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Magnetic Surface Modes: Classification, New Types, and Simple Method of Solution

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A physical interpretation of the eigenvalue equation is used to obtain the frequencies and spin configurations of ferromagnetic and antiferromagnetic surface modes and to provide a classification of these modes. A physical understanding of the features of surface modes also is provided. Since all spins precess at the same frequency in a normal mode, the frequency can be obtained from the equations of motion of one, or sometimes two or three, spins near the surface. One class of surface modes is the *changed-surface-parameter class*, in which the missing neighbors of a surface spin are compensated for by a changed parameter, such as an exchange constant, at the surface. Another is the *equivalent-layer class*, in which the crystal contains equivalent layers such that the net torque on each spin $\vec{S}$ on an equivalent layer exerted by all spins on the surface side of $\vec{S}$ is zero. Two new types of surface waves are studied. In the first, the spin precession changes phase as well as amplitude as a function of the distance from the surface. In the second, a surface layer of spins is antiparallel to the bulk spins.

1. INTRODUCTION

There have been a number of recent investigations of surface waves in magnetic systems.[1–7] In the present paper, a physical interpretation of the eigenvalue equation is used to obtain the normal-mode frequencies (eigenvalues) and the spin configurations (eigenvectors) of ferromagnetic and antiferromagnetic surface waves. An intuitive understanding of how the crystal can support the surface modes and of the physical features of the modes is afforded by the interpretation, which is used to classify surface modes and to study two new types of surface modes. Simple physical explanations of existing results also are afforded by the method, which is applicable to all presently known exponentially decaying modes, both acoustical and optical.

In a normal mode, all spins precess at the same frequency, by definition. By requiring the frequency of a surface spin to be the same as that of a bulk spin and assuming a form of the solution for the bulk spins (exponential for surface modes), the normal-mode frequencies can be obtained. In particular, a spin on the surface will have fewer neighbors than a corresponding spin in the bulk, and the missing torque on the surface spin resulting from the missing neighbors must be compensated for somehow. If some parameter, such as an exchange constant, anistropy field, etc., of the surface spins is different from that of the bulk spins, the frequency change of a surface spin resulting from the difference between the surface parameter and the bulk parameter sometimes can balance the frequency change from the missing neighbors. Such modes constitute the *changed-surface-parameter class* of surface modes.

When the surface parameters are the same as those of the bulk, a surface mode still can exist in some cases as follows: Assuming that the bulk spins in a crystal lie to the right of the surface plane, the net torque on a surface spin from all spins to the left is zero since there are no spins to the left. The spin arrangement sometimes may be such that the net torque on every spin in the second layer from all spins in the first layer is zero also. Similarly, the torque on each spin in the third layer from all spins in the first two layers is zero, etc. Thus, it is not necessary to compensate for the missing neighbors of the surface spin by changing some parameter at the surface, the spins on each layer already being equivalent to those in the surface layer.

Another possibility, which is common in antiferromagnetic crystals, is that the crystal can be divided into equivalent pairs of layers such that the torque on every spin in the third layer from all spins in the first two layers is zero, the torque on every spin in the fifth layer from all spins in the first four layers is zero, etc. For more complicated systems, the number of layers in the sets could be greater than two. All such modes will be said to be in the *equivalent-layer class*. Keffer[8] independently realized that the torque on the spins in every other layer of a simple cubic antiferromagnet with a (100) surface from the spins to left was zero for the surface waves of this system.

The first new type of mode, which will be called the *flip-surface type*, is assumed to have a ground state with the spins in the first layer antiparallel to the bulk spins, as illustrated schematically in Fig. 1(a). For the second type, which will be called the *oscillating-decaying type*, not only does the spin precession *amplitude* decay with distance away from the surface, but also the precessional *phase angle* changes with the distance from the surface, as illustrated schematically in Fig. 1(b).

Wallis, Maradudin, Ipatova, and Klochikhin[2] suggested that the characteristic required for the existence of surface modes with near-neighbor interaction and no change in the surface parameter probably was that the exchange interaction must couple spins whose line of centers is not normal to the surface. For the exponentially decaying states in a ferromagnet, this result is a special case of the equivalent-layer class. The requirement of non-normal coupling between the spins on different layers and the additional requirement that the wave vector $\vec{k}_f = (k_y, k_z)$ in the plane of the surface must be nonzero for near-layer coupling can be understood as follows: With only normal bonds, a spin $\vec{S}_2$ on the second layer is coupled to only one spin $\vec{S}_1$ on the surface layer; thus the torque on $\vec{S}_2$ from all coupled spins on the first layer ($\vec{S}_1$ in this case) cannot be zero unless $\vec{S}_1$ and $\vec{S}_2$ are parallel, i.e., the wave-vector component k_x along the normal to the surface is zero, and there is no decay of the amplitude as a function of the distance from the surface. If there is a non-normal coupling, $\vec{S}_2$ can be coupled to two or more surface spins whose vector sum is parallel to $\vec{S}_2$, thus allowing a nonzero k_x. If $\vec{k}_f = 0$, then all spins in the surface layer are parallel, and each surface spin is parallel to $\vec{S}_2$ when this vector sum is parallel to $\vec{S}_2$, again making $k_x = 0$.

