straight line is plotted through $T/T_N=1.000$ and (somewhat arbitrarily, although it makes little difference) the data point at $T=284.4°K$ for a number of β values. The deviation of the data points from this straight line is given in the lower part of Fig. 2 for five different β values. The best fit is for $\beta \cong 0.29$ which is the same conclusion found by correcting Fig. 1. Within experimental accuracy, set mainly by the tetragonality data, the β thus appears to assume an Ising value in agreement with the experimental specific-heat measurements in the critical region.[12]

We would especially like to thank Professor H. E. Stanley for many helpful discussions and for showing that the simple shift in $T_N(T)$ included in Fig. 1 gives the same result as Fig. 2. We are also indebted to M. B. Salamon for discussions of his work and ours and for his preprint. We would in addition like to thank J. Als-Nielsen, R. Harrison, L. P. Kadanoff, and C. B. Walker for stimulating discussions. This work was supported by the National Science Foundation to whom it is a pleasure to express our appreciation.

[1]H. P. Rooksby, Acta Cryst. 1, 226 (1948).
[2]R. C. Lambs and H. P. Rooksby, Nature 165, 442 (1950).
[3]J. Nagamiya, S. Saito, Y. Shimomura, and E. Uchida, J. Phys. Soc. Japan 20, 1285 (1965).
[4]P. C. Clapp and S. C. Moss, Phys. Rev. 142, 418 (1966); 171, 754 (1968).
[5]W. L. Roth, Phys. Rev. 111, 772 (1958).
[6]B. van Laar, Phys. Rev. 138, A584 (1965).
[7]S. Saito, K. Nakahigashi, and Y. Shimomura, J. Phys. Soc. Japan 21, 850 (1966).
[8]J. Als-Nielsen and O. W. Dietrich, Phys. Rev. 153, 717 (1967), on β'-CuZn; L. M. Corliss, A. Delapalme, J. M. Hastings, H. Y. Lau, and R. Nathans, J. Appl. Phys. 40, 1278 (1969), on $RbMnF_3$; J. C. Norvell, W. P. Wolf, L. M. Corlis, J. M. Hastings, and R. Nathans, Phys. Rev. 186, 557 (1969), on DyAlG where $\beta \cong \frac{1}{4}$ due to long-range dipolar forces. This is discussed at greater length by N. E. Frankel and A. E. Spargo, Phys. Letters 31A, 442 (1970).
[9]G. A. Baker, Jr., and J. W. Essam, Phys. Rev. Letters 24, 447 (1970).
[10]J. C. Norvell and J. Als-Nielsen, to be published.
[11]D. R. Chipman and C. B. Walker, Bull. Am. Phys. Soc. 15, 363 (1970).
[12]M. B. Salamon, to be published.
[13]R. Uno, J. Phys. Soc. Japan 18, 1686 (1963).
[14]B. van Laar, Phys. Rev. 141, 538 (1966).
[15]E. Uchida, R. Fukuoka, H. Kondoh, J. Takeda, Y. Nakazumi, and J. Nagamiya, J. Phys. Soc. Japan 19, 2088 (1964).
[16]D. Bloch, F. Chaisse, and R. Pauthenet, J. Appl. Phys. 37, 1401 (1966).
[17]P. Heller, Phys. Rev. 146, 403 (1966).

MACROSCOPIC AND MICROSCOPIC THEORIES OF DIPOLE-EXCHANGE SPIN WAVES IN THIN FILMS: CASE OF THE MISSING SURFACE STATES

T. Wolfram and R. E. De Wames
Science Center, North American Rockwell Corporation, Thousand Oaks, California 91360
(Received 18 March 1970)

Calculations of dipole-exchange spin waves in thin films using a microscopic model show no evidence of surface modes above the lowest bulk frequency, in contrast to recent results based on a macroscopic model. The apparent absence of the surface branch in the former model is explained in terms of the admixture of bulk and surface waves. Nearly exact quantitative agreement between the two models is obtained for wave vectors as large as 10^7 cm^{-1}.

