Surface Effects in the Heisenberg Antiferromagnet

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We have investigated the effect of a free (100) surface on low-temperature properties of a Heisenberg antiferromagnet of the CsCl structure. It is found that a surface magnon branch occurs in the excitation spectrum, with an excitation energy less than the frequency of a bulk magnon of the same wavelength. In the limit of infinite wavelength, the surface magnon frequency for this geometry is $(H_EH_A+H_A^2)^{1/2}$, compared to $(2H_EH_A+H_A^2)^{1/2}$ for bulk magnons of infinite wavelength. In the limit $H_E \gg H_A$, the surface magnon frequency is found to be insensitive to changes in exchange constants or anisotropy fields near the surface. The surface modes may be observed in the infrared absorption spectrum of the material, and will affect the low-temperature thermodynamic properties of the system. By means of a Green's-function method, we have examined the influence of the surface on the infrared absorption spectrum, the specific heat, and the low-temperature form of the parallel susceptibility and mean sublattice deviation. Numerical estimates indicate that the surface corrections of the thermodynamic quantities may be observable when $k_BT \ll (2H_EH_A)^{1/2}$.

I. INTRODUCTION

THERE has been a considerable amount of interest in the theoretical study of the influence of a free surface on the properties of crystals. One often finds that excitations localized near the surface occur. For example, many years ago, Lord Rayleigh[1] demonstrated that in the theory of elasticity, surface waves exist with displacement field localized near the surface. Wallis and co-workers[2] have recently examined the properties of short-wavelength surface waves in a simple crystal model. The properties of surface spin waves in ferromagnets have been discussed by several authors, in the long-wavelength limit, where the dominant contribution to the spin-wave energy comes from the dipolar interactions,[3] and in the short-wavelength region, where exchange interactions are important.[4,5]

The surface waves are eigenmodes of the Hamiltonian in a linearized theory. Consequently, thermally excited surface waves will contribute to the specific heat. In addition, the surface alters the distribution in frequency of the bulk waves. Thus the contribution to the specific heat from the bulk waves is changed by the presence of the surface. The low-temperature specific heat of a semi-infinite vibrating lattice has been studied by Onsager and co-workers,[6] by Stratton,[7] and by Maradudin and Wallis.[8] At low temperatures, a contribution proportional to the surface area, and to the square of the temperature, is obtained. In the high-temperature limit, Clark, Herman, and Wallis[9] have computed the mean-square displacement of an ion from its equilibrium position as a function of distance from the surface. The surface contribution to the magnetic specific heat of a ferromagnet, and the dependence of the mean spin deviation on distance from the surface, has also been studied recently.[10]

It has been possible to excite low-frequency surface vibrations directly,[11] and to study the vibrational properties of atoms in the surface layer by low-energy electron-diffraction techniques.[12] It appears that low-energy electron diffraction may also prove a useful technique for the study of magnetic properties of atoms in the surface layer of magnetic crystals.[13]

However, it has proved difficult to observe the surface contribution to thermodynamic properties of these systems. Indeed, we do not know of any unambiguous observation of the surface specific heat of a crystal, in experimental geometries similar to those employed in the model calculations described above. One difficulty is that in nonmagnetic crystals or in ferromagnetic arrays the bulk phonon or spin-wave energy tends to zero as the wavelength of the excitation becomes infinite.[14] Thus the contribution to the specific heat from the low-frequency bulk excitations generally dominates the surface contribution unless the temperature is very low, or the effective surface-to-volume ratio large.

For this reason, we have studied the influence of a free surface on the properties of a simple Heisenberg anti-

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[1] Lord Rayleigh, Proc. London Math. Soc. **17**, 4 (1887).
[2] D. C. Gazis, R. Herman, and R. F. Wallis, Phys. Rev. **119**, 533 (1960).
[3] J. Eshbach and R. Damon, Phys. Rev. **118**, 1208 (1960).
[4] R. F. Wallis, A. A. Maradudin, I. P. Ipatova, and A. A. Klochikhin, Solid State Commun. **5**, 89 (1967).
[5] D. L. Mills, in *Localized Excitations in Solids*, edited by R. F. Wallis (Plenum Press, Inc., New York, 1968), p. 426.
[6] M. Dupuis, R. Mazo, and L. Onsager, J. Chem. Phys. **33**, 1452 (1960).
[7] R. Stratton, J. Chem. Phys. **37**, 2972 (1962).
[8] A. A. Maradudin and R. F. Wallis, Phys. Rev. **148**, 947 (1966).
[9] C. Clark, R. Herman, and R. F. Wallis, Phys. Rev. **139**, A860 (1965).
[10] A. A. Maradudin and D. L. Mills, J. Phys. Chem. Solids **28**, 1855 (1967).
[11] R. M. Arzt, E. Salzmann, and K. Dransfeld, Appl. Phys. Letters **10**, 165 (1967).
[12] J. T. McKinney, E. R. Jones, and J. B. Webb, Phys. Rev. **151**, 467 (1966); **160**, 523 (1967).
[13] D. L. Mills, J. Chem. Phys. Solids **28**, 2245 (1967).
[14] Of course, a gap in the spin-wave spectrum may be introduced by an external magnetic field. However, from Refs. 3–5, one can see that surface magnons with frequency in the gap do not occur.

ferromagnet. When the exchange frequency ω_E is large compared to the anisotropy frequency ω_A, the minimum excitation energy $\omega_B^{(m)}$ for excitation of a bulk magnon is given by the well-known result $\omega_B^{(m)}=(2\omega_E\omega_A)^{1/2}$. The excitation energy $\omega_B^{(m)}$ is often quite large, the order of tens of degrees Kelvin in many instances. If, for a particular geometry, surface modes exist in the gap, then when $k_BT\ll\omega_B^{(m)}$, the ratio of the surface to the bulk contribution to the specific heat may be of observable magnitude, since the bulk contribution may be nearly frozen out in this range of temperature.

We have investigated the effect of a free (100) surface on the low-temperature properties of a simple Heisenberg antiferromagnet of the CsCl structure. The spins are assumed to interact via nearest-neighbor isotropic exchange interactions of antiferromagnetic sign. If it is assumed that spins in the surface layer experience the same anisotropy field as spins in the bulk, then the frequency of the surface branch becomes $(\omega_E\omega_A)^{1/2}$ in the limit as $\mathbf{k}\to 0$, if $\omega_E\gg\omega_A$. Thus the excitation energy of the long-wavelength surface waves lies in the gap below $\omega_B^{(m)}$. When $\omega_E\gg\omega_A$, the frequencies of the surface waves are found to be insensitive to changes in the exchange constants, and changes in the anisotropy field near the surface.

The surface spin-wave mode at $\mathbf{k}=0$ may be observed in the one-magnon absorption spectrum of the material. If the surface area of the sample is S, the volume V, and the lattice constant a, then the ratio of the integrated intensity of the line associated with the $k=0$ surface magnon to the integrated intensity of the bulk one-magnon absorption is found to be $(aS/2V)(\omega_E/\omega_A)^{1/2}$.

The purpose of this paper is to discuss the properties of the surface magnons in detail for the model described above, and to compute the effect of the surface on a number of low-temperature properties of the material. In Sec. II the properties of the surface magnons are studied by an equation-of-motion technique. In the spin-wave approximation, a number of thermodynamic properties of the crystal may be computed if certain single-particle Green's functions are known. In Sec. III, we define the Green's functions and solve for them from the appropriate equations of motion. The subsequent sections are devoted to a discussion of the one-magnon absorption spectrum, the surface contribution to the specific heat, the parallel magnetic susceptibility, and the variation of the temperature-dependent contribution to the sublattice spin deviation with distance from the surface.

The paper employs a picture of the surface that is certainly oversimplified. However, for the model considered, the expressions for the various quantities assume a reasonably simple form. We believe that the results provide a reliable estimate of the nature and magnitude of the surface effects. It is straightforward in principle to include the effect of changes in the anisotropy field and exchange constants in the surface. To include these effects makes the details of the algebra considerably more complex. As mentioned above, the results are expected to be insensitive to changes in ω_E near the surface, when $\omega_E\gg\omega_A$. We believe that these effects may be included in a later study, hopefully after some experimental information is available that will enable one to assess more readily the relative importance of the various complications.

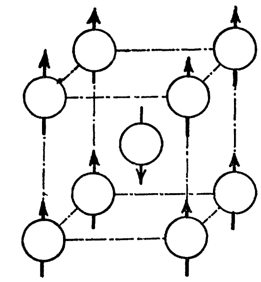

FIG. 1. The two-sublattice antiferromagnet considered in the present paper.

II. PROPERTIES OF SURFACE MODES

As mentioned in the Introduction, we shall confine our discussion to a particularly simple geometry, a two sublattice antiferromagnet of the CsCl structure. In the infinitely extended medium, a spin on a given sublattice is located at the body center of a cube of edge length a, with an antiparallel spin of the other sublattice at each cube corner. We assume nearest-neighbor antiferromagnetic exchange coupling between a spin and its eight nearest neighbors, in the bulk medium. Notice that each sublattice is a simple cubic lattice of lattice constant a. The model crystal is illustrated in Fig. 1.

In this section, we consider a semi-infinite antiferromagnetic array, with a (100) surface. The surface layer then consists of a layer of spins on just one sublattice. For definiteness, we suppose the surface layer consists of A spins, pointing in the $+z$ direction. It will also be assumed an external magnetic field H is applied parallel to the direction of the sublattice magnetizations.

For the moment, consider the Hamiltonian of the infinitely extended medium. One has (with $\hbar=1$)

$$H=-(\omega_A+\omega_H)\sum_{\mathbf{l}_a}S_z(l_a)+(\omega_A-\omega_H)\sum_{\mathbf{l}_b}S_z(l_b)+J\sum_{\mathbf{l}_a}\sum_{\boldsymbol{\delta}}\mathbf{S}(l_a)\cdot\mathbf{S}(\mathbf{l}_a+\boldsymbol{\delta}), \quad (1)$$

where ω_A is the frequency of precession of a spin in the local uniaxial anisotropy field ω_A, ω_H is the precession frequency in the external magnetic field, and J is the nearest-neighbor exchange integral. The sign convention is chosen so that J is a positive number. The first sum is over the sites of sublattice A, on which the spins point in the $+z$ direction. The second sum is over the sites of sublattice B, and in the third, for a given value of $\mathbf{l}_a$, one sums over the eight sites at $\{\mathbf{l}_a+\boldsymbol{\delta}\}$ adjacent to $\mathbf{l}_a$. Then assuming the temperature is low compared to the Néel temperature, we make the Holstein-Primakoff transformation to boson variables, retaining only terms quadratic in the spin deviation annihilation and creation operators.[15] Then, ignoring constant terms,

$$H=(\omega_A+\omega_E-\omega_H)\sum_{\mathbf{l}_b} b_{\mathbf{l}_b}{}^\dagger b_{\mathbf{l}_b}+(\omega_A+\omega_E+\omega_H)\sum_{\mathbf{l}_a} a_{\mathbf{l}_a}{}^\dagger a_{\mathbf{l}_a} +\tfrac{1}{8}\omega_E\sum_{\mathbf{l}_a}\sum_{\delta}(a_{\mathbf{l}_a}b_{\mathbf{l}_a+\delta}+a_{\mathbf{l}_a}{}^\dagger b_{\mathbf{l}_a+\delta}{}^\dagger). \quad (2)$$

We have defined the exchange frequency $\omega_E=8JS$. Recall the commutation relations

$$[a_{\mathbf{l}_a}, a_{\mathbf{l}_a'}{}^\dagger]=\delta_{\mathbf{l}_a\mathbf{l}_a'}, \qquad [b_{\mathbf{l}_b}, b_{\mathbf{l}_b'}{}^\dagger]=\delta_{\mathbf{l}_b\mathbf{l}_b'},$$

where all commutators not exhibited vanish.

The spin-wave frequencies associated with the bulk material may be derived in the standard manner. One obtains the equation of motion for the spin deviation operators $a_{\mathbf{l}_a}$ and $b_{\mathbf{l}_b}{}^\dagger$. Then it is assumed the solutions vary in time like $\exp(i\Omega t)$, and a Fourier transformation with respect to the spatial variables $\{\mathbf{l}_a\}$, $\{\mathbf{l}_b\}$ is carried out. For a given value of the wave vector $\mathbf{k}$, there exist two spin-wave modes of frequency[16]

$$\Omega_B(\mathbf{k})=\omega_H\pm[(\omega_A+\omega_E)^2-\omega_E^2\gamma(\mathbf{k})^2]^{1/2} \equiv\omega_H\pm\omega(\mathbf{k}), \quad (3)$$

where $\gamma(\mathbf{k})=\cos(\frac{1}{2}k_xa)\cos(\frac{1}{2}k_ya)\cos(\frac{1}{2}k_za)$ for the structure considered.

To include the effect of a free surface, the Hamiltonian of Eq. (2) must be modified to include the influence of the surface on the motion of the surface spins. The crystal will be assumed to lie in the region $z>0$ with a (100) surface of A spins in the x-y plane. The Hamiltonian may then be written in the form

$$\begin{aligned}H=&[\omega_A+\omega_E-\omega_H]\sum_{\mathbf{l}_b} b_{\mathbf{l}_b}{}^\dagger b_{\mathbf{l}_b}\\&+\sum_{\mathbf{l}_a}[\omega_A+\omega_E+\omega_H+\Delta(l_a{}^z)(\delta\omega_A-\tfrac{1}{2}\omega_E)]a_{\mathbf{l}_a}{}^\dagger a_{\mathbf{l}_a}\\&+\tfrac{1}{8}\omega_E\sum_{\mathbf{l}_a}[1-\Delta(l_a{}^z)]\sum_{\delta}[a_{\mathbf{l}_a}b_{l_a+\delta}+\text{H.c.}]\\&+\tfrac{1}{8}\omega_E\sum_{\mathbf{l}_a}\Delta(\mathbf{l}_a{}^z)\sum_{\delta;\delta_z>0}[a_{\mathbf{l}_a}b_{\mathbf{l}_a+\delta}+\text{H.c.}], \quad (4)\end{aligned}$$

where

$$\Delta(l_a{}^z)=1 \quad l_a{}^z=0$$
$$=0 \quad l_a{}^z\neq 0.$$

The sums in Eq. (4) extend only over sites in the upper half-space, with $l_a{}^z\geq 0$. From the second term in the first line, one notes the surface spins see only half the exchange field seen by an interior spin. The term in the third line occurs because a surface spin is "bonded" only to half as many B spins as an interior spin. For the moment, we ignore changes in the exchange constants near the surface. We shall return to consider the effect of altering the exchange constants on the solutions described below.

The quantity $\delta\omega_A$ is the change in anisotropy field in the surface layer. We assume that the change in anisotropy field near the surface is parallel to the direction of magnetization, so only the magnitude of ω_A differs in the surface, compared to its value in the bulk. For one model, if our anisotropy field is dipolar in origin and the sublattice magnetization is parallel to the surface, then this assumption is valid. For this case, we estimate $\delta\omega_A\cong-0.3\,\omega_A$. The change in anisotropy field one layer from the surface is quite negligible for this picture.

