Surface-spin-wave bound states in the Heisenberg ferromagnet*

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It is established that there exist one s-wave and one d-wave bound state of a pair of surface spin waves below the two-surface-spin-wave band in a semi-infinite simple-cubic Heisenberg ferromagnet with uniaxial exchange anisotropy, by using the Rayleigh-Ritz variational method. The qualitative spectra of the surface-spin-wave bound states are also obtained. I find that, for small exchange anisotropy, the surface-spin-wave bound states exist for the total wave vector $\vec{K}_\perp$ larger than some threshold value. For large exchange anisotropy, however, the surface-spin-wave bound states may exist for each value of the total $\vec{K}_\perp$. The prospect of observing the surface spin waves and their bound states is discussed.

I. INTRODUCTION

In recent years there has been increasing interest in surface effects on the properties of solids, as well as the catalytic processes on solid surfaces. Surface spin waves are one of the subjects which has attracted considerable attention, because the existence of these states can lead not only to strong effects on the low-temperature properties of magnetic materials,[1] especially those with large surface-to-volume ratio, but can also affect the catalytic activity on the magnetic surfaces.[2] Surface spin waves (SSW) in Heisenberg magnets are characterized by exponentially decaying amplitude with increasing depth into the crystal; they are Bloch waves, characterized by a two-dimensional wave vector $\vec{k}_\perp = (k_x, k_y)$ along the two directions parallel to the surface. The surface-spin-wave energies are usually lower than those of bulk spin waves (BSW) with the same $\vec{k}_\perp$.

It has been shown by Dyson[3] that two bulk spin waves in the Heisenberg ferromagnet interact via an attractive potential. The two-bulk-spin-wave bound states and resonances have subsequently been investigated by many authors.[4–8] Wortis[4] has discussed the bulk-spin-wave bound states in detail. He found that, for total wave vector $\vec{K}_\perp$ larger than a threshold, there are d bulk-spin-wave bound states below the two-bulk-spin-wave band in a d-dimensional hypercubic lattice. He also incorporated uniaxial exchange anisotropy in the Hamiltonian and studied its effect on the bulk-spin-wave bound-state spectra. He found that the bulk-spin-wave bound states may exist over the entire Brillouin zone for large enough anisotropy. Later, Fukuda and Wortis[5] reformulated the problem by focusing on construction of the wave function, based directly on the Schrödinger equation. They found the bulk-spin-wave bound-state function is sharply localized at nearest-neighbor separation at the zone boundary, but is quite spread out at the threshold.

The purpose of this paper is to show that two ferromagnetic *surface* spin waves, if they exist, form bound states below the two-surface-spin-wave band. For this reason, I consider a semi-infinite Heisenberg ferromagnet with uniform uniaxial exchange anisotropy. I show that the interaction between two surface spin waves is attractive. I follow the spirit of Fukuda and Wortis's formulation[5] to construct the Schrödinger equation for the two-spin-deviation states. By using the Rayleigh-Ritz variational method,[9] I establish the existence of one s-wave and one d-wave bound state of two surface spin waves. By choosing appropriate forms for the trial wave functions for the surface-spin-wave bound states, I am able to obtain the qualitative features of their spectra. I find that, for small exchange anisotropy (near the Heisenberg limit), surface-spin-wave bound states exist for the total wave vector $\vec{K}_\perp$ larger than some threshold value. For large exchange anisotropy (near Ising limit), however, the surface-spin-wave bound states may exist below the surface-spin-wave band for each value of the total wave vector $\vec{K}_\perp$.

In Sec. II I set up the Schrödinger equation for two-spin-deviation states. In Sec. III I establish the existence of surface-spin-wave bound states and obtain their qualitative spectra. In Sec. IV I discuss the results and the prospects of observing the surface spin waves and their bound states.

II. SCHRÖDINGER EQUATION

I consider a semi-infinite simple-cubic Heisenberg ferromagnet with free (100) surface defined as the $z=0$ plane. The crystal is assumed to occupy the half-space $z \geq 0$. The system is described by the Hamiltonian

$$\mathcal{H} = -\sum_{i,j} J(i,j)[\sigma(S_i^x S_j^x + S_i^y S_j^y) + S_i^z S_j^z], \qquad (1)$$

where

$$J(i,j)=J>0 \text{ if } i,j \text{ are nearest neighbors and both are in the crystal,}$$
$$=0 \quad \text{otherwise;} \tag{1a}$$

