

Temperature variation of surface-spin-wave frequencies in the Heisenberg ferromagnet: The exchange-dominated regime

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(Received 16 June 1983; revised manuscript received 30 November 1983)

We develop the theory of the temperature variation of surface-spin-wave frequencies in the semi-infinite Heisenberg ferromagnet, for modes with wavelength sufficiently short that exchange interactions provide the dominant contribution to the excitation energy. Magnon-magnon interactions are the source of the temperature variation of the surface-spin-wave energies, and our attention is directed toward the leading contribution in the limit of low temperatures. We find a $T^{5/2}$ correction to the $T=0$ spin-wave frequencies, so the leading term exhibits the same temperature variation as found for bulk spin waves.

I. INTRODUCTION

The influence of a surface on the properties of a semi-infinite ferromagnet has been the topic of numerous theoretical studies.¹ At temperatures T that are low compared to the Curie temperature T_c , spin-wave theory may be employed in such analyses, and one encounters surface spin waves,² which may be viewed as a magnetic analogue of the Rayleigh surface waves encountered in surface-lattice dynamics.

Surface spin waves have been studied in recent years by the technique of inelastic light scattering (Brillouin scattering).³ The modes excited here have wavelengths very long compared to a lattice constant, with the consequence that their excitation energy is dominated by the Zeeman and dipolar terms in the spin Hamiltonian; exchange interactions play only a minor role. In many of the microscopic theories cited above, short-wavelength exchange-dominated surface spin waves enter importantly, but with the exception of one microwave resonance study,⁴ we know of no experimental study of these modes.

A very beautiful study⁵ of a ferromagnetic alloy surface by means of spin-polarized electron scattering shows that the surface magnetization obeys the same $T^{3/2}$ law as that in the bulk when $T \ll T_c$, although the coefficient is found to be larger. This is in good accord with an early theoretical prediction⁶ but, as argued previously⁶⁻⁸ the surface-spin-wave contribution to the temperature variation of the surface magnetization is in fact canceled to leading order by a deficit which arises from a surface-induced "hole" in the bulk spin-wave density of states. Thus, the surface magnetization data cannot be viewed as providing even indirect evidence for the existence of exchange-dominated surface spin waves. In general, similar (possibly incomplete) cancellations will occur in any of the contributions to the thermodynamic quantities that scale as the surface area.

Some years ago, one of us proposed that inelastic electron scattering may serve as a means of detecting exchange-dominated surface spin waves on single-crystal surfaces of ferromagnets;⁹ the calculations show that the spectra will contain lines produced by surface spin waves, and energy-loss bands with origin in scattering from bulk

spin waves, by events which fail to conserve wave vector normal to the surface. Very similar results emerge from a study of electron scattering from the surface of a model antiferromagnet.¹⁰ Until recently, such large deflection-angle energy-loss studies have failed to make their appearance, but this method has been used very recently to explore the dispersion relation of surface phonons on a clean Ni(100) surface.¹¹ In principle, this method may be applied very much as in Ref. 11 to the study of spin waves on surfaces, provided the energy-loss cross section is sufficiently large. Finally, glancing incidence neutron scattering has been suggested as a means of probing surface spin waves; our earlier analysis¹² of this possibility suggests that an experimental problem will be whether the surface-spin-wave loss feature can be resolved from a broad peak produced by inelastic scattering off of the bulk spin waves.

If surface-spin-wave dispersion relations can be studied by a method such as electron-energy-loss spectroscopy, then the temperature variation of the surface-spin-wave frequency provides one with access to interactions between spin waves, when they are in the very near vicinity of the surface. In this paper, we present a theoretical study of the leading contribution to the temperature variation of the surface-spin-wave dispersion relation, for short-wavelength modes whose excitation energy is dominated by the exchange interactions between the spins. Just as the leading term in the surface magnetization has the same temperature variation ($T^{3/2}$ law) as that in the bulk, we find the first correction term in the surface-spin-wave dispersion relation has the $T^{5/2}$ temperature variation characteristic of that found for bulk spin waves.¹³

As the above remarks imply, the present paper examines only exchange-dominated surface spin waves. The reader may note that an earlier paper¹⁴ explores the temperature variation of the frequency of the (dipole-dominated) surface spin waves explored in the light scattering studies cited earlier.

II. THE CALCULATION

A. Outline of the procedure

We shall need a specific model crystal for the analysis, and we choose an fcc lattice of spins S with a (100) sur-

face, and there are nearest-neighbor exchange interactions of strength J between the spins. Nearest-neighbor spin pairs in and near the surface are coupled with interactions identical in strength to the bulk. This is one of many model geometries that admits surface spin waves.^{15,16} While the model may seem rather specific in nature, in fact numerous discussions show the qualitative features of the surface-spin-wave spectrum are insensitive to the details of the surface geometry,^{15,16} with the exception of the special case where the surface exchange is so strong, modes are pushed out *above* the bulk-spin-wave bands in frequency.¹⁷ Furthermore, the low-temperature behavior of the mean spin deviation

$$\Delta_{l_z}(T) = S - \langle S_z(\vec{I}_{||l_z}) \rangle_T$$

is also model insensitive. (Here the location of a spin is denoted by $\vec{I} = \vec{I}_{||} + \hat{z}l_z$, with l_z an integer that counts the layers of the crystal. We assume the crystal lies in the upper half space, with the surface layer labeled by $l_z = 1$.) Thus, while the calculations in the present paper have been carried through for the model just outlined, we believe the principal conclusions will be quite similar for other choices of crystal structure or surface geometry.

