such partial stopping powers in hydrogen gas and the contributions of charge-changing collisions to the total stopping losses.[27,31,32] Unfortunately, the technique used in these studies probably cannot be applied to solid absorbers. It would be of interest to see if differences persist between gas and solid absorbers with similar chemical binding at higher velocities where charge exchange effects are supposedly negligible.

It is unfortunate that low-energy alpha-particle data are even more scant than those for protons. More information is required to determine whether cross sections for gases are greater than those for solids also in the case of helium ions for energies somewhat above the maximum in the stopping-cross-section curve. In addition, a comparison of results on gases and solids with similar chemical binding should be worthwhile.

Table V illustrates the complex situation in regard to proton stopping in hydrocarbons. It is evident that a considerable amount of additional data would be required for any hope to gain an understanding of the details. Increased precision of measurements is highly desirable since the effects of physical state and other differences are not large. The situation is even worse for stopping with respect to helium beams because there are fewer empirical data.

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The continued support of the U. S. Atomic Energy Commission without which this work could not have been conducted is greatly appreciated.

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[32] M. N. Huberman, Phys. Rev. 127, 799 (1962).

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Long-Range Exchange Interactions from Spin-Wave Resonance

R. Weber and P. E. Tannenwald

Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts*

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The presence of the quartic term, in addition to the usual quadratic term, in the spin-wave dispersion relation has been established to within 3% accuracy by spin-wave resonance experiments in a number of Permalloy films. High-precision measurements at 70 Gc/sec permitted observation of spin wavelengths as short as 225 Å. The coefficient of the quartic term, which involves the fourth moment of the exchange interaction, leads to a long-range interaction $\langle\rho^2\rangle=120\,r_1^2$, where r_1 is the nearest-neighbor distance; this is in good agreement with the range determined previously from spin-wave interaction measurements on a similar sample. Since the films are predominantly polycrystalline, anisotropy in the exchange could not be distinguished. $\langle\rho^2\rangle$ determined from the coefficient of the $T^{5/2}$ term in the magnetization, measured by ferromagnetic resonance in the same type of sample, is smaller by a factor of about five. However, with such a long range, the validity of the usual expansion of the spin-wave energy to describe the magnetization is doubtful.

INTRODUCTION

THE basic result of spin-wave resonance experiments in magnetic films has been the determination of the ferromagnetic exchange constant.[1] However, improvements in experimental techniques have made possible measurements of second order exchange effects, such as the determination of spin-wave interactions,[2] that are of great importance to the theory of magnetism. The present experiment is concerned with a second refinement of spin-wave theory in a metal, namely, the establishment of the existence of the quartic term in the spin-wave dispersion relation

$$\omega=ak^2-bk^4\,, \qquad (1)$$

where a and b are constants to be determined. This form is a special case of the general dispersion relation from spin-wave theory of a ferromagnet for small k numbers,

$$\mathcal{E}_k=2S\left[\frac{1}{2}\sum_r(\mathbf{k}\cdot\mathbf{r})^2J_r-\frac{1}{24}\sum_r(\mathbf{k}\cdot\mathbf{r})^4J_r+\cdots\right], \qquad (2)$$

where $\mathcal{E}_k$ is the energy of a noninteracting spin wave of wave vector k, S is the spin per atom, and J_r is the exchange integral between spins separated by distance r. As will be discussed in detail, the magnitude of b depends on the direction of spin-wave propagation with respect to the crystallographic axes in a single crystal,

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* Operated with support from the U. S. Air Force.

[1] Z. Frait, Phys. Status Solidi 2, 1417 (1962).

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or is an appropriate average of the anisotropic quartic term in a polycrystal. An indication of the magnitude of the quartic term has already been seen from pyromagnetic-magnetization measurements in nickel[3] and Permalloy,[4] in which, in addition to the usual Bloch $T^{3/2}$ term, a $T^{5/2}$ term fits the data better than a T^2 itinerant-model term. The presence of a $T^{5/2}$ term in the magnetization is the consequence of a k^4 term in the dispersion relation. The present experiment, in which the dispersion relation is measured directly in Permalloy films, is sufficiently accurate to allow a self-consistent determination of coefficients a and b as well as the exponents of the k-vector terms in Eq. (1). The ratio of coefficients of the quartic term to quadratic term, b/a, leads to an effective long-range interaction $\langle\rho^2\rangle$, in terms of the ratio of fourth moment to second moment of the exchange interaction range[5]; i.e.,

$$b/a \propto \langle\rho^2\rangle \equiv \sum_r J_r r^4 / \sum_r J_r r^2 .$$

This simple proportionality can be expressed only for a completely random polycrystalline sample; in the general case, it will depend on the direction of spin wave propagation and crystal structure, and the summations will involve different weightings of the J_r's.

This same ratio $\sum_r J_r r^4/\sum_r J_r r^2$ appears in the coefficient of temperature variation of the exchange interaction measured in a previous spin-wave resonance experiment on a similar Permalloy sample.[2] Since the results of the two experiments are in good agreement, it might appear that the Heisenberg-Bloch-Dyson localized spin model—upon which the interpretation of the spin wave interaction experiment rests—is applicable in the case of Permalloy. However, this may be too strong a conclusion, since the spin-wave-interaction expression with which comparison is made is but the first term in a series expansion.[6] In addition, to determine whether other interactions simulate a temperature dependence of the range factor in the dispersion relation, the present experiment, which was carried out at room temperature, will be extended to low temperatures.