Elementary examples of the equivalent-layer and changed-surface-parameter classes of surface modes are given in Sec. 2 in order to illustrate the method. The remaining sections are devoted to the oscillating-decaying and the flip-surface modes. The crystal will be considered as a semi-infinite medium, and only nearest-neighbor exchange interactions will be considered. The re-

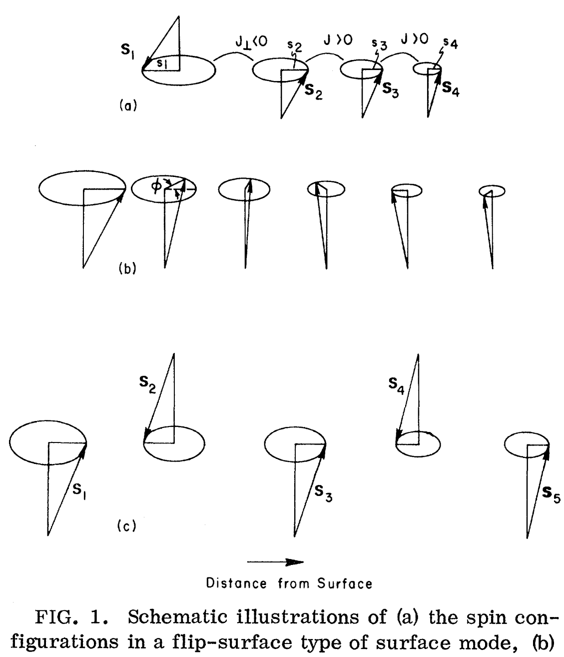

FIG. 1. Schematic illustrations of (a) the spin configurations in a flip-surface type of surface mode, (b) the spin configurations in an oscillating-decaying type of surface mode, and (c) the spins in a surface mode of a simple cubic antiferromagnetic having a (111) surface for $\phi = 0$.

sults should apply to a sample of finite thickness for modes whose decay length $1/k_x$ is short with respect to the sample thickness. The effects of dipole-dipole interactions and of imperfect surfaces are neglected in the present paper. The method also may be useful in studying other surface modes, such as elastic and magnetoelastic ones.

2. CHANGED-SURFACE-PARAMETER AND EQUIVALENT-LAYER CLASSES

Consider the changed-surface-parameter surface modes in a simple cubic ferromagnet having near-neighbor exchange coupling and having a (100) plane as its surface.[3,7] The exchange constant is assumed to be equal to J for all pairs of spins except those pairs having both spins in the surface, for which the exchange constant has the value $J_{\parallel}$. A few spins of a surface wave are illustrated schematically in Fig. 2(a) for the case of $k_y \neq 0$. Figure 2(b) illustrates that the torque which would be expected on $\vec{S}_1$ from the missing neighbor $\vec{S}_{\text{miss}}$ is in the opposite direction from that of the two neighboring surface spins $\vec{S}_{1u}$ and $\vec{S}_{1d}$. Thus, the value of $J_{\parallel}$ must be less than J in order to have the torque on $\vec{S}_1$ unchanged. There are no acoustical surface modes for this system if $J_{\parallel} \geq J$.

Writing $J_{\parallel} = J - \Delta J$ and equating the torque from $-\Delta J$ to that from the missing spin and using $s_{\text{miss}} = e^{k_x a}\ s_1$, where a is the lattice constant, gives

$$e^{k_x a} = 1 + 4\ (\Delta J/J)\ \Lambda_k\ , \qquad (2.1)$$

where s_1 and s_{miss} are the transverse components of the spins ($\vec{S} = S_z\hat{z} + \vec{s}$), and

$$\Lambda_k \equiv 1 - \tfrac{1}{2}\ (\cos k_y a + \cos k_z a)\ . \qquad (2.2)$$

The torques can be written simply by using the method presented in the Appendix.

The results of Filipov[3] for $J_{\parallel} > J$ correspond to growing exponentials.[7] This is intuitively clear from the simple physical argument given above, or from (2.1).

In the long-wavelength limit $k_j a \ll 1$, for $j = x, y, z$, Eq. (2.1) gives

$$k_x a = (\Delta J/J)\ (k_f a)^2\ , \qquad (2.3)$$

where $k_f^2 \equiv k_y^2 + k_z^2$. The usual frequency $|\gamma| Dk^2$, with an imaginary component ik_x of $\vec{k}$ corresponding to the exponential decay, is

$$\omega = |\gamma| D(k_f^2 - k_x^2)\ . \qquad (2.4)$$

Combining (2.3) and (2.4) gives

$$\omega = |\gamma| Dk_f^2 - |\gamma| D[(\Delta J/J)a]^2 k_f^4 \qquad (2.5)$$

and the surface mode has a lower frequency than that of the bulk mode, the first correction to the bulk-mode frequency being proportional to k_f^4.

Note that by considering the equation of motion of the surface spin, the results (2.1) and (2.5) were obtained directly, a simple physical explanation of why $J_{\parallel}$ must be less than J was afforded, and an intuitive explanation of how the surface wave is supported was obtained. The environments of spins $\vec{S}_2$, $\vec{S}_3$, ... in Fig. 2(a) are the same, and the difference in the environment of the surface spin $\vec{S}_1$ (i.e., the missing neighbor) is compensated for by the changed surface exchange.

