Influence of Surface Anisotropy and Next-Nearest-Neighbor Coupling on Surface Spin Waves

J. C. Levy, E. Ilisca, and J. L. Motchane
Laboratoire de Magnétisms des Surfaces, Université Paris VII, Paris, France
(Received 6 April 1971)

The spin-wave spectrum of a Heisenberg ferromagnet is derived in the framework of the method of De Wames and Wolfram, with the next-nearest-neighbor coupling, the surface and bulk exchange, and the surface anisotropy taken into account. The calculations are performed for a {100} surface in a simple-cubic structure. When the next-nearest-neighbor coupling is strong enough, the character of the surface modes is shown to change from acoustical to optical (or vice versa) as the components (k_x, k_y) of the wave vector parallel to the surface vary. This transition from acoustical to optical character occurs for a definite value λ_k^0 of the parameter $\lambda_k = 1 - \frac{1}{2}(\cos k_x a + \cos k_y a)$, where a is the lattice parameter. For this value λ_k^0, the effective coupling between the spin deviation in different bulk layers is shown to vanish, so that each of the bulk modes corresponds to vibrations of the spins of one single layer and to a surface spin wave strictly localized on the first layer. Moreover, the equilibrium spin configuration near the surface could be modified by the existence of the surface anisotropy.

I. INTRODUCTION

The dynamical properties of the surface spins in ferromagnetic substances are involved in various important phenomena such as ferromagnetic resonance,[1] magnetic scattering of low-energy electrons,[2] and magnetocatalytic effects.[3] Thus there has been a number of recent investigations concerning bulk and surface spin waves in a Heisenberg ferromagnet.

Using a continuous model, Kittel[4] and Sparks[5] have made a theoretical study of the spin wave. In order to explain the pinning effects they introduce surface-anisotropy fields. Eschbach and Damon[7] have demonstrated that the dipolar interactions act only on the long-wavelength spin waves. In the exchange-dominated part, Maradudin and Mills[8] and others[9] have shown that the coupling between next nearest neighbors (nnn) is sufficient to cause the existence of surface spin waves. The change of exchange integrals near the surface has been taken into account by De Wames and Wolfram.[10] However, the existence of the surface breaks the symmetry, and therefore a new term appears in the Hamiltonian, i.e., the uniaxial surface anisotropy, defined as

$$-\frac{1}{2}\sum_{f,f}I_{f,f'}\, S_f^z S_{f'}^z,$$

where $I_{f,f'}$, is the surface-anisotropy integral between the two electronic spins located on sites f and f', and the z axis is taken normal to the surface. As shown by Ilisca and Motchane,[11] the surface anisotropy affects the whole spectrum.

In this paper, it is assumed that the nnn coupling the surface-induced exchange perturbation, and the surface anisotropy are competitive. In other words, the purpose of this work is to study, using a simple model, how a slight electronic delocalization, a perturbation of the exchange integrals at the surface, and the surface anisotropy influence the surface spin-wave spectrum. For simplicity, we assume no bulk anisotropy. The calculations are performed for a {100} surface of a simple-cubic Heisenberg ferromagnet. In our model, in the ground state, all spins are normal to the surface. Moreover, the surface-anisotropy integrals $I_{f,f'}$ are supposed to be equal to $\frac{1}{4}A$ when the sites f and f' are two nearest neighbors (nn) located on the first layer and negligible elsewhere.

The resolvent-matrix method, used by De Wames and Wolfram,[10] gives in this case a simple equation from which both the surface spin-wave energy and the eigenvectors are easily deduced. For strong nnn coupling, when the components (k_x, k_y) of the wave vector parallel to the surface vary, the character of the surface mode changes from acoustical, where the spin deviations in two adjacent layers are in phase, to optical, where the spin deviations in two adjacent layers are 180° out of phase. This transition occurs for a definite value λ_k^0 of the parameter $\lambda_k = 1 - \frac{1}{2}(\cos k_x a + \cos k_y a)$, where a is the lattice parameter. For $\lambda_k = \lambda_k^0$, the bulk modes are degenerate with respect to the z component of the wave vector. Indeed, the effective coupling between the spin deviations of adjacent bulk layers then falls down to zero, and the spin motions in different layers become unconnected. The surface mode is strictly localized on the first layer and its energy differs from that of the bulk modes. This type of mode will be called a "two-dimensional localized mode."

When the surface-anisotropy integral $I_{f,f'}$ is negative, some surface spin wave would have a negative energy. This means that there is a spin rearrangement. The calculation of the free energy shows that the spins near the surface are tilted.[12] On the contrary, a positive value of the surface-anisotropy integral induces a stiffening of the surface, i.e., an increase of the exictation energy of surface spin waves.