Finally, previous measurements of the temperature dependence of the saturation magnetization of Permalloy films by ferromagnetic resonance[2] have been analyzed in detail and have been found to lead to an effective $\langle\rho^2\rangle$ which is a factor of five smaller than the $\langle\rho^2\rangle$ determined from the quartic term in the spin-wave dispersion relation. There are both points of agreement and disagreement with the pyromagnetic magnetization measurements of the IBM group, as will be taken up in detail in the Discussion. However, both results indicate that the $T^{5/2}$ coefficient in the magnetization in Permalloy is much smaller than would be expected on the basis of the observed quartic term in the dispersion relation. Neither the dispersion relation nor the magnetization temperature variation depends on the Heisenberg model for an interpretation, but only on spin-wave theory, so that a basic anomaly could be present. On the other hand, it may be that interactions not accounted for in the statistical treatment of the magnetization give rise to a $T^{5/2}$ dependence which is effective in reducing the coefficient of the observed $T^{5/2}$ term.

EXPERIMENTAL CONSIDERATIONS

To observe the quartic term in the spin-wave dispersion relation, spin waves of large wave number must be excited. An upper limit is set for obtainable k numbers by the extent of the magnetic-field interval bounded by the demagnetizing field and the field for resonance of the uniform mode. Since the latter varies directly as the microwave frequency, it is evidently necessary to turn to high frequencies.

To determine *accurately* the exponent of the "quartic" term and its coefficient a high density of resolvable resonance peaks within the large allowed magnetic field interval is desirable. Since the separation between adjacent peaks varies as the reciprocal of the square of the film thickness, as thick films as are compatible with the electromagnetic situation should be used.

Permalloys of varying compositions (55 Ni—45 Fe to 78 Ni—22 Fe) were vacuum evaporated ($\approx 5\times10^{-6}$ mm Hg) to thicknesses of 2000 Å to 7000 Å onto heated ($\approx$200°C) glass substrates. The compositions were obtained by spectrophotometric analyses of the actual samples used. The thickness range was based on a compromise between the desire for a high density of peaks and the limitations imposed by skin-depth considerations in the microwave region.

Standard microwave-cavity techniques were extended to 4 mm (70 Gc/sec), and phase-sensitive detection methods were incorporated into the system, presenting the derivative of the absorbed microwave power to be recorded as a function of the swept dc magnetic field supplied by a 22-in. Varian electromagnet. To obtain a recorded spin-wave spectrum, the dc field was swept at approximately 500 Oe per minute. The signal to the lock-in detector was derived from modulating the dc magnetic field at 100 cps with an amplitude corresponding to a few percent of the linewidth. The precise location of each peak was determined by adjusting the dc field to fall exactly on the peak and measuring the field by nuclear magnetic resonance. The magnetic field could be measured to ±0.5 Oe, and the peaks could generally be found to within ±5 Oe.

The measurements were carried out at room temperature. Magnetic film samples were selected on the basis that the saturation magnetization and the g factors be very close to their bulk values and that they exhibit a

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[3] B. E. Argyle, S. H. Charap, and E. W. Pugh, Phys. Rev. **132**, 2051 (1963).

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[5] W. Marshall, *Proceedings Eighth International Conference on Low Temperature Physics, London 1962* (Butterworths Scientific Publications, Ltd., London, 1963).

[6] F. Keffer and R. Loudon, J. Appl. Phys. **32S**, 2*S* (1961).

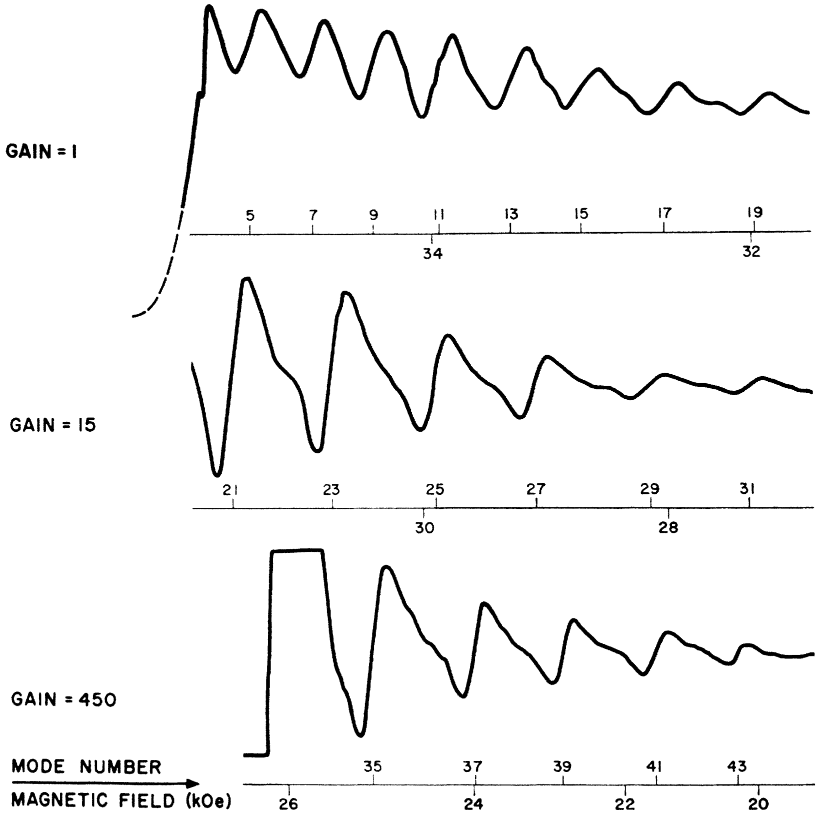

FIG. 1. Derivative trace of spin-wave resonance (SWR) absorption spectrum at 71.7 Gc/sec in a 78–22 Permalloy film of 5150 Å thickness. The large absorption near 26 kOe is due to a paramagnetic impurity in the glass substrate.

large number of well-resolved spin-wave peaks over a wide magnetic-field interval.

