Surface spin waves in Heisenberg ferromagnets with nonuniaxial single-ion anisotropy

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A Green-function formalism is employed to study the effect of nonuniaxial single-ion anisotropy on surface spin waves in semi-infinite Heisenberg ferromagnets with nearest-neighbor exchange interactions and at low temperatures $T \ll T_c$. Results are deduced for the dispersion relations of surface spin waves in terms of the exchange and anisotropy parameters, which are assumed different at the surface compared with those of the bulk. The spin-correlation functions are also calculated as functions of the layer distances from the surface layer.

I. INTRODUCTION

It is well known that at the surface of an ordered magnetic material there may be localized surface spin waves in addition to the bulk spin waves. In recent years there has been considerable interest, both theoretical and experimental, in the properties of surface spin waves and the applications have included light scattering and spin-wave resonance (e.g., see Refs. 1–3 for reviews). The purpose of this paper is to extend previous calculations for Heisenberg magnetic materials with uniaxial anisotropy by including the effects of nonuniaxial anisotropy.

In general, nonuniaxial anisotropy can arise in at least two ways—through nonuniaxial single-ion anisotropy (as in the ferromagnet $CrBr_3$ and in the antiferromagnet NiO) or through nonuniaxial anisotropic exchange (as in some of the rare-earth orthoferrites). The resulting modifications to the spin-wave properties have been studied by light scattering in several bulk systems (e.g., see Ref. 4). In the present paper we concentrate on ferromagnets with nonuniaxial single-ion anisotropy, but the results may be extended to ferromagnets with anisotropic exchange and to antiferromagnets. Some comments concerning nonuniaxial anisotropic exchange and also dipole-dipole anisotropy are given later.

In the case of bulk Heisenberg ferromagnets, the effects of nonuniaxial ("easy-plane") single-ion anisotropy on the spin waves have been calculated using Green-function methods.[5–7] We write the Hamiltonian representing the single-ion anisotropy in a material where the total symmetry is noncubic as

$$\mathcal{H}_A = -\sum_i D_i (S_i^z)^2 - \sum_i F_i [(S_i^x)^2 - (S_i^y)^2] \; . \tag{1}$$

Here $\mathbf{S}_i$ is the spin operator at site i in the ferromagnet, and D_i and F_i are anisotropy parameters (at site i) for the uniaxial and "easy-plane" terms, respectively. In the present work we retain the site dependence of D_i and F_i because these quantities depend on the crystalline electric fields, which may be different near the surface of a ferromagnet compared with the bulk. The total Hamiltonian $\mathcal{H}$ is taken as

$$\mathcal{H} = -\tfrac{1}{2}\sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - g\mu_B H_0 \sum_i S_i^z + \mathcal{H}_A \; , \tag{2}$$

where J_{ij} is the exchange interaction between sites i and j, H_0 is an applied magnetic field along the z direction (the direction of static magnetization), and $\mathcal{H}_A$ is given by Eq. (1).

In the present paper we confine ourselves to the low-temperature regime of $T \ll T_c$. This allows us to transform the Hamiltonian to boson operators ($b_i^\dagger$ and b_i) using the Holstein-Primakoff representation and, in the usual linear spin-wave approximation of retaining terms up to second order in the operators, we obtain

$$\mathcal{H} = \sum_i \left[g\mu_B H_0 + 2D_i S\eta + S\sum_j J_{ij} \right] b_i^\dagger b_i - S\sum_{i,j} J_{ij}\, b_i^\dagger b_j - S\eta^{1/2}\sum_i F_i (b_i^\dagger b_i^\dagger + b_i b_i) \; , \tag{3}$$

where we define $\eta = (1 - 1/2S)$. We notice that for $S = \frac{1}{2}$ we have $\eta = 0$ and so the single-ion terms correctly vanish in Eq. (3). For $S > \frac{1}{2}$ the single-ion terms are nonzero, and we see that the F_i anisotropy leads to a term in $\mathcal{H}$ proportional to $(b_i^\dagger b_i^\dagger + b_i b_i)$. It is this operator term that makes the calculations for spin waves in nonuniaxial systems more complicated, and in semiclassical terminology it makes the spin precession elliptical rather than circular. The solution of Eq. (3) for the spin waves in an *infinite* system can be found straightforwardly by Fourier transforming from site labels to a three-dimensional wave vector $\mathbf{k}$ and then "diagonalizing" the Hamiltonian (e.g., see Ref. 8). We now show how the Green-function calculation can be done for spin waves in a semi-infinite ferromagnet, where the absence of translational symmetry in the direction perpendicular to the surface complicates the analysis and leads to the possibility of surface spin waves. The general formalism is presented in Sec. III, including the case where the D and F anisotropy parameters may be different at the surface compared with the bulk. In Secs. III and IV we give results for the spin-wave energies and spectral intensities, respectively. Further discussion and conclusions are contained in Sec. V.

In some materials there is evidence that the anisotropy may be nonuniaxial at the surface while being uniaxial in the bulk, that is, F is zero everywhere except at the surface. This represents a special case of the more general results presented here (where there may be different anisotropies in the surface and bulk, and both may be nonuniaxial). The special case and related references are discussed in more detail in Sec. V.