The creation operators $\alpha_\eta{}^\dagger$ associated with the ηth normal mode of the semi-infinite crystal may be written in the form

$$\alpha_\eta=\sum_{\mathbf{l}_a}A_\eta(\mathbf{l}_a)a_{\mathbf{l}_a}{}^\dagger+\sum_{\mathbf{l}_b}B_\eta(\mathbf{l}_b)b_{\mathbf{l}_b}.$$

The coefficients $A_\eta(\mathbf{l}_a)$ and $B_\eta(\mathbf{l}_b)$ will be normalized so that

$$[\alpha_\eta, \alpha_\eta{}^\dagger]=1.$$

The eigenfrequencies Ω_η and the coefficients may be obtained from the eigenvalue equation

$$\begin{aligned}\Omega_\eta\alpha_\eta{}^\dagger&=[H, \alpha_\eta{}^\dagger]\\&=\sum_{\mathbf{l}_a}A_\eta(\mathbf{l}_a)[H, a_{\mathbf{l}_a}{}^\dagger]+\sum_{\mathbf{l}_b}B_\eta(\mathbf{l}_b)[H, b_{\mathbf{l}_b}].\end{aligned}$$

By first commuting this equation with $a_{\mathbf{l}_a}$, then with $b_{\mathbf{l}_b}{}^\dagger$ one derives two equations involving only the c-number amplitudes:

$$\Omega_\eta A_\eta(\mathbf{l}_a)=\sum_{\mathbf{l}_a'}A_\eta(\mathbf{l}_a')[[a_{\mathbf{l}_a'}{}^\dagger, H], a_{\mathbf{l}_a}] +\sum_{\mathbf{l}_b}B_\eta(\mathbf{l}_b)[[b_{\mathbf{l}_b}, H], a_{\mathbf{l}_a}]$$

and

$$\Omega_\eta B_\eta(\mathbf{l}_b)=\sum_{\mathbf{l}_a}A_\eta(\mathbf{l}_a)[b_{\mathbf{l}_b}{}^\dagger, [a_{\mathbf{l}_a}{}^\dagger, H]] +\sum_{\mathbf{l}_b}B_\eta(\mathbf{l}_b')[b_{\mathbf{l}_b}{}^\dagger, [b_{\mathbf{l}_b'}, H]].$$

Evaluation of the double commutators then gives

$$\Omega_\eta B_\eta(\mathbf{l}_b)=-(\omega_A+\omega_E-\omega_H)B_\eta(\mathbf{l}_b)+\tfrac{1}{8}\omega_E\sum_{\delta}A_\eta(\mathbf{l}_b+\boldsymbol{\delta}), \quad (5)$$

$$\begin{aligned}\Omega_\eta A_\eta(\mathbf{l}_a)=&[\omega_A+\omega_E+\omega_H+\delta\omega\Delta(l_a{}^z)]A_\eta(l_a{}^z)\\&-\tfrac{1}{8}\omega_E[1-\Delta(l_a{}^z)]\sum_{\delta}B_\eta(\mathbf{l}_a+\boldsymbol{\delta})\\&-\tfrac{1}{8}\omega_E\Delta(l_a{}^z)\sum_{\delta_z>0}B_\eta(l_a+\delta),\end{aligned}$$

where we have defined $\delta\omega=\delta\omega_A-\frac{1}{2}\omega_E$.

Because translational invariance with respect to displacements in the x and y directions (parallel to the surface) has not been destroyed, the solutions to Eqs.

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[15] C. Kittel, *Quantum Theory of Solids* (John Wiley & Sons, Inc., New York, 1963), p. 58.

[16] The excitation energies of the modes are $\omega_H+\omega(\mathbf{k})$ and $-\omega_H+\omega(k)$, respectively. The excitation energies are both positive, provided that $\omega_H<\omega_H{}^{(c)}=(2\omega_A\omega_E+\omega_A{}^2)^{1/2}$. When $\omega_H>\omega_H{}^{(c)}$, the ground state is unstable, and a spin-flop transition occurs in the bulk material. See S. Foner, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. I.

(5) have the Bloch form

$$A_\eta(\mathbf{l}_a)=\alpha_\eta(l_a{}^z)\exp(i\mathbf{k}_{||}\cdot\mathbf{l}_a),$$
$$B_\eta(\mathbf{l}_b)=\beta_\eta(l_a{}^z)\exp(i\mathbf{k}_{||}\cdot l_b),$$

where $\mathbf{k}_{||}=\hat{x}k_x+\hat{y}k_y$.

Substitution of this form into Eqs. (5) then yields equations for $\alpha_\eta(l_a{}^z)$ and $\beta_\eta(l_a{}^z)$:

$$\Omega_\eta\beta_\eta(l_b{}^z)=-(\omega_A+\omega_E-\omega_H)\beta_\eta(l_b{}^z)+\tfrac{1}{2}\omega_E\lambda(\mathbf{k}_{||})[\alpha_\eta(l_b{}^z+\tfrac{1}{2}a)+\alpha_\eta(l_b{}^z-\tfrac{1}{2}a)],\quad(6a)$$

$$\Omega_\eta\alpha_\eta(l_a{}^z)=[\omega_A+\omega_E+\omega_H+\delta\omega\Delta(l_a{}^z)][\alpha_\eta(l_a{}^z)]-\tfrac{1}{2}\omega_E\lambda(\mathbf{k}_{||})[1-\Delta(l_a{}^z)][\beta_\eta(l_a{}^z+\tfrac{1}{2}a)+\beta_\eta(l_a{}^z-\tfrac{1}{2}a)]-\tfrac{1}{2}\omega_E\lambda(\mathbf{k}_{||})\Delta(l_a{}^z)\beta_\eta(\tfrac{1}{2}a),\quad(6b)$$

with $\lambda(\mathbf{k}_{||})=\cos(\tfrac{1}{2}k_xa)\cos(\tfrac{1}{2}k_ya)$.

The first of Eqs. (6) may now be employed to eliminate the β_η's from the second of Eqs. (6) to give an equation for the α_η's alone. One finds

$$[(\Omega_\eta-\omega_H)^2-(\omega_A+\omega_E)^2]\alpha_\eta(l_a{}^z)+\tfrac{1}{4}\omega_E{}^2\lambda^2(\mathbf{k}_{||})\times[\alpha_\eta(l_a{}^z+a)+\alpha_\eta(l_a{}^z-a)+2\alpha_\eta(l_a{}^z)][1-\Delta(l_a{}^z)]=\Delta(l_a{}^z)\{\delta\omega(\Omega_\eta-\omega_H+\omega_A+\omega_E)\alpha_\eta(0)-\tfrac{1}{4}\omega_E{}^2\lambda^2(\mathbf{k}_{||})[\alpha_\eta(0)+\alpha_\eta(a)]\}.\quad(7)$$

This equation determines the quantities $\alpha_\eta(l_a{}^z)$. The amplitude $\beta_\eta(l_a{}^z)$ of the spin deviation on the B sublattice associated with a particular normal mode may be determined from Eq. (6a).

In this section, we shall be interested in studying the solutions of Eq. (7) for which the spin deviation is localized near the surface. Consider a mode in which the spin deviation decays exponentially into the sample:

$$\alpha_s(l_a{}^z)=\exp(-ql_a{}^z).\quad(8)$$

Substitution of Eq. (8) into Eq. (7) with $l_a{}^z\neq0$ yields an expression for the frequency Ω_s of the surface wave in terms of the attenuation constant q. One finds

$$(\Omega_s-\omega_H)^2=(\omega_A+\omega_E)^2-\omega_E{}^2\lambda^2(k_{||})\cosh^2(\tfrac{1}{2}qa).\quad(9)$$

For a given value of $\mathbf{k}_{||}$, the attenuation constant q is found by requiring the solution to satisfy Eq. (7) for the case $l_a{}^z=0$. It is necessary to require that the real part of the quantity q be positive, so that $\alpha_s(l_a{}^z)\to0$ as $l_a{}^z\to\infty$. From the equation for $l_a{}^z=0$, one finds

$$1+\exp(qa)=-[4\delta\omega/\omega_E{}^2\lambda^2(\mathbf{k}_{||})](\Omega_s-\omega_H+\omega_A+\omega_E).\quad(10)$$

Equations (9) and (10) may now be combined to yield an equation for Ω_s as a function of $\mathbf{k}_{||}$. At this point, the algebra is simplified greatly by ignoring the change in the anisotropy field in the surface layer. We shall return to discuss the effect of including an additional pinning field on the surface spins after first considering the case $\delta\omega_A=0$. Then $\delta\omega=-\tfrac{1}{2}\omega_E$.

From Eq. (10), after a bit of rearrangement, one finds

$$\cosh^2(\tfrac{1}{2}qa)=\frac{4}{\lambda^4\omega_E{}^2}\frac{(\Omega_s-\omega_H+\omega_A+\omega_E)^2}{(2/\lambda^2\omega_E)(\Omega_s-\omega_H+\omega_A+\omega_E)-1}.$$

Insertion of this result into Eq. (9) leads to a quadratic equation for the quantity $(\Omega_s-\omega_H)$:

$$(\Omega_s-\omega_H)^2+\tfrac{1}{2}\omega_E(1-\lambda^2)(\Omega_s-\omega_H)-(\omega_A-\omega_E)[\omega_A+\tfrac{1}{2}\omega_E(1-\lambda^2)]=0.\quad(11)$$

The two solutions to this equation are

$$\Omega_s=\omega_H-\tfrac{1}{4}\omega_E(1-\lambda^2)\pm\tfrac{1}{2}\{\tfrac{1}{4}\omega_E{}^2(1-\lambda^2)^2+4(\omega_A+\omega_E)[\omega_A+\tfrac{1}{2}\omega_E(1-\lambda^2)]\}^{1/2}.\quad(12)$$

As $\mathbf{k}_{||}\to0$, $\lambda\to1$, and one finds

$$\Omega_s=\omega_H\pm[\omega_A(\omega_A+\omega_E)]^{1/2}.$$

When $\omega_E\gg\omega_A$, with $\omega_H=0$, one has

$$\Omega_s=\pm(\omega_A\omega_E)^{1/2}.$$

For a given value of $\mathbf{k}_{||}$, the surface magnon excitation energy lies in the gap below the excitation spectrum of the bulk waves.

Only one of the two solutions to Eq. (12) satisfies the requirement $q>0$. For simplicity, consider the case $\mathbf{k}_{||}=0$. Then from Eq. (10), with $\delta\omega=-\tfrac{1}{2}\omega_E$,

$$1+\exp(qa)=+(2/\omega_E)\{\omega_E+\omega_A\pm[\omega_A(\omega_E+\omega_A)]^{1/2}\}.\quad(13)$$

The right side of Eq. (13) must be greater than 2 if $q>0$. This requires that the upper sign in Eqs. (12) and (13) be chosen. The solution corresponding to the lower sign corresponds to a wave with amplitude that grows exponentially from the surface. Thus there exists a single, nondegenerate surface magnon branch. The frequency Ω_s of a surface wave with a given value of $\mathbf{k}_{||}$ is

$$\Omega_s=\omega_H+\{(\omega_A+\omega_E)[\omega_A+\tfrac{1}{2}\omega_E(1-\lambda^2)]+\tfrac{1}{16}\omega_E{}^2(1-\lambda^2)^2\}^{1/2}-\tfrac{1}{4}\omega_E(1-\lambda^2).\quad(14)$$

A plot of the surface magnon dispersion relation, along with the dispersion relation of bulk waves propagating parallel to the surface, is given in Fig. 2.

The magnetic field dependence of the surface magnon frequency exhibited in Eq. (14) has some interesting consequences. If the direction of the magnetic field is parallel to the direction of the A spins that lie in the surface layer ($\omega_H>0$), then the application of a field "stiffens" the surface mode by increasing its frequency. Application of a field antiparallel to the A spin direction ($\omega_H<0$) "softens" the mode. The dependence of the sign of the shift in frequency of the surface mode on the orientation of the external magnetic field relative to the direction of surface-spin alignment will strongly influence the sign of the contribution of the layers near

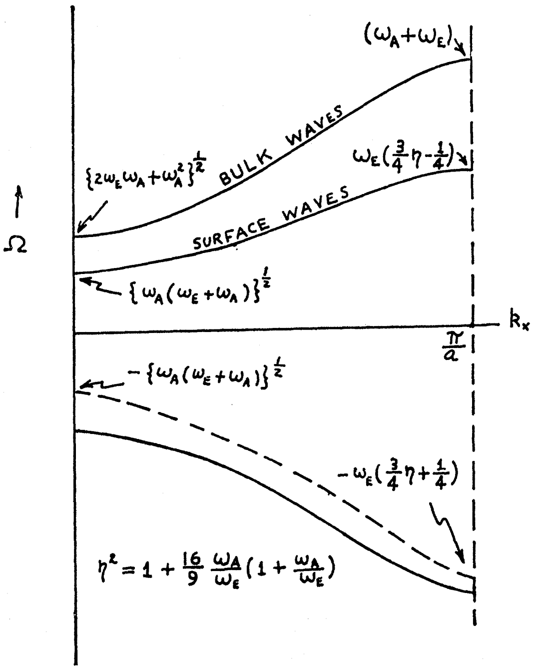

FIG. 2. The dispersion relations for surface magnons and bulk magnons propagating parallel to the x axis, which lies in the sample surface. The external magnetic field is assumed zero. The unphysical solution to Eq. (12) is indicated by the dashed line.

the surface to the parallel susceptibility. This will be discussed in detail in a subsequent section.

If $\omega_H<0$, as $|\omega_H|\rightarrow\omega_H^{(c)}=[\omega_A(\omega_A+\omega_E)]^{1/2}$ from below, the excitation energy of the $\mathbf{k}=0$ surface mode approaches zero. For $\omega_H>\omega_H^{(c)}$, the antiferromagnetic ground state is unstable with respect to a new ground state in which the spins near the surface rotate through an angle of roughly 90°. A detailed description of this "surface spin-flop" transition has been given in a paper by one of us.[17]

The discussion above neglects the effects of changes in the anisotropy field in the surface layer. We shall examine the sensitivity of the frequency of the surface mode at $\mathbf{k}_{||}=0$ to changes in the surface anisotropy field. Let $\omega_A^{(s)}$ be the anisotropy field seen by a spin in the surface layer. Define

$$\delta=2(\omega_A-\omega_A^{(s)})/\omega_E.$$

It will be assumed that $\delta\ll1$. If $\omega_E\gg\omega_A$, and the anisotropy field is largely dipolar in origin, then the discussion earlier in this section indicates that this assumption is reasonable. In terms of the parameter δ, Eq. (10) assumes the form (with $\mathbf{k}_{||}=0$ and $\omega_H=0$)

$$1+\exp(qa)=2(1+\delta)[(\omega_E+\omega_A-\Omega_s)/\omega_E].$$

Using this result, one finds the $\mathbf{k}_{||}=0$ surface magnon frequency is given as the solution of the quadratic equation

$$(1+\delta)\Omega_s^2+\tfrac{1}{2}\omega_E[(1+\delta)^2-1]\Omega_s-(\omega_A+\omega_E)[(1+\delta)\omega_A-\tfrac{1}{2}\omega_E\delta^2]=0, \quad (15)$$

which corresponds to $q>0$.

In the limit $\delta\ll1$, the change in Ω_s to first order in δ may be found. One finds that the fractional change in Ω_s is related to the fractional change in surface anisotropy field as follows:

$$\delta\Omega_s/\Omega_s^{(0)}=\{\omega_A/[\omega_A(\omega_A+\omega_E)]^{1/2}\}[(\omega_A^{(s)}-\omega_A)/\omega_A].$$

Thus, in the limit $\omega_E\gg\omega_A$,

$$(\delta\Omega_s/\Omega_s^{(0)})\ll1 \quad \text{if} \quad [(\omega_A^{(s)}-\omega_A)/\omega_A]\approx1.$$

In the case where $\omega_E\gg\omega_A$, the reason for the insensitivity of the surface magnon mode to changes in the surface anisotropy field may be seen by noting from Eq. (13) that $qa\cong2(\omega_A/\omega_E)^{1/2}$ at $k=0$. The spin deviation associated with the surface mode extends roughly $(\omega_E/\omega_A)^{1/2}$ atomic layers into the crystal. Since the mode involves the motion of spins in many layers, changes in the anisotropy field seen by the surface spin results in only a small shift in the frequency of the mode.

