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Magnetic Thin Films II

Spin Wave Resonance in Magnetic Films

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The suggestion and observation that standing spin waves can be directly excited in magnetic films by microwave resonance have led to a number of measurements significant to magnetism. Certain questions crucial to the interpretation of spin wave resonance spectra are discussed. The principal results obtainable by the spin wave resonance method are pointed out. New experiments are discussed which offer the possibility of studying second-order exchange effects and spin wave-phonon interactions. Finally the question of observing spin wave resonance in antiferromagnetic systems is taken up.

Introduction

The background for spin wave resonance was laid when it was realized that exchange effects might play a role in ferromagnetic resonance[1,2]. Subsequently, the series of experiments carried out by Rado and co-workers[3,4,5] demonstrated that the exchange field played an important and measurable part, even at room temperature, in ferromagnetic resonance in bulk metals. In later ferromagnetic resonance experiments in thin magnetic films, subsidiary resonances were observed, and it was pointed out that these might be the result of standing waves due to exchange modes[6]. On three different occasions an essentially similar problem was solved — the Bloch equation of motion with exchange plus Maxwell's equations with boundary conditions—each calculation suiting the purposes of the particular experiment[1,4,6]. In the third instance, where the magnetic field and the propagation vector were taken to be perpendicular to the film surface, the calculations led to entirely too small effects[6]. The breakthrough was provided by Kittel[7] when he pointed out that standing spin waves could be excited by a uniform r. f. field if the spins at the surface of thin films were considered as fixed or pinned. From that point on, observation of spin wave resonance became prevalent, useful results were obtained on the magnitude of the exchange constant, and the electromagnetic calculations were repeated with appropriate boundary conditions.

The purposes to which spin wave resonance has been applied are the following: measurement of exchange constant, measurement of surface anisotropy, measurement of second-order exchange interactions, and study of spin wave-phonon interactions. (Second-order exchange interactions refers to the correction terms in the energy of spin waves due to their mutual interaction.)

Spin wave resonance phenomenon

The crucial concept in the occurrence of spin wave resonance is the existence of some kind of boundary condition which makes the excitation of spin waves by a uniform r.f. field—that is, higher order modes in addition to the uniform mode—possible[7]. The subject has progressed in spite of the fact that understanding of conditions at the boundary developed slowly beyond Kittel's original suggestion that pinning might be due to a Néel-type surface anisotropy or Meiklejohn-Bean oxide layer, in magnitude of the order of one tenth to one hundredth of the exchange field. Clarification of the matter has been provided by an analysis of boundary conditions[5], by an estimate of the surface anisotropy[8], and by experimental work physically altering the boundary state*[9].

1. Excitation

As far as pinning at the boundaries is

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* Kindly communicated by the authors prior to publication.

concerned, there are three extreme cases possible. If the spins on both sides of the film are pinned, the film thickness must be an integral number of half-wavelengths, i.e., $L=n(\lambda/2)$. One pinned and one free surface leads to $L=(2n+1/2)(\lambda/2)$; and both surfaces free gives $L=n(\lambda/2)$. Since in the third case the excursions of the spins just balance out, no net energy transfer can take place, and this case is ruled out theoretically. Experimentally only an odd number of half-wavelengths is predominantly excited. This eliminates case two, leaving case one—both sides pinned—as the common situation.**

2. Boundary Conditions

A quantitative discussion can be based on the Rado-Weertman general boundary condition[5]

$$\frac{2A}{M_s^2}\boldsymbol{M}\times\frac{\partial \boldsymbol{M}}{\partial n}+\boldsymbol{T}_s=0\,,$$

where $\boldsymbol{T}_s$ is the surface torque density, $\boldsymbol{n}$ a unit vector normal to the surface. For small excitation this becomes

$$\frac{2A}{M_s}\boldsymbol{n}\times\frac{\partial \boldsymbol{m}}{\partial n}+\boldsymbol{T}_s=0\,.$$