II. MATRIX FORMALISM FOR THE SURFACE GREEN FUNCTIONS

We consider a semi-infinite anisotropic ferromagnet occupying the half-space $z \geq 0$ and described by the boson Hamiltonian of Eq. (3). We assume that the magnetic ions form a simple-cubic structure (where a is the lattice parameter) with crystallographic axes parallel to the xyz axes. [Note that the coordination of the nonmagnetic ions in a compound may generally be such as to make the overall symmetry (and hence the crystalline fields) noncubic, as assumed for Eq. (1)]. Defining the retarded commutator Green functions $\langle\langle b_i^\dagger(t); b_j(t')\rangle\rangle$ and $\langle\langle b_i(t); b_j(t')\rangle\rangle$ for general site labels and time labels, we construct in the usual way,[9] the equations of motion for their frequency Fourier transforms $\mathcal{G}_{ij}(\omega)$ and $\mathcal{G}'_{ij}(\omega)$, respectively. This gives the following set of coupled finite-difference equations:

$$\left[\hbar\omega + g\mu_B H_0 + 2D_i S\eta + S\sum_m J_{mi}\right]\mathcal{G}_{ij}(\omega) - S\sum_m J_{mi}\mathcal{G}_{mj}(\omega) = -(\delta_{ij}/2\pi) + 2F_i S\eta^{1/2}\mathcal{G}'_{ij}(\omega) , \quad (4)$$

$$\left[-\hbar\omega + g\mu_B H_0 + 2D_i S\eta + S\sum_m J_{mi}\right]\mathcal{G}'_{ij}(\omega) - S\sum_m J_{mi}\mathcal{G}'_{mj}(\omega) = 2F_i S\eta^{1/2}\mathcal{G}_{ij}(\omega) . \quad (5)$$

To solve these equations we utilize the translational symmetry in the xy plane to define the Fourier transforms

$$\mathcal{G}_{ij}(\omega) = \frac{1}{N}\sum_{\mathbf{k}_\parallel}\exp[i\mathbf{k}_\parallel\cdot(\mathbf{r}_i - \mathbf{r}_j)]G_{n,n'}(\mathbf{k}_\parallel,\omega) \quad (6)$$

and a similar expression for $\mathcal{G}'_{ij}(\omega)$. Here $\mathbf{k}_\parallel = (k_x, k_y)$ is a two-dimensional wave vector parallel to the surface, while n and n' are positive integers labeling the lattice planes (parallel to the surface) that contain sites i and j, respectively. Hence, $n=1$ is the surface layer, $n=2$ the next layer, and so on. The normalization constant N in Eq. (6) denotes the number of sites in any lattice plane parallel to the surface. It is also convenient to introduce the following summations for the exchange interactions:

$$u_n(\mathbf{k}_\parallel) = \sum_{\delta_1}\exp(i\mathbf{k}_\parallel\cdot\boldsymbol{\delta}_1)J_{ij}(\mathbf{r},\mathbf{r}+\boldsymbol{\delta}_1) , \quad (7)$$

$$v_n(\mathbf{k}_\parallel) = \sum_{\delta_2}\exp(i\mathbf{k}_\parallel\cdot\boldsymbol{\delta}_2)J_{ij}(\mathbf{r},\mathbf{r}+\boldsymbol{\delta}_2) , \quad (8)$$

where $\boldsymbol{\delta}_1$ is a vector connecting any site i located at $\mathbf{r}$ in layer n to its nearest neighbors j in the same layer n, while $\boldsymbol{\delta}_2$ is a vector connecting any site at $\mathbf{r}$ in layer n with its nearest neighbors in layer $n+1$. If we assume for simplicity that only nearest-neighbor exchange interactions are important, having the value J_S if both spins are in the surface layer ($n=1$) and the bulk value J otherwise, then

$$u_n(\mathbf{k}_\parallel) = \begin{cases} 4J_S\gamma(\mathbf{k}_\parallel) \equiv u_1(\mathbf{k}_\parallel) & \text{for } n=1 \\ 4J\gamma(\mathbf{k}_\parallel) \equiv u_B(\mathbf{k}_\parallel) & \text{for } n \geq 2 \end{cases} \quad (9)$$

and

$$v_n(\mathbf{k}_\parallel) = J \equiv v_B(\mathbf{k}_\parallel) \quad \text{for all } n \geq 1 , \quad (10)$$

where

$$\gamma(\mathbf{k}_\parallel) = [\cos(k_x a) + \cos(k_y a)]/2 . \quad (11)$$

Equations (9)–(11) may readily be generalized to accommodate other assumptions concerning the exchange interactions (e.g., to include next-nearest-neighbor exchange or different lattice structures). For the single-ion anisotropy parameters, we assume that they may have the perturbed values D_S and F_S for spins in the surface layer ($n=1$), but otherwise they have the corresponding bulk values D and F, respectively.

The set of coupled equations (4) and (5) can then be rewritten more compactly in terms of $\infty \times \infty$ matrices as

$$(\Omega\underline{I} + \underline{A})\underline{G}(\mathbf{k}_\parallel,\omega) = d_F\underline{R}\underline{G}'(\mathbf{k}_\parallel,\omega) - \lambda\underline{I} , \quad (12)$$

$$(-\Omega\underline{I} + \underline{A})\underline{G}'(\mathbf{k}_\parallel,\omega) = d_F\underline{R}\underline{G}(\mathbf{k}_\parallel,\omega) , \quad (13)$$

where $\lambda = 1/[2\pi S v_B(\mathbf{k}_\parallel)]$ and we define the dimensionless quantities:

$$\Omega = \hbar\omega/Sv_B(\mathbf{k}_\parallel), \quad d_F = 2F\eta^{1/2}/v_B(\mathbf{k}_\parallel) . \quad (14)$$