ANALYSIS AND RESULTS

The expression for the discrete standing wave modes that can be excited by a uniform microwave magnetic field in a thin film is given by the boundary condition $k=(\pi/L)p$, where k is the spin wave vector, L is the film thickness, and p is an odd integer, combined with the ferromagnetic resonance condition including the spin-wave contribution, $\omega/\gamma=H-4\pi M+(2A/M)k^2$, where H is the dc magnetic field, $4\pi M$ is the saturation magnetization, γ is the gyromagnetic ratio, and A is the exchange stiffness constant. Thus the magnetic field spacing $\bar{H}$ between the $p=0$ mode and the pth mode at constant ω may be written as $\bar{H}=\alpha p^2$. If the quartic spin-wave term is included, we have $\bar{H}=\alpha p^2-\beta p^4$, where α and β are constants directly related to the original a and b of Eq. (1).

Figure 1 shows the derivative of the absorption spectrum for a 78–22 Permalloy film of thickness $L=5150\pm50$ Å, with the appropriate numbering scheme for the allowed odd modes. Owing to the presence of appreciable eddy-current damping of the lower order modes and the broad main ($p=0$) resonance linewidth, the $p=1$ and $p=3$ modes were not resolved. The first observable spin-wave resonance peak, labeled $p=5$, and the next few peaks fall on the tail of the uniform-mode resonance. The resulting distortion of the line shape would make it difficult to determine their positions precisely. However, since these peaks were used for counting purposes only (i.e., in determining the numbering scheme), this slight inaccuracy is of no importance in quantitative considerations of the higher order terms in the dispersion relation. The higher-order peaks suffered little distortion, and their positions were taken to be the midpoints of their excursions about the base line. The large peak between $p=31$ and $p=35$ is the derivative of the absorption due to a paramagnetic impurity present in the glass substrate. Similar spectra were obtained for a 6850-Å thick 63–37 Permalloy film and for a 4050-Å thick 55–45 Permalloy film which were used in this experiment. The even-mode peaks were clearly discernible over most of the spectrum of the 63–37 sample, which was the thickest film. For each of the three films, the wavelength of the highest order resolvable spin-wave peak was close to 225 Å at a separation of approximately 15 kOe from the uniform-precession peak.

As is well known, the uniform-resonance peak in a semi-infinite conducting medium undergoes two shifts when compared with an insulator whose dimensions are small with respect to the rf wavelength: an "exchange shift" due to the rf field gradient producing an effective torque acting on the spins; and an "electromagnetic shift" due to the complex behavior of the electromag-

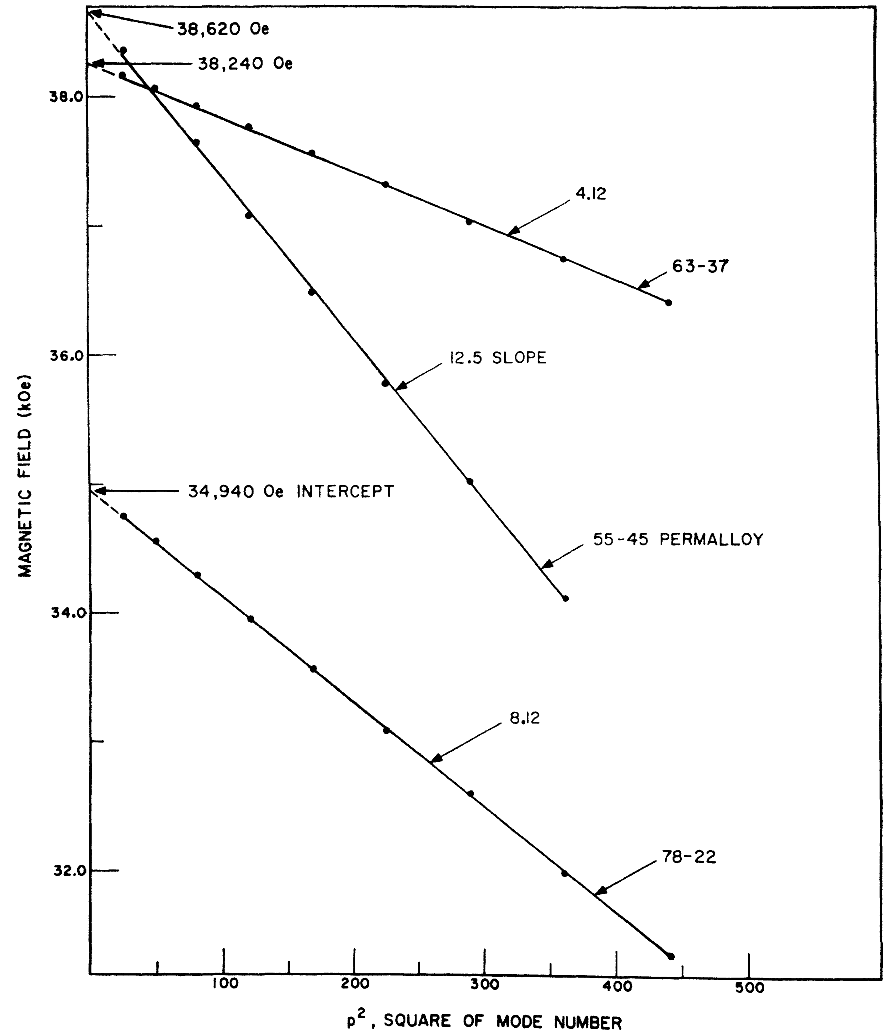

FIG. 2. Magnetic field for SWR versus square of mode number for three Permalloy samples. Slope of line determines exchange constant A, and intercept determines field for uniform precession resonance in the absence of exchange shift and electromagnetic propagation effects.