Next suppose that the exchange constants are changed near the surface. To be specific, suppose the exchange coupling between spins in the layer $l_a^z=0$ and $l_b^z=1$ is $J'\neq J$, and we define $\omega_E'=8J'S$. The effect of this perturbation on the $\mathbf{k}=0$ surface magnon frequency may be found by a method very similar to the preceding discussion of the effect of $\delta\omega_A$. Since the algebra involved is straightforward but tedious, we shall simply quote the result. To first order in $(\omega_E'-\omega_E)$ we find

$$\delta\Omega_s/\Omega_s^{(0)}=\tfrac{1}{2}(\omega_E'-\omega_E)/\omega_E.$$

This result may be understood qualitatively by the following simple argument. We have just seen that the effect of changing the anisotropy field in the surface layer shifts the surface mode frequency by an amount $\delta\omega_A$. If the only effect of the change in ω_E was to change the molecular field of a spin in the surface layer, then the shift in exchange constants described above would shift the frequency of the mode by an amount $\delta\omega_E=\omega_E'-\omega_E$. However, the change in ω_E can affect the frequency of the mode only if one has a nonzero value of the angle θ that measures the amount by which the angle between the A spins in the surface, and the B spins in the layer $l_b^z=1$, differs from 180°. When $\omega_A\ll\omega_E$, $\theta\approx(\omega_A/\omega_E)^{1/2}$. The frequency shift is thus reduced from the value $(\omega_E'-\omega_E)$ obtained from the molecular-field approximation to $(\omega_E'-\omega_E)\theta=(\omega_A/\omega_E)^{1/2}(\omega_E'-\omega_E)$. This is the result exhibited above, within a multiplicative constant of order unity. Notice that the frequency shift is proportional to θ rather than to θ^2 because of the low symmetry of the surface region.

It is difficult to estimate the magnitude of the changes in ω_E near the surface in a reliable way. However, it is

[17] D. L. Mills, Phys. Rev. Letters **20**, 18 (1968).

known[18] that in Ni the change Δa in the lattice constant near the surface is of the order of a few percent. In a crystal in which the magnetic ion is an S-state ion, the value of J may not be affected greatly by the lowered symmetry of the crystal field in the surface layer. If the change in J comes solely from the change in lattice constant near the surface, and if $\Delta a/a \cong 0.05$ is a typical change in a, then one might expect

$$(\omega_E' - \omega_E)/\omega_E \approx 0.05.$$

Thus changes in ω_E near the surface may shift Ω_s by a few percent, if the above estimates are correct.

In the subsequent discussion, we ignore changes in ω_E and ω_A on the quantities computed. In the spin-wave approximation, the surface magnon mode is an eigenmode of the Hamiltonian. In principle, one may observe a line from the surface mode in the infrared absorption spectrum of the material. Also, thermally excited surface modes will contribute to the specific heat, parallel susceptibility, and sublattice deviation. These effects are best discussed with a Green's-function technique, since in addition to the presence of the surface waves, it is necessary to realize that the bulk spin-wave frequency distribution function and eigenvectors are altered by the surface. Once certain Green's functions are known, a systematic discussion of the contribution of both the surface modes and the perturbed bulk waves to the various low-temperature properties of the system is possible.

In Sec. III, the relevant Green's functions are defined, and their properties discussed. In subsequent sections, the Green's functions will be employed to study some low-temperature properties of the system.

III. THERMODYNAMIC GREEN'S FUNCTIONS

We shall consider imaginary time Green's functions of the form

$$D^{(AB)}(\tau) = \langle T(A(\tau)B(0))\rangle \equiv D_{>}^{(AB)}(\tau)\theta(\tau) + D_{<}^{(AB)}(\tau)\theta(-\tau), \quad (16)$$

where

$$D_{>}^{(AB)}(\tau) = \langle A(\tau)B(0)\rangle$$

and

$$D_{<}^{(AB)}(\tau) = \langle B(0)A(\tau)\rangle.$$

The angular brackets denote an average of the enclosed operators over the appropriate finite-temperature statistical ensemble. For the system considered here,

$$\langle O\rangle \equiv \mathrm{Tr}(e^{-\beta H}O)/\mathrm{Tr}(e^{-\beta H}),$$

where H is the Hamiltonian of the antiferromagnet in the lowest-order Holstein-Primakoff transformation, and $\beta = 1/k_BT$. Also, $A(\tau) = e^{H\tau}A(0)e^{-H\tau}$.

It is by now well known that the various thermodynamic averages encountered in describing the properties of many-body systems may be conveniently related to the Fourier transform of the correlation function of Eq. (16). It will be useful to recall some properties of the correlation functions. From the periodicity property

$$D^{(AB)}(\tau+\beta) = D^{(AB)}(\tau),$$

one may expand D in a Fourier series of the form

$$D^{(AB)}(\tau) = (1/\beta)\sum_{n=-\infty}^{+\infty} D^{(AB)}(i\omega_n)\exp(i\omega_n\tau),$$

where $\omega_n = 2\pi n/\beta$. If we denote the analytic continuation of $D^{(AB)}(i\omega_n)$ off the imaginary axis into the appropriate half-plane by $D^{(AB)}(z)$, then it is useful to introduce the spectral density $\Gamma^{(AB)}(\Omega)$, defined by

$$\Gamma^{(AB)}(\Omega) = (1/2\pi i)[D^{(AB)}(\Omega+i\epsilon) - D^{(AB)}(\Omega-i\epsilon)]. \quad (17)$$

One may express a number of expectation values in terms of $\Gamma^{(AB)}(\Omega)$. In particular, if

$$n(\Omega) = [\exp(\beta\Omega) - 1]^{-1}$$

is the Bose-Einstein function, then

$$\langle AB\rangle = \int_{-\infty}^{+\infty} d\Omega\, n(\Omega)\Gamma^{(AB)}(\Omega)$$

and

$$\langle A[B,H]\rangle = -\langle[A,H]B\rangle = \int_{-\infty}^{+\infty} d\Omega\, \Omega n(\Omega)\Gamma^{(AB)}(\Omega).$$

The correlation functions may in principle be computed from the equations of motion, which describe the time development of D. One has

$$[\partial/\partial\tau]D^{(AB)}(\tau) + \langle T\{[A(\tau),H]B(0)\}\rangle = \langle[A(0),B(0)]\rangle\delta(\tau).$$

The Green's functions of interest in the present work are the functions

$$D^{(aa)}(\mathbf{l}_a, \mathbf{l}_a'; \tau) = \langle T(a_{\mathbf{l}_a}^{\dagger}(\tau)a_{\mathbf{l}_a'}(0))\rangle,$$
$$D^{(ba)}(\mathbf{l}_b, \mathbf{l}_a; \tau) = \langle T(b_{\mathbf{l}_b}(\tau)a_{\mathbf{l}_a}(0))\rangle,$$
$$D^{(bb)}(\mathbf{l}_b, \mathbf{l}_b'; \tau) = \langle T(b_{\mathbf{l}_b}^{\dagger}(\tau)b_{\mathbf{l}_b'}(0))\rangle.$$

In Sec. II, where the properties of the surface magnons were deduced from the operator equations of motion, the Hamiltonian employed described a semi-infinite array of spins. In order to obtain expressions for the Green's functions just defined, a different procedure will be convenient. We follow Maradudin and Wallis[8] and begin with a large macroscopic cube of perfect crystal with sides of length L. The periodic boundary conditions are imposed on all physical quantities, in the standard manner. Two free (100) surfaces are then created by setting to zero all interactions between spins in two adjacent, parallel (100)

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[18] A. U. MacRae, Science **139**, 379 (1963).

planes. Specifically, we shall set equal to zero the exchange couplings between B spins in the plane with z coordinate zero, and A spins in the plane with z coordinate $+\frac{1}{2}a$. With this procedure, one (100) surface of A spins and one (100) surface of B spins are obtained.

If desired, additional pinning fields, along with changes in the exchange constants near the surface, may be included in the Hamiltonian in a straightforward manner. As mentioned earlier, we believe that the insensitivity of the surface magnon frequency to changes in these quantities when $\omega_E \gg \omega_A$ suggests that a reliable estimate of the surface effects may be obtained by ignoring these complications. This approximation will greatly simplify the algebra in the remainder of the paper.

With the preceding remarks in mind, we write the Hamiltonian in the form

$$H = H_0 + V,$$

where H_0 is defined in Eq. (2), and

$$V = -\tfrac{1}{2}\omega_E \sum_{\mathbf{l}_a} a_{\mathbf{l}_a}{}^\dagger a_{\mathbf{l}_a} \Delta(l_a{}^z - \tfrac{1}{2}a) - \tfrac{1}{2}\omega_E \sum_{\mathbf{l}_b} b_{\mathbf{l}_b}{}^\dagger b_{\mathbf{l}_b} \Delta(l_b{}^z) - \tfrac{1}{8}\omega_E \sum_{\mathbf{l}_a} \sum_{\delta} [a_{\mathbf{l}_a} b_{\mathbf{l}_a+\delta} + a_{\mathbf{l}_a}{}^\dagger b_{\mathbf{l}_a+\delta}{}^\dagger] \Delta(l_a{}^z - \tfrac{1}{2}a) \Delta(\delta_z + \tfrac{1}{2}a). \quad (18)$$

The definition of the function $\Delta(x)$ is given after Eq. (2).

We shall consider in detail the functions $D^{(aa)}(\mathbf{l}_a, \mathbf{l}_a'; \tau)$ and $D^{(ba)}(\mathbf{l}_b, \mathbf{l}_a; \tau)$, since it will become apparent in the subsequent discussion that knowledge of these two functions will enable us to discuss the properties of the system of interest in this study. By employing the equation of motion obtained from differentiating the functions with respect to the imaginary time variable τ, then introducing the Fourier transform with respect to τ, one finds

$$[i\omega_n - (\omega_H + \omega_A + \omega_E)] D^{(aa)}(\mathbf{l}_a, \mathbf{l}_a'; i\omega_n) - \tfrac{1}{8}\omega_E \sum_{\delta} D^{(ba)}(\mathbf{l}_a + \boldsymbol{\delta}, \mathbf{l}_a'; i\omega_n) = -\Delta(\mathbf{l}_a - \mathbf{l}_a') - \tfrac{1}{2}\omega_E \Delta(l_a{}^z - \tfrac{1}{2}a) D^{(aa)}(\mathbf{l}_a, \mathbf{l}_a'; i\omega_n) - \tfrac{1}{8}\omega_E \Delta(l_a{}^z - \tfrac{1}{2}a) \sum_{\delta} \Delta(\delta^z + \tfrac{1}{2}a) D^{(ba)}(\mathbf{l}_a + \boldsymbol{\delta}, \mathbf{l}_a'; i\omega_n),$$

$$[i\omega_n + (\omega_A + \omega_E - \omega_H)] D^{(ba)}(\mathbf{l}_b, \mathbf{l}_a, i\omega_n) + \tfrac{1}{8}\omega_E \sum_{\delta} D^{(aa)}(\mathbf{l}_b + \boldsymbol{\delta}, \mathbf{l}_a; i\omega_n) = +\tfrac{1}{2}\omega_E \Delta(l_b{}^z; 0) D^{(ba)}(\mathbf{l}_b, \mathbf{l}_a, i\omega_n) + \tfrac{1}{8}\omega_E \Delta(l_b{}^z) \sum_{\delta} \Delta(\delta^z - \tfrac{1}{2}a) D^{(aa)}(\mathbf{l}_b + \boldsymbol{\delta}, \mathbf{l}_a'; i\omega_n). \quad (19)$$

In the subsequent discussion, the quantity $i\omega_n$ will be replaced by the complex variable z, since it will be convenient to employ values of $D^{(aa)}(\mathbf{l}_a, \mathbf{l}_a'; z)$ for z near the real axis.

Since the translational invariance of the system with respect to (discrete) translations parallel to the x and y axes has not been destroyed by creating the surfaces, $D^{(aa)}(\mathbf{l}_a, \mathbf{l}_a'; z)$ is a function only of $(l_a{}^y - l_a'{}^y)$ and $(l_a{}^x - l_a'{}^x)$. However, $D^{(aa)}$ depends on both $l_a{}^z$ and $l_a'{}^z$. We introduce the following Fourier expansion with respect to the spatial variables:

$$D^{(aa)}(\mathbf{l}_a, \mathbf{l}_a'; z) = N^{-1} \sum_{\mathbf{k}_{||}} \sum_{k_z k_z'} \exp[i\mathbf{k}_{||} \cdot (\mathbf{l}_a' - \mathbf{l}_a) + ik_z' l_a'{}^z - ik_z l_a{}^z] D^{(aa)}(\mathbf{k}_{||} k_z k_z'; z). \quad (20)$$

Recall that $\mathbf{k}_{||} = \hat{x}k_x + \hat{y}k_y$. The function $D^{(ba)}(\mathbf{l}_b, \mathbf{l}_a; z)$ will be Fourier analyzed in the same manner. From Eqs. (19), one may obtain equations satisfied by the Fourier transforms introduced in Eq. (20). We find

$$[z - (\omega_H + \omega_A + \omega_E)] D^{(aa)}(\mathbf{k}_{||} k_z k_z'; z) - \omega_E \lambda(\mathbf{k}_{||}) \cos(\tfrac{1}{2}ak_z) D^{(ba)}(\mathbf{k}_{||} k_z k_z'; z) = -\Delta(k_z - k_z') - (\omega_E/2L) \exp[i\tfrac{1}{2}(ak_z)] \sum_{k_z''} \exp[-i\tfrac{1}{2}(ak_z'')] D^{(aa)}(\mathbf{k}_{||} k_z'' k_z'; z) - (\omega_E/2L) \lambda(\mathbf{k}_{||}) \exp[i\tfrac{1}{2}(ak_z)] \sum_{k_z''} D^{(ba)}(\mathbf{k}_{||} k_z'' k_z'; z) \quad (21a)$$

and

$$[z + (\omega_A + \omega_E - \omega_H)] D^{(ba)}(\mathbf{k}_{||} k_z k_z'; z) + \omega_E \lambda(\mathbf{k}_{||}) \cos(\tfrac{1}{2}ak_z) D^{(aa)}(\mathbf{k}_{||} k_z k_z'; z) = +(\omega_E/2L) \sum_{k_z''} D^{(ba)}(\mathbf{k}_{||} k_z'' k_z'; z) + (\omega_E/2L) \lambda(\mathbf{k}_{||}) \sum_{k_z''} \exp[-i\tfrac{1}{2}(ak_z'')] D^{(aa)}(\mathbf{k}_{||} k_z'' k_z'; z), \quad (21b)$$

where

$$\lambda(\mathbf{k}_{||}) = \cos(\tfrac{1}{2}akx) \cos(\tfrac{1}{2}ak_y).$$

From Eqs. (21a) and (21b), it is evident that the Green's functions in the presence of the external magnetic field may be obtained from the zero-field functions by replacing the frequency variable z by $z - \omega_H$. In the discussion, we shall set $\omega_H = 0$, and take note of the preceding remarks. The functions $D^{(bb)}$ and $D^{(ab)}$ satisfy a set of coupled equations similar in structure to Eqs. (21), but the effect of finite ω_H is to replace z by $z + \omega_H$ rather than $z - \omega_H$.