The elements of matrices $\underline{G}$ and $\underline{G}'$ are the Green functions $G_{n,n'}(\mathbf{k}_\parallel,\omega)$ and $G'_{n,n'}(\mathbf{k}_\parallel,\omega)$ defined as in Eq. (6), $\underline{I}$ is the unit matrix, and $\underline{R}$ is equal to the unit matrix with the (1,1) element replaced by $\nu \equiv F_S/F$. The quantity $\underline{A}$ is a tridiagonal matrix, which can be split into two parts as

$$\underline{A} = \underline{A}_0 + \underline{\Delta} , \quad (15)$$

where

$$\underline{A}_0 = \begin{bmatrix} d & -1 & 0 & \cdots & & \\ -1 & d & -1 & 0 & \cdots & \\ 0 & -1 & d & -1 & 0 & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \end{bmatrix} , \quad (16)$$

$$\underline{\Delta} = \begin{bmatrix} \delta & 0 & \cdots \\ 0 & 0 & \cdots \\ \vdots & \vdots & \end{bmatrix} , \quad (17)$$

with

$$d = \{g\mu_B H_0 + 2DS\eta + S[u_B(0) - u_B(\mathbf{k}_\parallel)] + 2Sv_B(\mathbf{k}_\parallel)\}/Sv_B(\mathbf{k}_\parallel) , \quad (18)$$

$$\delta = \{2(D_S - D)S\eta + S[u_1(0) - u_1(\mathbf{k}_\parallel) - u_B(0) + u_B(\mathbf{k}_\parallel)] - Sv_B(\mathbf{k}_\parallel)\}/Sv_B(\mathbf{k}_\parallel) . \quad (19)$$

We choose to make this decomposition of $\underline{A}$ by analogy with earlier work[10,11] on uniaxial ferromagnets because the inverse of a matrix having the form of $\underline{A}_0$ is known analytically.

In the special case of uniaxial anisotropy (i.e.,

$F=F_S=0$), Eqs. (12)–(13) give simply $\underline{G}=-\lambda(\Omega\underline{I}+\underline{A})^{-1}$ and $\underline{G}'=\underline{0}$ for the matrix Green functions. This result is consistent with earlier Green-function calculations for uniaxial systems.[11,12] In the case of nonuniaxial anisotropy, $\underline{G}$ and $\underline{G}'$ are coupled and are both nonzero. After some algebraic manipulation and factorization of matrices, we find from Eqs. (12)–(13), the formal results that

$$\underline{G}(\mathbf{k}_{\parallel},\omega)=-\lambda(\underline{I}+\underline{A}_2^{-1}\underline{A}_1^{-1}\underline{D})^{-1}\times\underline{A}_2^{-1}\underline{A}_1^{-1}(-\Omega\underline{I}+\underline{A})\underline{R}^{-1}\ , \tag{20}$$

$$\underline{G}'(\mathbf{k}_{\parallel},\omega)=-\lambda d_F(\underline{I}+\underline{A}_2^{-1}\underline{A}_1^{-1}\underline{D}^T)^{-1}\underline{A}_2^{-1}\underline{A}_1^{-1}\ . \tag{21}$$

Here $\underline{A}_1$ and $\underline{A}_2$ are tridiagonal matrices defined as in Eq. (16) for $\underline{A}_0$, but with the diagonal element d replaced by $d_1=d+(\Omega^2+d_F^2)^{1/2}$ and $d_2=d-(\Omega^2+d_F^2)^{1/2}$, respectively, and $\underline{D}$ is an $\infty\times\infty$ matrix with only the following nonzero elements:

$$\begin{aligned}D_{11}&=(d+\delta)^2\nu^{-1}-d^2-\Omega^2(\nu^{-1}-1)-d_F^2(\nu-1)\ ,\\ D_{12}&=d-(d+\delta)\nu^{-1}+\Omega(\nu^{-1}-1)\ ,\\ D_{21}&=d-(d+\delta)\nu^{-1}-\Omega(\nu^{-1}-1)\ ,\\ D_{22}&=(\nu^{-1}-1)\ .\end{aligned} \tag{22}$$

The matrix $\underline{D}^T$ in Eq. (21) is the transpose of the matrix $\underline{D}$ defined above. A case of special interest occurs when the 'easy-plane' anisotropy coefficient in the surface layer is the same as in the bulk (i.e., $F_S=F$), whereupon Eq. (20) simplifies to

$$\underline{G}(\mathbf{k}_{\parallel},\omega)=-\lambda(\underline{I}+\underline{A}_2^{-1}\underline{\Delta})^{-1}\underline{A}_2^{-1}(\underline{I}+\underline{A}_1^{-1}\underline{\Delta})^{-1}\times\underline{A}_1^{-1}(-\Omega\underline{I}+\underline{A})\ . \tag{23}$$

The corresponding expression for the matrix Green function $\underline{G}'(\mathbf{k}_{\parallel},\omega)$ may be simply obtained from Eq. (21).

III. RESULTS FOR SPIN-WAVE FREQUENCIES

We now examine the Green-function results for the general case where F_S and F may be different. The spin-wave frequencies can be obtained directly from the poles of $\underline{G}(\mathbf{k}_{\parallel},\omega)$ or $\underline{G}'(\mathbf{k}_{\parallel},\omega)$, and corresponding to the term $(\underline{I}+\underline{A}_2^{-1}\underline{A}_1^{-1}\underline{D})^{-1}$ in Eq. (20) for $\underline{G}(\mathbf{k}_{\parallel},\omega)$ there are poles given by the determinantal condition:

$$\det(\underline{I}+\underline{A}_2^{-1}\underline{A}_1^{-1}\underline{D})^{-1}=0\ . \tag{24}$$

We show below that this has solutions (for Ω) that correspond to localized (or surface) spin waves. It can be verified that the other terms in Eq. (20) do not have surface spin-wave solutions.