σ is the parameter characterizing the exchange anisotropy, $1>\sigma>0$. The Heisenberg limit is $\sigma=1$, and the Ising limit is $\sigma=0$. The two-spin-deviation state may be written in general as

$$|\psi\rangle \equiv \frac{1}{2S}\sum_{i,i'} \Phi(i,i')S_i^- S_{i'}^- |g\rangle \ , \tag{2}$$

where S is the magnitude of each spin; the summation is over all lattice sites i and i', $\Phi(i,i')$ is the probability amplitude for creating two spin deviations at sites i and i'; S^- is the spin-lowering operator; and $|g\rangle$ is the ferromagnetic ground state. Note that

$$\Phi(i,i')=\Phi(i',i) \ . \tag{3}$$

Furthermore, $\Phi(i,i)$ is undefined for $S=\frac{1}{2}$, since two spin deviations are prevented from propagating onto the same site. Using the spin commutation relations and the identity

$$\langle g|S_j^+S_{j'}^+S_i^-S_{i'}^-|g\rangle = 4S^2(1-\delta_{ii'}/2S)(\delta_{ij}\delta_{i'j'}+\delta_{ij'}\delta_{i'j}) \tag{4}$$

the Schrödinger equation

$$\mathcal{H}|\psi\rangle = E|\psi\rangle \tag{5}$$

can be written in the following form:

$$[2JS(12-\delta_{z,0}-\delta_{z',0})-E]\phi(i,i') - 2JS\sigma\sum_{\delta}[\Theta(z+\delta_z)\phi(i+\delta,i')+\Theta(z'+\delta_z)\phi(i,i'+\delta)] - 2J\delta_{i,i'+\delta}\{\phi(i,i')-\tfrac{1}{2}\sigma[\phi(i,i)+\phi(i',i')]\}=0 \ , \tag{6}$$

where $\Theta(X)$ is the unit step function

$$\Theta(X)=0 \text{ if } X<0 \ ,$$
$$=1 \text{ if } X\geq 0 \ , \tag{7}$$

and

$$\phi(i,i')=[1-(1/2S)\delta_{ii'}]\Phi(i,i') \ . \tag{8}$$

Note that for $S=\frac{1}{2}$

$$\phi(i,i)=0 \ . \tag{9}$$

Equation (6) looks very similar to Eq. (7) in Ref. 5, which deals with the case of an infinite ferromagnet. The only difference is that in Eq. (6) of this paper there are additional terms $\delta_{z,0}$ and $\delta_{z',0}$ and additional factors $\Theta(z+\delta_z)$ and $\Theta(z'+\delta_z)$ due to the surface boundary condition contained in the Hamiltonian in Eq. (1). The last term in Eq. (6) is the manifestation of the attractive interaction between two spin waves (including surface and bulk spin waves). In the absence of this term, the equation would be just for two noninteracting spin waves.

Now we use Fourier transformation on $\phi(i,i')$ since the crystal is infinite in the x and y directions:

$$\phi(i,i')=\sum_{K_x,K_y}\exp\{\tfrac{1}{2}i[K_x(x+x')+K_y(y+y')]\} \times\phi_K(\bar{x},\bar{y},z,z') \ , \tag{10}$$

where K_x and K_y are the x and y components of the total wave vector $\vec{K}_\parallel$ of the pair of spin waves, and

$$\bar{x}=x-x' \ , \quad \bar{y}=y-y' \ . \tag{11}$$

Equation (6) can then be written as

$$(\tilde{\mathcal{H}}_0+\tilde{V})\phi_K(\bar{x},\bar{y},z,z')=E\phi_K(\bar{x},\bar{y},z,z') \ , \tag{12}$$

where

$$\begin{aligned}\tilde{\mathcal{H}}_0\phi_K(\bar{x},\bar{y},z,z') = {} & 2JS(12-\delta_{z,0}-\delta_{z',0})\phi_K(\bar{x},\bar{y},z,z') \\ & -4JS\sigma\{\alpha_x[\phi_K(\bar{x}+a,\bar{y},z,z')+\phi_K(\bar{x}-a,\bar{y},z,z')]+\alpha_y[\phi_K(\bar{x},\bar{y}+a,z,z')+\phi_K(\bar{x},\bar{y}-a,z,z')]\} \\ & -2JS\sigma\{(1-\delta_{z,0})\phi_K(\bar{x},\bar{y},z-a,z')+\phi_K(\bar{x},\bar{y},z+a,z')+(1-\delta_{z',0})\phi_K(\bar{x},\bar{y},z,z'-a) \\ & +\phi_K(\bar{x},\bar{y},z,z'+a)\}\end{aligned} \tag{12a}$$

and

$$\tilde{V}\phi_K(\bar{x},\bar{y},z,z')=-2J\delta_{\bar{x},\delta_x}\delta_{\bar{y},\delta_y}\delta_{z-z',\delta_z}\{\phi_K(\bar{x},\bar{y},z,z') - \sigma\cos[(\tfrac{1}{2}\vec{K}_\parallel)\cdot\vec{\delta}]\phi_K(0,0,z,z)\} \ , \tag{12b}$$