The results[7] for the optical modes of this system can be obtained by the present technique. With the sign changes appropriate to an optical mode, (2.1) becomes

$$(J_{\parallel} - J)/J = \tfrac{1}{4}(1 + e^{k_x a})/\Lambda_k\ .$$

The minimum value of $J_{\parallel}/J = \frac{5}{4}$ in this result corresponds to $k_x = 0$ and $k_y = k_z = \pi/a$. For this configuration, all five neighbors of a surface spin $\vec{S}_1$ have $s_{nx} = -s_{1x}$, and all six neighbors of a bulk spin $\vec{S}_b$ have $s_{nx} = -s_{bx}$. Since the four neighbors of $\vec{S}_1$ which are in the surface layer have exchange constant $J_{\parallel} = \frac{5}{4} J$ and the other neighbor has ex-

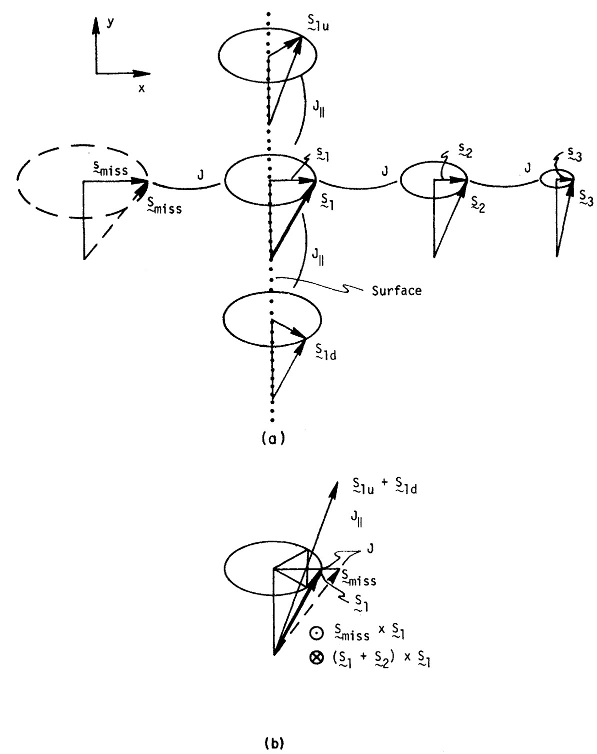

FIG. 2. (a) Spin configurations are shown for several spins near the surface for a simple cubic ferromagnetic with a (100) surface. The spin $\vec{S}_{\text{miss}}$ can be considered as having been removed in order to form the surface. (b) $\vec{S}_1$, $\vec{S}_{\text{miss}}$, and the two *surface* spins $\vec{S}_{1u}$ and S_{1d} to which $\vec{S}_1$ is coupled and is used in calculating the torques on $\vec{S}_1$ are shown.

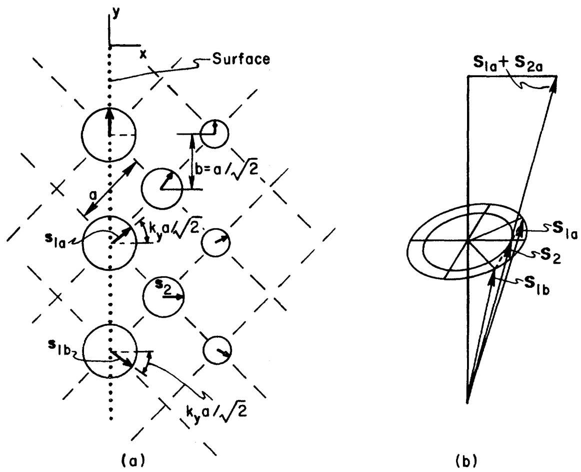

FIG. 3. (a) The transverse components of several spins in the first three layers of a simple cubic ferromagnetic having a (110) surface are shown. (b) $\vec{S}_2$ and the two spins $\vec{S}_{1a}$ and $\vec{S}_{1b}$ in the surface layer to which $\vec{S}_2$ is coupled and is used in calculating the torque on $\vec{S}_2$ are shown.

change constant J, the net exchange constant for the surface spin is $4(\frac{5}{4})\,J + J = 6J$. This value is the same as that of a bulk spin, thus giving equal frequencies of the surface and bulk spins, as required in a normal mode.

Next consider the equivalent-layer-surface modes in a simple cubic ferromagnet whose surface is a (110) plane.[2] The transverse components of several spins in the $z = 0$ plane are shown in Fig. 3(a) for the case of $k_y \neq 0$. The only surface spins to which $\vec{S}_2$ is coupled are $\vec{S}_{1a}$ and $\vec{S}_{1b}$. In order for the torque on $\vec{S}_2$ from $\vec{S}_{1a}$ and S_{1b} to be zero, $\vec{S}_{1a} + \vec{S}_{1b}$ must be parallel to $\vec{S}_2$. As seen in Fig. 3(b), this requires

$$s_2 = s_1 \cos k_y b \ , \qquad (2.6)$$

where $b = a/\sqrt{2}$, with a the lattice spacing. With $s_2 = e^{-k_x b}\, s_1$, this gives

$$e^{-k_x b} = \cos k_y b \ . \qquad (2.7)$$

It is seen from (2.7) or directly from Fig. 3(b) that there is no surface wave (no $e^{-k_x b} < 1$) unless $k_y \neq 0$. The condition $k_z \neq 0$ does not give a surface wave since the torque on a spin in the second layer is not affected by changing k_z.