Expressions for $\underline{A}_1^{-1}$ and $\underline{A}_2^{-1}$ can be written down by analogy with earlier work.[10,11] This involves introducing the complex variables x_1 and x_2, which satisfy $|x_1|\le 1$ and $|x_2|\le 1$, and are defined by

$$\begin{aligned}x_1+x_1^{-1}&=d+(\Omega^2+d_F^2)^{1/2}\ ,\\ x_2+x_2^{-1}&=d-(\Omega^2+d_F^2)^{1/2}\ .\end{aligned} \tag{25}$$

The matrix elements of $\underline{A}_\alpha^{-1}$ ($\alpha=1,2$) are then given by

$$(\underline{A}_\alpha^{-1})_{ij}=(x_\alpha^{i+j}-x_\alpha^{|i-j|})/(x_\alpha-x_\alpha^{-1})\quad(\alpha=1,2)\ . \tag{26}$$

As before,[10,11] the propagating bulk spin waves correspond to $|x_\alpha|=1$, whereas the localization condition for the existence of surface spin waves is $|x_\alpha|<1$. For the case of bulk spin waves we can write x_1 or x_2 as $\exp(ik_za)$, where k_z is a real wave vector component perpendicular to the surface, and Eq. (25) can be rearranged to give $\Omega=\Omega_B$ with

$$\Omega_B=\{[d-2\cos(k_za)]^2-d_F^2\}^{1/2}\ . \tag{27}$$

This is equivalent to the dispersion relation for a spin wave with three-dimensional wave vector $\mathbf{k}=(\mathbf{k}_{\parallel},k_z)$ in an *infinite* anisotropic ferromagnet, as can be seen directly by diagonalizing Eq. (3).

For surface spin waves in a semi-infinite ferromagnet we first rewrite Eq. (24) in terms of x_1 and x_2 using (25) and (26) to obtain the condition

$$(1+\delta x_1)(1+\delta x_2)+(1-\nu)\frac{d_F^2x_1x_2}{1-x_1x_2}[(1+\nu)+(1-\nu)x_1x_2]=0\ . \tag{28}$$

From the solutions for x_1 and x_2 that satisfy both this equation and the localization conditions $|x_1|<1$ and $|x_2|<1$, we can find the surface spin-wave frequencies by using Eq. (25). The solutions for x_1 and x_2 are conveniently obtained by first solving for a new variable y, defined by $y=x_1x_2$, which can be shown [using Eq. (28)] to satisfy the following cubic equation:

$$[d_F^2(1-\nu)^2-\delta^2]y^3+[2d_F^2(1-\nu)-2d\delta-1]y^2+[2d\delta+\delta^2+d_F^2(1-\nu^2)]y+1=0\ . \tag{29}$$

Some numerical results to illustrate this procedure are shown later, but it is helpful first to consider the special case of $F_S=F$ because the results simplify and provide us with more physical insight. From Eq. (23) the poles of $\underline{G}(\mathbf{k}_{\parallel},\omega)$ for this case are given by either $\det(\underline{I}+\underline{A}_1^{-1}\underline{\Delta})=0$ or $\det(\underline{I}+\underline{A}_2^{-1}\underline{\Delta})=0$. Since it is easily proved using Eqs. (17) and (26) that

$$\det(\underline{I}+\underline{A}_1^{-1}\underline{\Delta})=1+x_1\delta\ , \tag{30}$$

we conclude that either $x_1=-\delta^{-1}$ or $x_2=-\delta^{-1}$. This could also be deduced by putting $\nu=1$ in Eq. (28). Substitution of these values into Eq. (25) gives $\Omega=\Omega_S$ for the surface spin-wave frequencies:

$$\Omega_S=[(d+\delta+\delta^{-1})^2-d_F^2]^{1/2} \ . \qquad (31)$$

The localization condition $|x_1|<1$ (or $|x_2|<1$) corresponding to Eq. (31) becomes $|\delta|>1$ and a surface spin wave exists only when this inequality is satisfied. The attenuation length L (exponential decay length) is then

$$L=a(\ln|1/\delta|)^{-1} \ . \qquad (32)$$

If $|\delta|\to 1$ from above we have $L\to\infty$, so the surface mode becomes more deeply penetrating and eventually it becomes degenerate in frequency with a bulk spin wave. In general, the localization condition can be satisfied either by $-\infty<\delta<-1$ or $1<\delta<\infty$, which correspond, respectively, to an *acoustic* surface spin-wave branch coming below the bulk continuum or an *optic* branch coming above the bulk continuum. Using Eqs. (9), (10), and (19), the expression for δ can be rewritten explicitly as

$$\delta=2\eta\frac{D}{J}\left[\frac{D_S}{D}-1\right]+4[1-\gamma(\mathbf{k}_\parallel)]\left[\frac{J_S}{J}-1\right]-1 \ . \qquad (33)$$

Hence the existence condition depends on the ratios D/J, D_S/D, and J_S/J, as well as on the wave vector $\mathbf{k}_\parallel$ [through the structure factor $\gamma(\mathbf{k}_\parallel)$] and the spin quantum number S (through the η factor). For example, if $D_S/D<1$ and $J_S/J<1$ there is an acoustic surface spin wave for *all* value of $\mathbf{k}_\parallel$ (provided D and J are both positive).