It will be convenient to introduce the quantities

$$M^{(ba)}(\mathbf{k}_{||}k_z';z)=(1/L)\sum_{k_z''}D^{(ba)}(\mathbf{k}_{||}k_z''k_z';z)$$

and

$$M^{(aa)}(\mathbf{k}_{||}k_z';z)=(1/L)\sum_{k_z''}\exp[-ik_z''\tfrac{1}{2}a]\times D^{(aa)}(\mathbf{k}_{||}k_z''k_z';z).$$

Upon rearranging Eqs. (21), one may express the Green's functions in terms of $M^{(aa)}$ and $M^{(ba)}$. To simplify the expressions, let us introduce the quantities

$$\omega_m=\omega_A+\omega_E,$$

$$\omega_b=\omega_E\lambda(\mathbf{k}_{||}),$$

and

$$\omega(\mathbf{k})=[\omega_m^2-\omega_b^2\cos^2(\tfrac{1}{2}ak_z)]^{1/2}.$$

The function $\omega(\mathbf{k})$ is the frequency of a bulk spin wave of wave vector $\mathbf{k}$, and ω_m is the maximum spin-wave frequency in the infinitely extended medium. Then we find

$$D^{(aa)}(k_{||}k_zk_z';z)=\Delta(k_z-k_z')\frac{[z+\omega_m]}{\omega^2(\mathbf{k})-z^2}-\frac{1}{2}\frac{[\omega_b^2\cos(\frac{1}{2}ak_z)-\omega_E(z+\omega_m)\exp(i\frac{1}{2}ak_z)]}{\omega^2(\mathbf{k})-z^2}M^{(aa)}(\mathbf{k}_{||}k_z';z)$$
$$-\frac{1}{2}\frac{\omega_b[\omega_E\cos(\frac{1}{2}ak_z)-(z+\omega_m)\exp(i\frac{1}{2}ak_z)]}{\omega^2(\mathbf{k})-z^2}M^{(ba)}(\mathbf{k}_{||}k_z';z) \quad (22a)$$

and

$$D^{(ba)}(\mathbf{k}_{||}k_zk_z';z)=-\Delta(k_z-k_z')\frac{\omega_b\cos(\frac{1}{2}ak_z)}{\omega^2(\mathbf{k})-z^2}-\frac{1}{2}\frac{[\omega_b^2\cos(\frac{1}{2}ak_z)\exp(i\frac{1}{2}ak_z)+\omega_E(z-\omega_m)]}{\omega^2(\mathbf{k})-z^2}M^{(ba)}(\mathbf{k}_{||}k_z';z)$$
$$-\frac{1}{2}\frac{[\omega_E\cos(\frac{1}{2}ak_z)\exp(i\frac{1}{2}ak_z)+(z-\omega_m)]}{\omega^2(\mathbf{k})-z^2}M^{(aa)}(\mathbf{k}_{||}k_z';z). \quad (22b)$$

Two inhomogeneous equations for $M^{(ba)}$ and $M^{(aa)}$ may be obtained by multiplying Eq. (22a) by $\exp(-i\frac{1}{2}ak_z)$ and summing over k_z, then summing Eq. (22b) over k_z. This gives

$$\alpha(\mathbf{k}_{||},z)M^{(aa)}(\mathbf{k}_{||}k_z,z)+\beta(\mathbf{k}_{||},z)M^{(ba)}(\mathbf{k}_{||}k_z,z)=L^{-1}[(z+\omega_m)\exp(-i\tfrac{1}{2}ak_z)]/[\omega^2(\mathbf{k})-z^2]$$

and

$$\beta(\mathbf{k}_{||},-z)M^{(aa)}(\mathbf{k}_{||}k_z,z)+\alpha(\mathbf{k}_{||},-z)M^{(ba)}(\mathbf{k}_{||}k_z,z)=-L^{-1}[\omega_b\cos(\tfrac{1}{2}ak_z)]/[\omega^2(\mathbf{k})-z^2], \quad (23)$$

where

$$\alpha(\mathbf{k}_{||},z)=(1/L)\sum_{k_z}[(\omega_m+z)(\omega_m-\tfrac{1}{2}\omega_E-z)-\tfrac{1}{2}(\omega_b^2)\cos^2(\tfrac{1}{2}ak_z)]/[\omega^2(\mathbf{k})-z^2],$$

and

$$\beta(\mathbf{k}_{||},z)=(\omega_b/2L)\sum_{k_z}[\omega_E^2\cos^2(\tfrac{1}{2}ak_z)-(z+\omega_m)]/[\omega^2(\mathbf{k})-z^2].$$

Equation (23) may be solved for $M^{(aa)}$ and $M^{(ba)}$, then the result inserted in Eqs. (22a) and (22b) to find the Green's function. For the moment, we shall examine in detail the properties of the function $D^{(aa)}$, since a number of properties of the system may be described knowing only $D^{(aa)}$. Let us define the quantities

$$D_0(\mathbf{k}_{||}k_z;z^2)=[\omega^2(\mathbf{k})-z^2]^{-1},$$

$$F(\mathbf{k}_{||};z^2)=(1/L)\sum_{k_z}[\omega^2(\mathbf{k})-z^2]^{-1}.$$

Then

$$D^{(aa)}(\mathbf{k}_{||}k_zk_z';z)=\Delta(k_z-k_z')(z+\omega_m)D_0(\mathbf{k}_{||}k_z;z^2)+[D_0(\mathbf{k}_{||}k_z;z)\{\cos(\tfrac{1}{2}ak_z)\cos(\tfrac{1}{2}ak_z')N_1(\mathbf{k}_{||},z)$$
$$+[\cos(\tfrac{1}{2}ak_z)\exp(-i\tfrac{1}{2}ak_z')+\exp(i\tfrac{1}{2}ak_z)\cos(\tfrac{1}{2}ak_z')]N_2(\mathbf{k}_{||},z)$$
$$+\exp(i\tfrac{1}{2}ak_z)\exp(-i\tfrac{1}{2}ak_z')N_3(\mathbf{k}_{||},z)\}D_0(\mathbf{k}_{||}k_z';z)][\alpha(\mathbf{k}_{||},z)\alpha(\mathbf{k}_{||},-z)-\beta(\mathbf{k}_{||},z)\beta(\mathbf{k}_{||},-z)]^{-1}, \quad (24)$$

where

$$N_1(\mathbf{k}_{||}, z) = \omega_b^2[\omega_E - \tfrac{1}{2}\omega_E^2(\omega_m + z)(1-\lambda^2)F], \quad (25a)$$

$$N_2(\mathbf{k}_{||}, z) = \tfrac{1}{2}\omega_E^2(\omega_m + z) \times [(\omega_m^2 - z^2)(1-\lambda^2)F - (1+\lambda^2)], \quad (25b)$$

and

$$N_3(\mathbf{k}_{||}, z) = (\omega_m + z)^2[\omega_E - \tfrac{1}{2}\omega_E^2(\omega_m - z)(1-\lambda^2)F]. \quad (25c)$$

From Eq. (17), and the equations that follow, it is evident that knowledge of the spectral function $\Gamma^{(aa)}(\Omega)$ enables a number of averages of physical quantities to be computed. This function is related to the singularities in $D^{(aa)}(\Omega)$ on the real axis. From Eq. (24), one sees that the function $D^{(aa)}(\mathbf{k}_{||}k_z k_z'; z)$ has singularities at $z = \omega(\mathbf{k}_{||}k_z)$ and $z = \omega(\mathbf{k}_{||}k_z')$. The contribution to $\Gamma(\Omega)$ from the first term will provide a description of the perfect extended medium, with the free surfaces absent. The contribution from the singularities at $\omega(\mathbf{k}_{||}k_z)$ and $\omega(\mathbf{k}_{||}k_z')$ in the second term will describe the change in the spectral function associated with the modification of the bulk modes, since these singularities occur in the range of frequencies associated with the extended spin-wave modes of the unperturbed host. We now show that the denominator $\alpha(\mathbf{k}_{||}, z)\alpha(\mathbf{k}_{||}, -z) - \beta(\mathbf{k}_{||}, z)\beta(\mathbf{k}_{||}, -z)$ has a zero at the frequency of the surface spin-wave mode of wave vector $\mathbf{k}_{||}$.

Let z lie on the real axis, below the bulk spin-wave frequencies associated with the bulk waves of wave vector $\mathbf{k}_{||}$. We replace z by Ω in what follows, where $|\Omega| < (\omega_m^2 - \omega_b^2)^{1/2}$. The function F may then be evaluated explicitly:

$$F(\mathbf{k}_{||}, \Omega^2) = [(\omega_m^2 - \Omega^2)(\omega_m^2 - \omega_b^2 - \Omega^2)]^{-1/2}, \quad |\Omega| < (\omega_m^2 - \omega_b^2)^{1/2}.$$

After a bit of rearranging, α and β may be expressed in terms of F. We find

$$\alpha(\mathbf{k}_{||}, \Omega) = \tfrac{1}{2} + \tfrac{1}{2}(\omega_m + \Omega)(\omega_m - \omega_E - \Omega)F,$$

$$\beta(\mathbf{k}_{||}, \Omega) = -(1/2\lambda)[1 - (\omega_m + \Omega)(\omega_m - \omega_E\lambda^2 - \Omega)F]. \quad (26)$$

Then after some algebra, one finds

$$d(\mathbf{k}_{||}, \Omega) \equiv \alpha(\mathbf{k}_{||}, \Omega)\alpha(\mathbf{k}_{||}, -\Omega) - \beta(\mathbf{k}_{||}, \Omega)\beta(\mathbf{k}_{||}, -\Omega) = (1/2\lambda^2)\{[(\omega_m^2 - \Omega^2)(1+\lambda^2) - 2\omega_m\omega_E\lambda^2]F - (1-\lambda^2)\}. \quad (27)$$

Now consider the quantity

$$\left\{(\omega_m^2 - \Omega^2)(1+\lambda^2) - 2\omega_m\omega_E\lambda^2 + \frac{(1-\lambda^2)}{F}\right\}\left[2\lambda^2\frac{d(\mathbf{k}_{||}, \Omega)}{F}\right] = [(\omega_m^2 - \Omega^2)(1+\lambda^2) - 2\omega_m\omega_E\lambda^2]^2 - (1-\lambda^2)^2(\omega_m^2 - \Omega^2)(\omega_m^2 - \omega_b^2 - \Omega^2).$$

After manipulating the right side of this last equation and utilizing Eq. (11) for the case $\omega_H = 0$, we find

$$\frac{d(\mathbf{k}_{||}, \Omega)}{2F}\left[(\omega_m^2 - \Omega^2)(1+\lambda^2) - 2\omega_m\omega_E\lambda^2 + \frac{(1-\lambda^2)}{F}\right] = (\Omega^2 - \Omega_{s1}^2)(\Omega^2 - \Omega_{s2}^2), \quad (28)$$

where Ω_{s1} and Ω_{s2} are the two solutions of Eq. (11). Notice from Eq. (27) that $d(\mathbf{k}_{||}, \Omega)$ is a function only of Ω^2. The right side of Eq. (28) has a zero when $\Omega^2 = \Omega_{s1}^2$ and $\Omega^2 = \Omega_{s2}^2$. Let us label the roots of Eq. (11) so that $\Omega_{s2}^2 \geq \Omega_{s1}^2$. Now recall from the discussion of Sec. II that the root Ω_{s2} corresponds to an unphysical solution to Eq. (11) in which the spin deviation increases exponentially as one moves away from the surface. We now show from Eqs. (27) and (28) that $d(\mathbf{k}_{||}, \Omega)$ has a zero only at the physical root Ω_{s1}^2.

In Fig. 3, we have plotted the functions $\pm(1-\lambda^2)/F$ and $(\omega_m^2 - \Omega^2)(1+\lambda^2) - 2\omega_m\omega_E\lambda^2$ for $0 < \Omega^2 < \omega_m^2 - \omega_b^2$. From the figure, one sees that the function $d(\mathbf{k}_{||}, \Omega^2)$ has only a *single* zero in the interval $0 < \Omega^2 < \omega_m^2 - \omega_b^2$. From Eq. (28), it is then apparent that the quantity in square brackets on the left side of Eq. (28) has a zero in the interval. Consulting Fig. 3 once again, it is evident that the zero of $d(\mathbf{k}_{||}, \Omega)$ occurs at a *smaller* value of Ω^2 than the zero of this last-mentioned quantity. From Eq. (28), it follows that $d(\mathbf{k}_{||}, \Omega)$ has a zero at the physical surface magnon frequency Ω_{s1}^2, while the quantity in square brackets on the left side of Eq. (28) has a zero at the unphysical frequency Ω_{s2}^2.

We conclude this section with one final remark. In the discussion of the semi-infinite sample given in Sec. II, the surface magnon branch was found to be nondegenerate. For a given value of λ, the Green's function discussed in this section has one pole at $\Omega = \Omega_{s1}$, and $\Omega = -\Omega_{s1}$, so the surface mode is twofold degenerate.

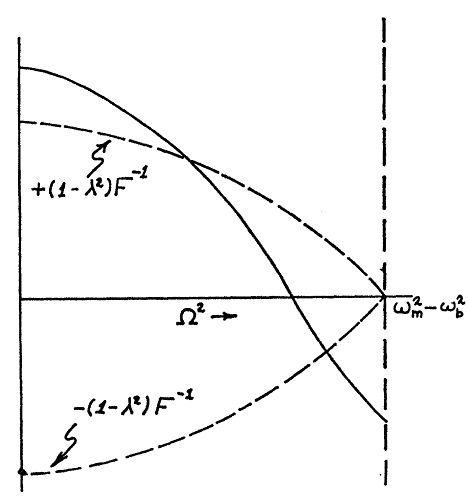

FIG. 3. Graphical representation of the functions encountered in the demonstration that $d(\mathbf{k}_{||}, \Omega)$ has a zero only at the physical surface magnon frequency. The solid line is a sketch of the function $[\omega_m^2 - \Omega^2](1+\lambda^2) - 2\omega_m\omega_E\lambda^2$.

The reason for this is that by "cutting the bonds" in the manner described at the beginning of the present section, two free surfaces were formed. Thus for a given λ, two modes occur, one localized about each surface. One surface layer consists of A spins, pointing upward. A positive frequency mode is associated with this surface. The second surface consists of B spins pointing downward, and gives rise to a negative frequency mode. These statements may be verified by examining the equations of motion for the two geometries that result from the bond-cutting procedure.

IV. SURFACE CONTRIBUTION TO THE SPECIFIC HEAT

The Green's functions discussed in Sec. III will now be applied to compute the contribution to the internal energy and specific heat proportional to the surface area. As mentioned earlier, there are two contributions to the surface specific heat. First, a contribution from the thermally exited surface modes is obtained. It must also be realized that the distribution in frequency of the bulk modes is altered by the presence of the surfaces. The change in the frequency distribution of the bulk waves also produces a change in the thermodynamic functions proportional to the surface area.

Before embarking on a discussion of the details of the calculation, we first make some comments on some approximations employed below. The Green's functions exhibited in Eqs. (22) and (24) have been obtained exactly, within the framework of the spin-wave theory of the Hamiltonian of the model crystal. In order to simplify the results obtained in the remainder of the paper, we shall assume that the exchange frequency ω_E is large compared to the anisotropy frequency ω_A. If ω_E and ω_A are comparable in magnitude, the results assume a more complex appearance. In addition, the discussion in Sec. II indicates that when ω_E and ω_A are comparable, one must consider the effect of changes in the anisotropy field near the surface in order to obtain a realistic description of the surface effects.

