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¹See, for instance, E. B. Tucker, in *Physical Acoustics*, edited by W. P. Mason (Academic, New York, 1966), Vol. 4B, Chap. 2.

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⁵In this paper the high-temperature limit is taken to mean temperatures high enough so that only spin-phonon mechanisms which contribute to the acoustic attenuation

as T^{-1} dominate.

⁶The notation for the phonons used in this paper is the same as in P. C. Kwok, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1967), Vol. 20.

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Pseudohelicoidal Surface Spin Waves

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A simple theoretical semiclassical calculation of the surface ferromagnetic equilibrium is given. The equilibrium orientation of the magnetization in the layers which are near the surface is tilted from the direction of the magnetization in the bulk. This rotation depends on both surface and bulk anisotropies. Thus, some instabilities in the spectrum disappear, and optical surface spin waves are found to be less energetic than what is usually calculated. Moreover, a simple interpretation of a possible origin of the pinning of surface spins is given.

There have been recently a number of theoretical investigations of surface spin waves in Heisenberg ferromagnets,¹⁻⁷ where the existence of surface modes is related, in the case of nearest-neighbor exchange, to the variation of the bulk exchange parameter at the surface. Nevertheless, an important question which remains open is that of the direction of the magnetization at the surface. In this paper a simple theoretical calculation of the surface equilibrium configuration is given. The most interesting result is that when the spin layer draws near to the surface, the spin magnetization rotates, and a so-called "pseudohelicoidal" structure is found in the vicinity of the surface. Consequently, a simple interpretation of the origin of pinning effects is given.

Moreover, the surface spin-wave spectrum is found to be perturbed by the existence of surface anisotropy. A treatment dealing with the existence of optical surface spin waves has already been given in the special case where the bulk and the surface magnetizations are parallel.⁸ However, the aim of this work is not to demonstrate that the surface and the bulk spectra are model dependent, but to lay emphasis on the fact that the magnetic structure of the surface often differs qualitatively from that of the bulk.

The direction of the bulk magnetization \vec{M} is assumed to be determined by a total energy balance (bulk anisotropy, sample configuration, . . .) and

thus it is independent of the surface anisotropy parameters. Let α be its angle with the axis Oz , which is perpendicular to the surface. In order to describe the surface effects we introduce the following Hamiltonian^{9,10}:

$$H = -\frac{1}{2} \sum_{f_1, f_2, \alpha} J(f_1 - f_2) S_{f_1}^\alpha S_{f_2}^\alpha - \frac{1}{2} \sum_{f_1, f_2} I(f_1 - f_2) S_{f_1}^z S_{f_2}^z. \quad (1)$$

The bulk anisotropy is not included in H ($\alpha = x, y, z$). The magnetocrystalline anisotropy integrals $I(f_1 - f_2)$ differ from zero near the surface only, where we make the assumption that $I(f_1 - f_2)$ is of the same order of magnitude as $J(f_1 - f_2)$. The isotropic exchange $J(f_1 - f_2)$ will also be perturbed. The penetration of the perturbation, i. e., the range of the helicoidal structure is given by the range where the I factors are nonzero, which is different from the surface spin-wave penetration. In order to simplify the calculation, we assume that only the first surface layer is perturbed. We define thus the following parameters:

$$J_{\parallel} = J(f_1 - f_2), \quad I_{\parallel} = I(f_1 - f_2),$$

where f_1 and f_2 are two nearest neighbors located on the surface layer and

$$J_{\perp} = J(f_1 - f_2), \quad I_{\perp} = I(f_1 - f_2),$$

where f_1 and f_2 are two nearest neighbors located on the first and second layers. Let us assume that

when neither f_1 nor f_2 are located in the first layer $J(f_1 - f_2)$ equals J and $I(f_1 - f_2)$ equals 0, where J is the isotropic exchange value in the bulk. The calculations are performed for cubic (100) structures.

We consider now the equilibrium spin configuration in a semiclassical way as described by Tyablikov.⁹ It is given by

$$\frac{\partial H}{\partial S_n^x} = a_n S_n^x, \quad (2)$$

where S_n^x is the component of the spin \vec{S}_n located on the n th layer from the surface (Ox being parallel to the surface), and a_n is a constant. Equation (2) means that the net torque acting on \vec{S}_n equals 0.⁷ All the spins which belong to the same layer are parallel. For the bulk layers Eq. (2) is equivalent to

$$\vec{S}_{n-1} + \vec{S}_{n+1} = f_n \vec{S}_n, \quad (3)$$

where f_n is a constant and thus \vec{S}_n belongs to the plane $(\vec{S}_{n-1}, \vec{S}_{n+1})$. Furthermore, as the surface anisotropy is directed along Oz , \vec{S}_1 belongs to the plane (Oz, \vec{M}) . Let Oy be perpendicular to this plane ($S_n^y = 0$). For the first two surface layers, Eq. (2) can be written