Some examples to illustrate these predicted dispersion relations for the special case of F_S equal to F are presented in Fig. 1, where we plot the spin-wave frequency (in terms of the dimensionless quantity Ω) against $|\mathbf{k}_\parallel a|$. The propagation wave vector $\mathbf{k}_\parallel$ is along the [100] direction and we consider spins with $S=1$. The bulk spin waves appear as a continuum in this kind of plot (with upper and lower edges corresponding to $k_z=\pi/a$ and $k_z=0$, respectively). Several surface spin-wave branches are shown corresponding to different assumed values for the exchange and anisotropy parameters at the surface compared to their values in bulk. It is seen that for the case $D_S/D<1$ and for $J_S/J>1$ there is a surface spin-wave branch below the bulk continuum (an acoustic branch) for $|\mathbf{k}_\parallel a|\lesssim 0.3\pi$ and an optic branch for $|\mathbf{k}_\parallel a|\gtrsim 0.6\pi$. In the case when $D_S/D>1$ there is an acoustic branch for $J_S/J<1$ and an optic branch for $J_S/J>1$, and both these spin-wave branches occur only above a certain value of $|\mathbf{k}_\parallel a|$.

The surface spin-wave modes for the general case of $F_S\neq F$ are obtained numerically by first solving for y from Eq. (29) and then looking for solutions for x_1 and x_2 that satisfy both $|x_1|<1$ and $|x_2|<1$. The spin-wave frequencies are then obtained using Eq. (25) and plots of these against $|\mathbf{k}_\parallel a|$ are presented in Fig. 2 assuming $F_S/F=2$ and for propagation vector along [110]. The other anisotropy and exchange parameters are taken to be the same as for Fig. 1. We notice that the spin-wave modes show a qualitatively similar behavior to the case of $F_S=F$, although their frequencies are altered.

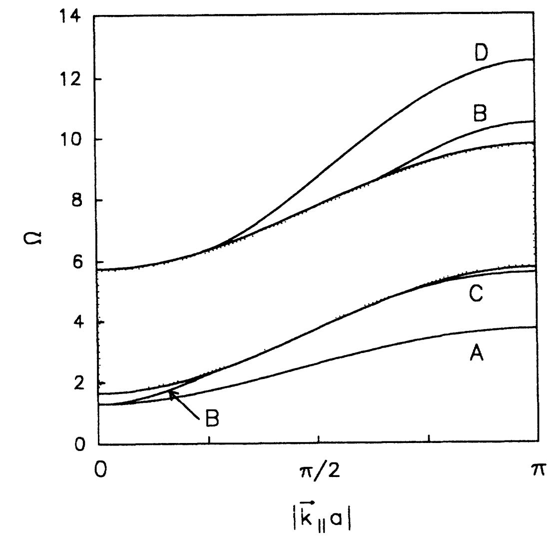

FIG. 1. A plot of the spin-wave frequencies (in units of SJ) against $|\mathbf{k}_\parallel a|$ in a semi-infinite $S=1$ Heisenberg ferromagnet with nonuniaxial anisotropy and for propagation wave vector $\mathbf{k}_\parallel$ along [100]. The applied magnetic field is such that $g\mu_B H_0/SJ=0.3$ and the anisotropy parameters are chosen to be $D/J=1.5$, $F/J=0.5$, and $F_S/F=1$. The bulk spin-wave continuum is shown together with surface spin-wave branches corresponding to A, $D_S/D=0.5$, $J_S/J=0.5$; B, $D_S/D=0.5$, $J_S/J=2.0$; C, $D_S/D=1.5$, $J_S/J=0.5$; D, $D_S/D=1.5$, $J_S/J=2.0$.

IV. RESULTS FOR SPECTRAL INTENSITIES

The Green-function results of Sec. II can also be used to discuss the spectral intensities of the surface spin waves. We introduce here the transverse spin-correlation functions $\langle S_i^-(t)S_j^+(t')\rangle$ for spin operators at sites i and j, which in the linear spin-wave approximation takes the form $2S\langle b_i^\dagger(t)b_j(t')\rangle$.

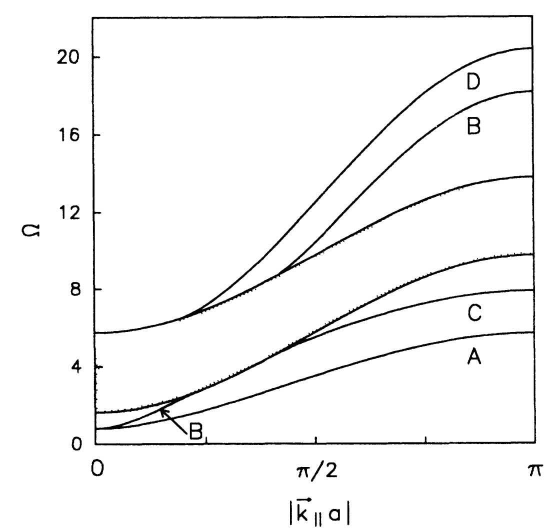

FIG. 2. The spin-wave frequencies (in units of SJ) plotted against $|\mathbf{k}_\parallel a|$ for $F_S/F=2$ and for propagation wave vector $\mathbf{k}_\parallel$ along the [110] direction. The other exchange and anisotropy parameters are as for Fig. 1.

For simplicity we outline the calculations for the special case of $F_S=F$, where the Green function $\underline{G}(\mathbf{k}_\parallel,\omega)$ is given by Eq. (23) and can be rewritten as

$$\underline{G}(\mathbf{k}_\parallel,\omega)=\frac{\underline{N}(x_1,x_2,\Omega)}{(1+x_1\delta)(1+x_2\delta)} . \tag{34}$$

Here matrix $\underline{N}$ is

$$\underline{N}=-\lambda\,\mathrm{adj}(\underline{I}+\underline{A}_2^{-1}\underline{\Delta})\underline{A}_2^{-1} \times \mathrm{adj}(\underline{I}+\underline{A}_1^{-1}\underline{\Delta})\underline{A}_1^{-1}(-\Omega\underline{I}+\underline{A}) \tag{35}$$

with adj denoting the matrix adjoint. Using Eqs. (34) and (31) and the definitions of x_1 and x_2, we have

$$\underline{G}(\mathbf{k}_\parallel,\omega)=\frac{(\delta^{-1}+x_1^{-1})(\delta^{-1}+x_2^{-1})\underline{N}(x_1,x_2,\Omega)}{(\Omega_S^2-\Omega^2)} , \tag{36}$$

which shows explicitly the poles for Ω at $\pm\Omega_S$, where Ω_S is the surface spin-wave frequency.