A simple ferromagnetic insulator is characterized by short-range exchange interactions and long-range dipole-dipole interactions. The Heisenberg exchange model (valid for large wave vectors) predicts surface spin-wave modes with frequencies below the bulk modes.[1-3] In contrast, magnetostatic theory (valid for small wave vectors) includes only dipolar effects and predicts a spin-wave surface mode above the lowest bulk frequency.[4] The behavior of surface and bulk spin waves in the transition region for which both dipolar and exchange effects are important has been a subject of considerable interest recently.[5-13] Mills[14] stated that the magnetostatic Damon-Eshbach (DE) surface branch would decrease with increasing wave vector and exit below the bulk band to join smoothly with the Heisenberg surface branch. The results of a microscopic theory in which the discrete exchange and dipolar interactions of a 30-atomic-layer system were numerically calculated were reported by Benson and Mills[8] (BM). These authors reported that

they were unable to find evidence of surface modes above the lowest bulk frequency. They commented that all surface modes found in their work had frequencies below the bulk modes except in the long-wavelength limit. These results were compatible with the original suggestion that except for the very-long-wavelength limit in which magnetostatic theory is valid, surface spin waves occur only below the bulk frequencies.

More recently a macroscopic theory based on the solutions of a sixth-order differential equation derived from Maxwell's equations together with the equations of motion for the magnetization has been studied by Wolfram and De Wames.[5-10] Detailed studies of the bulk and surface spin waves in the transition region have been reported. The correct eigenstates are admixtures of the magnetostatic DE surface waves and bulk waves. Spin-wave states with localized surface character were found for a system of approximately 1500 atomic layers[5] (see Fig. 2 of Ref. 5) at frequencies above the lowest bulk states. In addition the frequency of the surface branch increases with increasing wave vector in contrast to the suggestion of Mills.

The origin of the qualitative differences between microscopic and macroscopic theories was not evident. Since the microscopic theory was presumably the more accurate method, some doubt existed as to the validity of applying Maxwell's equations to very thin films.

The purpose of this paper is to show that no qualitative discrepancy exists and that the microscopic and macroscopic theories are in reasonable quantitative agreement. In particular, we show that the BM spin-wave states are admixtures of bulk and DE surface states and that the strong mixing of the surface states with the bulk states accounts for the distortion of the magnetization from a simple cosinusoidal shape.

A detailed discussion of the macroscopic theory has been given previously.[9] We present here a very brief description.

Consider a ferromagnetic film extending from $y=0$ to $y=s$ and infinite in the x-z plane with a constant applied magnetic field H_0 in the z direction. Maxwell's equations $\nabla\times\vec{h}=0$ and $\nabla\cdot(\vec{h}+4\pi\vec{m})=0$ together with the equation of motion for the magnetization,

$$-i\omega\vec{m}=\gamma\vec{M}_s\times\left[\vec{h}-\left(\frac{H_i}{M_s}-D\nabla^2\right)\vec{m}\right]=0, \tag{1}$$

lead to a sixth-order differential equation for the magnetic potential ψ ($h=-\nabla\psi$),

$$\left\{(\mathcal{O}^2-\Omega^2+\mathcal{O})\nabla^2-\mathcal{O}\frac{\partial^2}{\partial z^2}\right\}\psi=0. \tag{2}$$

The operator $\mathcal{O}=\Omega_H-(D/4\pi)\nabla^2$ and the dimensionless parameters are $\Omega_H=(H_i/4\pi M_s)$ and $\Omega=(\omega/4\pi\gamma M_s)$. In the above equations $\vec{h}$ and $\vec{m}$ are the small components of the magnetic field and the magnetization which vary in time as $\exp(-i\omega t)$, $\vec{M}_s$ is the saturation magnetization directed along the z axis, H_i is the internal field which in this case is equal to H_0, and D is the exchange parameter. Outside of the film $\nabla^2\psi=0$. The potential ψ is of the form $\exp(ik_x x+ik_z z)f(y)$. The essential physics involved is more apparent if we write Eq. (2) in terms of three second-order differential operators:

$$\mathcal{O}_1\mathcal{O}_2\mathcal{O}_3\psi=0, \tag{3}$$

where $\mathcal{O}_\alpha=(D/4\pi)\partial^2/\partial y^2+r_\alpha$ and the variables r_α are the three roots of the cubic equation

$$r^3+r^2[2\Omega_k+1+(D/4\pi)k^2]+r\{\Omega_k{}^2+\Omega_k-\Omega^2+[(2\Omega_k+1)k^2-k_z{}^2](D/4\pi)\}$$
$$+[(\Omega_k{}^2+\Omega_k-\Omega^2)k^2-\Omega_k k_z{}^2](D/4\pi)=0, \tag{4}$$

where $\Omega_k=\Omega_H+(D/4\pi)k^2$ and $k^2=k_x{}^2+k_y{}^2$.

The solutions of Eq. (2) may be constructed from the three characteristic types of solutions,

$$\psi=\exp(ik_x x+ik_z z)\sum_{\alpha=1}^{3}(A_\alpha\psi_\alpha{}^{+}+B_\alpha\psi_\alpha{}^{-}) \quad (0<y<s), \tag{5}$$

where $\mathcal{O}_\alpha\psi_\alpha{}^{\pm}=0$ and the functions $\psi_\alpha{}^{\pm}$ are just the plane waves $\exp\{\pm[4\pi r_\alpha/D]^{1/2}y\}$. The solutions outside of the film are

$$\psi=\exp(ik_x x+ik_z z)C\exp(-ky), \quad y>s,$$
$$=\exp(ik_x x+ik_z z)D\exp(ky), \quad y<0. \tag{6}$$

A study of Eq. (4) reveals that one of the roots r_1 is real and produces a bulk wave similar to the magnetostatic bulk wave, while the other two roots r_2 and r_3, are imaginary. The r_2 wave is similar to the DE surface wave. The r_3 root corresponds to a very highly localized surface wave which has no analog in magnetostatic theory.

The admixture amplitudes A_α and B_α (also C and D) which enter into the eigenstates are determined by the boundary conditions. The tan-

gential components of $\vec{h}$ and the normal components of $\vec{h}+4\pi\vec{m}$ must be continuous across the surfaces at $y=0$ and $y=s$. This gives only four of the eight required equations because the tangential boundary conditions are redundant. Four additional conditions are obtained which include microscopic surface perturbations as well as dipolar effects by starting from the linearized microscopic equations of motion and passing to a continuum representation. This procedure leads to equations containing the values of $\vec{m}$ and its normal derivatives at the surfaces. These detailed boundary equations have been given elsewhere[9] but are not essential to the present discussion. Numerical calculations have verified the fact that both the eigenvalues and eigenvectors are relatively insensitive to large variations in the latter type of boundary conditions for $k \lesssim 10^7\ \mathrm{cm}^{-1}$. These microscopically derived boundary conditions control principally the amplitude of the highly localized r_3 wave. Variations in the r_3 wave cause changes in the magnetization eigenfunction only very near the surfaces and cause little change in the gross shape.[8,9]

Imposition of the boundary conditions on the solutions leads to a matrix eigenvalue equation. We have solved these equations numerically for the case of a film whose thickness, exchange constant, and saturation magnetization corresponds to the parameters used by BM. The corresponding parameters are $s=30$, $D/4\pi=(J_1+4J_2)/(4\pi M_s)=3/2.5$, $4\pi M_s=2.5$, $H_0=1.0$, $\varphi_x=k_x$, $\varphi_y=k_y$, and $\varphi_z=k_z$. Lengths are measured in units of the lattice spacing a, wave vectors in units of a^{-1}, and energies in units of $4\pi\gamma M_s$, where γ is the gyromagnetic ratio. These parameters are not typical of those used in previous calculations and lead to very large surface-bulk admixtures.[15]