$$J_{\perp} \frac{S_2^x}{S_1^x} = 4I_n + (I_1 + J_{\perp}) \frac{S_2^x}{S_1^x}, \quad (4a)$$

$$J_{\perp} \frac{S_1^x}{S_2^x} + J \frac{S_3^x}{S_2^x} = (J_{\perp} + I_{\perp}) \frac{S_1^x}{S_2^x} + J \frac{S_3^x}{S_2^x}. \quad (4b)$$

We examine now the simple ferromagnetic case where all the bulk spins are parallel (and of unit length). Thus

$$\vec{S}_2 = \vec{S}_3 = \dots = \vec{S}_n.$$

In this simple case Eqs. (4a) and (4b) are found to be

$$\mu \frac{S_2^x}{S_1^x} = 4\lambda + (1 + \mu) \frac{S_2^x}{S_1^x}, \quad (5a)$$

$$\mu \frac{S_1^x}{S_2^x} = (1 + \mu) \frac{S_1^x}{S_2^x}, \quad (5b)$$

where $\lambda = I_n/I_1$ and $\mu = J_{\perp}/I_1$ are the only parameters which enter in Eqs. (5a) and (5b). We may notice that the bulk exchange J has disappeared.

Let us define $\alpha_1, \alpha_2, \dots, \alpha_n$ as the angles of the spins $\vec{S}_1, \vec{S}_2, \dots, \vec{S}_n$ with Oz . As stated before,

$$\alpha_2 = \alpha_3 = \dots = \alpha,$$

and therefore (5a) and (5b) may be written

$$\sin(\alpha + \alpha_1) = -(2\mu + 1) \sin(\alpha - \alpha_1), \quad (6a)$$

$$\sin(\alpha + \alpha_1) = -2\lambda \sin 2\alpha_1. \quad (6b)$$

From Eqs. (6a) and (6b) we finally obtain

$$\cos(\alpha_1 - \alpha) = -\left(\frac{1}{4\lambda} + \frac{4\lambda\mu(\mu+1)}{(1+2\mu)^2}\right), \quad (7a)$$

$$f(\lambda, \mu, \alpha) = \frac{1}{\mu^2} \sin^2\alpha + \left(\frac{1}{1+\mu}\right)^2 \cos^2\alpha - \left(\frac{4\lambda}{1+2\mu}\right)^2 = 0. \quad (7b)$$

Equation (7a) gives the rotation $\alpha_1 - \alpha$ of the surface spin \vec{S}_1 with respect to \vec{S}_2 . The choice of λ and μ may be restricted by Eq. (7b), which is a relation between the bulk and the surface anisotropy parameters. In fact this restriction is not so drastic when one remembers that generally the surface perturbation is not limited to the first layer.

The rotation of the magnetization given by Eq. (7a) is a direct consequence of the surface anisotropy. Obviously such an effect cannot be deduced from Osborne's model and results.³ In Ref. 3 the bulk anisotropy, which is the only one taken into account, introduces no qualitative change in the equilibrium spin configuration or in the spin-wave surface spectrum. Quantitatively, the energy levels and cutoff values are shifted as well as the penetration depth.

Let us give now a few numerical applications of Eqs. (7a) and (7b).

(i) Let us consider the case $\alpha = 0$ where all the bulk spins are perpendicular to the surface. Equations (6a) and (6b) imply that $\alpha_1 = 0$ or $\alpha_1 = \pi$. (The surface and bulk anisotropies have the same axis of symmetry.)

(ii) When all the spins are parallel to the surface, $\alpha = \frac{1}{2}\pi$, one of the solution corresponds to $\mu = -1$ and $|\lambda| \geq \frac{1}{4}$. α_1 is given by $\sin\alpha_1 = -1/4\lambda$. For example, $\lambda = \frac{1}{2}$ gives $\alpha_1 = -\frac{1}{6}\pi$, $\lambda = 1/2\sqrt{3}$ gives $\alpha_1 = -\frac{1}{3}\pi$, and $\lambda = -\frac{1}{4}$ gives $\alpha_1 = \alpha$.