If now $\boldsymbol{T}_s$ is along $\boldsymbol{n}$, and the external d.c. field along $\boldsymbol{n}$ (z coordinate), $2A(\partial \boldsymbol{m}/\partial z)-K_s\boldsymbol{m}=0$, where K_s is the surface anisotropy energy density. Since experiments show that pinning does indeed predominate, $k\boldsymbol{m}/(\partial \boldsymbol{m}/\partial z)\ll 1$, or $K_s\gg 2Ak$. Soohoo[8] has used this relation to estimate that the normal surface anisotropy must be greater than

$$2Ak\simeq 2\times10^{-6}\ \text{erg/cm}\times 6\times10^{5}\ \text{cm}^{-1}$$
$$\simeq 1.2\ \text{erg/cm}^2\,.$$

Let us briefly mention the situation in which the surface anisotropy is normal but the external field is applied along the surface. The general boundary condition leads to $\partial m_x/\partial n=0$, resulting in no net excitation. This explains why spin wave resonances are not generally excited in the parallel case. In actuality, the surface anisotropy might be different for spins lined up parallel or perpendicular to the surface.

3. Electromagnetic problem

In order to understand details of the spin wave spectrum and account for deviations from the simple dispersion relation, the exact electromagnetic problem has to be examined. Various aspects of this problem have been treated on different occasions in addition to references already cited[10,11], the most complete treatment for spin waves propagating parallel to the applied d.c. field having been given by Seavey[12]. Nevertheless the electromagnetic theory of spin wave resonance has only been used as a guide to qualitative understanding and has not been applied to a detailed comparison with experiment.

An electromagnetic theory treatment of spin wave resonance involves the calculation of (a) propagation constants, (b) modes, (c) field strengths, and (d) power absorbed. Inclusion of the exchange term in the equation of motion, solved together with Maxwell's equation, produces a dispersion relation quadratic in k^2. Each circular mode is split into two, having different propagation constants; of the two resonant modes, one is an electromagnetic mode the other a spin wave mode.

Seavey has derived the following conclusions:[12]

1. A correction to the standing wave condition due to spin wave attenuation results predominantly from relaxation($1/\gamma T$, Bloch-Bloembergen type) and not eddy current damping. However, the resultant shift of spin wave peaks toward higher magnetic field values is insignificant unless $1/\gamma T$ were so large that the peaks would be unresolved anyway.
2. For some typical $1/\gamma T$ values, as encountered in insulators, peaks up to $n=3$ or 5 can be shown to be unresolved.
3. In general, when power is incident evenly on both sides of the film, even modes due to the skin depth effect are not excited from the symmetry of the geometry. Usually experimental conditions represent some sort of intermediate case, with some power incident on the sample from both sides.
4. Depending on the r.f. field configuration as determined by the geometry of the experiment, odd peaks always retain the same field position; even peaks can vary. The main peak is an integral n number when power is incident from one side, but not otherwise.

Exchange constant

The first and principal result obtained by

** For controlled exceptions, see ref. 9.

the spin wave resonance method is the determination of exchange constant, which has been done in permalloy[13], nickel[14], cobalt[15,16], and Cu-Ni[17] and Pd-Ni[18] alloys. In effect the measurement has been reduced to a problem of magnetic spectroscopy, while other methods involve a fairly indirect procedure of extracting the exchange constant from the temperature variation of magnetization or specific heat measurement. While there is no doubt that the exchange constant A is of fundamental significance to ferromagnetism[19], there exist serious theoretical difficulties of how to relate this constant to quantities calculable from first principles—a problem which has beset ferromagnetism from the very beginning.

No detailed discussion of exchange constant measurements will be taken up here because the past and current literature, as cited in the references and in this Conference, deals with this subject for specific cases. However, it is obviously of great importance —both for calculation of the exchange constant as well as establishment of the spin wave dispersion relation—that the $\omega \propto k^2$ relation can be demonstrated to be valid. Deviations from the k^2 law that can not be accounted for by the electromagnetic calculations occur commonly, but *only* for the first few spin wave peaks near the main resonance line. Since these deviations near the main resonance line appear randomly in different films, they should not be associated with any fundamental property of the spin wave dispersion relation. When a sufficiently large number of spin wave peaks is observable, the peaks invariably follow a k^2 dependence.