Section II is devoted to the general theoretical features of the model, in Sec. III we study the occurrence and properties of the "two-dimensional localized" spin waves, and we give several numerical applications. Section IV deals with specific surface-anisotropy effects, and particularly with surface rearrangements induced by a negative value of the surface-anisotropy integral. Concluding remarks are given in Sec. V.

II. SURFACE SPIN-WAVE ENERGY SPECTRUM

Let us introduce the following Hamiltonian[11,13]:

$$H = -\frac{1}{2}\sum_{f,f',\alpha} J_{f,f'} S_f^\alpha S_{f'}^\alpha - \frac{1}{2}\sum_{f,f'} I_{f,f'} S_f^z S_{f'}^z , \qquad (1)$$

where the z axis is normal to the surface and S^α ($\alpha = x, y, z$) are the spin components. $I_{f,f'}$ and $J_{f,f'}$ are, respectively, the surface anisotropy and exchange integrals between electronic spins located on sites f and f'. From symmetry considerations it is obvious that $I_{f,f'}$ and $J_{f,f'}$ depend only on $|f-f'|$ and on f_z and f'_z, the z components of the vector $\vec{f}$. In this paper, we assume that the coupling integrals $J_{f,f'}$ and $I_{f,f'}$ differ from their value in the bulk only when the two coupled spins belong to the first layers. The nnn and the nn coupling for the exchange integrals $J_{f,f'}$ and only the nn coupling for the anisotropy integrals $I_{f,f'}$ are taken into account. From now on we shall restrict ourselves to the consideration of a simple-cubic structure with lattice parameter a and its free surface along a $\{100\}$ plane. Tables I and II give the values of the different integrals $I_{f,f'}$ and $J_{f,f'}$ in terms of J, the value of the exchange in the bulk. Moreover, we shall assume that the equilibrium magnetization is parallel to the z axis everywhere and does not depend on the location of the site (this assumption will be relaxed in Sec. III).

Let us set

$$\langle S_z \rangle = S . \qquad (2)$$

Then the equation of motion of $S_F^+ = S_f^x + iS_f^y$ reads in the random-phase approximation (RPA)[10,13]

$$i\frac{d}{dt} S_f^+ = S\sum_{f'} (J_{f,f'} + I_{f,f'})\, S_f^+ - S\sum_{f'} J_{f,f'}\, S_{f'}^+ . \qquad (3)$$

In order to take advantage of the translational invariance parallel to the surface and of the linearity of Eq. (3) in the time derivative, we introduce the Fourier transform

$$S_f^+(t) = \int_{-\infty}^{+\infty} e^{-i\omega t} \sum_{\vec{k}} e^{-i\vec{k}\cdot\rho_f} u_n(k,\omega)\, d\omega , \qquad (4)$$

where $\vec{k}$ is a wave vector of the (k_x, k_y) reciprocal plane associated with the (x,y) surface plane, and $\vec{\rho}_f$ and f_z are, respectively, the components of f parallel and normal to the surface.

Equation (3) then gives

$$-u_{n+1}\sum_{f'_+} J_{f,f'} \cos(\vec{k}\cdot\vec{\rho}_{f'}) + u_n\Big(-\frac{\omega}{S} + \sum_{f'} I_{f,f'} + \sum_{f'_0} J_{f,f'}\,(1-\cos\vec{k}\cdot\vec{\rho}_{f'}) + \sum_{f'_-,f'_+} J_{f,f'}\Big) - u_{n-1}\sum_{f'_-} J_{f,f'}\cos(\vec{k}\cdot\vec{\rho}_{f'}) = 0 . \qquad (5)$$

The summations are performed for a given site f in the nth layer. This site f is chosen on the z axis. $\sum_{f'_+}$, $\sum_{f'_-}$, and $\sum_{f'_0}$ mean summations over all the nn and the nnn f' of the site f located, respectively, in the $(n+1)$th, $(n-1)$th, and the nth layer. $\sum_{f'}$ means the complete summation over these three neighboring layers. Equation (5) has the same form as the equation of motion of Ref. 10:

$$g_{-1} u_{n-1} + g_0 u_n + g_1 u_{n+1} = 0 . \qquad (5')$$

However, it should be noticed that, owing to the presence of the surface anisotropy, this correspondence is only a formal one, since the detailed structure of the spectrum depends strongly on the precise form of g_{-1}, g_0, and g_1.