Our Hamiltonian is then

$$H = -\frac{J}{2} \sum_{\vec{I}} \sum_{\vec{\delta}} \vec{S}(\vec{I}) \cdot \vec{S}(\vec{I} + \vec{\delta}), \quad (2.1)$$

where the sum over $\vec{\delta}$ covers the nearest neighbors of the spin at site \vec{I} ; a spin in any layer with $l_z \geq 2$ has twelve nearest neighbors, and a surface spin with $l_z = 1$ has only eight. The four "broken bonds" are responsible for softening the restoring forces on the surface spin, and this leads to the surface-spin-wave branch of the excitation spectrum.¹⁵

If one introduces the Holstein-Primakoff transformation truncated as follows:

$$\begin{aligned} S_+(\vec{I}) &= S_x(\vec{I}) + iS_y(\vec{I}) \\ &= (2S)^{1/2} \left[a_{\vec{I}} - \frac{1}{4S} a_{\vec{I}}^\dagger a_{\vec{I}} a_{\vec{I}} + \dots \right], \end{aligned} \quad (2.2a)$$

$$\begin{aligned} S_-(\vec{I}) &= S_x(\vec{I}) - iS_y(\vec{I}) \\ &= (2S)^{1/2} \left[a_{\vec{I}}^\dagger - \frac{1}{4S} a_{\vec{I}}^\dagger a_{\vec{I}}^\dagger a_{\vec{I}} + \dots \right], \end{aligned} \quad (2.2b)$$

and

$$S_z(\vec{I}) = S - a_{\vec{I}}^\dagger a_{\vec{I}}, \quad (2.2c)$$

with $a_{\vec{I}}$ and $a_{\vec{I}}^\dagger$, boson operators, the Hamiltonian is decomposed into piece quadratic in the boson operators, and a quartic piece:

$$H = H_2 + H_4, \quad (2.3a)$$

where

$$H_2 = SJ \sum_{\vec{I}} \sum_{\vec{\delta}} (a_{\vec{I}}^\dagger a_{\vec{I}} - a_{\vec{I}}^\dagger a_{\vec{I} + \vec{\delta}}) \quad (2.3b)$$

and

$$\begin{aligned} H_4 = & -\frac{J}{2} \sum_{\vec{I}} \sum_{\vec{\delta}} (a_{\vec{I}}^\dagger a_{\vec{I} + \vec{\delta}}^\dagger a_{\vec{I}} a_{\vec{I} + \vec{\delta}} - \frac{1}{2} a_{\vec{I}}^\dagger a_{\vec{I}}^\dagger a_{\vec{I}} a_{\vec{I} + \vec{\delta}} \\ & - \frac{1}{2} a_{\vec{I} + \vec{\delta}}^\dagger a_{\vec{I}}^\dagger a_{\vec{I}} a_{\vec{I}}). \end{aligned}$$

It is possible, in principle, to diagonalize the quadratic terms exactly, for the semi-infinite magnet. Indeed, in a study⁷ some years ago of the origin of the intimate relationship between the surface-spin-wave contributions to the thermodynamic properties, and those from surface-induced redistribution of the bulk modes in frequency, this procedure was carried through explicitly for a finite ferromagnetic film of thickness L , with the limit $L \rightarrow \infty$ achieved at the end. Translational symmetry dictates the normal modes are characterized by a wave vector $\vec{k}_{||}$ parallel to the surface, and for each choice of $\vec{k}_{||}$, we have a sequence of eigenmodes denoted by $\alpha = 1, 2, 3, \dots, n$, where n is the number of layers in a finite, but very thick sample. If we denote the eigenvectors by $\phi_{\vec{k}_{||}\alpha}(l_z)$, and suppose the sample is finite in thickness for the moment, then we have the orthonormality condition

$$\sum_{l_z=1}^n \phi_{\vec{k}_{||}\alpha}^*(l_z) \phi_{\vec{k}_{||}\alpha'}(l_z) = \delta_{\alpha\alpha'}. \quad (2.4)$$

We may then introduce a unitary transformation between the local spin-deviation operators $a_{\vec{I}}^\dagger$, and the operators $a_{\vec{k}_{||}\alpha}^\dagger$ which create an eigenmode of the finite film,

$$a_{\vec{I}} = \frac{1}{\sqrt{N_s}} \sum_{\vec{k}_{||}\alpha} e^{i\vec{k}_{||} \cdot \vec{I}_{||}} \phi_{\vec{k}_{||}\alpha}(l_z) a_{\vec{k}_{||}\alpha}, \quad (2.5)$$

with a similar relation for $a_{\vec{I}}^\dagger$. We apply periodic boundary conditions in the two directions parallel to the surface, and N_s is the number of spins in the layers parallel to the surface, in the fundamental quantization area.

In what follows, we shall require the explicit form of $\phi_{\vec{k}_{||}\alpha}(l_z)$ only for the surface spin wave whose renormalized frequency we wish to calculate. For our model, the surface-spin-wave frequency may be written in the form¹⁶

$$\Omega_s(\vec{k}_{||}) = A(\vec{k}_{||}) - \frac{1}{2} B(\vec{k}_{||}) \left[\frac{1}{\gamma(\vec{k}_{||})} + \gamma(\vec{k}_{||}) \right], \quad (2.6)$$

where, with $\vec{k}_{||} = \hat{x}k_x + \hat{y}k_y$, we have

$$A(\vec{k}_{||}) = 8SJ, \quad (2.7a)$$

$$B(\vec{k}_{||}) = 8SJ \cos \left[\frac{a_0}{2} k_x \right] \cos \left[\frac{a_0}{2} k_y \right], \quad (2.7b)$$

and

$$\gamma(\vec{k}_{||}) = \cos \left[\frac{a_0}{2} k_x \right] \cos \left[\frac{a_0}{2} k_y \right], \quad (2.7c)$$

with the geometry illustrated in Fig. 1. For each choice of $\vec{k}_{||}$, the surface-spin-wave frequency lies below the bulk-spin-wave frequencies associated with the wave vector $\vec{k}_{||}$. These are given by, for the model,

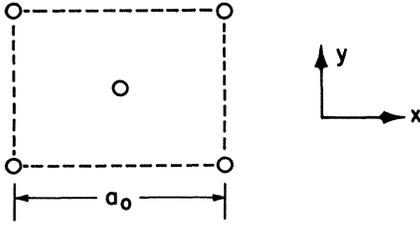


FIG. 1. Basic geometrical unit of the (100) plane of a fcc crystal.