netic propagation vector as one passes through ferromagnetic resonance (this latter shift is even more pronounced in insulators). As a matter of fact, before the advent of spin-wave resonance the exchange shift in a ferromagnetic metal[7] was used to obtain the Landau exchange stiffness constant A. The existence of propagation effects in ferromagnetic resonance has been realized since the early experiments: When the sample is not small compared with an electromagnetic wavelength, they give rise to such phenomena as the size effect, wall effect, and dimensional resonances. What applies to the present discussion is that electromagnetic propagation effects lead to maximum power absorption, not at the peak of the μ'' line (imaginary component of the permeability), but at a shifted value corresponding to an equivalent permeability combination of μ'' and μ' (the dispersive component of the permeability).[8] In principle, these effects can be calculated precisely, but in practice uncertain factors such as film conductivity, microwave cavity and film geometry, the amount of Bloch-Bloembergen phenomenological damping admixture, and the applicability of a plane, semi-infinite medium model only allow for a qualitative estimate. However, in the following method the unshifted main resonance is determined from the spin-wave resonance peaks themselves. It can be shown that the first few spin-wave peaks also undergo similar, but small, shifts if they fall on the tail of the main resonance[9,10]; however, such shifts become totally negligible after a few peaks. Now, the exchange forces determine the magnetic-field separation of the spin-wave peaks from the unshifted main resonance. With these facts in mind, then, the position of the unshifted main resonance peak may be determined by a linear plot of the magnetic field for resonance for a particular spin-wave peak versus the square of the associated mode number, i.e., $H=\omega/\gamma+4\pi M-\alpha p^2$. Such a construction is informative. First of all, as is clear from Fig. 2, the lower-order peaks follow

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[7] G. T. Rado and J. R. Weertman, Phys. Rev. 94, 1386 (1954).

[8] See, e.g., M. H. Seavey and P. E. Tannenwald, Lincoln Laboratory, M.I.T., Technical Report No. 143, 1957 (unpublished).

[9] P. Pincus, Phys. Rev. 118, 658 (1960).

[10] M. H. Seavey, Jr., Lincoln Laboratory, M.I.T., Technical Report No. 239, 1961 (unpublished).

TABLE I. Measured parameters and experimental results of three Permalloy films.

Composition	L (Å)	$4\pi M$ (Oe)	g	$A\times10^{-6}$ (erg/cm)	$\langle\rho^2\rangle/r_1^2$
78–22	5150	10 490±30	2.09	0.91±0.03	120±15
63–37	6850	14 300±30	2.13	1.11±0.02	125±10
55–45	4050	14 950±50	2.14	1.20±0.05	170±45
±1	±50 Å		±0.02		

a quadratic dispersion law. Even the lowest-order peaks are very nearly quadratic. The slope of each line is the coefficient, α, of p^2 for that particular film; this allows the exchange stiffness constant A to be calculated once the saturation magnetization $4\pi M$ is determined. The intercept of each line yields the value of the magnetic field $H_\perp$ at 71.7 Gc/sec for resonance for the unshifted uniform precessional mode. It was not possible to determine the resonance field $H_{\parallel}$ for the unshifted uniform mode in the parallel configuration because no spin-wave resonance spectra were observable for the three films in this particular geometry. Therefore, the measured parallel fields were adjusted by the same number of oersteds (≈ 100 Oe) that the perpendicular fields were shifted by the exchange and electromagnetic effects. Only the g factor and $4\pi M$ were affected at all by this approximation, and only by about 0.5%. From the $H_\perp$ and $H_{\parallel}$ data, $4\pi M$ and the g factor were calculated; and A was determined from the optically measured film thickness and the above-mentioned slope α. These results are presented in the first five columns of Table I.

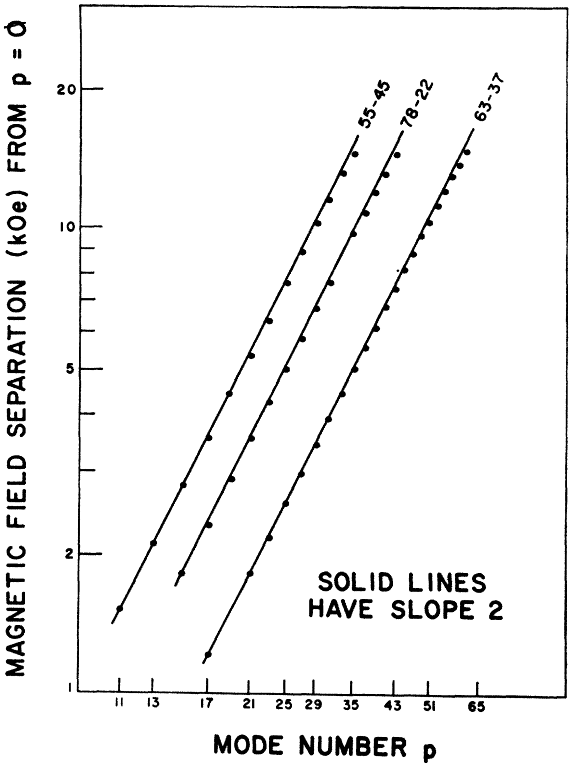

FIG. 3. Quadratic plus quartic region of SWR spectrum. The deviations from quadratic behavior become quite pronounced at the highest p numbers (~300 Oe).