Let us define

$$\cos\theta = \frac{JS(2+8\sigma+4\lambda_k+4\sigma\Lambda_k)-\omega}{JS(1+4\sigma-4\sigma\lambda_k)} \qquad (6)$$

and

$$d = \frac{4\lambda_k(\epsilon_\parallel - 1) + 4\Lambda_k(\sigma_\parallel - \sigma) + A - 1 - 4\sigma}{1+4\sigma-4\sigma\lambda_k} , \qquad (7)$$

where

$$\lambda_k = 1 - \tfrac{1}{2}(\cos k_x a + \cos k_y a) ,$$
$$\Lambda_k = 1 - \cos k_x a \cos k_y a . \qquad (8)$$

Equation (5) has two different forms, depending on whether n represents the surface or not. At the surface, it reads

$$u_1\,(2\cos\theta + d) - u_2 = 0 , \qquad (9)$$

and for the nth bulk layer it reads

$$-u_{n-1} + u_n\,(2\cos\theta) - u_{n+1} = 0 . \qquad (10)$$

These reduced equations (9) and (10) are similar to those which describe the vibrations of a semi-infinite linear chain of classical oscillators, with a perturbation at its free end.[14]

Equations (9) and (10) can be rewritten in matrix form[10]:

$$(D_N + \Delta D)_{n,m}\, u_m = 0 , \qquad (11)$$

where D_N corresponds to Eq. (10), i.e., to the bulk interactions, and ΔD is, in our case, a 1×1 matrix:

$$(\Delta D)_{n,m} = \delta_{n,1}\delta_{m,1} d \ . \tag{11a}$$

A. Bulk Modes

To the lowest order, and following the procedure used by De Wames and Wolfram,[10] the bulk spin waves are obtained from Eq. (11) by neglecting ΔD. The first approximation will be improved in a future publication.

Let us set

$$u_n = e^{ik_z a} u_{n-1} \ . \tag{12}$$

The Eq. (9) implies

$$\cos\theta = \cos k_z a \ . \tag{13}$$

Here k_z is a real number, and therefore θ is also real. This corresponds to a bulk-mode propagation along the z axis. The energy spectrum of bulk spin waves deduced from Eq. (6) is

$$E = \omega/JS = 2 + 8\sigma + 4\lambda_k + 4\sigma\Lambda_k - 2\cos\theta\,(1 + 4\sigma - 4\sigma\lambda_k) \ . \tag{14}$$

The limiting values of $\cos\theta$ are obtained for the two modes near the edges of the Brillouin zone in the k_z direction: $\cos\theta = -1$, then $\theta = \pi$, and $u_{n+1} = -u_n = (-1)^n u_1$. Spin deviations in adjacent layers are 180° out of phase: $\cos\theta = +1$, then $\theta = 0$, and $u_{n+1} = u_n = u_1$. Spin deviations in adjacent layers are in phase.

B. Surface Modes

A multiplication of Eq. (11) by the resolvent matrix D_N^{-1} yields

$$(I_N + D_N^{-1}\,\Delta D)\,u_n = 0 \ , \tag{15}$$

where I_N is the $N\times N$ identity matrix.

As can be checked, the exact value of the matrix elements $(D_N^{-1})_{n,m}$ is given by

TABLE I. When $\vec{f}$ or $\vec{f}'$ (or $\vec{f}$ *and* $\vec{f}'$) does not belong to the first layer, the exchange and surface-anisotropy integrals J and I are given as functions of the distance $\vec{f}-\vec{f}'$. σ characterizes the nnn exchange coupling in the bulk. There is no anisotropy coupling between nnn in the bulk or on the surface layer.

$\lvert\vec{f}-\vec{f}'\rvert$ \ J, I	$J(\vec{f}, \vec{f}')$	$I(\vec{f}, \vec{f}')$
a	J	0
$a\sqrt{2}$	σJ	0
$>a\sqrt{2}$	0	0

TABLE II. When $\vec{f}$ and $\vec{f}'$ belong to the first layer, the exchange and surface-anisotropy integrals J and I are given as a function of the distance $\vec{f}-\vec{f}'$. A characterizes the surface anisotropy. $\sigma_{\parallel}$ characterizes the nnn exchange coupling for the spins on the surface layer.

$\lvert\vec{f}-\vec{f}'\rvert$ \ J, I	$J(\vec{f}, \vec{f}')$	$I(\vec{f}, \vec{f}')$
a	J	$\frac{1}{4}JA$
$a\sqrt{2}$	$\sigma_{\parallel}J$	0
$>a\sqrt{2}$	0	0

$$(D_N^{-1})_{n,m} = \frac{e^{2i(N+1-r)\theta} - 1}{e^{2i(N+1)\theta} - 1}\,(2i\sin\theta)^{-1} \times (e^{i|n+m|\theta} - e^{i|n-m|\theta}) \ , \tag{16}$$

where N is the number of layers and r is equal to the highest value of (n, m). When $N\to\infty$ and $|e^{i\theta}| \neq 1$, the limiting value of $(D_N^{-1})_{n,m}$ is given by (see Ref. 10)

$$(D_N^{-1})^{\infty}_{n,m} = (2i\sin\theta)^{-1}\,(e^{i|n+m|\theta} - e^{i|n-m|\theta}). \tag{16a}$$