$$\Omega_B(\vec{k}_{||}, k_z) = A(\vec{k}_{||}) - B(\vec{k}_{||}) \cos(\frac{1}{2} k_z a_0), \quad (2.8)$$

where the maximum value of $k_z a_0$ is 2π . The eigenvector $\phi_{\vec{k}_{||} s}(l_z)$ of the surface wave, suitably normalized, is given by

$$\phi_{\vec{k}_{||} s}(l_z) = (e^\kappa - 1)^{1/2} e^{-(\kappa/2)l_z}, \quad (2.9)$$

where

$$\kappa = 2 \ln \frac{1}{\gamma(\vec{k}_{||})}. \quad (2.10)$$

One proceeds to generate the first correction $\delta\Omega_s(\vec{k}_{||})$ to the surface wave frequency as follows. The quartic terms in the spin-wave Hamiltonian are expressed in terms of the true normal-mode annihilation and creation operators through use of Eq. (2.5). Then the expectation value of H_4 is taken in a state in which the various modes are characterized by a well-defined set of occupation numbers $\{n_{\vec{k}_{||}\alpha}\}$. The result has the form

$$\langle H_4 \rangle = \frac{J}{2N_s} \sum_{\vec{k}_{||}, \alpha'} \sum_{\vec{q}_{||}, \alpha} \Gamma(\vec{k}_{||}, \alpha'; \vec{q}_{||}, \alpha) n_{\vec{k}_{||}\alpha'} n_{\vec{q}_{||}\alpha}. \quad (2.11)$$

Then

$$\delta\Omega_s(\vec{k}_{||}) = \frac{\delta\langle H_4 \rangle}{\delta n_{\vec{k}_{||} s}} = \frac{J}{N_s} \sum_{\vec{q}_{||}, \alpha} \Gamma(\vec{k}_{||}, s; \vec{q}_{||}, \alpha) n_{\vec{q}_{||}\alpha}, \quad (2.12)$$

where now $n_{\vec{q}_{||}\alpha}$ is replaced by the appropriate Bose-Einstein distribution function to generate the temperature-dependent contribution to the surface wave frequency. In terms of the eigenamplitudes $\phi_{\vec{k}_{||}\alpha}(l_z)$ introduced above,

$$\begin{aligned} \delta\Omega_s(\vec{k}_{||}) = & -\frac{J}{2N_s} \sum_{l_z, \vec{\delta}} \sum_{\vec{q}_{||}\alpha} n_{\vec{q}_{||}\alpha} \{ \phi_{\vec{k}_{||} s}^*(l_z) \phi_{\vec{k}_{||} s}(l_z) \phi_{\vec{q}_{||}\alpha}^*(l_z + \delta_z) \phi_{\vec{q}_{||}\alpha}(l_z + \delta_z) \\ & + \phi_{\vec{k}_{||} s}^*(l_z + \delta_z) \phi_{\vec{k}_{||} s}(l_z + \delta_z) \phi_{\vec{q}_{||}\alpha}^*(l_z) \phi_{\vec{q}_{||}\alpha}(l_z) \\ & - \phi_{\vec{k}_{||} s}^*(l_z) \phi_{\vec{k}_{||} s}(l_z) [\phi_{\vec{q}_{||}\alpha}^*(l_z + \delta_z) \phi_{\vec{q}_{||}\alpha}(l_z) e^{-i\vec{q}_{||} \cdot \vec{\delta}_{||}} \\ & + \phi_{\vec{q}_{||}\alpha}^*(l_z) \phi_{\vec{q}_{||}\alpha}(l_z + \delta_z) e^{+i\vec{q}_{||} \cdot \vec{\delta}_{||}}] \\ & + \phi_{\vec{k}_{||} s}^*(l_z) \phi_{\vec{k}_{||} s}(l_z + \delta_z) e^{i\vec{k}_{||} \cdot \vec{\delta}_{||}} [\phi_{\vec{q}_{||}\alpha}^*(l_z + \delta_z) \phi_{\vec{q}_{||}\alpha}(l_z) e^{i\vec{q}_{||} \cdot \vec{\delta}_{||}} - \phi_{\vec{q}_{||}\alpha}^*(l_z) \phi_{\vec{q}_{||}\alpha}(l_z)] \\ & + \phi_{\vec{k}_{||} s}^*(l_z + \delta_z) \phi_{\vec{k}_{||} s}(l_z) e^{-i\vec{k}_{||} \cdot \vec{\delta}_{||}} \\ & \times [\phi_{\vec{q}_{||}\alpha}^*(l_z) \phi_{\vec{q}_{||}\alpha}(l_z + \delta_z) e^{+i\vec{q}_{||} \cdot \vec{\delta}_{||}} - \phi_{\vec{q}_{||}\alpha}^*(l_z) \phi_{\vec{q}_{||}\alpha}(l_z)] \}. \end{aligned} \quad (2.13)$$

We shall use the expression in Eq. (2.9) for the amplitude of the surface spin wave, and the amplitudes of the thermally excited spin waves may be eliminated through resort to certain Green's functions given some time ago.^{6,16} In Ref. 16, these were written in a rather general form which may be applied directly to the present geometry. The notation used in the following is that of Ref. 16. The spin-wave Green's function, in the site representation, is denoted by $G(\vec{l}, \vec{l}'; \Omega - i\eta)$, with η a positive infinitesimal. The link to the present problem is provided by the identity [Eq. (2.10) of Ref. 16]

$$\int_0^\infty \frac{d\Omega}{i\pi} n(\Omega) \text{Im}[G(\vec{l}, \vec{l}'; \Omega - i\eta)] = \frac{1}{N_s} \sum_{\vec{q}_{||}, \alpha} n_{\vec{q}_{||}\alpha} e^{-i\vec{q}_{||} \cdot (\vec{l}_{||} - \vec{l}'_{||})} \phi_{\vec{q}_{||}\alpha}^*(l_z) \phi_{\vec{q}_{||}\alpha}(l'_z). \quad (2.14)$$

Then Eq. (2.13) may be written directly in terms of the Green's function.

Translational invariance allows one to write the Green's function in the form

$$G(\vec{l}, \vec{l}'; \Omega - i\eta) = \frac{1}{N_s} \sum_{\vec{q}_{||}} G(\vec{q}_{||}; l_z, l'_z; \Omega - i\eta) \times e^{i\vec{q}_{||} \cdot (\vec{l}_{||} - \vec{l}'_{||})}, \quad (2.15)$$

and explicit expressions for $G(\vec{q}_{||}; l_z, l'_z; \Omega - i\eta)$ are found in Ref. 16. This function consists of two pieces:

$$G(\vec{q}_{||}; l_z, l'_z; \Omega - i\eta) = G_0(\vec{q}_{||}; |l_z - l'_z|; \Omega - i\eta) + \Delta G_s(\vec{q}_{||}; l_z + l'_z; \Omega - i\eta). \quad (2.16)$$

The piece $G_0(\vec{q}_{||}; |l_z - l'_z|; \Omega - i\eta)$ describes spin-wave propagation in a crystal of infinite spatial extent. If only this piece is retained in Eq. (2.15), the result obtained would describe renormalization of the surface mode by interaction with bulk waves, described as plane waves, totally unaffected by the surface. The function $\Delta G_s(\vec{q}_{||}; l_z - l'_z; \Omega - i\eta)$ describes modification of the excitation spectrum by the surface. There is a pole at $\Omega_s(\vec{q}_{||})$, the surface-spin-wave frequency associated with the wave vector $\vec{q}_{||}$. This leads to a contribution which describes renormalization of the surface spin wave by thermally excited surface spin waves.