To obtain a comprehensive view of the experimental data in terms of the variables in the dispersion relation, a log-log plot was made of the separation in magnetic field between the unshifted uniform resonance peak and a particular spin-wave resonance peak, $\bar{H}\equiv H_\perp - H_p$, versus the p-number of the spin-wave peak. Figure 3 exhibits a quadratic region and a region which deviates from quadratic behavior for the high p numbers (i.e., for large k values). The direction of curvature indicates that a term must be subtracted from the simple quadratic dispersion relation. Even though the effect appears to be small, for the highest p numbers realized here, the effect is at least 300 Oe for each film, or one and a half orders of magnitude larger than the total experimental uncertainty.

The data for the 63–37 film were computer-programmed for a least-squares fit to the function $\bar{H}=\alpha p^2 - \beta p^x$, with x, α, and β to be determined. This film was selected because of the high density of spin-wave resonance peaks measured over a large magnetic field interval (29 odd peaks over approximately 15 kOe). Since the dispersion was purely quadratic within the precision of the measurements for the lower p values, the range of the mode number p was restricted to the interval $p=23$ to $p=61$. The results were $x=4.01$, $\alpha=4.12$ Oe, and $\beta=3.51\times10^{-5}$ Oe. The maximum deviation between the computed values and the experimental values was 6.6 Oe over the whole range of p.

When the experimental errors in measuring the location of each spin-wave peak and in locating the uniform mode are accounted for, the results must be modified to be $x=4.0\pm0.1$, $\alpha=4.12\pm0.02$ Oe, and $\beta=3.5\pm0.2\times10^{-5}$ Oe.

The data for the remaining two films were treated graphically. The power law was quartic in each case, with the precision decreasing with decreasing film thickness. The primary reason for this is that the density of spin-wave peaks per dc magnetic field interval is less the thinner the film. Figure 4 shows graphically the quartic contribution to the spin-wave dispersion relation for each film. The coefficient of the fourth power term β was determined by fitting lines of slope 4.0 to the data.

The explicit relationship between the experimental coefficients α and β in $\bar{H}=\alpha p^2-\beta p^4$ and the moments of the exchange interaction is as follows: From the dispersion relation, the ratio of the quartic to quadratic term is

$$\frac{(1/24)\sum_r(\mathbf{k}\cdot\mathbf{r})^4J_r}{(1/2)\sum_r(\mathbf{k}\cdot\mathbf{r})^2J_r}=\frac{1}{4}\,\frac{ck^4\sum_r J_rr^4}{k^2\sum_r J_rr^2}=\tfrac{1}{4}p^2\left(\frac{\pi}{L}\right)^2\frac{c\sum_r J_rr^4}{\sum_r J_rr^2},$$