The assumption that $|e^{i\theta}| \neq 1$ eliminates the bulk solution; then Eq. (15) yields

$$\det|I_N + D_N^{-1}\,\Delta D| = 0 \tag{17}$$

or

$$1 + e^{i\theta} d = 0 \ . \tag{18}$$

Then, from Eqs. (6), (7), and (18) the energy spectrum of the surface spin wave is

$$E = \omega/JS = 1 + 4\sigma + 4\sigma\Lambda_k + 4\epsilon_{\parallel}\lambda_k + A'(\Lambda_k) - \frac{(1 + 4\sigma - 4\sigma\lambda_k)^2}{1 + 4\sigma - 4\lambda_k(\epsilon_{\parallel} - 1) - A'(\Lambda_k)} \ , \tag{19}$$

with $A = 4I_{\parallel}/J$ and

$$A'(\Lambda_k) = A + 4\Lambda_K\,(\sigma_{\parallel} - \sigma) \ . \tag{19a}$$

In order to define the eigenvectors, one must solve Eq. (13); i.e.,

$$u_n = \textstyle\sum_p - (D_N^{-1}\,\Delta D)_{n,p} u_p \ . \tag{20}$$

Let us set

$$x = e^{-i\theta} \ , \tag{21}$$

where, according to Eq. (18), x is real. Equation (20) takes the form

$$u_n = x^{-(n-1)} u_1 \ . \tag{22}$$

Since the spin amplitude cannot grow indefinitely, we must have

$$|x| > 1 \ . \tag{23}$$

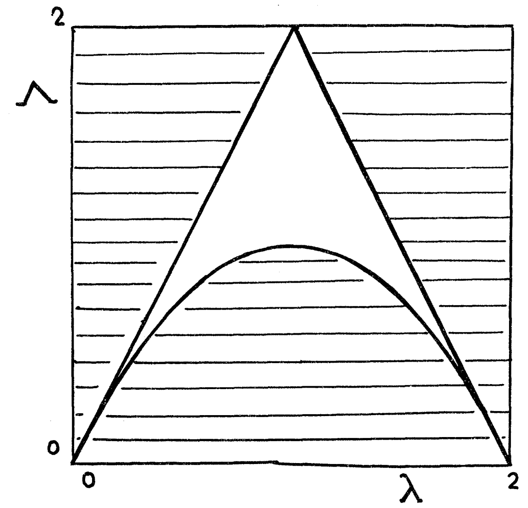

FIG. 1. Domain describing the inside of the Brillouin zone is not shaded.

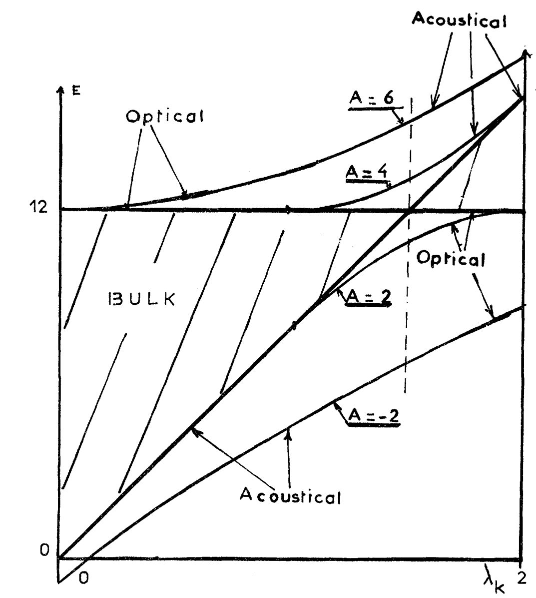

FIG. 3. Dispersion curves and two-dimensional localized modes when $\sigma > \frac{1}{4}$, $\sigma = \frac{1}{2}$, $\Lambda_k = \delta_{\parallel} = 0$.

Equations (22) and (23) show that the surface modes are exponentially damped, with a penetration depth δ given by

$$\delta = \frac{a}{\ln |x|} = \frac{a}{\ln |d|} \quad . \tag{24}$$

Two cases may occur which satisfy the inequality (23): (i) $x > 1$, $\mathrm{Re}\,\theta = 0$, and the corresponding modes are seen to be of acoustical character; (ii) $x < -1$, $\mathrm{Re}\,\theta = \pi$, u_n and u_{n-1} have opposite signs, and the modes are of optical character.