where

$$\alpha_\mu=\cos(\tfrac{1}{2}K_\mu a) \ , \quad \mu=x,y \tag{13}$$

and a is the lattice constant. The expression $\tilde{\mathcal{H}}_0\phi_K$ describes a pair of noninteracting spin waves with total wave vector (K_x,K_y), and $\tilde{V}\phi_K$ describes the attraction between the pair. This interaction term vanishes except when the two spin deviations are at adjacent sites. From Eq. (12a) we see that the excitation energy for two noninteracting surface spin waves is

$$E=4JS\left(5-\sigma^2-2\sum_{\mu=x,y}\alpha_\mu\cos\kappa_\mu a\right) \ , \tag{14}$$

and the corresponding wave function is

$$\phi_K(\bar{x},\bar{y},z,z')=e^{i\kappa_x\bar{x}+i\kappa_y\bar{y}}e^{-q(z+z')} \ , \tag{15}$$

where

$$e^{-qa} = \sigma \; ; \tag{16}$$

κ_x, κ_y are the x and y components of half the relative wave vector of the two surface spin waves. The band of two noninteracting surface spin waves is shown by the hatched area in Fig. 1. Since the interaction between two spin waves is present, we should solve Eqs. (12) for given values of α_x, α_y, and σ. For $\alpha_x = \alpha_y = 0$ (the zone boundary), I find that the binding energy of the SSW bound state is equal to $2J$. That is, its excitation energy is

$$E = 4JS(5-\sigma^2) - 2J \, . \tag{17}$$

Furthermore, its wave function is

$$\phi(\bar{x},\bar{y},z,z') = e^{-q(z+z')}\delta_{\bar{x},\delta_x}\delta_{\bar{y},\delta_y}\delta_{z-z',\delta_z} \, , \tag{18}$$

where q satisfies Eq. (16). For general values of α_x and α_y, I use the Rayleigh-Ritz variational method[9] to establish the existence of the SSW bound states and obtain their qualitative spectra in Sec. III.

III. SURFACE-SPIN-WAVE BOUND STATES: EXISTENCE AND SPECTRA

In this section I choose appropriate forms of the trial wave functions for a bound pair of surface spin waves. I then calculate the expectation value, $\langle \mathcal{H} \rangle$, of the Hamiltonian, Eq. (1), in such trial states. By varying the parameters for the trial functions, I obtain the absolute minimum value of $\langle \mathcal{H} \rangle$. Based on the Rayleigh-Ritz variational principle,[10] I thus establish the existence of the SSW bound states. In Appendix A I show that the amplitudes of the two bound states of a pair of bulk spin waves in an infinite square lattice possess s and d symmetry, respectively. These amplitudes decay with increasing separation of the two spin deviations. In Appendix B, it is shown that the amplitude of the SSW bound state in a semi-infinite chain decays exponentially with increasing separation and also with their individual distances from the surface of the two spin deviations.

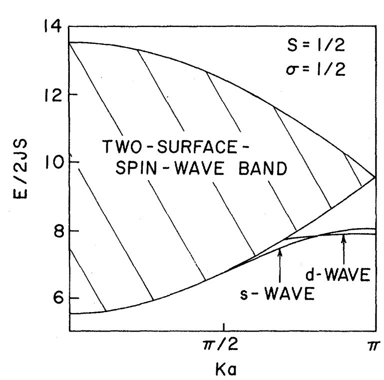

FIG. 1. Two-surface-spin-wave spectra for simple-cubic lattic for the case of $S = \frac{1}{2}$, $\sigma = \frac{1}{2}$ (small exchange anisotropy), and $K_x = K_y = K$. The hatched area shows the two-surface-spin-wave band. The two lines below the band show the energy spectra of the s-wave and d-wave surface-spin-wave bound states, respectively.

Since the crystal considered is infinite in the x and y directions and semi-infinite in the z direction, I choose the following two forms of the trial functions for the SSW bound states. For the s-wave state

$$\phi_K = \varphi_s(\bar{x},\bar{y},z,z')$$

$$= p(S) \quad \text{if } \bar{x} = \bar{y} = z - z' = 0 \, , \tag{19a}$$

$$= \exp[-\tfrac{1}{2}m(z+z') - \tfrac{1}{2}n|z-z'| - \beta|\bar{x}| - \beta|\bar{y}|] \quad \text{otherwise,} \tag{19b}$$

where

$$p(S) = 0 \quad \text{only when } S = \tfrac{1}{2} \, . \tag{19c}$$

For the d-wave states,

$$\phi_K = \varphi_d(\bar{x},\bar{y},z,z')$$

$$= 0 \quad \text{if } \bar{x} = \bar{y} = z - z' = 0 \, , \tag{20a}$$

$$= \exp[-\tfrac{1}{2}m(z+z') - \tfrac{1}{2}n|z-z'|][\exp(-\beta|\bar{x}| - \gamma|\bar{y}|) - \exp(-\gamma|\bar{x}| - \beta|\bar{y}|)] \quad \text{otherwise.} \tag{20b}$$