Suppose we now introduce a spectral function $\xi_n(\mathbf{k}_\parallel,\omega)$, defined in terms of the correlation function by

$$\langle b_n^\dagger(t)b_n(t')\rangle_{\mathbf{k}_\parallel}=\int_{-\infty}^{\infty}\xi_n(\mathbf{k}_\parallel,\omega)\exp[-i\omega(t-t')]d\omega , \tag{37}$$

where the operators refer to layer n and we seek the contribution from surface spin waves with wave vector component $\mathbf{k}_\parallel$. The spectral function can now be deduced from the imaginary part of the Green function $\underline{G}(\mathbf{k}_\parallel,\omega)$ by using the fluctuation-dissipation theorem.[9] With the aid of Eq. (36) this leads to

$$\xi_n(\mathbf{k}_\parallel,\omega)=[\pi(\delta^{-1}+x_1^{-1})(\delta^{-1}+x_2^{-1})f(\Omega)N_{n,n}(x_1,x_2,\Omega)/\Omega_S][\delta(\Omega_S+\Omega)-\delta(\Omega_S-\Omega)] , \tag{38}$$

where

$$f(\Omega)=[\exp(\Omega SJ/k_BT)-1]^{-1} . \tag{39}$$

When this is substituted into Eq. (37) we can immediately carry out the integration over ω (by using properties of the δ function) to obtain the transverse spin-correlation functions $\langle S_n^-(t)S_n^+(t)\rangle_{\mathbf{k}_\parallel}$ at equal times. Likewise we can also deduce $\langle S_n^+(t)S_n^-(t)\rangle_{\mathbf{k}_\parallel}$ and, hence, the quantity $\langle (S_n^x)^2+(S_n^y)^2\rangle_{\mathbf{k}_\parallel}$ which represents a mean-squared amplitude of the surface spin-wave precession.

We notice also that the correlation function

$$\langle S_n^+(t)S_{n'}^+(t')\rangle_{\mathbf{k}_\parallel}\cong 2S\langle b_n(t)b_{n'}(t')\rangle_{\mathbf{k}_\parallel}$$

can similarly be obtained from the Green function $\underline{G}'(\mathbf{k}_\parallel,\omega)$ of Eq. (21). The equal-time correlation function, when combined with $\langle S_n^-(t)S_n^-(t)\rangle_{\mathbf{k}_\parallel}$, can be used to show that

$$\langle (S_n^x)^2-(S_n^y)^2\rangle_{\mathbf{k}_\parallel}=S[(\delta^{-1}+x_1^{-1})(\delta^{-1}+x_2^{-1})/\Omega_S] \times[2f(\Omega_S)+1]N'_{n,n}(x_1,x_2) , \tag{40}$$

where the matrix $\underline{N}'$ is given by

$$\underline{N}'=d_F\mathrm{adj}(\underline{I}+\underline{A}_2^{-1}\underline{\Delta})\underline{A}_2^{-1}\mathrm{adj}(\underline{I}+\underline{A}_1^{-1}\underline{\Delta})\underline{A}_1^{-1} . \tag{41}$$

The quantity $\langle (S_n^x)^2-(S_n^y)^2\rangle_{\mathbf{k}_\parallel}$ provides a measure of the ellipticity of the surface spin-wave precession and reduces to zero in the absence of the F anisotropy.

Some numerical examples of these thermally averaged quantities $\langle (S_n^x)^2+(S_n^y)^2\rangle_{\mathbf{k}_\parallel}$ and $\langle (S_n^x)^2-(S_n^y)^2\rangle_{\mathbf{k}_\parallel}$ are plotted in Fig. 3 as functions of the layer index n, for particular values of $\mathbf{k}_\parallel$ along [100], and for some exchange and anisotropy parameters selected to correspond to the dispersion curves of Fig. 1. It is seen that these amplitudes decay more rapidly into the sample when they correspond to surface spin-wave frequencies located away from the bulk continuum (see the solid circles in Fig. 3) than when they correspond to surface spin-wave frequencies close to the bulk continuum (see open circles in Fig. 3). This is consistent with the attenuation length of Eq. (32). The lines connecting the points in Fig. 3 are just guides to the eye and do not have any physical significance.

V. DISCUSSION AND CONCLUSIONS

We have obtained the dispersion relations for the spin waves in a semi-infinite Heisenberg ferromagnet with nearest-neighbor exchange interaction and with nonuniaxial single-ion anisotropy. This description is confined to low temperatures $T \ll T_c$; otherwise the decoupling schemes used in the equations of motion for the Green functions will no longer be valid. Although we have carried out the calculations explicitly for the case where the