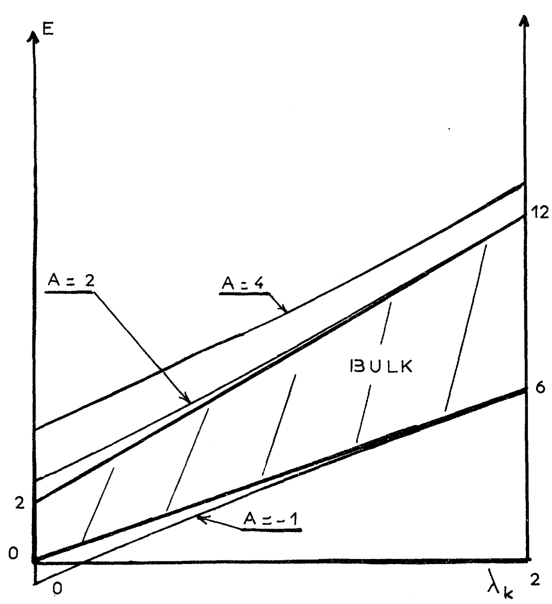

FIG. 2. Dispersion curves when $\sigma < 0$, $\sigma = -\frac{1}{8}$, $\Lambda_k = \delta_{\parallel} = 0$. When A' varies from 0 to 1, no surface spin wave is allowed.

In Eqs. (14) and (19) the energy E has been written as a function of the two variables $(\lambda_{k'}, \Lambda_k)$. Figure 1 shows the region of the (λ_k, Λ_k) plane described while the wave vector $\vec{k}(k_x, k_y)$ is taken in the whole Brillouin zone. As one restricts this Brillouin zone to the $k_x < k_y$ region, the correspondence between the couples (k_x, k_y) and (λ_k, Λ_k) becomes a "one-to-one correspondence." In order to describe the spin-wave energy spectrum $E(\lambda_{k'}, \Lambda_k)$, we draw the section of $E(\lambda_{k'}, \Lambda_k)$ by a plane $\Lambda_k = \text{const}$. Figure 1 shows that to every value of $\Lambda_{k'}$ corresponds an interval of values for λ_k. For instance, for $\Lambda_k = 0$ this interval reduces to the two allowed values $\lambda_k = 0$ and $\lambda_k = 2$.

When Λ_K changes, for instance, from Λ_k^1 to Λ_k^2, the energy spectrum of the bulk modes $E(\lambda_k, \Lambda_k^1)$ simply becomes $E(\lambda_{k'}, \Lambda_k^1) + 4\sigma\Lambda_k^2$, while the energy spectrum of the surface modes changes in a more complicated way described by Eq. (19). Figures 2 and 3, which represent the energy spectrum versus λ_k, are drawn for $\Lambda_k = 0$. From the above considerations, the curves representing the energies of the bulk and the surface modes versus λ_k for every value of Λ_k can be easily deduced.

III. TWO-DIMENSIONAL LOCALIZED MODES

Let us now come back to Eq. (5) and (5′). A glance at this equation of motion shows that a very particular situation arises when the coefficients g_1 and g_{-1} are both equal to zero; e.g.,

$$g_1 = g_{-1} = \sum_{f'_+} J_{f,f'} \cos(\vec{k}\cdot\vec{\rho}_{f'})$$

$$= \sum_{f'} J_{f,f'} \cos(\vec{k}\cdot\vec{\rho}_{f'}) = 0 \; . \tag{25}$$

When the condition (25) is fulfilled, the propagation of a wave in the nth layer exerts no torque on the $n+1$ and $n-1$ layers. There is then a complete decoupling of the spin motions in different layers. Therefore, the corresponding modes do not propagate along the z direction, and in the RPA (which is equivalent to the harmonic approximation for phonons), the variation of their amplitude along $0z$ is indeterminate. The same effect was shown to occur in the phonon case.[15] Within our assumptions, Eq. (25) may be rewritten

$$g_1/J = g_{-1}/J = 1 + 4\sigma - 4\sigma\lambda_k = 0. \tag{26}$$

The two-dimensional localized modes occur for a well-determined value λ_k^0 of λ_k, which is the solution of Eq. (26).

One can distinguish two cases:

(*a*.) *Weak nnn coupling*, $|\sigma| < \frac{1}{4}$ (Fig. 2). Equation (26) has no solution which satisfies the condition $0 < \lambda_k < 2$ and no two-dimensional localized modes can exist.

(*b*.) *Strong nnn coupling*, $|\sigma| > \frac{1}{4}$. Equation (26) has a physical solution. Two cases happen.