The parameters m, n, α, β are all positive. Note that

$$\varphi_s(\bar{x},\bar{y},z,z') = \varphi_s(\bar{y},\bar{x},z,z') \tag{21a}$$

and

$$\varphi_d(\bar{x},\bar{y},z,z') = -\varphi_d(\bar{y},\bar{x},z,z') \, . \tag{21b}$$

It is easily shown that these trial states are orthogonal to the ferromagnetic ground state. The expectation values of $\mathcal{H}$ in the trial states with the amplitudes in Eqs. (19) and (20) are denoted $\langle E \rangle_s$ and $\langle E \rangle_d$, respectively. I evaluate $\langle E \rangle_s$ and $\langle E \rangle_d$ in Appendix C. Here I present the results for $\alpha_x = \alpha_y = \alpha$. For the case of $S = \frac{1}{2}$, the expectation value of $\mathcal{H}$ in the s state is

$$\langle E \rangle_s = 24JS(1-\sigma\alpha B) + 8JS\sigma\alpha B^3(1+NM)/W - 4J(B^2+NM)(1-NM)(1-B^2)^2/W - 4JS(1+M)[1 - M + 2\sigma(NM)^{1/2}](1-NM/W) \, , \tag{22}$$

where

$$W = 1 + NM - (1-NM)(1-B^2)^2 \, . \tag{23}$$

The generalization to the cases of $S > \frac{1}{2}$ is straightforward, merely including one additional variational parameter, $p(S)$. The expectation value of $\mathcal{H}$ in the d state is

$$\langle E \rangle_d = 24JS - 4JS\sigma\alpha(B+C)(1+BC)(2-BC) - 2J(1+M)[1 - M + 4S\sigma(NM)^{1/2}]/(1+NM) - 4JS(1-NM)(1-B^2)(1-C^2)(1-BC)^2/(1+NM) \, . \tag{24}$$

In Eqs. (22)–(24),

$$N=e^{-na}\ ,\quad M=e^{-ma}\ ,$$
$$B=e^{-\beta a}\ ,\quad C=e^{-\gamma a}\ . \tag{25}$$

Since m, n, β, γ are positive, Eqs. (25) lead to the condition

$$1>N,M,B,C>0\ . \tag{26}$$

Now the task is to look for the absolute minima of $\langle E\rangle_s$ and $\langle E\rangle_d$ for B, C, M, N satisfying Eq. (26). The following conditions are necessary, although not sufficient, for $\langle E\rangle_s$ and $\langle E\rangle_d$ to take on absolute minimum values:

$$\frac{d\langle E\rangle_\mu}{dB}=0\ , \tag{27a}$$

$$\frac{d\langle E\rangle_\mu}{dM}=0\ , \tag{27b}$$

$$\frac{d\langle E\rangle_\mu}{dN}=0\ \text{ for } \mu=s,d\ , \tag{27c}$$

$$\frac{d\langle E\rangle_d}{dC}=0\ . \tag{27d}$$

Equations (27b) and (27c) lead to the following relations among the parameters for $S=\frac{1}{2}$:

$$\sigma^2 N=M \tag{28a}$$

for both s and d states, and

$$(1+M)(M-\sigma^2)+2M(1-B^2)(1-C^2)(1-BC)^2=0 \tag{28b}$$

for the d state. It is easily shown that

$$\frac{d^2\langle E\rangle}{dM^2}>0\ , \tag{29a}$$

$$\frac{d^2\langle E\rangle}{dN^2}>0 \tag{29b}$$

for N and M satisfying Eqs. (28) for fixed values of B and C. By substituting Eq. (28a) into Eq. (22) I obtain an expression for $\langle E\rangle_s$ in terms of M and B:

$$\begin{aligned}\langle E\rangle_s &= 12J(1-\sigma\alpha B)+4J\alpha B^3\sigma(\sigma^2+M^2)/W_1\\ &\quad -4J(B^2+M^2/\sigma^2)(\sigma^2-M^2)(1-B^2)^2/W_1\\ &\quad -2J(1+M^2)[\sigma^2-(\sigma^2-M^2)(1-B^2)^2]/W_1\ ,\end{aligned} \tag{30}$$