We begin with some remarks on the formalism. From the Hamiltonian of Eq. (4), one finds the identity

$$H=\sum_{\mathbf{l}_a} a_{\mathbf{l}_a}{}^{\dagger}[a_{\mathbf{l}_a}, H]-\sum_{\mathbf{l}_b}[b_{\mathbf{l}_b}{}^{\dagger}, H]b_{\mathbf{l}_b}.$$

Then employing an identity exhibited after Eq. (17), the internal energy $U(T)$ of the spin system at temperature T may be written

$$U(T)=\int_{-\infty}^{+\infty} d\Omega\, n(\Omega)\Omega\Big[\sum_{\mathbf{l}_a}\Gamma^{(aa)}(\mathbf{l}_a, \mathbf{l}_a, \Omega)+\sum_{\mathbf{l}_b}\Gamma^{(bb)}(\mathbf{l}_b, \mathbf{l}_b, \Omega)\Big],$$

where the spectral functions $\Gamma^{(aa)}(\mathbf{l}_a, \mathbf{l}_a, \Omega)$ and $\Gamma^{(bb)}(\mathbf{l}_b, \mathbf{l}_b, \Omega)$ are related to the Green's functions in the manner indicated in Eq. (17). In zero magnetic field, symmetry considerations imply that the contribution to U from the B sublattice is identical to the contribution of the A sublattice. Then

$$U(T)=2\int_{-\infty}^{+\infty} d\Omega\, n(\Omega)\Omega\sum_{\mathbf{l}_a}\Gamma^{(aa)}(\mathbf{l}_a, \mathbf{l}_a, \Omega). \quad (29)$$

In the antiferromagnet, it is well known that even at $T=0$, the spins are not perfectly aligned parallel or antiparallel to the z axis, but execute zero-point motion about this axis. Thus the energy of spin motion $U(T)$ is finite at $T=0$, since the zero-point motion elevates the energy of the ground state above the value expected for the perfectly aligned state. Since short-wavelength magnons contribute to the zero-point energy, an analytic calculation of the surface contribution to this quantity will be difficult. To compute the specific heat, only the temperature-dependent portion of Eq. (29) is required. When $T\ll T_N$, only long-wavelength magnons contribute to the temperature-dependent part of $U(T)$. Noting that $n(-\Omega)=-1-n(\Omega)$, Eq. (29) may be written in the alternative form

$$U(T)=2\int_0^{\infty} d\Omega\,\Omega\sum_{\mathbf{l}_a}\Gamma^{(aa)}(\mathbf{l}_a, \mathbf{l}_a, -\Omega)$$
$$+2\int_0^{\infty} d\Omega\,\Omega n(\Omega)\sum_{\mathbf{l}_a}[\Gamma^{(aa)}(\mathbf{l}_a, \mathbf{l}_a, \Omega)+\Gamma^{(aa)}(\mathbf{l}_a, \mathbf{l}_a, -\Omega)]$$
$$=U_0+\Delta U(T). \quad (30)$$

The first term on the right side of Eq. (30) is the temperature-independent zero-point energy of the system, while the remaining terms, which vanish as $T\to 0$, represent the contribution from the thermally excited spin waves. In the remainder of this section, we discuss only $\Delta U(T)$. Upon introducing the Fourier transform with respect to the spatial variables, Eq. (20), one has

$$\Delta U(T)=2\sum_{\mathbf{k}_{||}}\sum_{k_z}\int_0^{\infty} d\Omega\,\Omega n(\Omega)$$
$$\times[\Gamma^{(aa)}(\mathbf{k}_{||}k_zk_z', \Omega)+\Gamma^{(aa)}(\mathbf{k}_{||}k_zk_z, -\Omega)]. \quad (31)$$

The first term in the Green's function of Eqs. (22a) and (22b) gives the contribution to ΔU for the infinitely extended medium. We denote this contribution to $\Gamma^{(aa)}$ by $\Gamma_0{}^{(aa)}$. One easily finds

$$\Gamma_0{}^{(aa)}(\mathbf{k}_{||}k_zk_z', \Omega)$$
$$=[(\omega_m+\Omega)/2\omega(k)][\delta(\Omega-\omega(\mathbf{k}))-\delta(\Omega+\omega(\mathbf{k}))],$$

where $\omega(\mathbf{k})$ is the bulk magnon frequency given in Sec. III. The contribution to $\Delta U_0(T)$ from $\Gamma_0{}^{(aa)}$ is then the standard result

$$\Delta U_0(T)=2\sum_{\mathbf{k}}\omega(\mathbf{k})n(\omega(\mathbf{k})). \quad (32)$$

In the subsequent discussion, it will prove useful to employ the expression for $\Delta U(T)$ in the two temperature regions $k_BT\ll(2\omega_E\omega_A)^{1/2}$ and $(2\omega_E\omega_A)^{1/2}\ll k_BT\ll k_BT_N$. We briefly discuss these regimes separately.

(1) $k_BT\ll(2\omega_E\omega_A)^{1/2}$: The expression in Eq. (32)

may be evaluated employing an expression for $\omega(\mathbf{k})$ valid for $ka \ll 1$:

$$\omega(\mathbf{k}) = (2\omega_E\omega_A)^{1/2} + Dk^2,$$

where $D = \frac{1}{16}\sqrt{2}\omega_E(\omega_E/\omega_A)^{1/2}a^2$. Then the dominant term in $\Delta U_0(T)$ is

$$\Delta U_0(T) = [16V/(2^{1/2}\pi^3)^{1/2}a^3]\omega_A(\omega_A/\omega_E)^{1/4}(k_BT/\omega_E)^{3/2} \times \exp[-(2\omega_E\omega_A)^{1/2}/k_BT], \quad (33)$$

where V is the volume of the crystal.

(2) $(2\omega_E\omega_A)^{1/2} \ll k_BT \ll k_BT_N$: One may let $\omega_A \to 0$ in this regime, since k_BT is large compared to the energy gap $(2\omega_E\omega_A)^{1/2}$. Then the magnon dispersion relation is roughly linear for frequencies the order of k_BT. For $ka \ll 1$, one has

$$\omega(\mathbf{k}) \cong \tfrac{1}{2}\omega_E ak.$$

Then

$$\Delta U_0(T) = (8V/\pi^2a^3)[(k_BT)^4/\omega_E^3]\Gamma(4)\zeta(4), \quad (34)$$

where $\Gamma(x)$ and $\zeta(x)$ are the gamma function and Riemann zeta function, respectively.

To evaluate the surface contribution to $\Delta U(T)$, let us denote the part of $D^{(aa)}(\mathbf{k}_{||}k_zk_z, z)$ that results from the presence of the surfaces by $\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z, z)$. Then performing some manipulations on Eq. (24) yields

$$\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z; z) = +L^{-1}\frac{[(z+\omega_m)(1+\lambda^2)-\omega_b\lambda]-(z+\omega_m)(1-\lambda^2)(\omega_m^2-z^2-\frac{1}{2}\omega_b^2)F}{2\lambda^2 d(\mathbf{k}_{||}, z)[\omega^2(\mathbf{k})-z^2]}$$
$$+L^{-1}\frac{(z+\omega_m)\{2\omega_m\omega_b\lambda-[1+\lambda^2](\omega_m^2-z^2)\}+(z+\omega_m)(\omega_m^2-z^2)(\omega_m^2-\omega_b^2-z^2)(1-\lambda^2)F}{2\lambda^2 d(\mathbf{k}_{||}, z)[\omega^2(\mathbf{k})-z^2]^2}. \quad (35)$$

This result may be obtained from Eq. (24) by employing the identity

$$\omega_b^2\cos^2(\tfrac{1}{2}ak_z)/[\omega^2(\mathbf{k})-z^2] = (\omega_m^2-z^2)/[\omega^2(\mathbf{k})-z^2]-1.$$

At this point one may compute the contribution $\Delta\Gamma^{(aa)}(\mathbf{k}_{||}k_zk_z; z)$ from the presence of the surfaces utilizing Eq. (35). However, it will be more convenient to first sum the result of Eq. (35) over k_z, then compute the discontinuity of the result across the real axis. To do this, it will be convenient to consider two regimes of frequency separately.

(a) The case $0 < \Omega^2 < \omega_m^2 - \omega_b^2$, where $z = \Omega \pm i\epsilon$. The contribution of the surface modes to the function $\Delta\Gamma^{(aa)}$ will come from this region. Upon summing both sides of Eq. (35) over k_z, one obtains

$$\sum_{k_z}\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z; z) = [2\lambda^2 d(\mathbf{k}_{||}, z)]^{-1}\{[(z+\omega_m)(1+\lambda^2)-\omega_b\lambda-(z+\omega_m)(1-\lambda^2)(\omega_m^2-\tfrac{1}{2}\omega_b^2-z^2)F]F$$
$$+[(z+\omega_m)(2\omega_m\omega_b\lambda-(1+\lambda^2)(\omega_m^2-z^2))+(z+\omega_m)(\omega_m^2-z^2)(\omega_m^2-\omega_b^2-z^2)(1-\lambda^2)F]\partial F/\partial(z^2)\}. \quad (36a)$$

Now notice that

$$\partial F/\partial(z^2) = (\omega_m^2-\tfrac{1}{2}\omega_b^2-z^2)F^3.$$

Then one has

$$\sum_{k_z}\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z; z) = [2\lambda^2 d(\mathbf{k}_{||}, z)][(z+\omega_m)\{(1+\lambda^2)F-[(\omega_m^2-z^2)(1+\lambda^2)-2\omega_m\omega_b\lambda]\partial F/\partial(z^2)\}-\omega_b\lambda F]. \quad (36b)$$

From Eq. (27), it follows that

$$2\lambda^2[\partial/\partial(z^2)]d(\mathbf{k}_{||}, z) = [(\omega_m^2-z^2)(1+\lambda^2)-2\omega_m\omega_b\lambda]\partial F/\partial(z^2)-(1+\lambda^2)F.$$

Then finally

$$\sum_{k_z}\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z; z) = -\frac{(z+\omega_m)}{d(\mathbf{k}_{||}, z)}\frac{\partial d(\mathbf{k}_{||}, z)}{\partial(z^2)} - \omega_b\lambda\frac{F}{d(\mathbf{k}_{||}, z)}. \quad (36c)$$

From the discussion of Sec. III, it is apparent that $d(\mathbf{k}_{||}, z)$ has only a single zero on the real axis, for $0 < \Omega^2 < \omega_m^2 - \omega_b^2$. This zero occurs at $z^2 = \Omega_s^2$, where Ω_s is the surface magnon frequency. Then for z^2 near Ω_s^2, we have

$$d(\mathbf{k}_{||}, z) = [z^2 - \Omega_s^2]\partial d(\mathbf{k}_{||}, z)/\partial z^2$$

and

$$\sum_{k_z}\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z; z) = \frac{\omega_m+z}{\Omega_s^2-z^2} + \left(\frac{\omega_b\lambda}{\partial d/\partial z^2}\right)\frac{F}{\Omega_s^2-z^2}.$$

Using this form, the surface magnon mode contribution to the spectral density is found to be

$$\Delta\Gamma_s^{(aa)}(\mathbf{k}_{||}, \Omega) + \Delta\Gamma_s^{(aa)}(\mathbf{k}_{||}, -\Omega) = \delta(\Omega - \Omega_s(\mathbf{k}_{||})) + \delta(\Omega + \Omega_s(\mathbf{k}_{||})),$$

where

$$\Delta\Gamma_s^{(aa)}(\mathbf{k}_{||}, \Omega) = (1/2\pi i) \times \sum_{k_z}[\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z; \Omega+i\epsilon) - \Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z; \Omega-i\epsilon)]$$

for

$$0<\Omega^2<\omega_m^2-\omega_b^2.$$

The contribution of the surface spin-wave modes to the temperature-dependent part of the internal energy is then

$$\Delta U_s(T)=2\sum_{\mathbf{k}_{||}}\Omega_s(\mathbf{k}_{||})n(\Omega_s(\mathbf{k}_{||})). \quad (37)$$

This is the result that one would obtain in a straightforward manner from elementary considerations. The factor of 2 is present because the bond-cutting procedure employed in this calculation produces two free surfaces. For each value of $\mathbf{k}_{||}$, there are two surface modes, one localized about each surface.

To evaluate the contribution to $\Delta U(T)$ from the surface modes, it is again convenient to calculate $\Delta U_s(T)$ separately, as we did earlier in this section when the contribution from the unperturbed bulk waves were discussed.

(i) $k_BT\ll(\omega_E\omega_A)^{1/2}$. In the limit that $\omega_A\ll\omega_E$, for small values of $k_{||}a$, one has

$$\Omega_s(\mathbf{k}_{||})\cong(\omega_E\omega_A)^{1/2}+Dk_{||}^2,$$

where

$$D=\tfrac{1}{16}\omega_E(\omega_E/\omega_A)^{1/2}a^2.$$

The sum over $\mathbf{k}_{||}$ in Eq. (37) may be converted to an integral in the standard manner. In the low-temperature limit, the dominant term in $\Delta U_s(T)$ is

$$\Delta U_s(T)=4S/\pi a^2(\omega_A/\omega_E)k_BT\exp[-(\omega_E\omega_A)^{1/2}/k_BT].$$

In this expression, the total area of the two free surfaces is S. The specific heat associated with the surface waves may be obtained by differentiating this result with respect to temperature. One has

$$C_s(T)=k_B\frac{4S}{\pi a^2}\left(\frac{\omega_A}{\omega_E}\right)\frac{(\omega_E\omega_A)^{1/2}}{k_BT}\exp[-(\omega_E/\omega_A)^{1/2}/k_BT]. \quad (38)$$

We shall make a numerical estimate of the magnitude of the surface wave contribution later in this section.

(ii) $(\omega_E/\omega_A)^{1/2}\ll k_BT\ll k_BT_N$. The dispersion relation for the surface magnons varies in a linear manner with $|\mathbf{k}_{||}|$ when $\hbar\Omega_s(\mathbf{k}_{||})\cong k_BT$. One has

$$\Omega_s(\mathbf{k}_{||})\cong\tfrac{1}{4}\sqrt{2}\omega_Eak_{||}.$$

Then

$$\Delta U_s(T)\cong 8S/\pi a^2[(k_BT)^3/\omega_E^2]\zeta(3)$$

when $\omega_E\gg\omega_A$. Consequently,

$$C_S(T)=k_B(24S/\pi a^2)(k_BT/\omega_E)^2\zeta(3). \quad (39)$$

We now turn to the evaluation of the change in internal energy that results from the redistribution of the bulk waves in frequency. It is necessary to consider the properties of $\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z;z)$ for $\omega_m^2-\omega_b^2<\Omega^2<\omega_m^2$, where $z=\Omega\pm i\epsilon$.