(iii) In the special case where $I \ll J$, $\mu \gg 1$ and two solutions are found for α_1 : $\alpha_1 = \alpha$ or $\alpha_1 = \alpha + \pi$. When $\alpha = \frac{1}{2}\pi$, Eq. (7b) leads to $\lambda = \pm \frac{1}{2}$. For $\lambda = \frac{1}{2}$, $\alpha_1 = \alpha$, which is the classical solution found by de Wames.⁶ For $\lambda = -\frac{1}{2}$, $\alpha_1 = \alpha + \pi$, which is the new type of solution found by Sparks,⁷ where all the surface spins are antiparallel to the bulk spins (antiferromagnetic surface coupling).

Similar calculations can be performed for helical and antiferromagnetic structures [see Figs. 1(b) and 1(c)]. In the antiferromagnetic case a surface antiferro-spin-flop state is obtained, as described by Mills,¹¹ without applying any magnetic field. The same calculation could be performed when the axis of the surface anisotropy is not Oz but Ox (in the surface plane). In this case the surface spins rotate in the (Ox, \vec{M}) plane of an angle $\phi_1 - \phi = \alpha_1 - \alpha$ given by Eq. (7a).

The first important consequence of this calculation is that the surface spin-wave spectrum differs in most cases from those previously calculated, for example, by de Wames.⁶ In a semiclassical

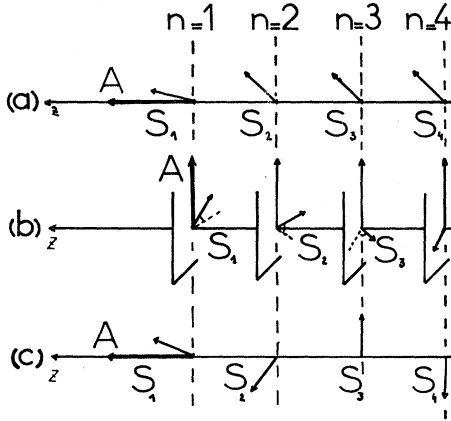


FIG. 1. The direction of the surface spins at equilibrium is in the plane given by the surface anisotropy axis A and the magnetization \vec{M} in the bulk. (a) Ferromagnetic case, where \vec{A} is perpendicular to the surface. (b) Helicoidal case, where \vec{A} is parallel to the surface. (c) Antiferromagnetic case, where \vec{A} is perpendicular to the surface. A surface spin-flop state is obtained.

calculation, we assume that

$$\langle S_1^f \rangle / \langle S_n^f \rangle = \cos \alpha_1,$$

and thus de Wames's matrix elements⁶ become

$$d_{11} = \left(\frac{2\Lambda_q J_{\parallel}}{J} + \frac{4I_{\parallel}}{J} \right) \cos \alpha_1 + \frac{J_{\perp} + I_{\perp}}{J} - 2(1 + \Lambda_q), \quad (8a)$$

$$d_{22} = \frac{J_{\perp} + I_{\perp}}{J} \cos \alpha_1 - 1, \quad (8b)$$

$$d_{12} = 1 - \frac{J_{\perp}}{J} \cos \alpha_1, \quad (8c)$$

$$d_{21} = 1 - \frac{J_{\perp}}{J}, \quad (8d)$$

and

$$b = d_{11} + d_{22}, \quad (9a)$$

$$c = d_{12} + d_{21} + d_{11} d_{22} - d_{12} d_{21}, \quad (9b)$$

$$d = d_{22}, \quad (9c)$$

Λ_q being related to the wave vector \vec{q} by

$$\Lambda_q = 2 - (\cos a q_x + \cos a q_y).$$

The energy of the surface spin waves is obtained by the following relation⁶:

$$E/4JS = 1 + \Lambda_q - \frac{1}{2}(x + 1/x), \quad (10)$$

where $S = \langle S_n^f \rangle$ and x is a root of the simple-cubic equation

$$x^3 + bx^2 + cx + d = 0 \quad (11a)$$

submitted to the condition

$$|x| \geq 1. \quad (11b)$$

Some particular results are given in Ref. 8: In the case where $J_{\parallel} = J_{\perp} = J$, no optical surface spin wave is found. Moreover, the surface branches are found to be parallel to the bulk ones, and thus the numerous cutoffs disappear. For example, when

$$J_{\parallel} \geq 2.5 J, \quad J_{\perp} = \frac{5}{8} J, \quad \mu = \frac{3}{2}, \quad \lambda = -\frac{1}{2},$$

which leads to

$$\alpha_1 - \alpha \approx \frac{1}{12} \pi.$$

We do find optical surface spin waves, but they are less energetic than those of Ref. 6, where the surface anisotropy was not considered. These results are summarized in Fig. 2. When surface spin waves propagate owing to the surface magnetocrystalline anisotropy, in some cases negative energies are found and there is thus an instability of the solution of Ref. 8. In fact, this calculation shows that the surface equilibrium configuration is modified so as to cancel this instability (Fig. 2).