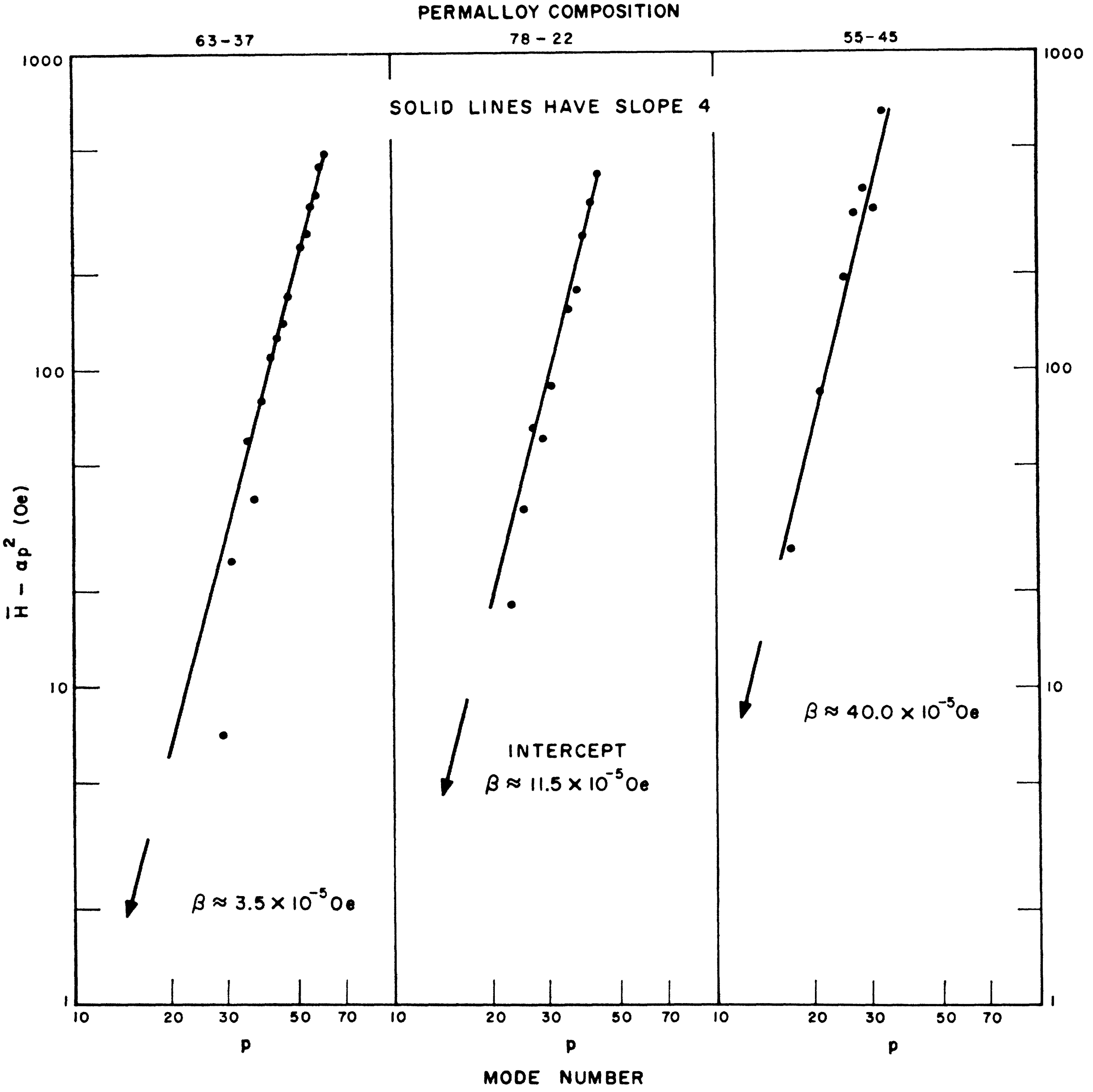

FIG. 4. Quartic contribution to spin-wave dispersion relation. Because ordinate represents differences between two large numbers, low p-number points fall on line within experimental uncertainty of ±10 Oe.

where

$$c \equiv \langle \sum_r J_r r_{\hat{k}}^4 \rangle / \sum_r J_r r^4 .$$

Here $r_{\hat{k}}$ denotes the components of r in the crystallite in the direction of k, and the brackets indicate that an appropriate average is to be taken. Hence

$$\frac{\beta}{\alpha} = \tfrac{1}{4}((\pi/L)^2 c \sum_r J_r r^4 / \sum_r J_r r^2).$$

The value of c evidently depends on the nature of the film and the geometry of the experiment.

It may be remarked that the deviations reported by Frait[11] at 36 Gc/sec in the spin-wave resonance spectrum of a 64–34 (nominal) Permalloy film for the higher p numbers can most likely be attributed to onset of the quartic term rather than the suggested magnon-phonon interaction.

DISCUSSION OF RESULTS

Dispersion Relation

As stated earlier, the dispersion relation for small k numbers is

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[11] Z. Frait, Phys. Status Solidi 3, K408 (1963).

$$\epsilon_k=2S\left[\frac{1}{2}\sum_r(\mathbf{k}\cdot\mathbf{r})^2J_r-\frac{1}{24}\sum_r(\mathbf{k}\cdot\mathbf{r})^4J_r+\cdots\right].$$

The spatial average of $\sum_r(\mathbf{k}\cdot\mathbf{r})^2J_r$ can be readily carried out and yields $\frac{1}{3}k^2\sum_r J_r r^2$. Because of the appearance of anisotropy, $\sum_r(\mathbf{k}\cdot\mathbf{r})^4J_r$ cannot be summed in closed form and be expressed in terms of a fourth moment of J_r except for random polycrystalline averaging, in which case the sum becomes $\frac{1}{5}k^4\sum_r J_r r^4$. For the moment, however, let us write the dispersion relation as

$$\epsilon_k=2S[\tfrac{1}{6}k^2\sum_r J_r r^2-(c/24)k^4\sum_r J_r r^4]\equiv\hbar[ak^2-bk^4],$$

in order to compare it with the experimental results. Since the data accurately fit the relation $\omega=ak^2-bk^4$ (even to the extent of determining the exponent in the k^4 term), the spin-wave dispersion relation is seen to be verified at room temperature in a metal, under conditions of selective excitation of specific spin wave vectors. The next higher order term, that is one involving k^6, would not be detectable since the shortest wavelengths excited in this experiment are of the order of 225 Å.

Long-Range Interactions Results

a. From Dispersion Relation

From the experimentally determined values for α and β, we find values for the long-range factor $\langle\rho^2\rangle/r_1^2=(\sum_r J_r r^4)/(r_1^2\sum_r J_r r^2)$, where r_1 is the nearest-neighbor distance. Our films are predominantly polycrystalline; for this case $c=\frac{1}{5}$, as mentioned before. Values for the long-range factor $\langle\rho^2\rangle/r_1^2$ for the three different films are given in the last column of Table I, assuming complete polycrystallinity.

In order to evaluate $\langle\sum_r J_r r_{\hat{k}}^4\rangle/\sum_r J_r r^4$ in the general case, the strength of the exchange integral J_r between neighbors, next-nearest neighbors, etc., must be known. There obviously exists no simple and unique way for doing this, but the large ratio of the fourth to second moment of the exchange interactions is clearly a manifestation of long-range exchange. For the case of nearest-neighbor interaction only, $\sum_r J_r r^4/\sum_r J_r r^2$ reduces to r_1^2 and the long-range factor $\langle\rho^2\rangle/r_1^2$ reduces to unity.

b. From Spin-Wave Interactions

Another manifestation of long-range interactions has been found in an experiment which measures spin-wave interactions.[2] The theoretically expected temperature variation of the exchange integral J is given by[5]

$$J(T)=J_0\left[1-\frac{\pi}{NS}\zeta(5/2)\frac{\sum_r J_r r^4}{\sum_r J_r r^2}\left(\frac{3kT}{4\pi S\sum_r J_r r^2}\right)^{5/2}\right].\quad(3)$$