By the method given in the Appendix, the equation of motion of $\vec{S}_{1b}$ is

$$-(\omega/SJ)\,s_1 = 4s_1 - 2s_1 \cos k_z a - 2s_2 \cos k_y b \ .$$

Eliminating s_2 by using (2.6) gives

$$-(\omega/2SJ) = 1 + \sin^2(k_y a/\sqrt{2}\,) - \cos k_z a \qquad (2.8)$$

in agreement with the results of Wallis and coworkers.[2] Note that the torque on $\vec{S}_{1b}$ from all of the spins on the second layer is not zero even though the torque on $\vec{S}_2$ from all spins in the surface layer is zero.

3. FERROMAGNETIC OSCILLATING-DECAYING MODE

The surface modes for a simple cubic ferromagnet having (100) and (110) surfaces have been reported in the literature.[2,7] We now show that the surface modes for a (111) surface with no change in surface parameters have an interesting new feature which has not been reported previously. In addition to the usual exponential decay of the precession amplitude as a function of the distance from the surface, these surface modes have a bulk-wave-type phase change from layer to layer as illustrated schematically in Fig. 1(b). In order to have the torque on a spin $\vec{S}_2$ in the second layer from all spins in the first layer be zero, the vector sum of the three surface spins $\vec{S}_{1n}$ which are coupled to $\vec{S}_2$ must be parallel to $\vec{S}_2$. The projections of the positions of these three spins on the surface plane are shown in Fig. 4(a) and the spins are shown in perspective in Fig. 4(b) for the case of $k_y = 0$.

The y component of $\sum \vec{S}_{1n}$ can be written from inspection of Fig. 4(b) and the corresponding figure for $k_y \neq 0$:

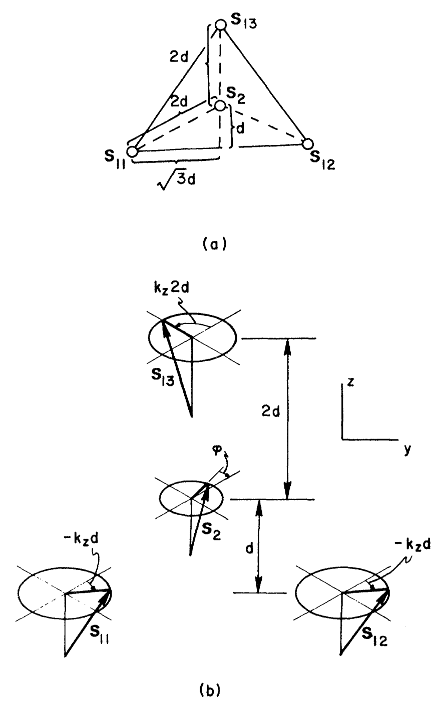

FIG. 4. Positions of $\vec{S}_2$ and the three surface spins $\vec{S}_{11}$, $\vec{S}_{12}$, and $\vec{S}_{13}$ to which $\vec{S}_2$ is coupled are shown for a simple cubic ferromagnetic having a (111) surface in the (a). The same four spins are shown in (b) for the case of $k_y = 0$.

$$\sum_{n=1}^{3} \vec{S}_{1n} \cdot \hat{y} = \sum_{n=1}^{3} \vec{s}_{1n} \cdot \hat{y} = s_1 [\sin 2k_z d - \sin(k_z d + \sqrt{3} k_y d) + \sin(k_z d - \sqrt{3} k_y d)] ,$$

which can be written as

$$\sum_{n=1}^{3} \vec{S}_{1n} \cdot \hat{y} = s_1 (\sin 2k_z d - 2 \sin k_z d \cos \sqrt{3} k_y d) , \tag{3.1}$$

where $2\sqrt{3}d = \sqrt{2}a$ and a is the lattice constant.

In all exchange surface waves considered in the past, the symmetry has been such that the y component of the net torque on $\vec{S}_2$ was zero. Thus the precessional phase angle ϕ in Fig. 1(b) was zero. For the present case, the y component is nonzero (therefore $\phi \neq 0$) unless $k_x = 0$ or $\pm\sqrt{3}k_y$.