Second-order exchange interactions

By second-order exchange interactions it is meant that the exchange field or exchange integral is no longer assumed constant with respect to temperature. A straightforward, but not self-consistent, approach is to use the localized electron model in a semi-classical spin wave picture—one that is frequently employed in ferromagnetism in metals and is justified only because more complicated theoretical analysis do not necessarily lead to a more discriminate fitting of experimental data. On such a simple basis, the exchange integral J has been shown[15] by a spin wave resonance experiment in cobalt to be independent of temperature between 4°K and 293°K.

A more satisfactory approach conceptually is provided by a physical model of spin wave interactions[20], which specifically predicts a temperature variation of the exchange parameter D on the basis of the same model of ferromagnetism assumed above. The exchange parameter D is related to the better known exchange constant A through the relation $D/\gamma\hbar = 2A/M$. The essence of this analysis is that, as the temperature is raised and additional spin waves are excited, the exchange field will decrease, not as the angle between a particular spin and the average magnetization, but as the angle between a particular spin and its neighbor. The result yields the conclusion that $D \propto T^{5/2}$. When this is re-inserted into the first-order Bloch law $M \propto T^{3/2}$, it leads to a magnetization law previously derived by Dyson by a more complicated treatment: $M = M_0[1 - A_0 T^{3/2} - A_3 T^4]$, plus other structure dependent terms. It is clearly better to attempt a verification of the spin wave interaction term, not by a measurement of the temperature variation of the magnetization, but through a measurement of the temperature variation of D itself.

In spin wave resonance experiments, the magnetic field separation between odd peaks is given by

$$H_n - H_{n+1} = \frac{4D}{\gamma\hbar}\left(\frac{\pi}{L}\right)^2 [n+1] .$$

If it is assumed that the g-factor is temperature independent and that no other temperature dependent corrections enter into the spin wave dispersion relation, then a straightforward measurement of the separation of two spin wave peaks as a function of temperature would yield the temperature variation of D. Nevertheless such an experiment does not give unequivocal results without some difficulty. Samples were selected which have a series of well-defined spin wave peaks. The peaks occur around 10,000 gauss with a separation of about 500 gauss and a change in separation between 4°K and room temperature of only about 50 gauss. Experimentally then, the small temperature variation of the difference between two large quantities must

be measured. The resultant uncertainty precludes an unambiguous interpretation (at the time of this writing) other than that the data can be fitted satisfactorily by a $T^{5/2}$ dependence. In one experimental run, $T^{5/2}$ gave a slightly better fit than $T^{3/2}$, and the results on another sample could not be fitted well on a log-log plot with a slope less than 5/2.

Although good agreement cannot really be expected because of the idealized nature of the model compared to the complex situation prevailing in a ferromagnetic metal, one can nevertheless compute the theoretical coefficient in the second-order exchange relation[20]

$$D=D_0\left[1-\pi\rho^2\frac{g\beta}{M_s}\xi\left(\frac{5}{2}\right)\left(\frac{kT}{4\pi D_0}\right)^{5/2}\right]$$

and compare it with the coefficient when the data is forced to fit a $T^{5/2}$ dependence. The experimental result gives $D=D_0[1-7\times10^{-8}T^{5/2}]$ from one set of measurements, while the computed relation is $D=D_0[1-0.2\times10^{-8}T^{5/2}]$. The discrepancy is obviously large. Evidently the state of affairs leaves something to be desired, with the burden at the moment falling on the experimenter to provide more accurate data to establish the uniqueness of the $T^{5/2}$ unambiguously. Work is in progress to measure the temperature variation of D more accurately, possibly to an order of magnitude improvement.

Spin wave-phonon interactions

At various times it has been suggested that ferromagnetic spin precession is coupled to the crystal lattice by way of magnetostriction and, in fact, damping of the precession and thus line width of the resonance has been considered to be due to such a mechanism. More recently the energy transfer between spin waves and phonons has been considered[21,7,22,23], giving rise to the theoretical prediction that a strong coupling exists between spin waves and phonons provided that both their wavelengths and frequencies are equal. A further study of this cross-over point (equal λ and equal ω) has shown that the normal modes in the strongly coupled region are a mixture of spin wave and phonon modes and has led to a dispersion relation characteristic of two coupled systems. Experimentally the excitation of phonons by ferromagnetic resonance, and the inverse, has been demonstrated[24], but not specifically at the cross-over point.