(*i*) $\sigma < -\frac{1}{4}$. There is a strong negative nnn coupling and we have $0 < \lambda_k^0 < 1$. In the absence of an external magnetic field, some of the bulk spin waves have a negative energy. It is well known that, in that case, the stable configuration is helimagnetic.[13–16] However, when a high magnetic field is applied along the z axis, the ferromagnetic spin configuration may be stable.

(*ii*) $\sigma > \frac{1}{4}$. There is a strong positive nnn coupling and we have $\lambda_k^0 > 1$ (Fig. 3). In that case, when $A'(\Lambda_k)$ varies from $-\infty$ to $+\infty$, going through the critical value $A_c' = (1+4\sigma)\,[1-(\epsilon_{\parallel}-1)/\sigma]$, the curve representing the surface mode goes from below the bulk spectrum region in the diagram E versus λ_k to above the bulk spectrum region. For $A'(\Lambda_k) = A_c'$ there is no surface mode.

Several remarks must be made about these two-dimensional localized modes:

(i) The existence of these localized modes is related to a delocalization of the electronic wave function, which makes the coupling between nnn important.

(ii) From Eqs. (5), (9), (10), and (25), it is seen that, for a wave vector (k_x, k_y) such as $\lambda_k = \lambda_k^0$, all bulk magnons have the same excitation energy; only the surface magnon has a different energy. This means that, in the E-versus-λ_k diagram, for λ_k^0 the bulk spectrum is squeezed down to a single point. An experimental test of this squeezing down could be a ferromagnetic resonance experiment in a geometry such as $\vec{k} = \vec{k}^0$, i.e., where the sample is not normal to the radio-frequency-field wave vector. In that case, all ferromagnetic resonance lines reduce to a single line.

(iii) As λ_k varies continuously, Eqs. (5) and (25) imply that the coupling constant $g_{\pm 1}$ between the spin deviations in adjacent layers goes through zero. g_{-1} for the surface mode being a function of λ_k only, it changes continuously from a positive to a negative value when λ_k goes through λ_k^0; on the other hand, the sign of g_0 does not change. If the spin deviations in the first and second layers are in phase before λ_k goes through λ_k^0, they become 180° out of phase afterwards. In other words, it is an acoustical to optical continuous transition.

(iv) It is worth noting that the spin deviations in a layer are enhanced when the spin wave is localized in this layer only. Therefore, it implies that in low-energy electron diffraction there is a peak when the electron beam meets the surface with the correct angle. Thus, a scanning could provide a test for the occurrence of such modes.

In order to illustrate the influence of the surface parameters on the energy spectrum, we have computed it in some simple cases of physical interest.

(*a*.) *"Free" surface* ($\epsilon_{\parallel} = 1$, $\sigma_{\parallel} = \sigma$, $A = 0$). The nnn and nn couplings are not changed by the presence of the surface and there is no surface anisotropy. This case has already been studied in Ref. 9. From (18), (19), and (21) we obtain

$$x = \frac{1+4\sigma}{1+4\sigma-4\sigma\lambda_k} \;, \tag{27}$$

$$E = 4\sigma\Lambda_k + 4\lambda_k(1+2\sigma) - \frac{16\sigma^2}{1+4\sigma}\lambda_k^2 \; . \tag{28}$$

The surface spin-wave energy curve is a truncated parabola, since $|x| > 1$.

(*b*.) *"Anirotropic" surface*. The nnn and nn couplings are equal in the surface layer and in the bulk ($\epsilon_{\parallel} = 1$, $\sigma_{\parallel} = \sigma$), but the surface anisotropy is nonzero:

$$x = \frac{1+4\sigma-A}{1+4\sigma-4\sigma\lambda_k} \;, \tag{29}$$

$$E = 4\sigma\Lambda_k + 4\lambda_k(1+2\sigma) - \frac{(A-4\sigma\lambda_k)^2}{1+4\sigma-A} . \tag{30}$$

The energy remains a parabolic function of λ_k.

(*c*.) *General surface*. The perturbation of the exchange integrals at the surface is taken into account ($\delta_{\parallel} = \epsilon_{\parallel} - 1$, $t_{\parallel} = \sigma_{\parallel} - \sigma$).

Then we have

$$x = \frac{1+4\sigma-A'-4\lambda_k\delta_{\parallel}}{1+4\sigma-4\sigma\lambda_k} \;, \tag{31}$$

$$E = 1+4\sigma+A'+4\lambda_k(1+\delta_{\parallel})+4\sigma\lambda_k - \frac{(1+4\sigma-4\lambda_k)^2}{1+4\sigma-A'-4\lambda_k\delta_{\parallel}} \;, \tag{32}$$

The surface spin-wave energy curve is then either one or two arcs of a hyperbola.