From Fig. 4(b) and the corresponding figure for $k_y \neq 0$, the x component of $\sum \vec{S}_{1n}$ is

$$\sum_{n=1}^{3} \vec{S}_{1n} \cdot \hat{x} = s_1 [\cos 2k_z d + \cos(k_z d + \sqrt{3} k_y d) + \cos(k_z d - \sqrt{3} k_y d)] ,$$

which can be written as

$$\sum_{n=1}^{3} \vec{S}_{ln} \cdot \hat{x} = s_l (\cos 2k_z d + 2 \cos k_z d \cos \sqrt{3} k_y d) . \tag{3.2}$$

We have written (3.2) for a general layer l, rather than for layer 1, since this result is true for all layers l by symmetry. The condition for $\sum \vec{S}_{1n}$ to be parallel to $\vec{S}_2$ is

$$s_2 / S = \left| \sum_{n=1}^{3} \vec{s}_{1n} \right| / 3S . \tag{3.3}$$

From (3.1) and (3.2), the value of $\sum \vec{s}_{1n}$ in (3.3) is

$$\left| \sum_{n=1}^{3} \vec{s}_{1n} \right| = \sigma s_1 , \tag{3.4}$$

where

$$\sigma \equiv [1 + 4 \cos\sqrt{3} k_y d (\cos\sqrt{3} k_y d + \cos 3 k_z d)]^{1/2} . \tag{3.5}$$

The equation of motion of a spin $\vec{S}_1$ on the surface is

$$-\omega s_1 = 3 S J s_1 - S J \left| \sum \vec{s}_{2n} \right| . \tag{3.6}$$

Using (3.4) (which is valid with 1 replaced by 2) to eliminate $|\sum \vec{s}_{2n}|$ in (3.6) and using (3.3) and (3.4) to eliminate s_2 gives the frequency

$$-\omega / S J = \tfrac{4}{3} [2 - \cos\sqrt{3} k_y d (\cos\sqrt{3} k_y d + \cos 3 k_z d)] . \tag{3.7}$$

For $k_y d \ll 1$ and $k_z d \ll 1$, (3.7) gives

$$-\omega / S J = (k_y^2 + k_z^2) a^2 . \tag{3.8}$$

The phase angle ϕ is given by the relation

$$e^{i\phi} = \frac{1}{\sigma} \sum_{n=1}^{3} \vec{S}_{1n} \cdot (\hat{x} + i\hat{y}) , \tag{3.9}$$

where the two sums on the right-hand side are given by (3.1) and (3.2) and σ is defined in (3.5). The value of k_x is given by the expression

$$|\vec{s}_2| / |\vec{s}_1| = |\vec{s}_3| / |\vec{s}_2| = \cdots = e^{-k_x a} = \tfrac{1}{3}\sigma . \tag{3.10}$$

The spin configurations (for the $\vec{s}$'s) are shown in Fig. 5 for the case of $k_z d = \pi/2$ and $k_y = 0$, which gives a nonzero ϕ.

4. ANTIFERROMAGNETIC OSCILLATING-DECAYING STATE

The spin arrangement for the simple cubic antiferromagnet with a (111) surface is similar to that of the simple cubic ferromagnet of Sec. 3, except that the spins on the second, fourth, etc., layers have negative z components. For the bulk mode with $\vec{k} = 0$, $\vec{s}_l = -\vec{s}_{l\pm 1}$, where l labels the layers. For the antiferromagnetic problem the torque on each spin in the third layer from all spins in the first two layers must be zero. This gives

$$s_3 = \tfrac{1}{3} \left| \sum_{n=1}^{3} \vec{s}_{2n} \right| \tag{4.1}$$

in analogy with (3.3) for the ferromagnetic case. The equations of motion for a spin on the first layer and one in the second layer are

$$-\bar{\omega} s_1 = (3 + \bar{\omega}_a) s_1 - \left| \sum_{n=1}^{3} \vec{s}_{2n} \right| , \tag{4.2a}$$

$$-\bar{\omega} s_2 = -(6 + \bar{\omega}_a) s_2 + \left| \sum_{n=1}^{3} \vec{s}_{1n} \right| + \left| \sum_{n=1}^{3} \vec{s}_{3n} \right| , \tag{4.2b}$$

where $\bar{\omega} = \omega / SJ$, $\bar{\omega}_a = \omega_a / SJ$, and ω_a is the anisotropy field expressed in units of frequency.

Using (3.4) reduces (4.2) to

$$\begin{bmatrix} 3 + \bar{\omega}_a + \bar{\omega} & -\sigma \\ \sigma & -6 - \bar{\omega}_a + \bar{\omega} + \tfrac{1}{3}\sigma^2 \end{bmatrix} \begin{bmatrix} s_1 \\ s_2 \end{bmatrix} = 0 . \tag{4.3}$$

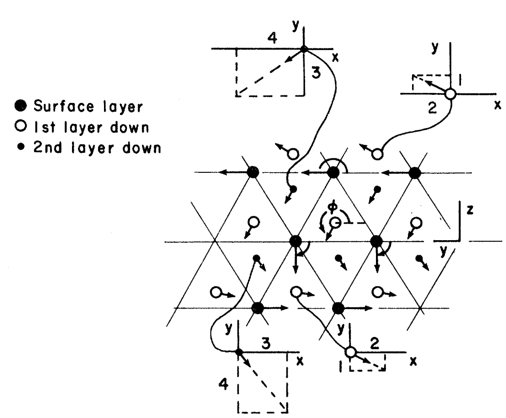

FIG. 5. Spins on the first three layers of a simple cubic ferromagnetic having a (111) surface. In order to show both the position of the spins and their transverse components $\vec{s}_n$, the spins have been rotated through 90° so that the positions are shown in the y-z plane and the spins are shown in the x-y plane.