In order to examine spin wave-phonon interactions in a spin wave resonance experiment, a new problem is solved in which a magnetoelastic term is added to the equation of motion containing exchange. The result consists of three circular rotating resonant modes —an electromagnetic, a spin wave and a phonon mode. Since the subject is treated in detail in the references we will state for brevity's sake only the physical results. Spin waves will be strongly coupled to phonons when their wavelengths and frequencies are equal. This intersection of the respective dispersion relations happens to fall into the upper microwave region. As a first consequence one would expect that, depending on the strength of the coupling,

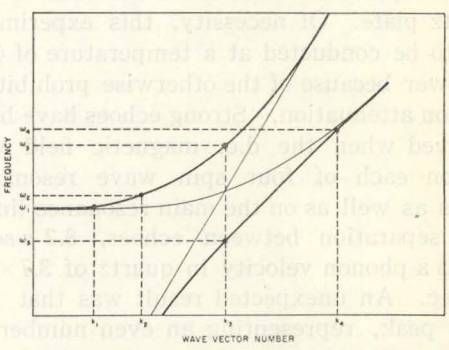

Fig. 1. Magneto-acoustic modes in the region where spin waves and phonons have equal ω and k, and the possible effect on the spin wave resonance spectrum.

mixed magnetoelastic modes will occur in the cross-over region. This could cause a deviation from the monotonic sequence of the spin wave peaks, as shown in Fig. 1 on a frequency vs. wave number plot. To detect such a deviation experimentally, a suitable operating temperature region has to be selected since the generally large attenuation of acoustic waves at microwave frequencies reduces the magnitude of the spin wave-phonon interaction. On the other hand, the sharpness of interaction probably depends on the difference of $\partial\omega/\partial k$ between spin waves and phonons at the cross-over region[25].

A second possible method of studying spin wave-phonon interactions in a spin wave reso-

nance experiment involves the use of sufficiently large r.f. driving fields so that phonons of detectably large amplitudes are generated on spin wave resonance. The films are evaporated onto single crystal quartz plates, the latter being polished to optical flatness and parallelism. Phonons generated through magnetostriction during an r.f. magnetic field pulse driving the spin precession are propagated down the quartz rod and are reflected from the far end. In contrast to other experiments[26], the quartz rod does not extend into a second cavity where phonons might be detected by the inverse piezo-electric effect; rather, the reflected phonon echo, in the relatively short quartz plate re-generates an r.f. magnetic field pulse in the same cavity, which is then detected by means of standard heterodyne techniques. Actually, a series of pulses is observed due to multiple reflections from the ends of the quartz plate. Of necessity, this experiment has to be conducted at a temperature of 4°K or lower because of the otherwise prohibitive phonon attenuation. Strong echoes have been received when the d.c. magnetic field was set on each of four spin wave resonance peaks as well as on the main resonance line*. The separation between echoes, 8.2 μ sec., yields a phonon velocity in quartz of 3.7×10^5 cm/sec. An unexpected result was that the $n=8$ peak, representing an even number of half-wavelengths, was larger than the $n=7$ and 9 peaks. This effect is not understood. For the film used, $\omega/\gamma-H_{cr}\sim90$ Oe, where H_{cr} is the calculated field at cross-over. Thus the expected H_{cr} fell on the tail of the main line where the excitation of spin waves with proper k-number is not sufficiently strong to couple to phonons. It is evidently desirable to repeat this experiment under circumstances where H_{cr} is well-separated from the main resonance and where appropriate frequency changes move the cross-over region through a spin wave peak.