This type of measurement was carried out in a very similar material (81–19 nominal Permalloy film) and, besides verifying the $T^{5/2}$ dependence at low temperatures, gave for the effective long-range factor $\sum_r J_r r^4/r_1^2\sum_r J_r r^2=112$.[12] This result agrees rather well with $\langle\rho^2\rangle/r_1^2=120$ for the 78–22 Permalloy sample determined in Sec. *a* assuming polycrystal average.

c. From Temperature Dependence of Magnetization

The temperature dependence of the saturation magnetization, in the spin-wave description, contains the well-known Bloch $T^{3/2}$ law plus a $T^{5/2}$ term which arises from retaining the k^4 term in the spin-wave dispersion relation. The detailed expression is

$$\frac{M_0-M(T)}{M_0}=\frac{\zeta(3/2)}{NS}\left(\frac{3kT}{4\pi S\sum_r J_r r^2}\right)^{3/2}+\frac{3\pi}{4NS}\zeta(5/2)\times\frac{\sum_r J_r r^4}{\sum_r J_r r^2}\left(\frac{3kT}{4\pi S\sum_r J_r r^2}\right)^{5/2}+\cdots.\quad(4)$$

The coefficient of the $T^{5/2}$ term in Eq. (4) is identical with the coefficient of the $T^{5/2}$ term in Eq. (3) except for the numerical factor $\frac{3}{4}$. The important point in comparing the two expressions is that even though their physical origin is different (spin-wave excitations leading to a reduction in saturation magnetization versus spin-wave interactions), both involve the same ratio of fourth to second moment of the exchange interactions, $\sum_r J_r r^4/\sum_r J_r r^2$, as a result of the same type of averaging occurring in the derivation. Consequently, comparison would provide an important check on the consistency of the model for a metal.

The temperature dependence data of the saturation magnetization of an 81–19 Permalloy film, measured by ferromagnetic resonance, of Ref. 2 has been analyzed in detail. Previously these data had only been used to evaluate the exchange integral J from the coefficient of the $T^{3/2}$ term. The $T^{3/2}$ term alone described the magnetization well only up to 80°K. If now, in addition, $4\pi[M_0-M(T)]-CT^{3/2}$ is plotted versus T on log-log paper, a well-defined, unique slope of 2.5 ± 0.1 results up to approximately 155°K (see inset of Fig. 5). A least-squares computer fit of the data to the function $4\pi[M_0-M(T)]=CT^{3/2}+ET^{5/2}$ yielded an excellent fit up to 150°K; that is, the calculated points deviated no more than experimental error (±5 Oe) from the measured points. The resulting fit is shown in Fig. 5. Inclusion of $T^{5/2}$ in the fitting reduced the previously computed coefficient of the $T^{3/2}$ term by 19%, bringing the corresponding calculated values of exchange parameter D, or exchange integral $J(D=2SJa^2$ at 0°K) into complete agreement with D or J measured from spin-

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[12] This value is twice the number reported in Ref. 2 because of an erroneous use of the cube edge instead of nearest-neighbor distance in the equation in paragraph 4.3 of Ref. 2.

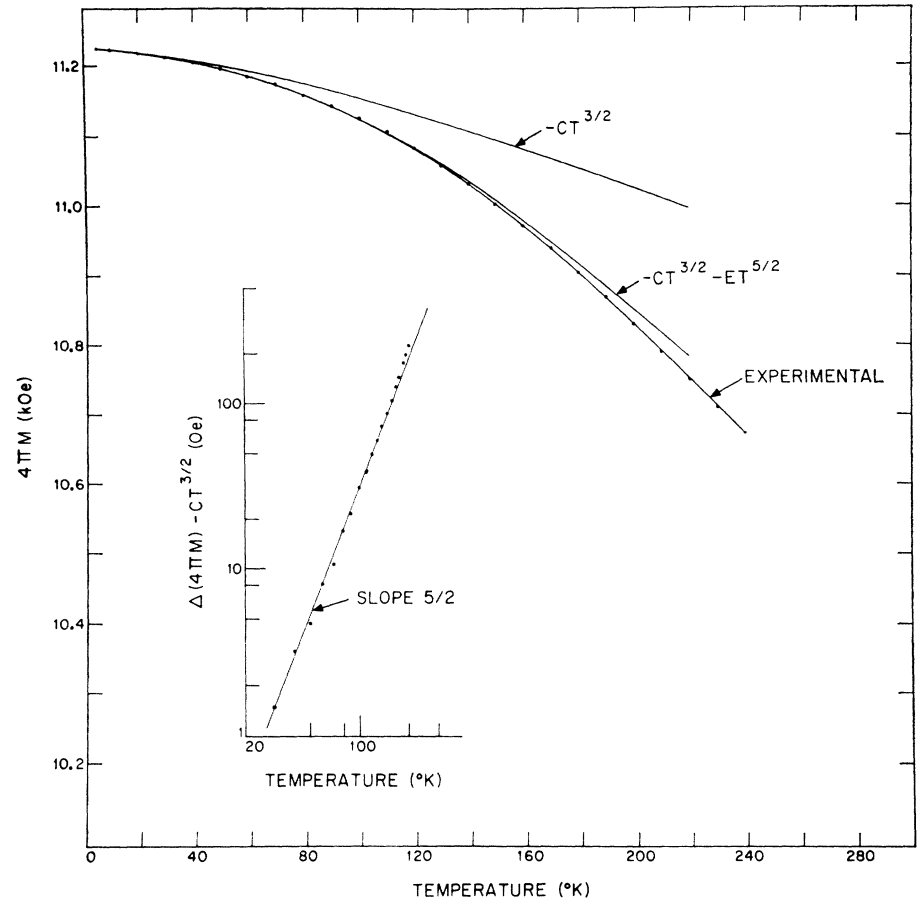

FIG. 5. Fit of saturation magnetization data of a nominal 81–19 Permalloy film to $-CT^{3/2}$ and to $-CT^{3/2}-ET^{5/2}$ law. Experimental points are good to ±5 Oe. Inset shows that $T^{3/2}$ term alone leaves a deviation that is fitted best with a $T^{5/2}$ term.

wave resonance on the same sample in Ref. 2.[13] The results are $J=280k$ from the Bloch law and $J=277k$ from spin-wave resonance. As a check, computer calculations over successively larger temperature intervals, beginning at 30°K, were made, and gave the same result.