The secular equation gives the frequency

$$-\omega/SJ=\tfrac{1}{2}(3-\tfrac{1}{3}\sigma^2)+[\tfrac{1}{4}(3-\tfrac{1}{3}\sigma^2)^2+(3-\tfrac{1}{3}\sigma^2+\bar{\omega}_a)(6+\bar{\omega}_a)]^{1/2}\ , \qquad (4.4)$$

where the positive sign of the square root was chosen to give exponentially decaying rather than increasing waves. The method has reduced this rather complicated problem to the simple two-spin problem (4.3). For $k_y=k_z=0$, (3.5) gives $\sigma=3$, and (4.4) gives

$$-\omega/SJ=[\bar{\omega}_a(6+\bar{\omega}_a)]^{1/2}\cong(6\bar{\omega}_a)^{1/2}\ . \qquad (4.5)$$

The approximate equality, which is valid for $\bar{\omega}_a\ll 1$, is a factor of $\sqrt{2}$ smaller than the $k_f=0$ bulk-mode result. This result and the physical reason for having to choose the positive square root in (4.4), while both roots are kept in the bulk-mode problem, can be seen easily as follows: The frequency of the $k_f=0$ bulk mode can be obtained by solving a two-spin problem, just as for the surface state. The equations of motion of an up spin $\vec{S}_1^+$ and a neighboring down spin $\vec{S}_2^-$ are

$$\begin{bmatrix} 6+\bar{\omega}_a+\bar{\omega} & -6 \\ -6 & -(6+\bar{\omega}_a)+\bar{\omega}\end{bmatrix}\begin{bmatrix} s_1^+ \\ s_2^-\end{bmatrix}=0\ . \qquad (4.6)$$

The secular equation gives

$$-\omega/SJ=\pm[12\bar{\omega}_a+\bar{\omega}_a^2]^{1/2}\simeq\pm(12\bar{\omega}_a)^{1/2}\ . \qquad (4.7)$$

The positive sign in (4.7) corresponds to a larger precessional amplitude of $\vec{S}_1^+$ than of $\vec{S}_2^-$, and the negative sign corresponds to a large precessional amplitude of $\vec{S}_2^-$. Both solutions are allowed, and the two are interchanged if the crystal is turned upside down. Now for the surface wave, we assumed that the $\vec{S}_1^+$ spin was on the surface. In order to have a decaying wave, the precessional amplitude of the spin $\vec{S}_2$ on the second layer must be less than that of $\vec{S}_1$ on the surface, thus explaining the choice of the positive sign on the right-hand side of (4.5). The spin configuration is illustrated schematically in Fig. 1(c) for the simple case of $\phi=0$.

With $k_f=0$, $\vec{S}_2$ and $\vec{S}_3$ must be antiparallel in order for there to be no torque on $\vec{S}_3$ from the spins in the second layer, and this also makes the torque on $\vec{S}_2$ from the spins in the third layer zero. Thus, $\vec{S}_2$ is effectively coupled only to the three spins of the first layer rather than to the six spins of the first and third layers as in the bulk mode. Replacing the factor of 6 in (4.6) by 3 gives a factor of 6, rather than 12, in (4.7), thus explaining the factor of $\sqrt{2}$ difference between the bulk and surface frequencies. It should be mentioned that for other spin configurations the spins on the third layer will exert a torque on $\vec{S}_2$, in general, and the ratio of bulk to surface frequencies is not directly related to the ratio of the numbers of neighbors of a surface spin to that of a bulk spin.

5. FLIP-SURFACE WAVES

In this section, we shall find the surface modes for a simple cubic ferromagnet whose surface is (100) plane, with exchange constant J for all near-neighbor pairs of spins except those having one spin in the surface and one in the second layer, for which the exchange constant $J_\perp$ is negative (antiferromagnetic). In the ground state, it is assumed that all spins on the first layer have $S_z=-S$, and all others have $S_z=+S$, as illustrated schematically in Fig. 1(a). The spin precession amplitudes s_2, s_3, ... decay exponentially, starting at the second layer.

The equation of motion of $\vec{S}_l$ for $l>2$ and the relation $s_{l\pm1}=e^{\mp k_x a}s_l$ give

$$\omega/SJ=2\cosh k_x a-2-4\Lambda_k\ , \qquad (5.1)$$

where $\Lambda_k\equiv1-\tfrac{1}{2}(\cos k_y a+\cos k_z a)$.

The value of $k_x a$ in (5.1), which is determined by the value of $J_\perp$, is found as follows: In order for the frequency of $\vec{S}_2$ to be the same as that of $\vec{S}_l$ for $l>2$, the torque on $\vec{S}_2$ from $\vec{S}_1$ must be the same as if $J_{12}=J$ and $\vec{S}_1$ fit into the exponential series (i.e., the same as if $S_{1z}=+S$ and $s_1=e^{k_x a}s_2$). Equating these two torques gives

$$J(e^{k_x a}-1)s_2=|J_\perp|(s_1-s_2)\ ,$$

which can be written as

$$s_2/s_1=|\epsilon_\perp|/(e^{k_x a}-1+|\epsilon_\perp|)\ , \qquad (5.2)$$

where $J_\perp\equiv\epsilon_\perp J$, with $\epsilon_\perp$ negative.