where

$$W_1=\sigma^2+M^2-(\sigma^2-M^2)(1-B^2)^2\ . \tag{31}$$

By performing numerical calculations of the values of $\langle E\rangle_s$ in Eq. (30) for the values of M and B in Eq. (26), I obtain the absolute minimum value of $\langle E\rangle_s$ for given values of σ and α for the case of $S=\frac{1}{2}$. The results are shown in Figs. 1 and 2. As for the d state, Eq. (28b) leads to

$$M=-\tfrac{1}{2}(1+2k-\sigma^2)+\tfrac{1}{2}[(1+2k-\sigma^2)^2+4\sigma^2]^{1/2}\ , \tag{32a}$$

where

$$k=(1-B^2)(1-C^2)(1-BC)^2\ . \tag{32b}$$

By substituting Eqs. (28a) and (32) into Eq. (24), and again using numerical methods, I calculate the values of $\langle E\rangle_d$ for the values of B and C in Eq. (26). I thus also determine the absolute minimum value of $\langle E\rangle_d$ for given values of σ and α. The results are also shown in Figs. 1 and 2 for $S=\frac{1}{2}$. Note that for $\sigma=\frac{1}{2}$, both $\langle E\rangle_{d,\min}$ and $\langle E\rangle_{s,\min}$ lie below the bottom of the SSW band for part of the Brillouin zone not including the zone center. For small values of σ, say, $\sigma=0.1$, however, bound states exist for each value of $\vec{K}_\perp$. Furthermore, at the zone boundary where $\alpha=0$, the minimum values of both $\langle E\rangle_s$ and $\langle E\rangle_d$ are slightly larger than the exact energy for exciting the SSW bound state, Eq. (17). For $\alpha=0$,

$$\langle E\rangle_{s,\min}\sim 8J\ ,\quad \langle E\rangle_{d,\min}\sim 7.82J\ ,$$

while

$$E_{\text{exact}}=7.5J\ \text{ for } \sigma=\tfrac{1}{2}\ ;$$
$$\langle E\rangle_{s,\min}\sim 8J\ ,\quad \langle E\rangle_{d,\min}\sim 7.99J\ , \tag{33}$$

while

$$E_{\text{exact}}=7.98J\ \text{ for } \sigma=0.1\ . \tag{34}$$

This indicates that the variational method with the reasonable trial functions gives rise to bound-state energies quite close to exact values.

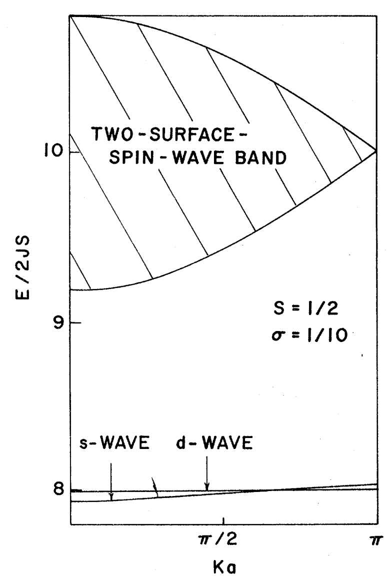

FIG. 2. Two-surface-spin-wave spectra for the case of $S=\frac{1}{2}$, $\sigma=\frac{1}{10}$ (large exchange anisotropy), and $K_x=K_y=K$. Note that both the s-wave and d-wave surface-spin-wave bound states lie below the two-surface-spin-wave band for each value of the total wave vector $\vec{K}_\perp$.

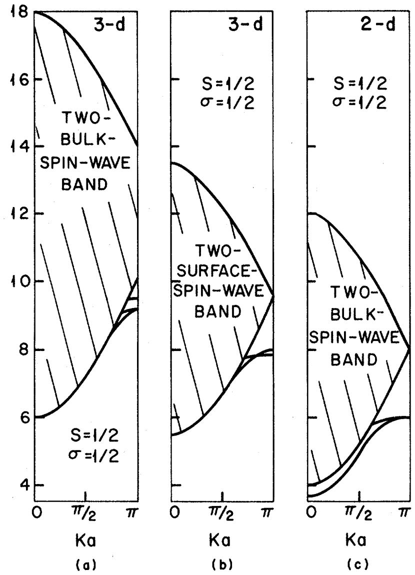

FIG. 3. (a) Two-bulk-spin-wave spectra for simple-cubic lattice for the case of $S=\frac{1}{2}$, $\sigma=\frac{1}{2}$, and the total wave vector $\vec{K}=K(1,1,0)$. (b) Same as Fig. 1. (c) Two-bulk-spin-wave spectra for two-dimensional square lattice for the case of $S=\frac{1}{2}$, $\sigma=\frac{1}{2}$, and the total wave vector $\vec{K}=K(1,1)$.