The second important consequence is that the rotation of the direction of the spins at equilibrium at the surface may in some cases explain the pinning of surface spins. Obviously, when $\alpha_1 - \alpha \neq 0$ (for example, $\alpha_1 = \alpha + \frac{1}{2}\pi$), the spins S_1 do not precess around their equilibrium positions. Bulk spin waves are damped by the pseudohelicoidal structure when they enter the vicinity of the surface. This has already been demonstrated from a macroscopic point of view by Pincus,¹⁰ who has investigated two simple cases (pseudodipolar and anisotropic exchange). For example, the case of

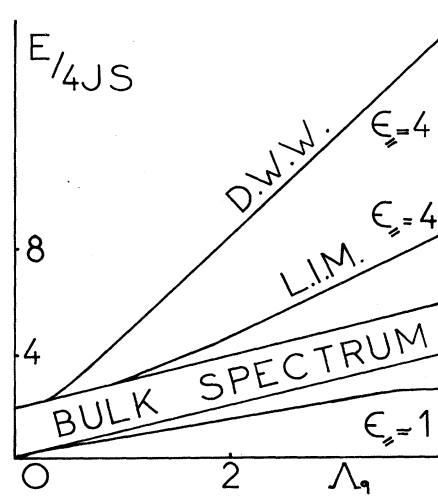


FIG. 2. Surface spin-wave spectrum: $E/4JS$ versus $\Lambda_q = 2 - (\cos a q_x + \cos a q_y)$. The branch $J_{\parallel} = 4J$ obtained here (L. I. M.) is compared to the one obtained by de Wames (D. W. W.). The branch $J_{\parallel} = J$ is shown starting at $E = 0$ and not at $E < 0$ as in Ref. 8. $\epsilon_{\parallel} = J_{\parallel}/J$.

an antiferromagnetic surface layer described by Meiklejohn¹² is a special pseudohelical structure where Pincus's calculation is still valid. Afterwards, Soohoo¹³ and Wigen¹⁴ confirmed and developed these surface spin pinning mechanisms in agreement with some experimental results.

However, the pseudohelical structure which generalizes the previous simple cases differs from a real helical one because a few layers only are perturbed and thus the structure is not periodical. The shorter the range of the pseudohelical structure is, the stronger the pinning.

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General Theorems on Ferromagnetism and Ferromagnetic Spin Waves*

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Ideal ferromagnetism in perfect crystals (and/or in free space), where spin-orbit interactions may be neglected, is investigated at zero temperature under the following conditions: The fermion system considered here should have the inversion symmetry of the space coordinates and the thermodynamic limit. Its ground state is nondegenerate for a fixed eigenvalue of S_z . In other respects the ferromagnets considered are quite general and may cover all possible types of ferromagnetism: insulators, metals, and free fermions. Dynamical spin-spin correlation functions are studied. Sum rules for them are developed so as to exclude the contributions from Stoner excitations. Spin waves are considered by means of these sum rules. In the case of complete ferromagnetism (all electron spins being aligned in one direction), it is shown rigorously that no consistent result can be obtained; the excitation energies of magnons cannot be finite in the form of Dq^2 , but are vanishing. This suggests that the complete ferromagnetism, if it could exist, must violate one of the above conditions.

I. INTRODUCTION

Ferromagnetism and ferromagnetic spin waves have been discussed for many years.^{1,2} The discussions have mostly concerned specific models, such as ideal Heisenberg ferromagnets and itinerant-electron models. If the spin-orbit interaction is neglected and hence the spontaneous magnetization of a ferromagnet may take any direction, then a well-defined acoustic spin-wave mode with frequency spectrum $\omega = Dq^2$ is always obtained for small values of the wave number q . This fact, which can be easily inferred from the results derived for the specific models, has also been discussed by some authors³ from the point of view of the Goldstone theorem⁴ relating the acoustic mode with symmetry breaking down. However, such discussions have again been confined to the models.

A simple and undoubtedly clear derivation of the magnon mode was given for the ideal Heisenberg ferromagnet,¹ which is, however, an oversimplification of real ferromagnets. Actually, the problems concerning the nonorthogonality and variety of ionic configurations of atoms in a solid must inevitably be kept in mind whenever we go beyond the simple-minded pictures in which we neglect the nonorthogonality and assume fixed atomic orbital configurations. These problems will destroy all the advantage of the Heisenberg model in its mathematical simplicity even in the case of ferromagnetic insulators.

In the spin-wave theory² of metallic ferromagnets, which was initiated by the famous work of Herring and Kittel, we have been dealing with some models which are again approximate pictures. Yet we have not been successful in rigorously de-