IV. SURFACE-ANISOTROPY EFFECTS

When the surface-anisotropy coupling constant A is negative, some surface spin waves have a negative energy (whatever the values are of σ, $\delta_{\parallel}$, and $\sigma_{\parallel}$, as may be seen from Eqs. (31) and (32) and from the condition $|x|>1$. Therefore, even at $T=0\,^{\circ}\mathrm{K}$, if A is negative, the ferromagnetic configuration of the spins near the surface is unstable. The surface spin waves of negative energy generally correspond to small values of k. For these modes the dipolar interactions, which are not *a priori* negligible before the surface-anisotropy energy, have the effect of shifting the whole energy spectrum slightly towards negative values, thus contributing to the instability of the ferromagnetic configuration. We shall therefore neglect them. In the absence of an external magnetic field, there is a spin rearrangement. The equilibrium spin configuration occurs for a non-negative minimum of the free energy.

Let us consider the problem from the semiclassical point of view of Refs. 12 and 13. The spin $\vec{S}_n$ of an atom belonging to the nth layer is then considered as a classical vector of constant length. Then one looks for the spin configuration which minimizes the semiclassical energy $\mathscr{E}$ at $T=0\,^{\circ}\mathrm{K}$. $\mathscr{E}$ is obtained by replacing the spin operators in the Hamiltonian of the system by the classical vectors $\vec{S}_n$. The energy extrema are given by the N equations

$$\frac{\delta\mathscr{E}}{\delta S_n^{\alpha}}=a_n S^{\alpha}\,,\quad n=1\cdots N\,,\quad \alpha=x,y,z \tag{33}$$

where N is the number of the layers and a_n are the Lagrange multipliers. Equation (33) means that no resulting torque is exerted on the spin S_n by the other spins in the equilibrium configuration.

We assume first that there is no bulk anisotropy and no nnn coupling, i.e., $\sigma_{\parallel}=\sigma=0$. We assume also that all spins in one layer are parallel to each other. For $n>1$, using Eq. (1) and Tables I and II, the explicit form of Eq. (33) is for the spins in the bulk

$$\vec{S}_{n-1}+\vec{S}_{n+1}=-2\,(a_n+2)\vec{S}_n\,. \tag{34}$$

Let the x axis be in the plane of $(S_1\,,z)$. For the spins at the surfaces Eq. (33) gives

$$\frac{S_2^x}{S_1^x}=A_1+\frac{S_2^z}{S_N^z}\,,\quad S_2^y=0,$$
$$\frac{S_{N-1}^x}{S_N^x}=A_N+\frac{S_{N-1}^z}{S_N^z}\,,\quad S_{N-1}^y=0. \tag{34a}$$

Here A_1 and A_N are the surface-anisotropy coupling constants of the two surfaces. From Eq. (34), $S_n^y=0$ and all the spins are parallel to the (x,z) plane. Let α_n be the angle between S_n and the z axis. Equation (34) and (34a) give

$$\alpha_n=\alpha_1+(n-1)\epsilon-2l\pi, \tag{35}$$

where l is an integer for the bulk layers, and

$$\sin 2\alpha_1=\frac{2\sin\epsilon}{A_1}\,,\qquad \sin 2\alpha_N=\frac{2\sin\epsilon}{A_N} \tag{35a}$$

for the two surface layers, where the angle ϵ between the directions of the spins in two adjacent layers is a constant given by the resolution of the system (35) and (35a) in which one replaces n by N. It gives

$$\sin 2(N-1)\epsilon=2\sin\epsilon\,\frac{[1-(4\sin^2\epsilon)^{1/2}/A_1^2]^{1/2}}{A_N}-\frac{[1-(4\sin^2\epsilon)]^{1/2}/A_N^2]^{1/2}}{A_1}\,. \tag{36}$$

When the film is nearly symmetrical, i.e., when $A_1\approx A_N$, one obtains

$$\epsilon\approx\frac{2l\,\pi}{N-1+2\,(1/A_N-1/A_1)}\quad (l=0,1,2,\cdots).$$

In ferromagnetic materials where the spins in the bulk are aligned along the z direction, this helicoidallike spin configuration still exists near the