As shown in Fig. 3, the present results for the two-surface-spin-wave spectra [see Fig. 1 or 3(b)] are compared with the two-bulk-spin-wave spectra obtained by Wortis[4] for (i) simple-cubic lattice with the total wave vector $\vec{K}=K(1,1,0)$ [see Fig. 3(a)], and (ii) two-dimensional square lattice with the total wave vector $\vec{K}=K(1,1)$ [see Fig. 3(c)]. For the same values of S, σ, and K, the spectra in Fig. 3(b) lie between those in Figs. 3(a) and 3(c). This is reasonable because it takes more energy to propagate two spin deviations infinitely far than a finite distance into the interior of the crystal. Furthermore, it takes the least energy to propagate two spin deviations only in the surface plane.

IV. CONCLUSION

A semi-infinite Heisenberg ferromagnet with uniform uniaxial exchange anisotropy is considered. The interaction between two surface spin waves is shown to be attractive. By using the Rayleigh-Ritz variational method, the existence of one s-wave and one d-wave bound state of a pair of surface spin waves is established. I have chosen appropriate forms for the trial functions of a bound pair of surface spin waves in order to obtain the qualitative spectra of the SSW bound states. It is found that, for small exchange anisotropy, the SSW bound states exist for the total wave vector $\vec{K}_\perp$ larger than some threshold value. For sufficiently large exchange anisotropy, however, the SSW bound states may exist below the band for each value of the total wave vector $\vec{K}_\perp$. The generalization to the cases of $S>\frac{1}{2}$ and more realistic surface boundary conditions is straightforward. The SSW bound states may also exist in the general cases as long as the interaction between surface spin waves remains attractive.

Mills[11] has discussed the possibility of observing surface spin waves in the Heisenberg ferromagnet by using the inelastic low-energy electron scattering. The best experimental resolution in the electron beam energy achieved so far is about 50 meV.[12] As soon as the resolution is improved to several meV or less, the inelastic low-energy electron scattering will be a promising probe for surface spin waves. For samples with large surface-to-volume ratio, the chance of observing surface spin waves may be good. Indeed, Wigen's group[13] and Tittman[14] recently observed surface spin waves in YIG films by using microwave resonance at finite applied field. Wigen and co-workers[13] also varied the thickness of the film and found that the intensity of the surface-spin-wave resonance relative to that of the bulk spin wave is stronger for thinner film. Besides low-energy electron scattering and microwave resonance, far-infrared absorption and surface phonons may also be possible probes for surface spin waves. The penetration depth of surface phonons which have been successfully generated is of the order of microns[15] (approximately a few thousand atomic layers). In the materials with strong spin-lattice coupling the surface phonon is probably a sensitive probe for the surface spin waves with comparable penetration depth.

Bulk spin wave and the BSW bound states have been observed in $CoCl_2 \cdot 2H_2O$ by Torrance and Tinkham[16] and Nicoli and Tinkham[17] using far-infrared absorption. As analyzed by Torrance and Tinkham,[16] $CoCl_2 \cdot 2H_2O$ can be approximated by an Ising ferromagnetic chain when the applied field is over 45 kG. The absorption energy versus applied field spectrum is in the form of a fan of lines originating from $E\simeq 2J_0^{zz}$, where J_0^{zz} is the intrachain coupling. The slopes of the lines are approximately $ng\mu_B$ ($n=1,2,\ldots$, etc.). These lines correspond to bulk spin waves and the multiple bulk-spin-wave bound states in the Ising chain. Now, consider a finite chain of Ising ferromagnets. The energy versus applied field spectrum of surface spin waves and the multiple surface-spin-wave bound states also have a "fan"-like structure originating from $E\simeq J_0^{zz}$. The relative absorption intensity associated with surface spin waves as compared with that of the bulk spin waves is estimated[18] to be of the order of $1/L$, where L is the number of atoms on each chain. As reported by Nicoli and Tinkham,[17] the absorption intensity can be measured to an accuracy of 10^{-3}. Therefore, it is hopeful to observe surface spin waves and the SSW bound states in very small samples of $CoCl_2 \cdot 2H_2O$ using far-infrared absorption techniques.

