de résonance magnétique. L'aimantation à saturation a été mesurée en fonction de l'épaisseur. On a fait une analyse théorique des effets d'échange en résonance ferromagnétique dans les couches minces, et divers critères pour l'observation de tels effets sont comparés avec les résultats expérimentaux préliminaires. Un nouveau phénomène a été observé, dans lequel un signal de résonance est obtenu aux extrémités de deux fils conducteurs soudés à la couche.

Abstract. — Thin films of permalloy have been investigated by microwave resonance techniques. The saturation magnetization has been measured as a function of thickness; harmonic generation has been considered. A theoretical analysis has been carried out of exchange effects in ferromagnetic resonance in thin films, and various criteria for observing such effects are compared with preliminary experimental results. A new phenomenon has been observed, in which a d.c. resonance signal is taken off two wires soldered to the film.

In addition to a long-standing interest in thin ferromagnetic films for investigations into basic magnetism, films have taken on added significance in recent years because of their potential use as fast switching devices. In this paper we will be concerned with fundamental magnetic properties of permalloy films (~80% Ni; 20% Fe) — in particular with the behavior as brought out by microwave resonance experiments. The original discovery of ferromagnetic resonance (FMR) was made in thin films of iron, nickel and cobalt [1], and subsequent resonance experiments in nickel films were carried out by Macdonald [2]. For some years afterwards, microwave resonance in ferromagnetic films was neglected in favor of bulk metals and ferrites. Recently there has taken place a revival of interest [3], and experience gained from other resonance experiments has been applied to thin films [4].

We have continued our thin film work by using the microwave resonance technique to study saturation magnetization, exchange effects, harmonic generation and d.c. generation by FMR effects. Each one of these topics will now be taken up separately.

\[ T = 4\text{K} \]
\[ T = 78\text{K} \]
\[ T = 300\text{K} \]

Fig. 1. — Magnetization as a function of thickness based on microwave measurements at 9 GHz.

A. Magnetization. — Microwave resonance affords a means of measuring the magnetization of thin films. Determination of $\Delta M$, as well as $g$,
is possible from two resonance measurements in two geometries — the dc field parallel and perpendicular to the plane of the film. More conventional techniques have been used previously for obtaining the magnetization [5-7].

The results for $4\pi M$ as a function of thickness in permalloy from microwave resonance at room temperature, at 78°K and at 4 °K are shown in figure 1. The room temperature curve has a sharp break at 60 Å, and 50°% of the bulk magnetization remains at 25 Å. Other details have appeared previously [8]. Comparison between spin-wave theory and experiment is still not quite satisfactory, as can be seen from a recent paper by Glass and Klein [9]. A different approach to obtain a theoretical expression for the magnetization of thin films has been used by Valenta [10]. His case is based on a generalization of the Weiss-Néel theory of molecular fields, in which the film is considered to be composed of a number of magnetic sublattices formed by atomic planes. It would indeed be interesting to apply the equation of motions for the magnetization to Valenta’s sublattice model in thin films — especially in view of the success enjoyed by the sublattice model in explaining ferri- and antiferromagnetic resonance. However, the difficulties in carrying out such a solution seem rather formidable.

B. Exchange Effects. — A phenomenon which may be studied by means of thin films is the effect of exchange anisotropy on the ferromagnetic resonance line. An exchange anisotropy field arises because of the small skin depth in metals and is physically due to the non-uniform variation of the spin precession angle in the direction of electromagnetic propagation. This effect leads to a damping of the resonance even in the absence of relaxation damping — essentially because the exchange forces prevent wide opening up of the precession angles. The final energy loss is due to eddy current dissipation.

The magnitude of the exchange effect was first estimated by Kittel and Herring [11]; however, these authors underestimated the significance of the effect. MacDonald [12] carried out a general solution of the exchange problem and pointed out the existence of such features as negative equivalent permeability and the difference in resonance line shapes due to power reflected from and transmitted through thin films. Rado and Weertman [13] showed that exchange effects should be observable at room temperature under suitable conditions and have demonstrated that their data can be fitted by the results of the analysis of Ament and Rado [14] (AR), including the occurrence of negative equivalent permeability.

AR and MacDonald both treat the case of the applied magnetic field parallel to the medium surface and perpendicular to the propagation direction. The present authors have found that the considerable mathematical complexity of the parallel case can be greatly reduced by treating the case of the magnetic field perpendicular to the medium surface. Consequently this situation has been applied now to thin films in order to determine the feasibility of exchange effect experiments.