(b) The case $\omega_m^2-\omega_b^2<\Omega^2<\omega_m^2$. The expression for $\sum_{k_z}\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z,z)$ exhibited in Eq. (36a) remains valid in this frequency range. However, the function F must be reevaluated. If $z=\Omega+i\epsilon$, and $\omega_m^2-\omega_b^2<\Omega^2<\omega_m^2$, then

$$F=i\,\mathrm{sgn}\Omega/[(\omega_m^2-\Omega^2)(\Omega^2-\omega_m^2+\omega_b^2)]^{1/2}\equiv if.$$

The relation

$$\partial F/\partial(\Omega^2)=(\omega_m^2-\tfrac{1}{2}\omega_b^2-\Omega^2)F^3$$

used to simplify the algebra in the preceding discussion remains valid. Using this result, and the expression in Eq. (27) for $d(\mathbf{k}_{||},\Omega)$, yields

$$\sum_{k_z}\Delta D^{(aa)}(\mathbf{k}_{||}k_zk_z;\Omega+i\epsilon)=\frac{f^3g(\mathbf{k}_{||},\Omega)\{[(\omega_m^2-\Omega^2)(1+\lambda^2)-2\omega_E\omega_m\lambda^2]f-i(1-\lambda^2)\}}{(1-\lambda^2)^2+f^2[(\omega_m^2-\Omega^2)(1+\lambda^2)-2\omega_m\omega_b\lambda]^2}, \quad (40)$$

where the expression has been rationalized to separate the function into its real and imaginary parts. We have defined

$$g(\mathbf{k}_{||},\Omega)=(\omega_m^2-\Omega^2)(\Omega^2-\omega_m^2+\omega_b^2)[(\Omega+\omega_m)(1+\lambda^2)-\omega_b\lambda]+(\omega_m+\Omega)[(1+\lambda^2)(\omega_m^2-\Omega^2)-2\omega_m\omega_b\lambda](\omega_m^2-\tfrac{1}{2}\omega_b^2-\Omega^2).$$

The expression in Eq. (40) has been derived for the case $z=\Omega+i\epsilon$. When $z=\Omega-i\epsilon$, one obtains the complex conjugate of the result in Eq. (40). Then for $\omega_m^2-\omega_b^2<\Omega^2<\omega_m^2$, the perturbation of the continuum modes by the surface produces the following change in the spectral function:

$$\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},\Omega)=(1/2\pi i)\sum_{k_z}[D^{(aa)}(\mathbf{k}_{||}k_zk_z;\Omega+i\epsilon)-D^{(aa)}(\mathbf{k}_{||}k_zk_z;\Omega-i\epsilon)]$$

or

$$\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},\Omega)=-[\mathrm{sgn}(\Omega)\,|f|^3/\pi]g(\mathbf{k}_{||},\Omega)/\{(1-\lambda^2)^2+f^2[(\omega_m^2-\Omega^2)(1+\lambda^2)-2\omega_m\omega_b\lambda]^2\}.$$

After some simplification, which employs some of manipulations similar to those described in Sec. III, we obtain

$$\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},\Omega)+\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},-\Omega)=\frac{\omega_E\Omega(1-\lambda^2)[2\omega_m(\omega_m^2-\tfrac{1}{2}\omega_b^2-\Omega^2)-\tfrac{1}{2}\omega_E(1+\lambda^2)(\omega_m^2-\Omega^2)]}{2\pi[(\omega_m^2-\Omega^2)(\Omega^2-\omega_m^2+\omega_b^2)]^{1/2}(\Omega^2-\Omega_{s1}^2)(\Omega^2-\Omega_{s1}^2)}. \quad (41)$$

Again we have introduced the solutions Ω_{s1} and Ω_{s1} of Eq. (11), for $\omega_H=0$.

The result exhibited in Eq. (41) is valid for arbitrary Ω and $\mathbf{k}_{||}$. So long as the temperature is low compared to the Néel temperature, only the low-frequency, long-wavelength modes will contribute to the internal energy. Thus we simplify Eq. (41) by assuming only frequencies Ω near $(\omega_a^2-\omega_b^2)^{1/2}$, and values of λ near unity are of interest. We introduce a quantity δ, defined by

$$\Omega^2=\omega_m^2-\omega_b^2+\omega_E^2\delta^2.$$

Then when $\delta^2\ll1$, and $1-\lambda^2\ll1$, the numerator of Eq. (41) may be simplified to write the expression in the form

$$\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},\Omega)+\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},-\Omega)\cong\frac{\omega_E^3\Omega(1-\lambda^2)[\omega_A+(\frac{1}{2}\omega_E-\omega_A)(1-\lambda^2)-2(\omega_A+\frac{1}{2}\omega_E)\delta^2]}{[(\omega_m^2-\Omega^2)(\Omega^2-\omega_m^2+\omega_b^2)]^{1/2}(\Omega^2-\Omega_{s1}^2)(\Omega^2-\Omega_{s2}^2)}.\tag{42}$$

Inside the square brackets in the numerator, terms of order $(1-\lambda^2)^2$ and $\delta^2(1-\lambda^2)$ have been ignored.

To proceed further, it will be convenient to restrict attention to the two temperature regimes considered in the computation of the contribution to $\Delta U(T)$ from the surface modes.

(i) The case $k_BT\ll(2\omega_E\omega_A)^{1/2}$. Again we suppose $\omega_E\gg\omega_A$. In the denominator of Eq. (42), in the long-wavelength limit, one has $(\Omega^2-\Omega_{s1}^2)(\Omega^2-\Omega_{s2}^2)\cong\omega_E^2\omega_A^2$. Also, $(\omega_m^2-\Omega^2)(\Omega^2-\omega_m^2+\omega_b^2)\cong\omega_E^4\delta^2$. Then one obtains the simple result

$$\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},\Omega)+\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},-\Omega)\cong(\sqrt{2}/\pi)(\omega_A\omega_E)^{-1/2}(1-\lambda)/\delta.$$

For the model employed in this work, $1-\lambda\cong\frac{1}{8}a^2k_{||}^2$. Then the change in the contribution to the internal energy from the continuum modes is, from Eq. (31),

$$\Delta U_c(T)=\frac{a^2}{2\pi(2\omega_E\omega_A)^{1/2}}\sum_{\mathbf{k}||}k_{||}^2\int_{(\omega_m^2-\omega_b^2)^{1/2}}^{\infty}\frac{d\Omega\,\Omega}{\delta}n(\Omega).$$

The upper limit of integration has been replaced by infinity, in the spirit of the low-temperature approximation. It is convenient to introduce a new variable k_z, defined by

$$\delta=\tfrac{1}{2}ak_z.$$

After some rearranging, one obtains the simple result

$$\Delta U_c(T)=\frac{a^3\omega_ES}{6\sqrt{2}}\left(\frac{\omega_E}{\omega_A}\right)^{1/2}\int\frac{d^3\mathbf{k}}{(2\pi)^3}k^2n(\omega(\mathbf{k})).$$

Again S is the total surface area, and $\omega(\mathbf{k})$ is the bulk magnon frequency, given by

$$\omega(\mathbf{k})\cong(2\omega_E\omega_A)^{1/2}+\tfrac{1}{16}\sqrt{2}\omega_E(\omega_E/\omega_A)^{1/2}a^2k^2.$$

Upon performing the integration over wave vector,

$$\Delta U_c(T)=(\sqrt{2}/\pi^3)^{1/2}(4S/a^2)\omega_E(\omega_A/\omega_E)^{3/4}(k_BT/\omega_E)^{5/2}\times\exp[-(2\omega_E\omega_A)^{1/2}/k_BT].$$

The contribution to the specific heat from the perturbation of the continuum modes is then

$$\Delta C_c(T)=\partial\Delta U_c(T)/\partial T\cong k_B(\sqrt{2}/\pi^3)^{1/2}(4\sqrt{2}S/a^2)(\omega_A/\omega_E)^{5/4}(k_BT/\omega_E)^{1/2}\times\exp[-(2\omega_E\omega_A)^{1/2}/k_BT].$$

(ii) The case $(2\omega_E\omega_A)^{1/2}\ll k_BT\ll k_BT_N$. Again employing $\omega_A\ll\omega_E$, one may obtain an expression for the spectral density by allowing $\omega_A\to0$. For $(1-\lambda)\ll1$ and $\delta\ll1$, we find

$$\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},\Omega)+\Delta\Gamma_c^{(aa)}(\mathbf{k}_{||},-\Omega)\cong\frac{(1-\lambda)[2(1-\lambda)+\delta^2]^{1/2}}{\pi\omega_E\delta}\frac{[(1-\lambda)-\delta^2]}{[(1-\lambda)+\delta^2]^2}.$$

In the expression for $\Delta U_c(T)$, the integral over Ω may be replaced by an integral over δ by noting $\Omega d\Omega=\omega_E^2\delta d\delta$. Then

$$\Delta U_c(T)=(2\omega_E/\pi)\sum_{\mathbf{k}||}(1-\lambda)\times\int_0^\infty d\delta\frac{[2(1-\lambda)+\delta^2]^{1/2}[(1-\lambda)-\delta^2]}{[(1-\lambda)+\delta^2]^2}\times n\{\omega_E[(1-\lambda^2)+\delta^2]^{1/2}\}.$$

Once again let $\delta=\frac{1}{2}ak_z$, and note $1-\lambda\cong\frac{1}{8}a^2k_{||}^2$. Then we find

$$\Delta U_c(T)=(a^2\omega_E/2L)\sum_{\mathbf{k}}|\mathbf{k}|(k^2-k_z^2)\times[(k^2-3k_z^2)/(k^2+k_z^2)^2]n(\tfrac{1}{2}\omega_Ea|\mathbf{k}|),$$

where the thickness of the macroscopic cube is L. Upon converting the sum to an integral, then defining $\nu=\cos\theta$, where θ is the angle between $\mathbf{k}$ and the k_z axis,

$$\Delta U_c(T)=\frac{a^2S\omega_E}{(4\pi)^2}\int_0^\infty dk\,k^3n(\tfrac{1}{2}\omega_Eak)\times\int_{-1}^{+1}d\nu\frac{(1-\nu^2)(1-3\nu^2)}{(1+\nu^2)^2}$$

$$=\frac{12}{\pi^2}\frac{S}{a^2}\omega_E\left(\frac{k_BT}{\omega_E}\right)^4\zeta(4)\int_0^{+1}d\nu\frac{(1-\nu^2)(1-3\nu^2)}{(1+\nu^2)^2}.$$

The integral over ν may be performed to yield

$$\Delta U_c(T)=(6/\pi^2)(10-3\pi)(S/a^2)\omega_E(k_BT/\omega_E)^4\zeta(4).$$

The contribution from the change in the frequency distribution of the bulk modes is thus

$$\Delta C_c(T)=(24/\pi^2)(10-3\pi)(S/a^2)(k_BT/\omega_E)^3$$

in the temperature regime $(2\omega_E\omega_A)^{1/2}\ll k_BT\ll k_BT_N$.

We now summarize the results obtained in this section, and provide some estimates of the magnitude of the surface contributions to the specific heat employing the parameters relevant to MnF_2. Again it will be convenient to consider the two temperature regimes discussed above separately.

(i) The case $k_BT\ll(\omega_E\omega_A)^{1/2}$. The total specific heat is given by

$$C(T)=C_0(T)+\Delta C_c(T)+C_s(T),$$

where $C_0(T)$ is found by differentiating the result of Eq. (33) with respect to temperature. Then we have found

$$C(T)=k_B\left(\frac{\sqrt{2}}{\pi^3}\right)^{1/2}\frac{16V}{a^3}\left(\frac{\omega_E}{k_BT}\right)^{1/2}\left(\frac{\omega_A}{\omega_E}\right)^{7/4}\times\left[1+\frac{1}{2}\frac{aS}{V}\frac{k_BT}{(2\omega_E\omega_A)^{1/2}}\right]\exp[-(2\omega_A\omega_E)^{1/2}/k_BT]$$
$$+k_B\frac{4S}{\pi a^2}\left(\frac{\omega_A}{\omega_E}\right)^{3/2}\left(\frac{\omega_E}{k_BT}\right)\exp[-(\omega_E\omega_A)^{1/2}/k_BT]. \quad (43)$$

Note that the term proportional to the surface area in the first line of Eq. (43) may be neglected, since it is always small compared to the term proportional to the volume. This term has its origin in the change in the frequency distribution of the continuum modes produced by the surface. Thus, when $k_BT\ll(\omega_E\omega_A)^{1/2}$, the dominant contribution to the specific heat proportional to the surface area comes from the surface magnons. Since the ratio of the surface term to the volume term contains the factor $\exp[(\sqrt{2}-1)(\omega_E\omega_A)^{1/2}/k_BT]$, when $k_BT\ll(\omega_E\omega_A)^{1/2}$ the contribution of the surface modes to the specific heat may be comparable to the contribution from the bulk excitations.

We shall compare the contribution from the surface modes with that from the bulk modes, employing parameters relevant to MnF_2. Consider a thin film consisting of N atomic layers. Then $V=\frac{1}{2}(NSa)$, where S is the total surface area. For a given value of N, the surface and bulk contributions are equal when the temperature T satisfies

$$N=\pi^{1/2}/2^{5/4}(\omega_E/k_BT)^{1/2}(\omega_E/\omega_A)^{1/4}\times\exp[(1-\tfrac{1}{2}\sqrt{2})(2\omega_E\omega_A)^{1/2}/k_BT].$$

For MnF_2, one has $(2\omega_E\omega_A)^{1/2}=12.6°K$, and $(\omega_E/\omega_A)^{1/2}\cong 8$.[19] Then for a film of MnF_2, one has the numerical relation

$$N\cong 18.5T^{-1/2}\exp(3.8/T).$$

For $T=1°K$, the bulk and surface contributions to the specific heat will be equal if $N=850$ layers.

This estimate indicates that at low temperatures, the surface spin-wave contribution to the specific heat of antiferromagnetic films or small particles is of an observable magnitude. Our estimate indicates that at 1°K, about 10% of the specific heat of a film of $MnF_2\approx$ 25 000 Å thick will come from the surface modes.

(ii) $(2\omega_E\omega_A)^{1/2}\ll k_BT\ll k_BT_N$. The total specific heat has been found to be

$$C(T)=k_B\left(\frac{8}{\pi}\right)^2\frac{3V}{a^3}\left(\frac{k_BT}{\omega_E}\right)^3\zeta(4)\times\left[1+\tfrac{1}{8}\pi\frac{\omega_E}{k_BT}\frac{\zeta(3)}{\zeta(4)}\frac{Sa}{V}+\tfrac{1}{8}\pi\left(\frac{10}{\pi}-3\right)\frac{Sa}{V}\right].$$

Since $k_BT\ll\omega_E$, the dominant contribution to the portion of the specific heat proportional to the surface area again comes from the surface magnons, rather than the term that arises from the redistribution of the bulk modes in frequency. It is interesting to note that in the previous studies of the surface specific heat of the vibrating lattice,[8] and the surface heat of the Heisenberg ferromagnet,[10] the change in the specific heat that results from the redistribution of the continuum modes is the same magnitude as that from the surface excitations.

Notice that, in the antiferromagnet, when

$$(2\omega_E\omega_A)^{1/2}\ll k_BT\ll kT_N,$$

the surface magnons give a contribution to $C(T)$ proportional to T^2. For this contribution to be observable in this temperature range, it is necessary to have $(aS/V)\omega_E/k_BT$ comparable to unity.

For a given surface-to-volume ratio, it seems possible that $C_s(T)$ may be easiest to observe when $k_BT\ll(2\omega_E\omega_A)^{1/2}$, where the bulk mode contribution will be frozen out and the surface magnon part exhibits the exponential temperature dependence.

V. TEMPERATURE-DEPENDENT PART OF THE MEAN SUBLATTICE DEVIATION

In this section, we study the quantity

$$\Delta_l^{(a)}=S-\langle S_{l_a}^z\rangle=\langle a_{l_a}^\dagger a_{l_a}\rangle$$

that describes the mean deviation from perfect alignment of an A spin with z coordinate $l_a^z=a(l+\frac{1}{2})$. From the expression that follows Eq. (17), one sees that

$$\Delta_l^{(a)}=(2a^2/S)\sum_{\mathbf{k}_{||}}\int_{-\infty}^{+\infty}d\Omega\, n(\Omega)\Gamma_l^{(a)}(\mathbf{k}_{||},\Omega), \quad (44)$$

where

$$\Gamma_l^{(a)}(\mathbf{k}_{||},\Omega)=(1/2\pi i)\{A_l(\mathbf{k}_{||},\Omega+i\epsilon)-A_l(\mathbf{k}_{||},\Omega-i\epsilon)\},$$

and

$$A_l(\mathbf{k}_{||},\Omega\pm i\epsilon)=(a/L)\sum_{k_zk_z'}\exp[i(k_z-k_z')l_a^z]\times D^{(aa)}(\mathbf{k}_{||}k_zk_z';\Omega\pm i\epsilon).$$

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[19] D. Sell, R. Greene, and R. White, Phys. Rev. **158**, 489 (1967).