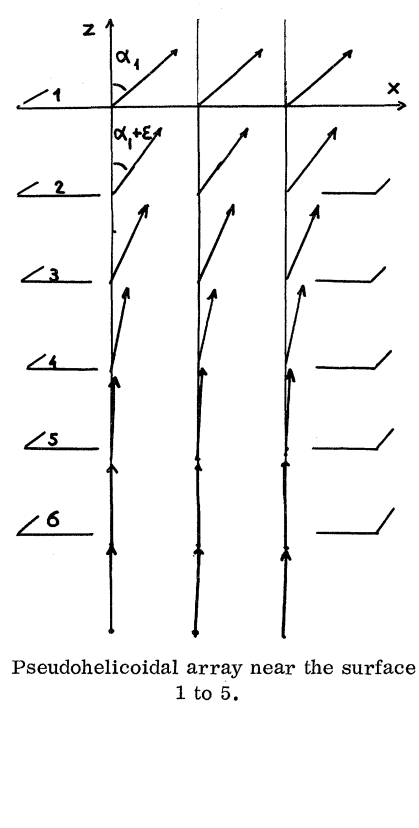

FIG. 4. Pseudohelicoidal array near the surface j layers 1 to 5.

surface, but is destroyed far from the surface by the bulk anisotropy which would tend to align the spins along the z direction. In that case ϵ would not be a constant any longer, but would be n dependent. This case is represented in Fig. 4. This is what we call a pseudohelicoidal spin configuration.

On the contrary, when the surface-anisotropy coupling constant is positive, the surface spin pattern is stiffer than in the absence of surface effects. Namely, the minimum energy required to set up a surface spin wave is nonzero. This energy gap for the excitation of the surface spin waves occurs at the "cutoff" value k_c where $|x| = 1$, as shown in Fig. 3. The equilibrium configuration is then ferromagnetic at the surface as well as in the bulk.

V. CONCLUDING REMARKS

In conclusion, within the framework of the above simple model, we have analyzed the effects of the surface anisotropy, the nnn coupling, and the nonuniform value of the exchange constants. Three properties have been shown. (i) The surface modes go through a transition from acoustical to optical character when the wave-vector parameter λ_k varies, going through λ_k^0. This transition is related to the existence of a nnn exchange coupling. (ii) For $\lambda_k = \lambda_k^0$, there are new types of modes for both bulk and surface spin waves. These modes are localized in one surface layer. (iii) Some rearrangements of the spins can occur near the surface, owing to the surface anisotropy.

The rearrangements of the surface spins occur when the surface-anisotropy constant A is negative. Then a special spin configuration is expected which is helicoidallike if there is no bulk anisotropy and pseudohelicoidal in the opposite case. This result extends the assumption of an antiferromagnetic first layer already used by Pincus,[16] Meiklejohn and Bean,[17] and Sparks.[18] As shown in Ref. 12, the spin-wave spectrum is changed when the equilibrium spin configuration is pseudohelicoidal. Moreover, the pinning conditions are modified because the propagation of the long-wavelength bulk modes is perturbed near the surface. On the other hand, when the surface-anisotropy coupling constant A is positive, the spin pattern is stiffened, since a large excitation energy is needed to create a surface spin wave. No rearrangement of the spins is expected in that case.

ACKNOWLEDGMENTS

The authors are indebted to Dr. Christiane Caroli, Dr. Danielle Lederer, and to the Alcibiades Group for interesting discussions.

[1] R. Weber, P. E. Tannenwald, and C. M. Bajorek, Appl. Phys. Letters 16, 35 (1970).

[2] D. L. Mills, J. Phys. Chem. Solids 28, 2245 (1967).

[3] E. Ilisca, Phys. Rev. Letters 24, 797 (1970).

[4] C. Kittel, Phys. Rev. 110, 1295 (1958).

[5] M. Sparks, Phys. Rev. B 1, 3831 (1970).

[6] H. S. Jarett and P. K. Waring, Phys. Rev. 111, 1223 (1958).

[7] J. Eschbach and R. Damon, Phys. Rev. 118, 1208 (1960).

[8] A. A. Maradudin and D. L. Mills, J. Phys. Chem. Solids 28, 1855 (1967).

[9] R. F. Wallis, A. A. Maradudin, I. P. Ipatova, and A. A. Klochikin, Solid State Commun. 5, 89 (1967).

[10] R. E. De Wames and T. Wolfram, Phys. Rev. 185, 720 (1969).

[11] E. Ilisca and J. L. Motchane, Phys. Letters 32A, 524 (1970).

[12] J. C. Levy, E. Ilisca, and J. L. Motchane, Phys. Rev. B (to be published).

[13] S. V. Tyablikov, *Methods in the Quantum Theory of Magnetism* (Plenum, New York, 1967), p. 155.

[14] C. Kittel, *Introduction to Solid State Physics*, 2nd ed. (Wiley, New York, 1961), p. 109.

[15] D. C. Gazos, R. Hermann, and R. F. Wallis, Phys. Rev. 119, 533 (1960).

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

[17] W. H. Meiklejohn and C. P. Bean, Phys. Rev. 102, 1413 (1956).

[18] M. Sparks, Phys. Rev. B 1, 4439 (1970).