The equation of motion of $\vec{S}_1$ gives

$$-\omega/SJ=-|\epsilon_\perp|(1-s_2/s_1)-4\Lambda_k\ . \qquad (5.3)$$

Equating the right-hand sides of (5.1) and (5.3) and using (5.2) gives

$$|\epsilon_\perp|=-\frac{2(e^{k_x a}-1)(1-\cosh k_x a+4\Lambda_k)}{1-e^{-k_x a}+8\Lambda_k}\ . \qquad (5.4)$$

For given values of $|\epsilon_\perp|$ and Λ_k, (5.4) is a cubic equation for $e^{k_x a}$. The root with $e^{k_x a}\geqq1$ must be chosen in order to have an exponentially decaying wave.

For $\vec{k}_f\equiv\hat{y}k_y+\hat{z}k_z=0$, the value of Λ_k is zero, and (5.4) reduces to

$$|\epsilon_\perp|=-2\,e^{k_x a}(1-\cosh k_x a)\ ,$$

which is easily solved to give

$$e^{k_x a}=1+\sqrt{|\epsilon_\perp|}\ \text{ for }\vec{k}_f=0\ , \qquad (5.5)$$

where the positive square root has been chosen to make $e^{k_x a} > 1$. With this value of $e^{k_x a}$ and with $\Lambda_k = 0$, (5.1) gives

$$\omega/SJ = |\epsilon_\perp|/(1+\sqrt{|\epsilon_\perp|}) \text{ for } \vec{k}_f = 0 . \quad (5.6)$$

For $\vec{k}_f \neq 0$, another useful form of (5.4) is

$$8\Lambda_k = 2\cosh k_x a - 2 - \frac{|\epsilon_\perp|(e^{k_x a}-1)}{e^{k_x a} - 1 + |\epsilon_\perp|} . \quad (5.7)$$

In order to illustrate the dispersion curve, first consider the case of $|\epsilon_\perp| = 1$, for which (5.7) gives

$$8\Lambda_k = 2e^{-k_x a} + e^{k_x a} - 3 ,$$

which can be solved to give

$$e^{k_x a} = \tfrac{1}{2}(3+8\Lambda_k) + \tfrac{1}{2}[(3+8\Lambda_k)^2 - 8]^{1/2} , \quad (5.8)$$

where the positive sign of the square root was chosen as usual. In Fig. 6, the curve marked $\epsilon_\perp = -1$ is a plot of (5.1) with $e^{k_x a}$ given by (5.8).

For $\epsilon_\perp = 0$ the surface layer is decoupled from the other layers, and the frequency of this layer has the same magnitude as that of the bulk mode with $k_x = 0$ (since the near-neighbor spins in adjacent layers are parallel when $k_x = 0$). The sign of ω is positive for the single-sheet surface wave ($S_z = -S$) and is negative for the $k_x = 0$ bulk wave ($S_z = +S$). The dispersion curve for this single-layer surface wave for $\epsilon_\perp = 0$ is included in Fig. 6.

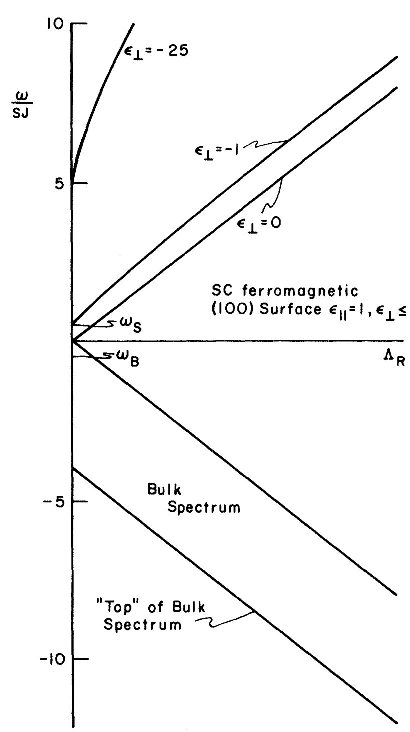

FIG. 6. Dispersion relations for the flip-surface waves of a simple cubic ferromagnetic with a (100) surface for various values of the surface exchange constant $\epsilon_\perp J$. The bulk-mode dispersion relations also are shown.

Finally, for $|\epsilon_\perp| \gg 1$, (5.7) gives

$$e^{k_x a} \cong (1+8\Lambda_k)^{1/2} |\epsilon_\perp|^{1/2} .$$

Using this result in (5.1) gives

$$\omega/SJ \cong (1+8\Lambda_k)^{1/2}|\epsilon_\perp|^{1/2} \text{ for } \epsilon_\perp \gg 1 . \quad (5.9)$$

A portion of this result (5.9) is illustrated in Fig. 6 for $\epsilon_\perp = -25$.

Note that there are surface curves which have values of $|\omega|$ which are equal to the value of $|\omega|$ for a bulk wave, as marked ω_S and ω_B in Fig. 6, for example. None of the previously reported surface waves have shown this type of degeneracy. The sign of ω is positive for the surface waves and negative for the bulk waves; that is, the spins rotate in different directions for the two types of waves.