The functional dependence of both $G_0(\vec{q}_{||}; |l_z - l'_z|; \Omega - i\eta)$ and $\Delta G_s(\vec{q}_{||}; l_z + l'_z; \Omega - i\eta)$ are sufficiently simple

that one may carry out the sums on l_z and $\vec{\delta}$ in Eq. (2.13) explicitly, once it is written in terms of the Green's function. Upon noting Eq. (2.15), we are left with the sum on $\vec{q}_{||}$ and the integral on frequency. The summand will be simplified by noting that when $T \ll T_C$, the thermally excited (bulk and surface) spin waves have a wavelength very long compared to a lattice constant. By exploiting this, the various contributions may be simplified to the point where simple expressions emerge from the analysis. We now turn to the details.

B. Mathematical details

It is convenient to introduce a new dimensionless frequency variable $\Omega' = [\Omega - A(\vec{q}_{||})]/B(\vec{q}_{||})$. As Ω is varied from $\Omega_m(\vec{q}_{||})$ to $\Omega_M(\vec{q}_{||})$, where $\Omega_m(\vec{q}_{||})$ and $\Omega_M(\vec{q}_{||})$ are the minimum and maximum bulk-spin-wave frequencies Ω' varies from -1 to $+1$. At the surface-spin-wave frequency, $\Omega' = \Omega'_s(\vec{q}_{||}) < -1$. Then the function $G_0(\vec{q}_{||}; |l_z - l'_z|; \Omega - i\eta)$ is given by

$$G_0(\vec{q}_{||}; |l_z - l'_z|; \Omega - i\eta) = \frac{\{i[-\Omega' - i(1 - \Omega'^2)^{1/2}]^{|l_z - l'_z|}\}}{B(\vec{q}_{||})(1 - \Omega'^2)^{1/2}}, \quad (2.17)$$

while (for l_z and l'_z both equal to or greater than unity),

$$\Delta G_s(\vec{q}_{||}; l_z + l'_z; \Omega - i\eta) = \frac{\{-\Omega' - i(1 - \Omega'^2)^{1/2}\}^{|l_z + l'_z| - 1}}{B(\vec{q}_{||})\sqrt{1 - \Omega'^2}} \left[\frac{1 + \gamma^2(\vec{q}_{||}) + 2\Omega'\gamma(\vec{q}_{||})}{[1 - \gamma^2(\vec{q}_{||})](1 - \Omega'^2)^{1/2} - i\{2\gamma(\vec{q}_{||}) + \Omega'[1 + \gamma^2(\vec{q}_{||})]\}} \right]. \quad (2.18)$$

As remarked earlier, ΔG_s has a pole at the surface-spin-wave frequency, Eq. (2.6). In the near vicinity of this pole, Eq. (2.18) may be arranged to read

$$\Delta G_s(\vec{q}_{||}; l_z + l'_z; \Omega - i\eta) = \frac{\{-\Omega'_s(\vec{q}_{||}) - [\Omega'_s(q_{||}) - 1]^{1/2}\}^{|l_z + l'_z| + 1}}{B(\vec{q}_{||})[\Omega'_s(q_{||})^2 - 1]} \frac{[1 - \gamma^2(\vec{q}_{||})]^3}{4\gamma^3(\vec{q}_{||})} \frac{1}{\Omega' - i\eta - \Omega'_s(\vec{q}_{||})}. \quad (2.19)$$

We now display the form of the various contributions to the frequency shift, once the sums on l_z and $\vec{\delta}$ have been carried out. First, consider the piece from $G_0(\vec{q}_{||}; |l_z - l'_z|; \Omega - i\eta)$, and call this $\delta\Omega_s^{(a)}(\vec{k}_{||})$. When Ω' lies between -1 and $+1$, it is useful to define $\xi(\vec{q}_{||}) = -\Omega' - i(1 - \Omega'^2)^{1/2}$, where the dependence on $\vec{q}_{||}$ arises from that in $A(\vec{q}_{||})$ and $B(\vec{q}_{||})$. We then let $\kappa_s = 2 \ln[1/\gamma(\vec{k}_{||})]$ be the attenuation constant of the surface spin wave whose renormalized frequency is under study. One then finds

$$\delta\Omega_s^{(a)}(\vec{k}_{||}) = -\frac{2J}{i\pi N_s} \sum_{\vec{q}_{||}} \int_{\Omega_m(\vec{q}_{||})}^{\Omega_M(\vec{q}_{||})} \frac{d\Omega n(\Omega)(e^{\kappa_s} - 1)}{B(\vec{q}_{||})(1 - \Omega'^2)^{1/2}} \text{Im} \left[i \left[[1 - 2\gamma(\vec{q}_{||})\xi(\vec{q}_{||})] e^{-\kappa_s} \right. \right. \\ \left. \left. + 2[\gamma(\vec{q}_{||} - \vec{k}_{||})\xi(\vec{q}_{||}) - \gamma(\vec{k}_{||})] e^{-(3/2)\kappa_s} \right. \right. \\ \left. \left. + \frac{1}{(e^{\kappa_s} - 1)} \{1 + 2[1 - 4\gamma(\vec{q}_{||})\xi(\vec{q}_{||})] e^{-\kappa_s} + e^{-2\kappa_s} \right. \right. \\ \left. \left. + 2[\gamma(\vec{k}_{||} - \vec{q}_{||})\xi(\vec{q}_{||}) - 2\gamma(\vec{k}_{||})] (e^{-(1/2)\kappa_s} + e^{-(3/2)\kappa_s}) \} \right] \right]. \quad (2.20)$$