It is well known that in antiferromagnets the spins are not perfectly aligned along the z axis at the absolute zero of temperature, since the spins execute zero-point motion about the z axis. Thus Δ_l is finite at $T=0$. To compute the amplitude of the zero-point motion, it is necessary to perform an integration over the entire first Brillouin zone. In the presence of the surfaces, the amplitude of the zero-point motion executed by a given spin will be a function of its distance from the surface. To compute the position dependence of the zero-point motion is not straightforward, since the functions to be integrated are quite complex.

In this section, only the temperature-dependent part of Δ_l will be computed. To compute $\Delta_l(T)-\Delta_l(0)$, one needs only to include the contribution from the low-frequency, long-wavelength spin-wave modes. We will also confine our attention to temperatures sufficiently low that $k_BT\ll(\omega_E\omega_A)^{1/2}$. The temperature-dependent part of the mean sublattice deviation near the surface then comes entirely from the thermally excited surface magnons. The discussion of Sec. IV implies that the influence of the surface on the spatial variation of Δ_l will be most pronounced in this region of temperature. Upon manipulating Eq. (40) a bit,

$$\delta\Delta_l^{(a)}(T)=\Delta_l^{(a)}(T)-\Delta_l^{(a)}(0)$$
$$=(2a^2/S)\int_0^\infty d\Omega\, n(\Omega)\times\sum_{\mathbf{k}_{||}}[\Gamma_l^{(a)}(\mathbf{k}_{||},\Omega)-\Gamma_l^{(a)}(\mathbf{k}_{||},-\Omega)].$$

We now separate $\delta\Delta_l(T)$ into the portion independent of l that characterizes the extended medium, and the spatially varying part produced by the presence of the surface:

$$\delta\Delta_l^{(a)}(T)=\delta\Delta_\infty^{(a)}(T)+d_l^{(a)}(T).$$

From the first term in the Green's function of Eq. (24), one finds

$$\delta\Delta_\infty^{(a)}(T)=(a^3/V)\sum_k[\omega_m/\omega(\mathbf{k})]n(\omega(\mathbf{k}))$$
$$\cong16(\sqrt{2}\pi)^{1/2}(\omega_A/\omega_E)^{1/4}(k_BT/\omega_E)^{3/2}\times\exp[-(2\omega_E\omega_A)^{1/2}/k_BT].$$

The last expression is valid in the low-temperature limit considered in the present section. Now write

$$A_l(\mathbf{k}_{||},\Omega\pm i\epsilon)=A_\infty(\mathbf{k}_{||},\Omega)+\delta A_l(\mathbf{k}_{||},\Omega),$$

where $\delta A_l(\mathbf{k}_{||},\Omega)$ is the change in the quantity $A_l(\mathbf{k}_{||},\Omega)$ produced by the presence of the surface.

From the Green's function exhibited in Eq. (24), we have

$$\delta A_l(\mathbf{k}_{||},\Omega)=[2d(\mathbf{k}_{||},\Omega)]^{-1}\times[(\tfrac{1}{2}N_1+N_2)F_lF_{l+1}+(\tfrac{1}{4}N_1+N_2+N_3)F_{l+1}^2+\tfrac{1}{4}N_1F_l^2],$$

where the definitions of N_1, N_2, and N_3 are given in Eqs. (25), $d(\mathbf{k}_{||},\Omega)$ is defined in Eq. (27), and

$$F_l=(1/L)\sum_{k_z}[\cos(k_zla)/(\omega^2(\mathbf{k})-\Omega^2)].$$

Since we are only interested in the surface magnon contribution to $d_l(T)$, the function F_l may be evaluated for $0<\Omega^2<\omega_a^2-\omega_b^2$. The integral is elementary:

$$F_l=\exp(-\beta\,|\,l\,|)/[(\omega_m^2-\Omega^2)(\omega_m^2-\omega_b^2-\Omega^2)]^{1/2},$$

with

$$e^{-\beta}=(2/\omega_b^2)\times\{\omega_m^2-\tfrac{1}{2}\omega_b^2-\Omega^2-[(\omega_m^2-\Omega^2)(\omega_m^2-\omega_b^2-\Omega^2)]^{1/2}\}.$$

We can then write (for $l>0$)

$$\delta A_l(\mathbf{k}_{||},\Omega)=\left[\frac{(\tfrac{1}{2}N_1+N_2)e^{-\beta}+(\tfrac{1}{4}N_1+N_2+N_3)e^{-2\beta}+\tfrac{1}{4}N_1}{2d(\mathbf{k}_{||},\Omega)(\omega_m^2-\Omega^2)(\omega_m^2-\omega_b^2-\Omega^2)}\right]e^{-2\beta l}.$$

It is now possible to obtain the surface magnon contribution to $\Gamma_l(\mathbf{k}_{||}\Omega)$ by recalling that $d(\mathbf{k}_{||},\Omega)$ has a zero at $\Omega^2=\Omega_{s1}^2$, where Ω_{s1} is the surface spin-wave frequency. Again, for Ω near Ω_{s1},

$$d(\mathbf{k}_{||},\Omega)=(\Omega^2-\Omega_{s1}^2)[\partial d(k_{||},\Omega)/\partial(\Omega^2)].$$

With the expression given in Sec. IV for $\partial d/\partial(\Omega^2)$, one has (still with $l>0$)

$$\delta\Gamma_l^{(a)}(\mathbf{k}_{||},\Omega)=\frac{\lambda^2[\tfrac{1}{4}N_1+(\tfrac{1}{2}N_1+N_2)e^{-\beta}+(\tfrac{1}{4}N_1+N_2+N_3)e^{-2\beta}]e^{-2\beta l}}{2\Omega_s\{(1+\lambda^2)F^{-1}-(\omega_m^2-\tfrac{1}{2}\omega_b^2-\Omega_s^2)[(\omega_m^2-\Omega_s^2)(1+\lambda^2)-2\omega_m\omega_b\lambda]F\}}[\delta(\Omega-\Omega_s)-\delta(\Omega+\Omega_s)].\quad(45)$$

Since only the long-wavelength surface modes will contribute to d_l at low temperatures, the quantity λ will be replaced by unity. Then with $\omega_E\gg\omega_A$, we find for $l>0$

$$\delta\Gamma_l^{(a)}(\mathbf{k}_{||},\Omega)-\delta\Gamma_l^{(a)}(\mathbf{k}_{||},-\Omega)\cong\tfrac{1}{4}\delta(\Omega-\Omega_s)\times[\tfrac{1}{2}\omega_E/\omega_A-(\omega_E/\omega_A+2)e^{-\beta}+(\tfrac{1}{2}\omega_E/\omega_A+4)e^{-2\beta}]e^{-2\beta l}.$$

From the definition of $e^{-\beta}$, with Ω replaced by Ω_s, one sees

$$e^{-\beta}\cong1-2(\omega_A/\omega_E)^{1/2}\quad\text{for}\quad\omega_A\ll\omega_E,$$

so that

$$\beta\cong2(\omega_A/\omega_E)^{1/2}.$$

Then

$$\delta\Gamma_l^{(a)}(\mathbf{k}_{||},\Omega)-\delta\Gamma_l^{(a)}(\mathbf{k}_{||},-\Omega)\cong\delta(\Omega-\Omega_s)\exp[-4(\omega_A/\omega_E)^{1/2}l]\qquad l\geq0.$$

To obtain this simple result, terms in ω_A/ω_E were assumed small compared to unity. Then the position-dependent portion of $\Delta_l(T)-\Delta_l(0)$ is given by

$$d_l(T)=\exp[-4(\omega_A/\omega_E)^{1/2}l](2a^2/S)\sum_{\mathbf{k}_{||}} n(\Omega_s(\mathbf{k}_{||})).$$

The sum over $\mathbf{k}_{||}$ may be converted to an integral, and the integral may be evaluated by employing the long-wavelength form of the surface magnon dispersion relation. The final result for the temperature-dependent part of the mean sublattice deviation is found to be

$$\Delta_l^{(a)}(T)-\Delta_l^{(a)}(0)=16(\sqrt{2}\pi)^{1/2}\left(\frac{\omega_A}{\omega_E}\right)^{1/4}\left(\frac{k_BT}{\omega_E}\right)^{3/2}$$
$$\times\exp[-(2\omega_E\omega_A)^{1/2}/k_BT]+\frac{8}{\pi}\left(\frac{\omega_A}{\omega_E}\right)^{1/2}\left(\frac{k_BT}{\omega_E}\right)$$
$$\times\exp[-4(\omega_A/\omega_E)^{1/2}\,|\,l\,|\,]\,\exp[-(\omega_E\omega_A)^{1/2}/k_BT]. \quad (46)$$

This expression is valid for $k_BT\ll(\omega_E\omega_A)^{1/2}$. Although the derivation was discussed in detail only for the case $l>0$, it may be seen that $\Delta_l(T)-\Delta_l(0)$ is an even function of l. Thus $|\,l\,|$ rather than l appears in Eq. (46).

When $(k_BT)\ll(\omega_E\omega_A)^{1/2}$, the result of Eq. (46) shows that $\Delta_l(T)-\Delta_l(0)$ is greatly enhanced near the surface, compared to its value in the bulk crystal.

The result in Eq. (46) gives the temperature-dependent part of the sublattice deviation of the A spins, for all values of l in the temperature range considered. In zero magnetic field, a symmetry argument may be employed to find the mean deviation on the B sublattice. One simply needs to note that magnitude associated with a plane of B spins located above the x–y plane at $l_b^z=al$ $(l=1, 2, \cdots)$, is the same as that of a plane of A spins below the x–y plane at $l_a^z=a(-l+\frac{1}{2})$.

The dependence of the result in Eq. (47) on $|\,l\,|$ may be understood in a simple manner. In Sec. II, we found that for a surface magnon with $\mathbf{k}_{||}=0$, the spin deviation was proportional to $\exp[-2(\omega_A/\omega_E)^{1/2}\,|\,l\,|]$. When $k_BT\ll(\omega_E\omega_A)^{1/2}$, the thermally excited surface modes all have a wavelength long compared to the lattice constant. Since the change in the mean value of S_z is proportional to the mean of the square of the transverse component of spin for small deviations, the spatial-dependent part of $\Delta_l(T)-\Delta_l(0)$ is proportional to $\exp[-4(\omega_A/\omega_E)^{1/2}\,|\,l\,|]$.

VI. SURFACE CORRECTIONS TO THE LOW-TEMPERATURE PARALLEL SUSCEPTIBILITY

Suppose a magnetic field is applied parallel to the z axis of the system. Let the field be parallel to the direction of the A spins, with the precession frequency of a free spin in the field denoted by ω_H. In Sec. III, we noted that the effect of a field was to replace the function $D^{(aa)}(\mathbf{l}_a\mathbf{l}_a', z)$ by $D^{(aa)}(\mathbf{l}_a\mathbf{l}_a', z-\omega_H)$. The change $\Delta_l^{(a)}(H, T)-\Delta_l^{(a)}(0, T)$ in the mean deviation of a plane of A spins produced by the field is then

$$\Delta_l^{(a)}(H, T)-\Delta_l^{(a)}(0, T)=(2a^2/S)$$
$$\times\sum_{\mathbf{k}_{||}}\int_{-\infty}^{+\infty}d\Omega\, n(\Omega)[\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega-\omega_H)-\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega)],$$

where $\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega)$ is defined in Sec. V. To first order in H,

$$\Delta_l^{(a)}(H, T)-\Delta_l^{(a)}(0, T)$$
$$=-\omega_H(2a^2/S)\sum_{\mathbf{k}_{||}}\int_{-\infty}^{+\infty}d\Omega\, n(\Omega)[\partial\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega)/\partial\Omega].$$

We define a susceptibility per atomic layer that relates the change in spin deviation to first order in the field to the applied magnetic field:

$$\Delta_l^{(a)}(H, T)-\Delta_l^{(a)}(0, T)=-\omega_H\chi_l^{(a)},$$

where

$$\chi_l^{(a)}=-(2a^2/S)\sum_{\mathbf{k}_{||}}\int_{-\infty}^{+\infty}d\Omega\, n(\Omega)[\partial/\partial\Omega]\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega).$$

After a partial integration is performed, and the result is rearranged, we find

$$\chi_l^{(a)}=(2a^2/Sk_BT)\sum_{\mathbf{k}_{||}}\int_0^{\infty}d\Omega\, n(\Omega)[1+n(\Omega)]$$
$$\times[\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega)+\Gamma_l^{(a)}(\mathbf{k}_{||}, -\Omega)]. \quad (47)$$

Since there is a gap in the spin-wave spectrum, it is evident that $\chi_l\to0$ as $T\to0$. As in Sec. V, the low-temperature region with $k_BT\ll(\omega_E\omega_A)^{1/2}$ will be considered, since the surface corrections to χ_l have their greatest relative importance in this temperature range. So long as $k_BT\ll(\omega_E\omega_A)^{1/2}$, we will have $n(\Omega)\ll1$ for the frequencies of interest. Hence

$$\chi_l^{(a)}\cong(2a^2/Sk_BT)\sum\int_0^{\infty}d\Omega\, n(\Omega)$$
$$\times[\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega)+\Gamma_l^{(a)}(\mathbf{k}_{||}, -\Omega)].$$

Following the development in Sec. V, $\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega)$ may be separated into a part independent of l that characterizes the bulk material, and an l-dependent part:

$$\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega)=\Gamma_\infty^{(a)}(\mathbf{k}_{||}, \Omega)+\delta\Gamma_l^{(a)}(\mathbf{k}_{||}, \Omega).$$

From the Green's function of Eq. (24), one has

$$\Gamma_\infty^{(a)}(\mathbf{k}_{||}, \Omega)+\Gamma_\infty^{(a)}(\mathbf{k}_{||}, -\Omega)$$
$$=\int_{-\pi/a}^{+\pi/a}(adk_z/2\pi)\delta(\Omega-\omega(\mathbf{k})),$$

where $\omega(\mathbf{k})$ is the bulk magnon frequency. We then write

$$\chi_l^{(a)}=\chi_\infty^{(a)}+\delta\chi_l^{(a)},$$

where

$$\chi_\infty=(a^3/Vk_BT)\sum_{\mathbf{k}}n(\omega(\mathbf{k}))$$

is the parallel susceptibility of the extended medium, and $\delta\chi_l$ is the position-dependent part of χ_l that results from the presence of the surfaces. In the previous equation, the sum over the wave vector may be converted to an integral, and evaluated in the standard manner. When $k_BT \ll (\omega_E\omega_A)^{1/2}$, the quantity χ_∞ is well approximated by

$$\chi_\infty^{(a)} = \frac{32}{\omega_E}\left(\frac{\pi}{\sqrt{2}}\right)^{1/2}\left(\frac{\omega_A}{\omega_E}\right)^{3/4}\left(\frac{k_BT}{\omega_E}\right)^{1/2} \times \exp[-(2\omega_E\omega_A)^{1/2}/k_BT]. \quad (48)$$

The quantity $\delta\Gamma_l^{(a)}(\mathbf{k}_{||},\Omega)$ required to compute the position-dependent part $\delta\chi_l$ of the parallel susceptibility is exhibited in Eq. (45). It is now necessary to take the sum $\delta\Gamma_l^{(a)}(\mathbf{k}_{||},\Omega)+\delta\Gamma_l^{(a)}(\mathbf{k}_{||},-\Omega)$ rather than the difference of these two quantities, as we did in Sec. V.

In the limit $\omega_E \gg \omega_A$ employed throughout this paper, we obtain the surface spin-wave contribution

$$\delta\Gamma_l^{(a)}(\mathbf{k}_{||},\Omega)+\delta\Gamma_l^{(a)}(\mathbf{k}_{||},-\Omega) = \mathrm{sgn}(l)\,\delta(\Omega-\Omega_s(\mathbf{k}_{||}))\exp[-4(\omega_A/\omega_E)^{1/2}|l|].$$

In the low-temperature limit, the corrections to χ_∞ from the presence of the surfaces will come almost entirely from the surface magnons, since the bulk mode contribution will be nearly completely frozen out.