Since the exchange anisotropy field is dependent on the skin depth and hence the propagation constant, a determination of the modes of propagation and permeabilities requires a simultaneous solution of Maxwell’s equations and the equation of motion of the magnetization vector. The second part of the analysis consists of a solution of the boundary value problem for sheets (or films) of arbitrary thickness. From such a solution, explicit expressions are readily obtained for the reflection, absorption and transmission coefficients. This analysis has been outlined previously [15]. The expressions, though explicit, are sufficiently complicated so that numerical calculations were carried out on the Lincoln IBM 704 computer. Curves have been constructed of the reflection, absorption and transmission coefficients as functions of magnetic field through resonance for the following parameters:

- Effective exchange constant
  
  $$A' = 4A/M_n$$
  
  $\delta = 0.1, \ 0.25, \ 0.5, \ 1.0$ ;

- Bloch-Bloembergen damping
  
  $$2\gamma T = 0, 10, \ 50, \ 200 \text{ oe}$$ ;

- Film thickness to classical skin depth ratio
  
  $$2d/\delta = 0.1, \ 0.2, \ 0.4, \ 1.0$$.

From such plots the exchange shift $\delta U$, the line width $\Delta H$, and the positions of possible subsidiary resonances may be obtained. That secondary resonances are at all conceivable arises form the following considerations. It is a result of the analysis that for one of the exchange propagation modes the skin depth is greater than the wavelength on the low field side of resonance. This means that interference effects could exist when an integral number of half-wavelengths fit into the film thickness, and thus give rise to subsidiary absorption peaks. If such peaks could be observed, they would be clear-cut evidence of exchange, since, for the ordinary case of a metal without exchange, the skin depth is always less than the wavelength. Because the modes cannot be observed independently, and because of the likely addition of relaxation damping, it is of course necessary to proceed through the computations indicated above, to see if the theory actually predicts such body resonances.

At the present time all the theoretical data have not been analyzed, and only preliminary experimental results are available. However, some com-
parison between theory and experiment can already be made of various exchange effect manifestations.

1. Exchange Shift $\delta H$. — This is generally small compared to the resonance field — being at most 150 oersteds.

2. Differences in Magnetic Field for Max. Absorption, Transmission and Reflection. — Even though $\delta H$ is small, it is not the same for the reflection and transmission coefficients. It is relatively easy to measure even very small field differences between the resonance peaks. So far agreement between theory and experiment is only qualitative.

3. Increase in Transmitted Power on Resonance. — The theory predicts that when exchange damping is dominant and the film is not too thin, the transmitted power should increase rather than decrease on resonance.

4. Subsidiary Peaks. — Highly damped peaks on the low field side of resonance appear in the theoretical reflection curves, and are much more sharply defined in the transmission plots. The position of these peaks checks with simple calculations of the wavelength to thickness ratios at which interference effects should occur. The experimental traces do indeed show numerous subsidiary peaks only on the low field side of resonance in both reflected and transmitted powers. The number of peaks decreases with decreasing film thickness and vanishes in films less than 1,000 Å, this also checks qualitatively with theory.

5. Line Width Variations. — The principal conclusions are that the width should increase with increasing frequency and film thickness.

A proposed plan now is to make measurements on a given film down to very low temperatures. Some data on conductivity as a function of temperature and film thickness in permalloy are available [16]. Thus the variation with temperature of the effective exchange constant $A'$ can be calculated, and the theory can be used to predict the behavior of above criteria 1 through 5 as a function of temperature. From matching the experimental curves to the appropriate theoretical plots, a value of the exchange constant $A$ may be deduced.

The results of our theory are exact solutions as far as the particular chosen model is concerned. As Gurevich [17] and Rado [18] have discussed, under certain conditions the anomalous skin effect should be considered in ferromagnetic resonance in metals. In our case of thin films we have the additional possibility of influences from the anomalous conductivity due to electron scattering from the surface.