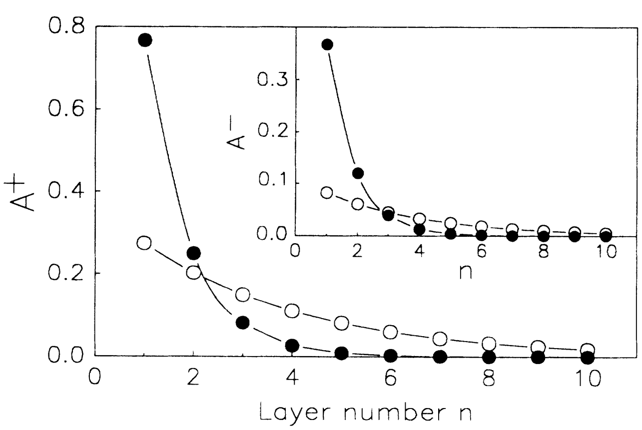

FIG. 3. Mean-squared amplitudes $A^+=\langle (S_n^x)^2+(S_n^y)^2\rangle_{\mathbf{k}_\parallel}$ and $A^-=\langle (S_n^x)^2-(S_n^y)^2\rangle_{\mathbf{k}_\parallel}$ (see inset) plotted against the layer index n for the exchange and anisotropy parameters of Fig. 1 and for different $\mathbf{k}_\parallel$ along [100]. The temperature is chosen such that $k_BT/SJ=0.1$. The open circles correspond to $\mathbf{k}_\parallel=\pi/4a$, $D_S/D=0.5$, and $J_S/J=2.0$; the solid circles correspond to $\mathbf{k}_\parallel=0$, $D_S/D=0.5$, and $J_S/J=0.5$.

magnetic ions form a simple-cubic lattice, the results are easily generalized to other structures (and to other orientations of the surface relative to the crystal axes). In some cases this would just involve redefining the exchange sums $u_n(\mathbf{k}_\parallel)$ and $v_n(\mathbf{k}_\parallel)$ and the structure factor $\gamma(\mathbf{k}_\parallel)$ in Eqs. (9)–(11).

We have also obtained explicit results for the spin-correlation functions $\langle S_n^-(t)S_n^+(t)\rangle_{\mathbf{k}_\parallel}$ and $\langle S_n^+(t)S_n^+(t)\rangle_{\mathbf{k}_\parallel}$ of the surface spin waves and these are important in determining the dynamic response of the system in various experiments (e.g., light scattering or spin-wave resonance) involving materials with nonuniaxial single-ion anisotropy.

As mentioned before, the results in this paper can also be generalized to include nonuniaxial anisotropic exchange, such as represented by an antisymmetric combination of the form

$$J_{ij}^D \mathbf{S}_i \times \mathbf{S}_j \tag{42}$$

in the spin Hamiltonian. Here J_{ij}^D is the Dzialoshinski-Moriya exchange interaction,[13,14] which is known to be important in some materials (e.g., $FeBO_3$, $CoCO_3$, and several rare-earth orthoferrites). The full Hamiltonian would still be expressible in boson operators in the same form as Eq. (3), but with modified coefficients. The matrix formalism of Sec. II could then be used to solve for the surface Green functions.

The matrix method would not, however, be appropriate for treating magnetic dipole-dipole anisotropy, which is nonuniaxial. This is because the dipole-dipole interactions are long range and would not lead to a tridiagonal matrix as in Sec. II. By contrast, surface spin waves in magnetic systems with dipole-dipole interactions can be conveniently treated by macroscopic methods involving Maxwell's equations (e.g., see Ref. 4 for a review).

In certain materials it is believed that the anisotropy may be nonuniaxial only at the surface, i.e., the case of $F=0$ but $F_S\neq0$ in our notation. This can occur because the surface lowers the symmetry and allows additional terms to occur in the Hamiltonian (e.g., see Refs. 15 and 16 for discussion). An example is provided by single-crystal {110} Fe films (e.g., see Refs. 17 and 18). When $F=0$ in the theory of Sec. II, it is easily shown that Eq. (29) for the surface spin-wave parameter y becomes replaced by

$$\{[4F_S^2\eta/v_B^2(\mathbf{k}_\parallel)]-\delta^2\}(y^2+y)-(2d\delta+1)y-1=0\ . \tag{43}$$

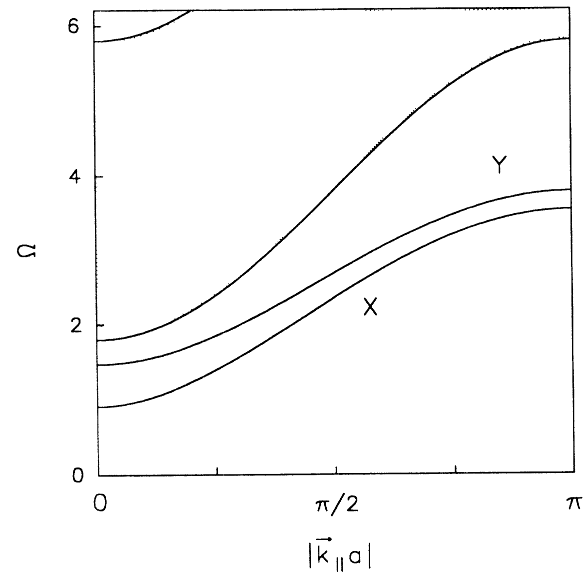

FIG. 4. The spin-wave frequencies (in units of SJ) plotted against $|\mathbf{k}_\parallel a|$ for the case of nonuniaxial anisotropy in the surface layer only ($F_S\neq0$, $F=0$) of a $S=1$ Heisenberg ferromagnet. Two surface spin-wave branches are shown (labeled X and Y), corresponding to $F_S/J=1.0$ and 0.1, respectively. The propagation wave vector $\mathbf{k}_\parallel$ is along the [100] direction and the other parameters are $g\mu_B H_0/SJ=0.3$, $D/J=1.5$, $D_S/D=0.5$, and $J_S/J=0.5$.