Spin wave resonance in antiferromagnets

It is natural to consider extension of spin wave resonance experiments to antiferromagnetic systems. The pertinent resonance relations were first given by Orbach and Pincus[27] and Cofta[28]

$$\omega/\gamma=\pm H_n+[H_A(H_A+2H_E)+H_E^2k_n^2a^2]^{1/2}\,.$$

For purposes of resolution—especially if the antiferromagnetic resonance line width is broad—it is desirable to have the spacings of the spin wave peaks as large as possible. The separation between odd peaks is given by $\sqrt{H_E/2H_A}\,2H_E(\pi a/L)^2(n+1)$, where a is the distance between spins. This relation can be contrasted with the separation between odd peaks in the ferromagnetic case, i.e., $2H_E(\pi a/L)^2(n+1)$. It is seen that the separation is more favorable in antiferromagnets by a factor $\sqrt{H_E/2H_A}$, which can be as large as 10. This circumstance contributes to a hopeful outlook for the observation of antiferromagnetic spin wave resonance since up to the present time it has been difficult either to obtain antiferromagnetic materials having line widths as narrow as ferromagnets or to prepare single crystal antiferromagnets sufficiently thin. In any event, one can expect that antiferromagnetic spin wave resonance will be observed in the millimeter wavelength region because the spin wave term makes an additional contribution to the already large effective field $(2H_EH_A)^{1/2}$, thus raising the resonance frequency even higher.

We can speculate how the spin wave resonance frequency might be raised in a ferromagnet. It is evident from the dispersion relation that either a higher wave order number or a thinner sample would result in a larger contribution from the exchange field. One is restricted to relatively low wave order numbers because of the net excitation requirement. On the other hand, ferromagnetic resonance can be readily observed in films as thin as 100 Å. Thus, assuming the usual boundary conditions were satisfied, spin wave resonance in such thin films could conceivably be observed up to 1000 kMcps. The separation between peaks would of course be enormous, and it is questionable whether a given spectroscopy technique would even be applicable to two adjacent peaks. However, the nature of the spin wave dispersion relation, and its likely deviation from the small angle approximation would be most interesting. For completeness sake, let us point out that the high-frequency limit of spin wave resonance would be

* The experiment was carried out by M. H. Seavey, Jr.

the case $\lambda/2=a$, the separation between adjacent spins. As far as is known, such an "exchange" resonance, which would fall in the infrared, does not exist for a single sublattice system consisting of identical spins. By way of contrast, there is no difficulty in exciting antiferromagnetic resonance by a uniform r.f. field because the oppositely directed spins on the two sublattices are driven in the same direction.

One other approach should be mentioned that has yielded the spin wave dispersion relation from a different viewpoint. Applying the equations of motions to Valenta's sublattice model of a thin film, Fraitová[29] has obtained a system of n Bloch equations for a film composed of n atomic planes, giving the resonance conditions as the characteristic roots of a determinant of the n-th degree. The usual spin wave dispersion relation, having been derived from a continuum hypothesis, results in a continuous k-vector spectrum.

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DISCUSSION

D. O. Smith: How do the values of the exchange constants determined from spin-wave resonance compare with the values found by measuring temperature dependence of the magnetization?

P. E. Tannenwald: For permalloy, I can only recall the results of Kondorski and Fedotov approximately, and would say about 20%. For cobalt, the nuclear magnetic

resonance measurement of the temperature dependence of the magnetization of Jaccarino agreed with SWR to about 15%. For Ni the results agree to 10%. For details of this comparison see the paper of Kimura and Nosé.

C. KITTEL: The temperature dependence of $D_0 \propto Ja^2$ because of thermal expansion should be considered, as this would appear to give effects of the order of these observed. If, for example, $\Delta J/J \approx 10\Delta a/a$, then

$$\frac{\Delta D_0}{D_0} \simeq 10\frac{\Delta a}{a} + 2\frac{\Delta a}{a}.$$

In the region below the Debye temperature the variation of a would not be linear in T.

It is also possible that a long-range indirect exchange interaction would enhance the temperature dependence of D because of the effective range factor ρ^2 in the theory. It would be valuable to correlate J as deduced from the Curie temperature with $J\rho^2$ from D_0 and ρ^2 from $D_0 - D$.

P. E. TANNENWALD: Thermal expansion should be considered. $\Delta a/a$ is probably not much larger than 0.1% over the temperature range considered, while $\Delta D/D_0$ changed by about 10%. In any case $\Delta a/a$ could be measured experimentally, or the effect of lattice expansion on D_0 could be appropriately incorporated into the theory.

A long-range indirect exchange interaction would increase the effective range factor and bring the calculated result into closer agreement with the experimental one.