Before describing the results at large values of k_x it is important to note that when $\varphi_x=\varphi_z=0$ there is no mixing between the bulk and surface waves and the solution of Eq. (2) yields for the bulk exchange eigenstates (in the units described above)

$$\omega_n=(4\pi M_s)[H_n(H_n+1)]^{1/2},$$
$$H_n=(1/4\pi M_s)[H_0+(J_1+4J_2)(n\pi/s)^2],$$
$$\varphi_y=(n\pi/s)\quad(n=1,2,3\cdots). \tag{7}$$

In this case the Kittel uniform mode is a mode belonging to the DE surface branch whose energy is given by Eq. (7) with $n=0$. It should be noted that the φ_y wave vectors are quite large for $s=30$ even for small n so that Eq. (7) is not a long-wavelength result. The accuracy of this macroscopic result is quite good. For example, it gives 1.8708, 1.9103, 2.0272, and 2.2181 for the first four lowest levels ($n=0$, 1, 2, and 3). By numerically summing the discrete dipole and exchange interactions, BM obtained the values 1.8725, 1.9101, 2.0297, and 2.2181. The magnetization wave functions are nearly cosinusoidal curves and are essentially identical for the two theories.

As φ_x increases the surface branch rises more rapidly than the bulk exchange branches because of dipolar effects,[4] and in the absence of the admixture effect it would cross the bulk exchange branches. Because of the mixing, the branches repel each other and do not cross. The spin-wave spectrum with and without the admixture effect is shown schematically in Fig. 1. For the parameters considered here the mixing is quite large. For example, at $\varphi_x=3(\pi/30)=0.1\pi$, $\varphi_z=0$, the first four levels are approximately equal admixtures of surface and bulk waves. In Fig. 2 we show the x component of the magnetization as a function of y across the film for the four lowest levels. The dashed curves are sketches of the BM results (Fig. 3, Ref. 8) which are normalized to our curves at one of the maxima. The percent-

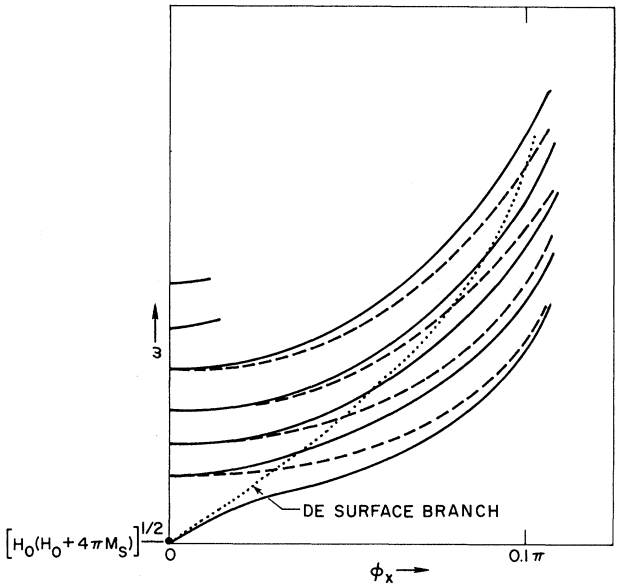

FIG. 1. A schematic of the spectrum for $\varphi_z=0$ as a function of φ_x. The dashed curves represent the bulk-exchange branches in the absence of the admixture effect. The dotted curve is the unmixed surface branch. The solid curves are the admixed bulk-surface eigenstates.