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APPENDIX: EQUATIONS OF MOTION

The equation of motion of a spin $\vec{S}_A = S_{Az}\hat{z} + \vec{s}_A$ coupled to $\vec{S}_B$ by the Hamiltonian $\mathcal{H} = -J\,\vec{S}_A \cdot \vec{S}_B$ is

$$d\,\vec{S}_A/dt = -J\,\vec{S}_B \times \vec{S}_A , \quad (A1)$$

where the exchange constant J is positive for ferromagnetic interactions and the caret denotes a unit vector. For circular precession, at the instant of time at which $\vec{s} = \hat{x}s$, the vector of $d\,\vec{S}_A/dt$ is given by

$$d\,\vec{S}_A/dt = \hat{y}\,\omega s_A . \quad (A2)$$

For up spins (i.e., $S_z = +S$) with $\vec{s} = \hat{x}\,s_x$, the angle between $\vec{S}_B$ and $\vec{S}_A$ is approximately equal to $(s_{Ax} - s_{Bx})/S$ when the usual linearization approximation $|s_x| \ll |S_z|$ is satisfied; thus (A1) and (A2) give

$$\omega s_A = SJ(s_{Bx} - s_{Ax}) .$$

The corresponding results for arbitrary values of S_{Az} and S_{Bz} are

$$-(\omega/SJ)\,s^{+}_{Ax} = \pm s^{+}_{Ax} - s^{\pm}_{Bx} , \quad (A3a)$$

$$-(\omega/SJ)\,s^{-}_{Ax} = \pm s^{-}_{Ax} + s^{\pm}_{Bx} , \quad (A3b)$$

where the $^+$ and $^-$ superscripts indicate $S_z = +S$ and $S_z = -S$, respectively. The first term on the right-hand side of (A3a) [or (A3b)] corresponds to $S_{Bz}\hat{z}\times\vec{s}_A$, and the second term corresponds to $\vec{s}_B \times S_{Az}\hat{z}$. Using these results, it is not difficult to show that for $J > 0$ and $S_{nz} = +S$ exponentially decaying spins $s_n = s_0 e^{-k_x a n}$ give a positive ω, while cosinusoidally varying spins $s_{nx} = s_0 e^{ik_x a n}$ give a negative ω.

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Nature of the Antiferromagnetic-Paramagnetic Transition in $MnCl_2 \cdot 4H_2O$†

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The heat capacity and magnetization at constant field versus temperature, as well as the adiabatic variation of temperature with magnetic field, have been determined in fields directed along the *c* axis of a large spherical single crystal of $MnCl_2 \cdot 4H_2O$ to virtually the limits of resolution of the dc methods used. In this study of the behavior of these quantities in the neighborhood of the antiferromagnetic-paramagnetic transition, one specific goal was to observe the (near) singularity in $(\partial M/\partial T)_H$ at $T_N(H)$. In addition, we sought to test the predictions that isentropes cross the phase boundary (defined as the locus of maxima in C_H) tangentially, and that this crossing point should prove to be the point of inflection of the isentropes provided C_H does not diverge too strongly. A test for determining the existence of a divergence without the necessity of measuring infinitely high values is outlined. The fact that the maximum in the zero-field adiabatic susceptibility occurs at a temperature $T_{max} > T_N(0)$ has been found to be reflected in the persistence of a minimum in plots of the isentropic variation of T versus H up to $T = T_{max}$. This curious behavior has led us to speculate on a larger coexistence region of somewhat different character than has heretofore seemed reasonable.

The bulk magnetothermodynamic properties of antiferromagnetic substances have for some time been the subject of intensive study. It has been the practice of those performing these experiments to summarize the salient features of the resultant data in a graphical display known as the phase diagram in the H-T plane. For a uniaxial antiferromagnet of weak anisotropy with the field applied along this axis, the resultant diagram will be similar to that shown in Fig. 1. The letters a, p, and b will be used to denote the antiferromagnetic, paramagnetic, and spin-flopped regions. Unfortunately, the phase diagrams derived from different types of measured data are not always congruent.

The heat capacities of these materials typically exhibit λ-shape anomalies. In analogy with the work of Buckingham and Fairbank on He, [1] the maxima in these curves are usually taken as the Néel temperature $[T_N(H)]$. The available data on the in-field heat capacities of oriented single-crystal antiferromagnets are sparse,[2–5] but that available data agree well with the estimates of $T_N(H)$ derived from optical[6] and radio-frequency[7] spectroscopic measurements although more comment is also necessary even here. Large discrepancies appear, however, in attempts at correlations with magnetization data where the maxima in the observed M versus T isoersteds have been used as a measure of $T_N(H)$.

The antiferromagnetic-paramagnetic (ap) transition in antiferromagnets is often referred to as being of second order. Using the Ehrenfest criteria, this implies that the second derivatives of the Gibbs function with respect to its intensive variables are discontinuous. Thus we might expect the trio

$$\left(\frac{\partial^2 G}{\partial T^2}\right)_H = C_H, \quad \left(\frac{\partial^2 G}{\partial H^2}\right)_T = \chi_T, \quad \left(\frac{\partial^2 G}{\partial H \partial T}\right) = \left(\frac{\partial M}{\partial T}\right)_H$$

to exhibit discontinuities if this scheme were applicable. Cooperative phenomena display, rather, a divergence of these Ehrenfest derivatives to hypothetically infinite values where the physical