The contribution from ΔG_s , in the frequency regime between $\Omega_m(\vec{q}_{||})$ and $\Omega_M(\vec{q}_{||})$ is in fact quite comparable to that displayed in Eq. (2.20). We call this contribution $\delta\Omega_s^{(b)}(\vec{k}_{||})$, and find it has the form

$$\begin{aligned}
\delta\Omega_s^{(b)}(\vec{k}_{\parallel}) = & \frac{2J}{i\pi N_s} \sum_{\vec{q}_{\parallel}} \int_{\Omega_m(\vec{q}_{\parallel})}^{\Omega_m(\vec{q}_{\parallel})} \frac{d\Omega n(\Omega)(e^{\kappa_s} - 1)[1 + \gamma^2(\vec{q}_{\parallel}) + 2\Omega'\gamma(\vec{q}_{\parallel})]}{B(\vec{q}_{\parallel})(1 - \Omega'^2)^{1/2}} \\
& \times \text{Im} \left\{ \frac{1}{([\gamma^2(\vec{q}_{\parallel}) - 1](1 - \Omega'^2)^{1/2} + i\{2\gamma(\vec{q}_{\parallel}) + \Omega'[1 + \gamma^2(\vec{q}_{\parallel})]\})} \right. \\
& \times \left\{ [\xi(\vec{q}_{\parallel}) - 2\gamma(\vec{q}_{\parallel})]\xi^2(\vec{q}_{\parallel})e^{-\kappa_s} \right. \\
& + 2\xi(\vec{q}_{\parallel})[\gamma(\vec{k}_{\parallel} - \vec{q}_{\parallel})\xi(\vec{q}_{\parallel}) - \gamma(\vec{k}_{\parallel})]e^{-(3/2)\kappa_s} \\
& + \frac{\xi^2(\vec{q}_{\parallel})}{[e^{\kappa_s} - \xi^2(\vec{q}_{\parallel})]} \left[\left[\xi^3(\vec{q}_{\parallel}) + \frac{1}{\xi(\vec{q}_{\parallel})} - 2\gamma(\vec{q}_{\parallel})[1 + \xi^2(\vec{q}_{\parallel})] \right] e^{-\kappa_s} \right. \\
& + \xi(\vec{q}_{\parallel})(1 + e^{-2\kappa_s}) + 2e^{-(1/2)\kappa_s} \{ \gamma(\vec{k}_{\parallel} - \vec{q}_{\parallel}) - \gamma(\vec{k}_{\parallel})\xi(\vec{q}_{\parallel}) \\
& + e^{-\kappa_s}\xi(\vec{q}_{\parallel})[\gamma(\vec{k}_{\parallel} - \vec{q}_{\parallel})\xi(\vec{q}_{\parallel}) \\
& \left. \left. \left. + \gamma(\vec{k}_{\parallel}) \right] \right\} \right] \left. \right\} \left. \right\}. \tag{2.21}
\end{aligned}$$

We are left with the contribution from the thermally excited surface spin waves. This we call $\delta\Omega_s^{(c)}(\vec{k}_{\parallel})$. In the algebra, one encounters the combination $-\Omega'_s - (\Omega_s'^2 - 1)^{1/2}$ which, upon using the surface-spin-wave dispersion relation, may be shown to equal $\gamma(\vec{q}_{\parallel})$. Then

$$\begin{aligned}
\delta\Omega_s^{(c)}(\vec{k}_{\parallel}) = & -\frac{J}{N_s} \sum_{\vec{q}_{\parallel}} \frac{n[\Omega_s(\vec{q}_{\parallel})](e^{\kappa_s} - 1)[1 - \gamma^2(\vec{q}_{\parallel})]^3}{\gamma^3(\vec{q}_{\parallel})[\Omega_s'(\vec{q}_{\parallel})^2 - 1]} \\
& \times \left\{ -\gamma^3(\vec{q}_{\parallel})e^{-\kappa_s} + e^{-2\kappa_s}\gamma(\vec{q}_{\parallel}) + 2\gamma(\vec{q}_{\parallel})e^{-(3/2)\kappa_s}[\gamma(\vec{q}_{\parallel})\gamma(\vec{q}_{\parallel} - \vec{k}_{\parallel}) - \gamma(\vec{k}_{\parallel})] \right. \\
& + \frac{\gamma^2(\vec{q}_{\parallel})}{[e^{\kappa_s} - \gamma^2(\vec{q}_{\parallel})]} \left[\left[\frac{1}{\gamma(\vec{q}_{\parallel})} - 2\gamma(\vec{q}_{\parallel}) - \gamma^3(\vec{q}_{\parallel}) \right] e^{-\kappa_s} + \gamma(\vec{q}_{\parallel})(1 + e^{-2\kappa_s}) \right. \\
& + 2e^{-(\kappa_s/2)} \{ \gamma(\vec{k}_{\parallel} - \vec{q}_{\parallel}) - \gamma(\vec{k}_{\parallel})\gamma(\vec{q}_{\parallel}) \\
& \left. \left. \left. + \gamma(\vec{q}_{\parallel})e^{-\kappa_s}[\gamma(\vec{q}_{\parallel})\gamma(\vec{k}_{\parallel} - \vec{q}_{\parallel}) - \gamma(\vec{k}_{\parallel})] \right\} \right] \left. \right\}. \tag{2.22}
\end{aligned}$$

It is possible to simplify the expression for $\delta\Omega_s^{(a)}(\vec{k}_{\parallel})$ considerably. We introduce $f_1(\kappa_s) = 16e^{-\kappa_s/2}$, $f_2(\kappa_s) = -8(1 + e^{-\kappa_s})$, and $f_0(\kappa_s) = f_1(\kappa_s) + f_2(\kappa_s)$, and after some algebra, we are led to the much simpler form

$$\delta\Omega_s^{(a)}(\vec{k}_{\parallel}) = \frac{J}{2\pi N_s} \sum_{\vec{q}_{\parallel}} \int_{\Omega_m(\vec{q}_{\parallel})}^{\Omega_m(\vec{q}_{\parallel})} \frac{d\Omega n(\Omega)}{(1 - \Omega'^2)^{1/2}} [f_0(\kappa_s) + \Omega'f_1(\kappa_s)\gamma(\vec{q}_{\parallel} - \vec{k}_{\parallel}) + \Omega'f_2(\kappa_s)\gamma(\vec{q}_{\parallel})]. \tag{2.23}$$