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APPENDIX A

In this appendix it is shown that the amplitudes of the two bound states of a pair of ferromagnetic bulk spin waves in an infinite square lattice possess s and d symmetry, respectively. These amplitudes decay with increasing separation of the two spin deviations. The equation satisfied by the amplitudes of the two-spin-deviation states, $\phi_K(\vec{R})$, in a Heisenberg ferromagnet is of the form

$$(\tilde{\mathcal{H}}_0+\tilde{V})\phi_K(\vec{R})=E\phi_K(\vec{R}) , \tag{A1}$$

where

$$\tilde{\mathcal{H}}_0\phi_K(\vec{R})=4JS\left(z\phi_K(\vec{R})-\sigma\sum_{\vec{\delta}}\cos(\tfrac{1}{2}\vec{K}\cdot\vec{\delta})\phi_K(\vec{R}+\vec{\delta})\right) \tag{A2}$$

and

$$\tilde{V}\phi_K(\vec{R})=-2J\delta_{\vec{R},\vec{\delta}}[\phi_K(\vec{\delta})-\sigma\cos(\tfrac{1}{2}\vec{K}\cdot\vec{\delta})\phi_K(\vec{0})] . \tag{A3}$$

By the Green's-function method, the solutions $\phi_K(\vec{R})$ are linear combinations of the Green's functions $G_K(\vec{R})$. That is

$$\phi_{\vec{K}}(\vec{R})=\sum_{\vec{R}'}A(\vec{R}')G_K(\vec{R}-\vec{R}') , \tag{A4}$$

where

$$G_K(\vec{R})=\frac{1}{N}\sum_{\vec{\rho}}\frac{e^{i\vec{\rho}\cdot\vec{R}}}{4JS[z-\sigma\sum_{\vec{\delta}}\cos(\frac{1}{2}\vec{K}\cdot\vec{\delta})\cos(\vec{\rho}\cdot\vec{\delta})]-E} \tag{A5}$$

and $A(\vec{R}')$ are the coefficients to be determined. Combining Eqs. (A3)–(A5), and making use of the inversion symmetry of the square lattice, we obtain a set of two equations for the two nonvanishing coefficients,

$$A_x=A(\vec{R}=(\pm a,0)) , \quad A_y=A(\vec{R}=(0,\pm a)) , \tag{A6}$$

where a is the lattice constant.

$$C_1A_x+C_2A_y=0 , \tag{A7a}$$

$$C_2A_x+C_1A_y=0 , \tag{A7b}$$

where

$$C_1=1+4J(\sigma\alpha D_1-D_{11}) \tag{A8a}$$

and

$$C_2=4J(\sigma\alpha D_1-D_{12}) ; \tag{A8b}$$

$$\alpha=\cos(\tfrac{1}{2}Ka) \tag{A9}$$

and K is the magnitude of each component of the total wave vector $\vec{K}$. Then

$$D_1=\frac{1}{N}\sum_{\rho_x,\rho_y}\frac{\cos(\rho_x a)}{8JS[2-\alpha\sigma(\cos\rho_x a+\cos\rho_y a)]-E} , \tag{A10}$$

$$D_{11}=\frac{1}{N}\sum_{\rho_x,\rho_y}\frac{\cos^2(\rho_x a)}{8JS[2-\alpha\sigma(\cos\rho_x a+\cos\rho_y a)]-E} , \tag{A11}$$

and

$$D_{12}=\frac{1}{N}\sum_{\rho_x,\rho_y}\frac{\cos(\rho_x a)\cos(\rho_y a)}{8JS[2-\alpha\sigma(\cos\rho_x a+\cos\rho_y a)]-E} . \tag{A12}$$

Equations (A7) would have a nontrivial solution if

$$\det=C_1^2-C_2^2=0 . \tag{A13}$$

That is,

$$C_1+C_2=0 , \tag{A14a}$$

and hence

$$A_x=A_y . \tag{A14b}$$

Or,

$$C_1-C_2=0 , \tag{A15a}$$

and hence

$$A_x=-A_y . \tag{A15b}$$

Equation (A14a) is equivalent to

$$1-4J(D_{11}+D_{12}-2\alpha\sigma D_1)=0 . \tag{A16a}$$

The corresponding amplitude is

$$\phi_K(\vec{R})=\frac{2A}{N}\sum_{\rho_x,\rho_y}\frac{e^{i\vec{\rho}\cdot\vec{R}}(\cos\rho_x a+\cos\rho_y a)}{8JS[2-\alpha\sigma(\cos\rho_x a+\cos\rho_y a)]-E} , \tag{A16b}$$

where E satisfies Eq. (A16a). This is the s-wave amplitude. Equation (A14b) is equivalent to

$$1-4J(D_{11}-D_{12})=0 . \tag{A17a}$$

The corresponding amplitude is

$$\phi_K(\vec{R})=\frac{2A}{N}\sum_{\rho_x,\rho_y}\frac{e^{i\vec{\rho}\cdot\vec{R}}(\cos\rho_x a-\cos\rho_y a)}{8JS[2-\alpha\sigma(\cos\rho_x a+\cos\rho_y a)]-E} , \tag{A17b}$$

where E satisfies Eq. (A17a). Note that

$$\phi_K(x,y)=-\phi_K(y,x) . \tag{A18}$$

This is the d-wave amplitude

From the asymptotic form of the lattice Green's function of the square lattice for a large distance at fixed energy obtained by Katsura and Inawashiro,[19] we see that both amplitudes (A16b) and (A17b) decay as the separation increases.