The quantity of primary interest in this discussion, however, is the computed value of $E=3.02\times10^{-4}$ Oe deg$^{-5/2}$, which leads to an effective long-range interaction factor $\sum_r J_r r^4/r_1^2 \sum_r J_r r^2=25$.

Discussion of Long-Range Interactions

We see thus that the dispersion relation results and spin-wave interaction measurements on the same type sample agree within experimental error (neglecting possible anisotropy in the exchange). The interpretation of spin-wave interactions is based on a Heisenberg model.[6] On the other hand, the effective long-range determined from magnetization measurements on the same sample is smaller by a factor of five than the value expected from the k^4 term in the dispersion relation, even though the connection between the dispersion relation and the magnetization law only involves a statistical treatment of spin-wave excitations. The same type of discrepancy, of roughly the same order of magnitude, was already pointed out for nickel,[3] where pyromagnetic magnetization measurements were compared with spin-wave interaction data from neutron diffraction. In Permalloy, however, the IBM magnetization data[4] gave for the coefficient of the $T^{5/2}$ term the value 0.15×10^{-8} deg$^{-5/2}$ compared with our value of $E/4\pi M_0=2.7\times10^{-8}$ deg$^{-5/2}$ for the 78–22 film. This is clearly an experimental disagreement, and not one of theory.

In considering the discrepancy of the effective range determined from magnetization measurements up to 150°K, it must be remembered that the simple spin-wave interaction picture was shown experimentally to break down at about 80°K, at which temperature the $T^{5/2}$ magnetization term plays a very small role. Furthermore, since the coefficients of the terms in the dispersion relation increase as some power of $\langle\rho^2\rangle$, terms higher than the k^2 term may make significant contributions to the energy for relatively small k-values, severely restricting the usefulness of the expansion of the energy in powers of k^2. In fact, given the present range, the k^2 and k^4 terms would become equal at about 40 Å.

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[13] It can be readily shown that a T^4 term need not be considered, inasmuch as its contribution to the decline of the saturation magnetization is negligible up to 150°K, even if experimental long-range were included in its coefficient.

Similarly, the magnetization expression becomes invalid at about 30°K. (The equivalent temperature of the spin waves excited in this experiment is about 2°K.) Thus comparison with the magnetization expression must be questioned since the validity of the spin-wave energy expansion becomes doubtful. This difficulty could ultimately be circumvented with numerical substitution of the measured dispersion relation into the Bose-Einstein function.

It may also be that modes of excitation other than those measured through the dispersion relation—perhaps due to polarized conduction electrons—contribute a cancelling $T^{5/2}$ term to the magnetization.

Measurement of the spin-wave dispersion relation inherently involves more detail concerning exchange interactions than can be provided by magnetization or spin-wave interactions measurements. Consequently, three new factors must be considered in some detail: the crystal structure of the film samples, the appropriate averaging process, and a model for the range dependence of the J_r's.

It seems clear that the polycrystalline averaging approach would be justified, provided the film consisted of randomly oriented crystallites and the average crystallite size is much smaller than the spin wavelength. This, in fact, is not the case. The microcrystal size distribution runs up to 1000 Å, with the average size being of the order of 200 Å. Magnetic films are frequently known to be partially oriented, that is, to have a preferred "fiber" axis normal to the film surface and a random orientation about this axis. X-ray diffraction analysis of our films does indeed show a partial [111] fiber axis, but quantitative measurements as to the fraction of crystallites that are oriented remain to be carried out. For the complete [111] fiber axis case, and spin-wave propagation along this direction, $c\sum_r J_r r^4$ becomes $(8/3)r_1^4[J_1+J_2+16J_3+\cdots]$ compared with $(12/5)r_1^4[J_1+2J_2+16J_3+\cdots]$ for the polycrystalline case. Evidently, quantitative comparison of the two expressions can only be made if some range dependence of the J_r's—possibly anisotropic—can be postulated. One possible model involves the Ruderman-Kittel type of long-range interaction with an anisotropic Fermi surface.[14] For nearest neighbors only, the coefficient c which is $\frac{1}{5}$ in the polycrystalline case, becomes 2/9 in the [111] case.

CONCLUSION

From the work that has been performed, it is clear that (1) the range of exchange interaction between spins is substantially greater than nearest-neighbor distance over a range of Permalloy compositions, (2) there is good agreement between the range found in the present work and the range determined from spin-wave interaction measurements, and (3) the saturation magnetization of Permalloy may be described by the usual Bloch term plus a $T^{5/2}$ term up to 150°K, the shorter range indicating that the coefficient of the $T^{5/2}$ term may be determined to some extent by other than spin-wave modes of excitation in the metal.

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[14] L. M. Roth, H. J. Zeiger, and T. A. Kaplan (private communication).