The integrals on frequency in Eq. (2.23) may be evaluated in closed form. We let $\tau = k_B T / \hbar$, and find

$$\int_{A-B}^{A+B} \frac{d\Omega}{(e^{\Omega/\tau} - 1)(1 - \Omega^2)^{1/2}} = \pi B \sum_{n=1}^{\infty} e^{-(nA/\tau)} I_0 \left[\frac{nB}{\tau} \right], \tag{2.24}$$

where $I_0(x)$ is the standard modified Bessel function of order zero. A similar analysis gives

$$\int_{A-B}^{A+B} \frac{d\Omega \Omega}{(e^{\Omega/\tau} - 1)(1 - \Omega^2)^{1/2}} = -\pi B \sum_{n=1}^{\infty} e^{-nA/\tau} I_1 \left[\frac{nB}{\tau} \right], \quad (2.25)$$

with $I_1(x)$ the modified Bessel function.

We now introduce a sequence of approximations that exploit the fact that only low temperatures $T \ll T_c$ are of interest. We make no assumption about the magnitude of $\vec{k}_{||}$, but the thermally excited waves will have $|\vec{q}_{||} a_0| \ll 1$ in this temperature regime. Thus, for example, we may expand $\gamma(\vec{k}_{||} - \vec{q}_{||})$ in powers of $\vec{q}_{||}$. This expansion assumes the form

$$\begin{aligned} \gamma(\vec{k}_{||} - \vec{q}_{||}) = & \gamma(\vec{k}_{||}) \left[1 - \frac{a_0^2}{8} q_{||}^2 + \cdots \right] + \frac{a_0}{2} \left[q_y \cos \left[\frac{a_0}{2} k_x \right] \sin \left[\frac{a_0}{2} k_y \right] + q_x \cos \left[\frac{a_0}{2} k_y \right] \sin \left[\frac{a_0}{2} k_x \right] \right] \\ & + \frac{a_0^2}{4} q_x q_y \sin \left[\frac{a_0}{2} k_x \right] \sin \left[\frac{a_0}{2} k_y \right] + \cdots, \end{aligned} \quad (2.26)$$

where the omitted terms are cubic or higher in q_x and q_y . The terms linear in $\vec{q}_{||}$ fail to contribute to the integral, and the same is true of the term proportional to $q_x q_y$. Thus, it is only the first term that need be retained. In the low-temperature limit, we also have $\tau \ll B$, so in Eq. (2.24) and (2.25), I_0 and I_1 may be replaced by their asymptotic forms for large argument:

$$I_0(z) = \frac{e^z}{\sqrt{2\pi z}} \left[1 + \frac{1}{8z} + \cdots \right], \quad (2.27a)$$

$$I_1(z) = \frac{e^z}{\sqrt{2\pi z}} \left[1 - \frac{3}{8z} + \cdots \right]. \quad (2.27b)$$

Finally, the sum over $\vec{q}_{||}$ may be replaced by an integral. When these steps are completed, we have with A the surface area and N_s the number of surface unit cells,

$$\begin{aligned} \delta\Omega_s^{(a)}(\vec{k}_{||}) = & \frac{JA\tau^{1/2}}{2(2\pi)^{3/2}N_s} \sum_{n=1}^{\infty} \frac{1}{n^{1/2}} \int_0^{\infty} \frac{dq_{||} q_{||}}{B^{1/2}} e^{-n(A-B)/\tau} \left[f_0(\kappa_s) - \gamma(\vec{k}_{||}) f_1(\kappa_s) - f_2(\kappa_s) \right] \\ & + \frac{\tau}{8nB} [f_0(\kappa_s) + 3\gamma(\vec{k}_{||}) f_1(\kappa_s) + 3f_2(\kappa_s)] \\ & + \frac{a_0^2 q_{||}^2}{8} [f_1(\kappa_s) \gamma(\vec{k}_{||}) + f_2(\kappa_s)] + \cdots \end{aligned} \quad (2.28)$$

where the terms retained will generate the leading correction to the dispersion relation. Explicit reference to the dependence of A and B on $\vec{q}_{||}$ in Eq. (2.28) has been suppressed.

It may be shown that the first term in square brackets on the right-hand side of Eq. (2.31) vanishes identically. Also, from Fig. 1, one sees that the ratio A/N_s equals $a_0^2/2$, so that

$$\begin{aligned} \delta\Omega_s^{(a)}(\vec{k}_{||}) = & \frac{Ja_0^2\tau^{3/2}}{32(2\pi)^{3/2}} [f_0(\kappa_s) + 3\gamma(\vec{k}_{||}) f_1(\kappa_s) + 3f_2(\kappa_s)] \sum_{n=1}^{\infty} \frac{1}{n^{3/2}} \int_0^{\infty} \frac{dq_{||} q_{||}}{B^{3/2}} e^{-[n(A-B)/\tau]} \\ & + \frac{Ja_0^4\tau^{1/2}}{32(2\pi)^{3/2}} [f_1(\kappa_s) \gamma(\vec{k}_{||}) + f_2(\kappa_s)] \sum_{n=1}^{\infty} \frac{1}{n^{1/2}} \int_0^{\infty} \frac{dq_{||} q_{||}^3}{B^{1/2}} e^{-[n(A-B)/\tau]}. \end{aligned} \quad (2.29)$$

In the spirit once again of the long-wavelength approximation, we may replace $A-B$ by $SJa_0^2 q_{||}^2$, while B itself is replaced by $8SJ$. The integrals on $q_{||}$ may then be performed:

$$\delta\Omega_s^{(a)}(\vec{k}_{||}) = \frac{J\zeta(\frac{5}{2})}{(2\pi)^{3/2}} \left[\frac{\tau}{8SJ} \right]^{5/2} \left\{ [f_1(\kappa_s) \gamma(\vec{k}_{||}) + f_2(\kappa_s)] + \frac{1}{8} [f_0(\kappa_s) + 3\gamma(\vec{k}_{||}) f_1(\kappa_s) + 3f_2(\kappa_s)] \right\}. \quad (2.30)$$