This quadratic equation for y can be solved, and the dispersion relations of the surface spin waves may then be found as before. Some numerical results to illustrate the predicted behavior for different values of F_S are shown in Fig. 4. Most of the experimental studies of nonuniaxial surface anisotropy to date have been for transition metals (see Refs. 17–19). It would be of considerable interest to have further experimental studies carried out for magnetic insulator or magnetic semiconductor materials that would be better described by the Heisenberg model employed in this paper. Suitable techniques for studying the excitations could include inelastic light scattering (Brillouin and Raman spectroscopy) and magnetic resonance (ferromagnetic resonance, spin-wave resonance, etc.). These methods have already been applied to investigate surface and bulk spin waves in various magnetic materials (see Refs. 2 and 3).

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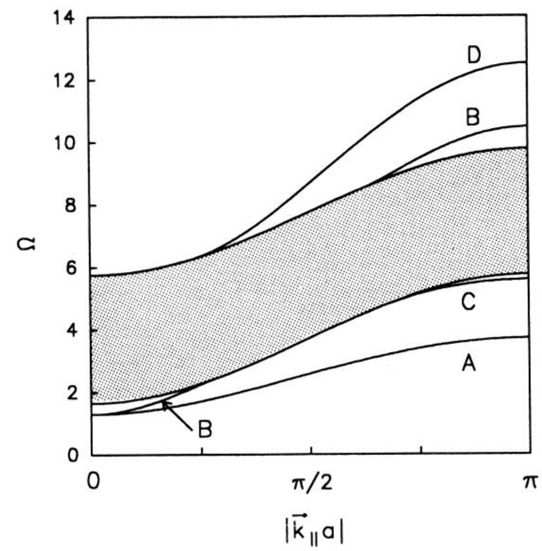

FIG. 1. A plot of the spin-wave frequencies (in units of SJ) against $|\mathbf{k}_{\parallel}a|$ in a semi-infinite $S=1$ Heisenberg ferromagnet with nonuniaxial anisotropy and for propagation wave vector $\mathbf{k}_{\parallel}$ along [100]. The applied magnetic field is such that $g\mu_B H_0/SJ=0.3$ and the anisotropy parameters are chosen to be $D/J=1.5$, $F/J=0.5$, and $F_S/F=1$. The bulk spin-wave continuum is shown together with surface spin-wave branches corresponding to A, $D_S/D=0.5$, $J_S/J=0.5$; B, $D_S/D=0.5$, $J_S/J=2.0$; C, $D_S/D=1.5$, $J_S/J=0.5$; D, $D_S/D=1.5$, $J_S/J=2.0$.

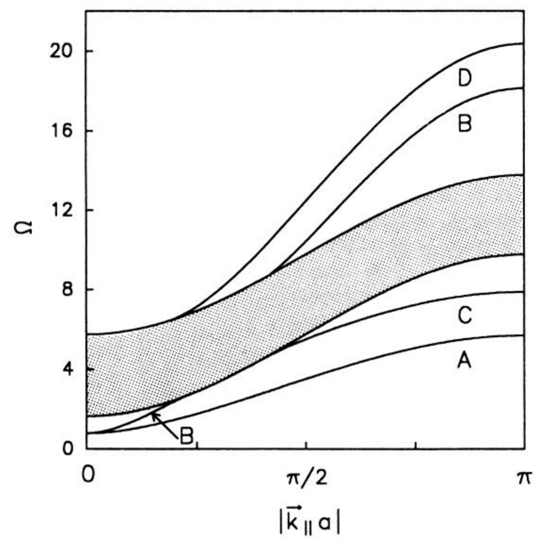

FIG. 2. The spin-wave frequencies (in units of SJ) plotted against $|\mathbf{k}_{\parallel} a|$ for $F_S/F=2$ and for propagation wave vector $\mathbf{k}_{\parallel}$ along the [110] direction. The other exchange and anisotropy parameters are as for Fig. 1.

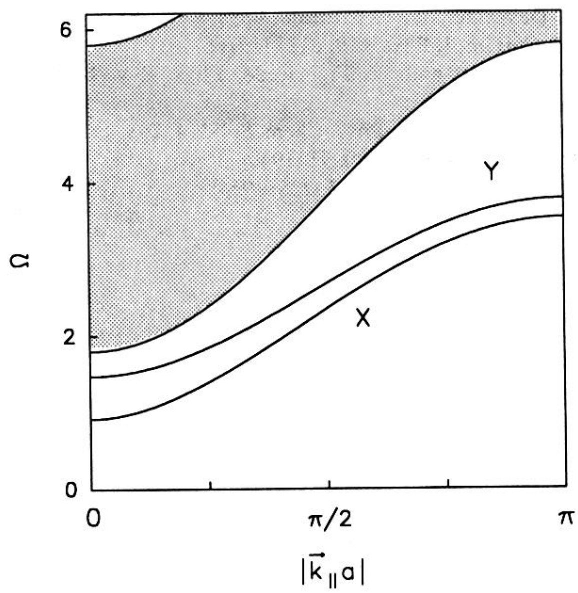

FIG. 4. The spin-wave frequencies (in units of SJ) plotted against $|\mathbf{k}_{\|}a|$ for the case of nonuniaxial anisotropy in the surface layer only ($F_S \neq 0$, $F=0$) of a $S=1$ Heisenberg ferromagnet. Two surface spin-wave branches are shown (labeled X and Y), corresponding to $F_S/J=1.0$ and 0.1, respectively. The propagation wave vector $\mathbf{k}_{\|}$ is along the [100] direction and the other parameters are $g\mu_B H_0/SJ=0.3$, $D/J=1.5$, $D_S/D=0.5$, and $J_S/J=0.5$.