APPENDIX B

The Schrödinger equation for the two-spin-deviation states in a semi-infinite one-dimensional ferromagnetic chain is of the form

$$\begin{aligned}&[2JS(4-\delta_{z,0}-\delta_{z',0})-E]\phi(z,z')\\&\quad-2J\sigma S[(1-\delta_{z,0})\phi(z-a,z')+\phi(z+a,z')\\&\quad+(1-\delta_{z',0})\phi(z,z'-a)+\phi(z,z'+a)]\\&\quad-2J\delta_{|z-z'|,a}\{\phi(z,z')-\tfrac{1}{2}\sigma[\phi(z,z)+\phi(z',z')]\}=0.\end{aligned} \quad (B1)$$

By solving Eq. (B1) exactly, I find that the wave function of the surface-spin-wave bound state is

$$\phi(z,z')=\exp[-q(z+z')-n|z-z'|], \quad (B2)$$

where

$$e^{qa}=(2-\sigma^2)^{1/2}/\sigma \quad (B3)$$

and

$$e^{na}=(2-\sigma^2)^{1/2}. \quad (B4)$$

The corresponding bound-state energy is

$$E=4JS(1-\sigma^2)/(2-\sigma^2). \quad (B5)$$

APPENDIX C

The expectation value of the Hamiltonian $\mathcal{H}$ is

$$\langle E\rangle_\mu=\sum_{|\bar{x}|,|\bar{y}|,z,z'}\varphi_\mu(\tilde{\mathcal{H}}_0+\tilde{V})\varphi_\mu\Big/\sum_{|\bar{x}|,|\bar{y}|,z,z'}(\varphi_\mu)^2, \quad (\mu=s,d). \quad (C1)$$

Using the trial functions in Eqs. (19), and through straightforward but very tedious algebra, we obtain

$$\begin{aligned}\sum_{|\bar{x}|,|\bar{y}|,z,z'}\varphi_s(\tilde{\mathcal{H}}_0+\tilde{V})\varphi_s&=\frac{24JSW}{K}-\frac{2J(B^2+NM)}{1-M^2}-8JS\alpha\sigma B\frac{3W-B^2(1+NM)}{K}\\&\quad-4JS[1-M+2\sigma(NM)^{1/2}](1+M)\frac{W-NM}{K}\end{aligned} \quad (C2)$$

and

$$\sum_{|\bar{x}|,|\bar{y}|,z,z'}(\varphi_s)^2=\frac{W}{K}, \quad (C3)$$

where

$$W=(1+NM)-(1-NM)(1-B^2)^2 \quad (C4)$$

and

$$K=(1-M^2)(1-B^2)^2(1-NM). \quad (C5)$$

Therefore,

$$\begin{aligned}\langle E\rangle_s&=24JS(1-\alpha\sigma B)-4J(B^2+NM)(1-NM)\frac{(1-B^2)^2}{W}\\&\quad+8JS\alpha\sigma B^3\frac{1+NM}{W}-4JS(1+M)[1-M\\&\quad+2\sigma(NM)^{1/2}]\frac{1-NM}{W}.\end{aligned} \quad (C6)$$

Using the trial functions in Eqs. (20) and through algebraic manipulations, we obtain

$$\begin{aligned}\sum_{|\bar{x}|,|\bar{y}|,z,z'}\varphi_d(\tilde{\mathcal{H}}_0+\tilde{V})\varphi_d&=\frac{24JSP}{Q}-8JS\frac{(B-C)^2}{(1-M^2)}\\&\quad-4J(1+M)[(1-M)+4\sigma S(NM)^{1/2}]\frac{(B-C)^2}{Q}\\&\quad-2JS(\alpha_x+\alpha_y)(B+C)(1+BC)(2-BC)\frac{P}{Q}\end{aligned} \quad (C7)$$

and

$$\sum_{|\bar{x}|,|\bar{y}|,z,z'}(\varphi_d)^2=\frac{P}{Q}, \quad (C8)$$

where

$$P=2(1+NM)(B-C)^2 \quad (C9a)$$

and

$$Q=(1-NM)(1-M^2)(1-B^2)(1-C^2)(1-BC)^2. \quad (C9b)$$

Hence

$$\begin{aligned}\langle E\rangle_d&=24JS-2JS(\alpha_x+\alpha_y)(B+C)(1+BC)(2-BC)\\&\quad-4JS(1-NM)(1-B^2)(1-C^2)\frac{(1-BC)^2}{1+NM}\\&\quad-2J(1+M)\frac{1-M+4\sigma S(NM)^{1/2}}{1+NM}.\end{aligned} \quad (C10)$$

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