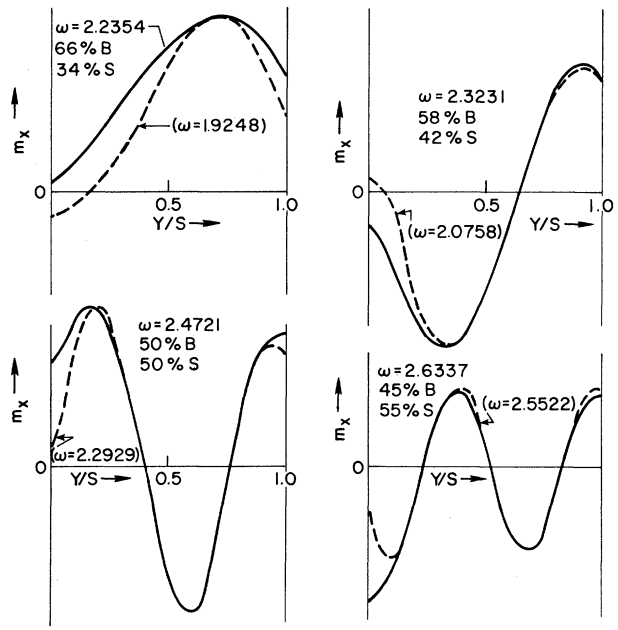

FIG. 2. The x component of the magnetization for the first four lowest levels with $\varphi_x = 0.1\pi$, and $\varphi_z = 0.0$. The solid curves are obtained from the macroscopic theory while a sketch of the microscopic BM results is indicated by the dashed curves. The BM eigenfrequencies are enclosed in parentheses. The percentage surface (S) and bulk (B) admixture for each state is indicated.

age surface (S) and bulk (B) amplitudes for each state are also indicated. The fact that no magnetization wave function is highly localized at the film surface is a result of the large admixture of the bulk wave. Nearly pure surface states (i.e., r_2 waves which are eigenstates) exist at smaller values of φ_x. The eigenvalues obtained by BM (shown in parentheses) appear to be converging to our values for the higher levels, but differ by about 15% for the first level. This discrepancy does not appear to be due to intrinsic differences between the microscopic and macroscopic theories but at the present time has not been resolved. Nevertheless, the qualitative agreement between the two approaches is good enough to assert the following conclusions:

(1) The microscopic and macroscopic theories are in general qualitative agreement. (2) Spin waves in the transiton region in which both dipole and exchange interactions are important are admixtures of surface and balk waves. (3) For $\varphi_x = 0$ and $\varphi_z = 0$ the low-lying spin-wave modes of films consisting of as few as 30 atomic layers may be calculated accurately from the simple macroscopic result of Eq. (7).

Recent conclusions of Sparks[16] based on erroneous interpretations and incorrect boundary conditions are contradicted by both the present results and the results of Benson and Mills.

[1]R. F. Wallis, A. A. Maradudin, I. P. Ipatova, and A. A. Klockikhin, Solid State Commun. 5, 89 (1967).
[2]R. E. De Wames and T. Wolfram, Phys. Rev. 185, 720 (1969).
[3]R. J. Jellitto, Z. Naturforsch. 19a, 1567 (1964).
[4]R. W. Damon and J. R. Eshbach, J. Phys. Chem. Solids 19, 308 (1960).
[5]T. Wolfram and R. E. De Wames, Solid State Commun. 8, 191 (1970).
[6]R. E. De Wames and T. Wolfram, Appl. Phys. Letters 15, 297 (1969).
[7]T. Wolfram and R. E. De Wames, Phys. Letters 30A, 3 (1969).
[8]R. E. De Wames and T. Wolfram, Solid State Commun. 7, 1451 (1969).
[9]R. E. De Wames and T. Wolfram, J. Appl. Phys. 43, 987 (1970).
[10]T. Wolfram and R. E. De Wames, Phys. Rev. (to be published).
[11]V. V. Gann, Fiz. Tverd. Tela 8, 3167 (1967) [Sov. Phys.–Solid State 8, 2537 (1967)].
[12]H. Benson and D. L. Mills, Phys. Rev. 178, 839 (1969).
[13]H. Benson and D. L. Mills, Phys. Rev. 188, 849 (1969).
[14]D. L. Mills, *Localized Excitations in Solids,* edited by R. F. Wallis (Plenum, New York, 1968), pp. 426–433 (see p. 430).
[15]For these parameters the exchange energy becomes equal to the dipolar energy for wave vectors of the order a^{-1} while for a material like yttrium iron garnet (YIG) the exchange and dipole energies are equal for wave vectors of the order of $10^{-2}a^{-1}$. In terms of the units used in the text $D/4\pi \approx 3\times10^{4}a^{-2}$ for YIG.
[16]M. Sparks, Phys. Rev. Letters 24, 1178 (1970).