The last step is to notice a relation between $f_0(\kappa_s)$, $f_1(\kappa_s)$, and $f_2(\kappa_s)$ and $\gamma(\vec{k}_{||})$ which follows from Eq. (2.10). This gives the final result for that portion of the renormalization contributed by $G_0(\vec{q}_{||}; |l_z - l'_z|; \Omega - i\eta)$:

$$\delta\Omega_s^{(a)}(\vec{k}_{||}) = -\frac{12J\zeta(\frac{5}{2})}{(2\pi)^{3/2}} \left[\frac{\tau}{8SJ} \right]^{5/2} [1 - \gamma^2(\vec{k}_{||})]. \quad (2.31)$$

The contribution to the frequency shift originating from Eq. (2.21) may be handled in a very similar fashion. Howev-

er, one must treat the integrand with considerable care. As pointed out earlier,^{7,15} when one considers the reflection of surface spin waves off the surface, there is a resonance in the scattering amplitude, for waves which strike the surface at near glancing incidence. If q_{\perp} is the wave-vector component of the wave normal to the surface and \vec{q}_{\parallel} that parallel to it, then the resonance occurs when $q_{\perp} \cong q_{\parallel}^2 a_0$. When any physical quantity is computed, one must take care to insure that the presence of this feature is accounted for. We proceed by writing $\Omega' = -1 + \epsilon$ in Eq. (2.21), and also we suppose $q_{\parallel} a_0 \ll 1$. After considerable algebra, we find the integrand may be written as

$$\delta\Omega_s^{(b)}(\vec{k}_{\parallel}) = + \frac{J[1-\gamma^2(k_{\parallel})]}{2N_s} \frac{1}{i\pi} \sum_{\vec{q}_{\parallel}} \int_0^{\infty} \frac{d\epsilon}{\sqrt{2\epsilon}} \left\{ \left[\exp \left[\frac{SJ}{\tau} (a_0^2 q_{\parallel}^2 + 8\epsilon) \right] - 1 \right]^{-1} \text{Im}[(iq_{\parallel}^2 a_0^2 - 8\sqrt{2\epsilon})] \right\}, \quad (2.32)$$

which becomes simply

$$\delta\Omega_s^{(b)}(\vec{k}_{\parallel}) = \frac{J[1-\gamma^2(k_{\parallel})]a_0^2}{2\pi N_s} \sum_{\vec{q}_{\parallel}} q_{\parallel}^2 \int_0^{\infty} \frac{d\epsilon}{\sqrt{2\epsilon}} \left[\exp \left[\frac{SJ}{\tau} (a_0^2 q_{\parallel}^2 + 8\epsilon) \right] - 1 \right]^{-1}. \quad (2.33)$$

The resonance just described leads to a factor of $iq_{\parallel}^2 a_0^2 + 8\sqrt{2\epsilon}$ in the denominator of the integrand. This, however, is canceled by an identical factor in the numerator, and as a consequence, the resonance plays no role in the present analysis. Upon evaluating Eq. (2.33) in the low-temperature limit, we find

$$\delta\Omega_s^{(b)}(\vec{k}_{\parallel}) = + \frac{2J\xi(\frac{5}{2})}{(2\pi)^{3/2}} \left[\frac{\tau}{8SJ} \right]^{5/2} [1-\gamma^2(\vec{k}_{\parallel})]. \quad (2.34)$$

We are left with the contribution from thermally excited surface spin waves. As remarked in Sec. I, this portion vanishes identically, in the first order of perturbation theory. To see this, refer to the quantity in curly brackets in Eq. (2.22) as $N(\vec{k}_{\parallel}, \vec{q}_{\parallel})$, and multiply it by $[e^{\kappa_s} - \gamma^2(\vec{k}_{\parallel})]/8\gamma(\vec{q}_{\parallel})$, to find after some algebra that

$$\frac{[e^{\kappa_s} - \gamma^2(\vec{k}_{\parallel})]}{8\gamma(\vec{q}_{\parallel})} N(\vec{k}_{\parallel}, \vec{q}_{\parallel}) = e^{-\kappa_s} + 2\gamma(\vec{q}_{\parallel})\gamma(\vec{k}_{\parallel} - \vec{q}_{\parallel})e^{-(\kappa_s/2)} - \gamma(\vec{k}_{\parallel})e^{-(\kappa_s/2)} - \gamma(\vec{k}_{\parallel})\gamma^2(\vec{q}_{\parallel})e^{-(\kappa_s/2)} - \gamma^2(\vec{q}_{\parallel})e^{-\kappa_s}. \quad (2.35)$$

Now for the purposes of calculating the frequency shift $\delta\Omega_s^{(c)}(\vec{k}_{\parallel})$, the quantity $\gamma(\vec{k}_{\parallel} - \vec{q}_{\parallel})$ only enters averaged over the direction of \vec{q}_{\parallel} . Calling this average $\langle \gamma(\vec{k}_{\parallel} - \vec{q}_{\parallel}) \rangle$, one may easily show the average equals the product $\gamma(\vec{q}_{\parallel})\gamma(\vec{k}_{\parallel})$. We are left with

$$\frac{[e^{\kappa_s} - \gamma^2(\vec{k}_{\parallel})]}{8\gamma(\vec{q}_{\parallel})} \langle N(\vec{k}_{\parallel}, \vec{q}_{\parallel}) \rangle = e^{-\kappa_s} + \gamma(\vec{k}_{\parallel})\gamma^2(\vec{q}_{\parallel})e^{-(\kappa_s/2)} - \gamma(\vec{k}_{\parallel})e^{-(\kappa_s/2)} - \gamma^2(\vec{q}_{\parallel})e^{-\kappa_s}. \quad (2.36)$$

Upon noting that $e^{-(\kappa_s/2)} \cong \gamma(\vec{k}_{\parallel})$, one sees the right-hand side of Eq. (2.36) vanishes, and thus $\delta\Omega_s^{(c)}(\vec{k}_{\parallel})$ does also.

Past discussions^{7,8} have pointed out that there is an intimate relationship between the contribution to surface properties of the Heisenberg ferromagnet from thermally excited surface spin waves, and that from the resonant scattering of glancing incidence bulk waves. The latter produces an "antiresonance" in the density of bulk-spin-wave eigenstates, which partially cancels surface-spin-wave contributions to surface thermodynamic properties. We have seen that there is no manifestation of the bulk-spin-wave scattering resonance in the present calculation, so it is perhaps reasonable for the surface wave contribution to vanish also.