C. Harmonic Generation. — From simple considerations of the precession model, it appears that a plane specimen should be particularly suited for generation of harmonics by ferromagnetic resonance. The precession path about an axis in the plane of the film is highly elliptical because of demagnetization effects. From a straightforward calculation based on the equations of motion for the uniform mode (non-spin-wave treatment), it can be shown that in a plane specimen an alternating component of magnetization is generated in the Z-direction (dc. field) at twice the exciting frequency, of magnitude

$$\delta m_z = \frac{4\pi M (H_R + \delta M) M \delta H}{4(H_R + 2\pi M)^2 (\Delta H^2)}.$$  

This shows the second power dependence on the exciting r.f. field $h_x$, as well as the inverse dependence on the line width $\Delta H$ in terms of applied field.

We can define an intrinsic conversion efficiency as the ratio of "harmonic susceptibility" $\frac{\delta m_z}{h_x}$ to the usual fundamental susceptibility $\frac{m_z}{h_x}$. Then

$$C. E. = \frac{\delta m_z}{m_z} = \frac{4\pi M h_x}{2(2H_R + \pi M)} (\Delta H^2)^{-1}.$$  

The maximum theoretical C. E. under our experimental conditions is approximately

$$\frac{10^4 \times 1}{2(2 \times 10^4 + 10^4) \times 10^{-2}} \approx \frac{1}{2} \times 10^{-2}.$$  

For a spherical sample, the C. E. would be much more unfavorable, namely

$$\frac{\delta m_z}{m_z} = \frac{1}{4} \frac{h_x}{H_R}.$$  

We have observed harmonic power at 18 GHz from magnetron excitation at 9 GHz of a dual mode cavity capable of oscillating at the fundamental and orthogonal harmonic frequency. Experiments are being carried out to determine the conversion efficiency. Our aim is to establish a definition of conversion efficiency which is an intrinsic property of the material — independent of a specific geometrical arrangement.

D. Generation of d.c voltage by FMR. — A new method of detecting ferromagnetic resonance (FMR) has been discovered in thin films which is about two orders of magnitude more sensitive than methods which depend on changes in cavity $Q$. Upon excitation of FMR, a d.c. signal can be taken off two wires soldered to the permalloy film. For detection purposes the incident microwaves should be amplitude modulated. Minimum power of approximately 0.5 watts peak incident on the
cavity is necessary, but the effect can also be observed in a wave-guide. Resonance from $10^{15}$ spins ($\Delta H = 100$) can be detected with such low noise level that $10^{13}$ spins ought to be readily detectable. The relative sensitivity increases for thinner films.

While these experiments were in progress, workers at the Polytechnic Research Institute of Brooklyn, U. S. A., already reported [19] discovery of the same effect. These authors have considered the Hall effect as a possible explanation of this phenomenon. Our evidence for the extraordinary microwave Hall effect is the following: 1) Proper geometry is necessary. 2) Polarity reversal of d.c. signal with reversal of d.c. magnetic field. 3) Phase reversal of signal in going through FMR. It is possible to derive a rudimentary expression based on the extraordinary Hall formula [20]

$$e_H = R_1 M J_f.$$  

The result is $e_H = \frac{R_1 x h^2}{4\pi \delta}$, where $R_1$ is the extraordinary Hall coefficient, $x$ the off-diagonal tensor susceptibility, $h$ the r.f. magnetic field, and $\delta$ the skin depth. The resulting voltage, of the order of millivolts per cm per r.f. oe., agrees with experimentally found magnitudes, but of course neglects all phase effects between the r.f. magnetic field and eddy currents. Physically, the product of two r.f. terms, namely $M(\omega) J(\omega)$, gives rise to the d.c. term as well as higher harmonics. The second harmonic has been observed to appear on the two wires.

REFERENCES


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DISCUSSION

Mr. Clogston. — Do you think that the effect you describe might also occur in ferrimagnetic materials exhibiting conductivity of the Verwey type; for instance, nickel ferrite containing ferrous iron?

Mr. Tannenwald. — As a matter of fact, the effect has also been observed by us in just this material. However, the geometry is not as straightforward. Furthermore, rectification phenomena probably play a significant part here.

Mr. Suhl. — Some years ago C. Herring suggested that spin waves in metals might be accompanied by a d.c. current. Observation of such currents might be facilitated by the thin-film geometry. Perhaps this is an alternative to the Hall-effect explanation of the voltage generated in ferromagnetic resonance.

Mr. Tannenwald. — It is appropriate to consider other possibilities. The above mentioned comment may provide a useful approach, which has not been considered by us. Of course we must obtain detailed predictions of the proposed mechanism before reaching further conclusions.