I do not see how the correlations mentioned would be sufficiently accurate to be of much significance.

R. D. LOWDE: At Harwell, J. F. Mattett has made a careful search for the neutron scattering from spin waves in nickel, and finds that it is absent. (More accurately he can say that it is a factor of at least 50 below what is predicted on the single exchange-coupled model). The spin-wave wavelengths to which his experiment is most sensitive are of the order of 200–300 Å, and thus are right in the region where they have been observed by ferromagnetic resonance. A similar situation is true for permalloys of approximately the composition you have studied particularly. We have recently searched for the spin-wave scattering as a function of composition across the Fe–Ni constitution diagram, and find that it decreases gradually to zero as nickel is added to iron. These results seem on present ideas to be in flat contradiction with the work on SWR, and we can think of only two explanations. One is that due to the "Van Vleck" character of the magnetism in nickel, the spin waves have a short coherence length, too short to give the appropriate neutron diffraction pattern; this explanation however is inconsistent with the line widths observed in SWR. The other explanation is that perhaps there are normally no spin waves in bulk nickel, and that in SWR experiment you create the spin waves. The idea would be that spin waves in nickel-rich alloys have a very short relaxation time, about 10^{-11} sec, but that there exists some bottleneck impeding their relaxation so that with RF you can drive spin system into the observed mode of oscillation. In any event, one may imagine that the true state of affairs is more complicated than is assumed in the simple models of SWR.

P. E. TANNENWALD: First, without taking account of the neutron scattering results, I would say that the coherence length for spin waves which we observe is approximately

$$l = v \times t = 10^5 \text{ cm/sec} \times 10^{-9} \text{ sec} \simeq 10^{-4} \text{ cm};$$

that is, an order of magnitude larger than the size of our samples. Second, there does not appear to be anything unusual happening to the SWR spectra in passing through the iron-nickel composition range of one Bohr magneton. Third, I would agree that the nature of SWR experiments is different from neutron scattering in

that absorption of energy by spin waves of a particular k-number can take place even though their thermal excitation is very small.

D. O. SMITH: One may ask whether the disappearance of neutron spin wave scattering for Ni-Fe alloys containing more than 85% Ni is related to the negative magnetostriction of these alloys.

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Surface Spin Pinning in Permalloy by an Oxide Layer

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Standing spin waves in thin magnetic films can be excited by uniform microwave fields if the surface spins are pinned. Observation of spin wave resonances by uniform microwave fields in thin permalloy (80% Ni-20% Fe) films in the oxidized and reduced state indicates that the major portion of the surface spin pinning is due to the oxide layer on the surface of the film. The oxidized films show strong resonances of odd numbered spin wave modes with even numbered spin wave modes almost totally absent. This indicates strong pinning at both surfaces. The reduced films show weak resonances of both odd and even spin waves. This is interpreted as indicating a strong pinning at the substrate surface of the film and a weak pinning at the free, reduced surface of the film.

We conclude that surface spin pinning in our permalloy films is due mainly to the effects of an oxide layer on the surface of the film, probably by the "exchange anisotropy" of Meiklejohn and Bean, rather than to the "surface anisotropy" proposed by Néel.

Introduction

Ferromagnetic resonance experiments in ferromagnetic films of certain thicknesses (generally in the range 3000 to 6000 Å) reveal a number of microwave power absorption peaks attributable to standing spin waves.[1,2] These are caused by pinning of the surface spins and excitation of the magnetization in simple sinusoidal modes analogous to the vibration of a string with fixed ends[3]. The propagation vector, k, must satisfy the relation $k=n\pi/L$ where n is an integer and L is the thickness of the film. In a uniform microwave field only the odd numbered spin wave modes are excited. However, as shown below, if the spin pinning is relaxed at one surface the even modes appear. This is the basis of the present experiment. We look for the amplitude of the even modes as a function of surface treatment in order to determine the mechanism of surface spin pinning.

The pinning of the surface spins has been attributed to a) an antiferromagnetic surface layer presumably an oxide of nickel or iron and b) a surface anisotropy due to the lower symmetry in which a surface spin is located as compared to an interior spin[3,4,5]. By ferromagnetic resonance experiments on oxidized and reduced permalloy films we have found

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