Our final result for the renormalized surface-spin-wave frequency is found by summing that in Eq. (2.31) and that in Eq. (2.34). Thus,

$$\delta\Omega_s(\vec{k}_{\parallel}) = - \frac{10J\xi(\frac{5}{2})}{(2\pi)^{3/2}} \left[\frac{\tau}{8SJ} \right]^{5/2} [1-\gamma^2(\vec{k}_{\parallel})]. \quad (2.37)$$

Note that in our discussion, we have made no assumption

about the magnitude of the wave vector of the surface spin wave, so Eq. (2.37) applies throughout the two-dimensional Brillouin zone.

III. SOME CONCLUDING REMARKS

The temperature variation we have found for the excitation energy of the surface spin wave is precisely the same as for bulk spin waves, which is also $T^{5/2}$ in leading order. Here we compare the magnitude of the effect for the two cases. There is particular interest in the limit $\vec{k}_{\parallel} \rightarrow 0$ since, in the absence of interactions, the term in the surface-spin-wave dispersion relation quadratic in k_{\parallel} is precisely the same as that of a bulk spin wave which propagates parallel to the surface. As discussed earlier,^{2,6} the "binding energy" of the surface spin wave resides entirely in the quartic terms, in this limit. Any difference in the coefficient in the dispersion relation produced by interactions between spin waves thus has an important influence on their binding, in the long-wavelength limit. Upon noting that as $k_{\parallel} \rightarrow 0$, $\gamma(\vec{k}_{\parallel}) \cong 1 - \frac{1}{8}a_0^2 k_{\parallel}^2$, one finds from Eq. (2.37)

$$\delta\Omega_s(\vec{k}_{||}) = -\frac{5}{2} \frac{J}{(2\pi)^{3/2}} (k_{||}a_0)^2 \left(\frac{\tau}{8SJ} \right)^{5/2} \zeta\left(\frac{5}{2}\right). \quad (3.1)$$

Expressions for the renormalized frequency of bulk spin waves have been given by a number of authors. Most particular, Ref. 14 has explored the role of nonlinearities in both the dipole-dipole and the exchange terms in renormalizing spin-wave frequencies. From the formulas given there,¹⁸ we find for bulk spin waves in the long-wavelength limit, for the fcc crystal with nearest-neighbor exchange,

$$\delta\Omega(\vec{k}) = -\frac{J}{(2\pi)^{3/2}} (ka_0)^2 \left(\frac{\tau}{8SJ} \right)^{5/2} \zeta\left(\frac{5}{2}\right). \quad (3.2)$$

Thus, interactions between spin waves affect the surface

mode more strongly than the bulk waves, in the long-wavelength limit, and produce a difference in the quadratic terms in the dispersion relation in this regime.

Note added in proof. Since this paper was submitted for publication, we have learned that Kontos and Cottam have also studied the renormalization of surface spin waves by thermally excited bulk and surface spin waves [D. E. Kontos and M. G. Cottam, paper presented at the Conference Internationale sur la Dynamique des Interfaces, Lille, France, September 1983 (unpublished)]. These authors obtain results only in the long-wavelength limit, $k_{||}a_0 \ll 1$, but allow for changes in surface exchange constants. With surface and bulk exchange equal, they find a $T^{5/2}$ law as we do, and there is no contribution from thermally excited surface spin waves. They conclude that altering surface exchange may lead to deviations from $T^{5/2}$ behavior.

¹For a review, see the article by D. L. Mills, in *Surface Excitations*, edited by V. M. Agranovich and R. Loudon (North-Holland, Amsterdam, in press), Chap. 5.

²R. F. Wallis, A. A. Maradudin, I. P. Ipatova, and A. A. Klochikhin, *Solid State Commun.* **5**, 89 (1967); B. M. Filipov, *Fiz. Tverd. Tela* **9**, 1339 (1967) [*Sov. Phys.—Solid State* **9**, 1048 (1967)]. The case of the itinerant ferromagnet has been discussed by J. Mathon, *Phys. Rev. B* **24**, 6588 (1981).

³See the discussion and references given by R. E. Camley, T. S. Rahman, and D. L. Mills, *Phys. Rev. B* **23**, 1226 (1961).

⁴J. T. Yu, R. A. Turk, and P. E. Wigen, *Phys. Rev. B* **11**, 420 (1975).

⁵D. T. Pierce, R. J. Celotta, J. Unguris, and H. C. Siegmann, *Phys. Rev. B* **26**, 2566 (1982).

⁶A. A. Maradudin and D. L. Mills, *J. Phys. Chem. Solids* **28**, 1855 (1967).

⁷D. L. Mills, *Phys. Rev. B* **1**, 264 (1970).

⁸D. L. Mills, *Comments Solid State Phys.* **4**, 28 (1971); *ibid.* **4**, 95 (1972).

⁹D. L. Mills, *J. Phys. Chem. Solids* **28**, 2245 (1967).

¹⁰B. Koiller and L. M. Falicov, *Phys. Rev. B* **27**, 346 (1983).

Evidently these authors were unaware of the work in Ref. 9.

¹¹S. Lehwald, J. M. Szeftel, H. Ibach, T. S. Rahman, and D. L. Mills, *Phys. Rev. Lett.* **50**, 518 (1983).

¹²P. Mazur and D. L. Mills, *Phys. Rev. B* **26**, 5175 (1982).

¹³C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1963), p. 71.

¹⁴Talat S. Rahman and D. L. Mills, *Phys. Rev. B* **20**, 1173 (1979); *Phys. Rev. B* **21**, 2046(E) (1980). Also, see Ref. 18 of the present paper.

¹⁵See Sec. II of Ref. 6.

¹⁶D. L. Mills, *J. Phys. (Paris) Colloq.* **31**, C1-33 (1970).

¹⁷Such modes have been discussed by T. Wolfram and R. E. de Wames, *Phys. Rev. B* **1**, 4358 (1970).

¹⁸This may be calculated from the second term in Eq. (2.33) of Ref. 14, after setting $\theta_k = 0$. Unfortunately, a multiplicative factor of z which appears in this expression should be removed. This same factor should also be omitted from Eq. (2.34c) of Ref. 14.