Inserting this result into the expression for $\chi_l^{(a)}$ gives

$$\delta\chi_l^{(a)} = \mathrm{sgn}(l)\,\frac{8}{\pi\omega_E}\left(\frac{\omega_A}{\omega_E}\right)^{1/2}\exp[-4(\omega_A/\omega_E)^{1/2}|l|] \times \exp[-(\omega_E\omega_A)^{1/2}/k_BT]. \quad (49)$$

The exponential dependence of $\delta\chi_l^{(a)}$ on $|l|$ is of the same form as the spatial dependence of the mean sublattice deviation discussed in Sec. V. The physical origin of this spatial dependence was discussed near the end of Sec. V.

One interesting feature of the form exhibited in Eq. (49) is that it is an odd function of l. Application of a magnetic field parallel to the $+z$ axis *decreases* the mean deviation Δ_l of a plane of A spins with positive z coordinate[20] while the sublattice deviation of a plane of A spins with negative z coordinate is *increased*.

This behavior of $\chi_l^{(a)}$ may be understood by referring to the discussion in Sec. II of the magnetic field dependence of the (nondegenerate) surface magnon mode associated with the semi-infinite medium. It was pointed out that if an external magnetic field is applied parallel to the direction of magnetization of the spins in the surface layer, the surface mode stiffens, i.e., its excitation energy increases. Thus the surface magnon contribution to the mean deviation Δ_l near the surface decreases. The application of the magnetic field has thus resulted in a negative $\delta\chi_l^{(a)}$. However, if the field is applied antiparallel to the direction in which the A spins point, the surface magnon frequency is decreased, and a positive $\delta\chi_l^{(a)}$ is obtained at low temperatures.

In the present model, we have formed two free surfaces by setting to zero exchange interactions between the B spins in the layer with z coordinate zero, and the A spins with z coordinate $+\frac{1}{2}a$. Application of a magnetic field parallel to the A spin direction stiffens the surface mode associated with the upper surface to produce a positive $\delta\chi_l^{(a)}$ for $l>0$, while the surface mode associated with the lower surface is softened. Hence $\delta\chi_l^{(a)}<0$ for $l<0$.

We have exhibited the form of $\delta\chi_l$ to first order in ω_H. So long as only the change in Δ_l first order in ω_H is considered, the surface magnon contribution to the change in Δ_l for a plane of A spins with z coordinate $a(l+\frac{1}{2})$ is the same as the surface magnon contribution to the change in sublattice deviation of a plane of B spins with z coordinate $-al$.

We conclude this section with an estimate of the importance of the surface-region contribution to the total moment induced by an external magnetic field applied parallel to the z axis. Consider the region above the x–y plane of the model crystal employed in this work. If there are N_s spins in one layer, the contribution of this region to the total moment induced by the field is proportional to

$$2N_s\sum_{l=0}\delta\chi_l^{(a)} = 16N_s/\pi\omega_E(\omega_A\omega_E)^{1/2} \times[1-\exp(-4(\omega_A/\omega_E)^{1/2})]\exp[-(\omega_E\omega_A)^{1/2}/k_BT] \cong (4N_s/\pi\omega_E)\exp[-(\omega_E\omega_A)^{1/2}/k_BT],$$

when $\omega_E \gg \omega_A$. The factor of 2 has been introduced because the B spin contribution to the moment equals the A spin contribution to first order in H.

This expression may be compared to the contribution from the bulk of the medium, which is equal to $2N_sL\chi_\infty$ if the material consists of L atomic layers.

Thus we have the following ratio:

$$\frac{\Delta M_s}{M_0} = \frac{\text{moment induced near one surface}}{\text{moment induced in the bulk}} = (16\pi L)^{-1}\left(\frac{\sqrt{2}}{\pi}\right)^{1/2}\left(\frac{\omega_E}{\omega_A}\right)^{3/4}\left(\frac{\omega_E}{k_BT}\right)^{1/2} \times\exp[(1-\tfrac{1}{2}\sqrt{2})(2\omega_E\omega_A)^{1/2}/k_BT].$$

If the parameters relevant to MnF_2 are employed in this expression (see Sec. IV), then we find

$$(\Delta M_s/M_0) \cong (2.8/LT^{1/2})\exp[3.8/T].$$

If we consider a film the order of 10 000 Å thick ($L\approx 2000$), then at 1°K,

$$\Delta M_s/M_0 \cong 0.05.$$

This estimate suggests that the experimental observation of the surface magnon contribution to the parallel susceptibility may be possible, if films the order of 1 μ in thickness are studied at temperatures $\cong$1°K.

[20] Note the sign convention employed in the definition χ_l.

VII. ONE-MAGNON ABSORPTION SPECTRUM

In this section, we shall consider the effect of the surface on the absorption spectrum of the antiferromagnetic array. It is well known that bulk antiferromagnetic crystals exhibit an absorption line at the bulk $\mathbf{k}=0$ magnon frequency, equal to $(2\omega_E\omega_A)^{1/2}$ in the limit $\omega_E \gg \omega_A$. The presence of the surface mode will introduce a new absorption line in the spectrum at the frequency $(\omega_E\omega_A)^{1/2}$ of the $\mathbf{k}_{||}=0$ surface magnon. The integrated strength of this line will clearly be proportional to the surface-to-volume ratio. The purpose of this section is to compute the strength of the surface mode absorption, and compare it to the strength of the main AFMR line.

Suppose a microwave field with frequency Ω is incident on the sample. Let the field be circularly polarized. Since in the frequency range of interest the wavelength of the radiation is long compared to the lattice constant, we suppose the wavelength of the radiation infinite. The interaction between the radiation and the crystal may be written in the form

$$V=-g\mu_B \exp(-i\Omega t)\left[\sum_{\mathbf{l}_a}\mathbf{h}\cdot\mathbf{S}(\mathbf{l}_a)+\sum_{\mathbf{l}_b}\mathbf{h}\cdot\mathbf{S}(l_b)\right]+\text{H.c.},$$

where we suppose $\mathbf{h}=\frac{1}{2}\sqrt{2}h(\hat{x}+i\hat{y})$. Then

$$V=-g\mu_B \exp(-i\Omega t)\tfrac{1}{2}\sqrt{2}h\left[\sum_{\mathbf{l}_a}S_{(+)}(\mathbf{l}_a)+\sum_{\mathbf{l}_b}S_{(+)}(\mathbf{l}_b)\right]+\text{H.c.}$$

After carrying out the Holstein-Primakoff transformation, and retaining only the lowest-order terms, we have

$$V=-g\mu_B hS^{1/2}\exp(-i\Omega t)A+\text{H.c.},$$

where the operator

$$A=\sum_{\mathbf{l}_a}a_{\mathbf{l}_a}+\sum_{\mathbf{l}b}b_{\mathbf{l}_b}{}^{\dagger}.$$

Then if we treat the effect of V by employing the Fermi golden rule of time-dependent perturbation theory, the system may be seen to absorb energy from the field at the rate

$$dE/dt=2\pi g^2\mu_B{}^2h^2S\Omega\Big\{\sum_{I,F}P_I\,|\,\langle F\,|\,A\,|\,I\rangle\,|^2\delta(E_F-E_I-\Omega)-\sum_{I,F}P_I\,|\,\langle F\,|\,A^{\dagger}\,|\,I\rangle\,|^2\delta(E_F-E_I+\Omega)\Big\}. \quad (50)$$

We assume the probability that the incident radiation encounters the system in state $|\,I\rangle$ is P_I. The radiation induces a transition to the final state $|\,F\rangle$. The energies of the initial and final states are E_I and E_F, respectively.

The expression for dE/dt may be expressed in convenient form by introducing the correlation function

$$D^{(A\dagger A)}(\tau)=\langle T(A^{\dagger}(\tau)A(0))\rangle,$$

where as before $A^{\dagger}(\tau)=\exp(H\tau)A^{\dagger}(0)\exp(-H\tau)$. As in Sec. III, the function $D^{(A\dagger A)}(\tau)$ is a periodic function of τ with period $\beta=1/k_BT$. Let $D^{(A\dagger A)}(i\omega_n)$ denote the appropriate Fourier transform with respect to τ (again we follow Sec. III), and $D^{(A\dagger A)}(z)$ the function obtained by analytically continuing $D^{(A\dagger A)}(i\omega_n)$ off the imaginary axis into the appropriate half-plane. Then introduce the spectral density

$$\Gamma^{(A\dagger A)}(\Omega)=(1/2\pi i)\left[D^{(A\dagger A)}(\Omega+i\epsilon)-D^{(A\dagger A)}(\Omega-i\epsilon)\right].$$

One has the explicit expression

$$n(\Omega)\Gamma^{(A\dagger A)}(\Omega)=(1/Z)\sum_{n,m}\exp(-\beta E_n)\,|\,\langle m\,|\,A\,|\,n\rangle\,|^2\delta(\Omega_{nm}-\Omega),$$

where

$$Z=\text{Tr}\rho=\sum_n\exp(-\beta E_n).$$

Note that

$$\Gamma^{(A\dagger A)}(\Omega)=-\Gamma^{(AA\dagger)}(\Omega). \quad (51)$$

If the identity in Eq. (51) is employed in Eq. (50), one finds the result

$$dE/dt=-2\pi g^2\mu_B{}^2h^2S\Omega\Gamma^{(A\dagger A)}(-\Omega).$$

If one employs the specific form of the operator A, then

$$\begin{aligned}D^{(A\dagger A)}(\tau)&=\sum_{l_al_a'}\langle T(a_{l_a}{}^{\dagger}(\tau)a_{l_a'}(0))\rangle+\sum_{l_al_b}\langle T(a_{l_a}{}^{\dagger}(\tau)b_{l_b}{}^{\dagger}(0))\rangle+\sum_{l_al_b}\langle T(b_{l_b}(\tau)a_{l_a}(0))\rangle+\sum_{l_al_b'}\langle T(b_{l_b}(\tau)b_{l_b'}{}^{\dagger}(0))\rangle\\&=\sum_{l_al_a'}D^{(a\dagger a)}(l_al_a',\tau)+\sum_{l_al_b}D^{(a\dagger b\dagger)}(l_al_b,\tau)+\sum_{l_al_b}D^{(ba)}(l_bl_a,\tau)+\sum_{l_bl_b'}D^{(bb\dagger)}(l_bl_b',\tau).\end{aligned}$$

In the superscripts associated with the correlation functions in the last expression, for the sake of clarity, we have introduced a notation slightly different from that employed in earlier sections of the paper.

Next we introduce Fourier transforms with respect to the spatial variables, in a fashion analogous to Eq. (20). Then we obtain (with N the total number of spins in the sample)

$$N^{-1}\Gamma^{(A\dagger A)}(-\Omega)=\Gamma_0{}^{(a\dagger a)}(-\Omega)+\Gamma_0{}^{(a\dagger b\dagger)}(-\Omega)+\Gamma_0{}^{(ba)}(-\Omega)+\Gamma_0{}^{(bb\dagger)}(-\Omega).$$

The subscript "0" indicates that the spectral density $\Gamma^{(a\dagger a)}(\Omega)$ is to be obtained from $D^{(a\dagger a)}(\mathbf{k}_{||}k_zk_z',z)$, with $\mathbf{k}_{||}=k_z=k_z'=0$.

Now, in a manner similar to Eq. (51), one has

$$\Gamma_0{}^{(a\dagger b\dagger)}(-\Omega)=-\Gamma_0{}^{(ab)}(\Omega),$$

and

$$\Gamma_0{}^{(bb\dagger)}(-\Omega)=-\Gamma_0{}^{(b\dagger b)}(\Omega).$$

Hence,

$$dE/dt=2\pi g^2\mu_B^2h^2NS\Omega \times[\Gamma_0^{(b\dagger b)}(\Omega)-\Gamma_0^{(a\dagger a)}(-\Omega)+\Gamma_0^{(ab)}(\Omega)-\Gamma_0^{(ba)}(-\Omega)].$$

The quantities $\Gamma_0^{(a\dagger a)}(\Omega)$ and $\Gamma_0^{(ba)}(\Omega)$ may be computed from the expressions given in Eqs. (22) and (24). Until this point, we have not needed the explicit form of $\langle T(b_{l_b}^\dagger(\tau)b_{l_b'}(0))\rangle$ and its Fourier transforms with respect to the space and time variables. One may compute these quantities in a fashion analogous to the discussion in Sec. III. The equation of motion of $\langle T(b_{l_b}^\dagger(\tau)b_{l_b'}(0))\rangle$ involves $\langle T(a_{l_a}(\tau)b_{l_b}(0))\rangle$. For these two functions, one obtains a closed set of equations similar in structure to Eqs. (19). Upon solving the equations, one sees that $\Gamma_0^{(b\dagger b)}(\Omega)=\Gamma_0^{(a\dagger a)}(\Omega)$. [This identity is obtained only in the limit when k_z and k_z' in $D^{(b\dagger b)}(\mathbf{k}_{||}k_zk_z', z)$ and $D^{(a\dagger a)}(\mathbf{k}_{||}k_zk_z', z)$ approach zero.]

Finally, one has

$$dE/dt=2\pi g^2\mu_B^2h^2NS\Omega \times[\Gamma_0^{(a\dagger a)}(\Omega)-\Gamma_0^{(a\dagger a)}(-\Omega)+\Gamma_0^{(ab)}(\Omega)-\Gamma_0^{(ba)}(-\Omega)].$$

To compute the strength of the bulk magnon line, one may derive $\Gamma_0^{(a\dagger a)}(\Omega)$ and $\Gamma_0^{(ba)}(\Omega)$ from the first term on the right side of Eqs. (20). The result is

$$\Gamma^{(a\dagger a)}(\Omega)-\Gamma^{(a\dagger a)}(-\Omega)=(1/2\Omega_0)(\omega_A+\omega_E)\delta(\Omega-\Omega_0)$$

and

$$\Gamma^{(ab)}(\Omega)-\Gamma^{(ba)}(-\Omega)=(-1/2\Omega_0)\omega_E\delta(\Omega-\Omega_0),$$

where $\Omega_0=(2\omega_E\omega_A+\omega_A^2)^{1/2}$ is the frequency of the bulk $\mathbf{k}=0$.

Then, for $\omega_E\gg\omega_A$,

$$dE_B/dt=\pi g^2\mu_B^2h^2NS\omega_A\delta(\Omega-(2\omega_E\omega_A)^{1/2}). \quad (52)$$

The result of Eq. (52) may also be obtained from the standard methods of spin-wave theory. In the presence of the surface, there will be corrections to the strength of the bulk magnon line proportional to the surface area. We have ignored such corrections in the result exhibited in Eq. (52). Note that in Eq. (52), the spin of an ion is S. This quantity must not be confused with the surface area, denoted by S in the earlier sections.

From the forms in Eqs. (22) and (24), one may compute the contribution to the absorption spectrum from the surface mode. Since algebraic manipulations with the surface corrections to the Green's functions have been described in detail in the preceding sections of the paper, we shall be content to simply exhibit the final result at this point.

In the limit that $\omega_A\ll\omega_E$, the surface mode contributions to $\Gamma_0^{(a\dagger a)}(\Omega)$ and $\Gamma_0^{(ba)}(\Omega)$ lead to (for $\Omega>0$)

$$\Gamma_0^{(a\dagger a)}(\Omega)-\Gamma_0^{(a\dagger a)}(-\Omega) =(1/2L)(\omega_E/\omega_A+1)\delta(\Omega-(\omega_E\omega_A)^{1/2})$$

and

$$\Gamma_0^{(ba)}(\Omega)-\Gamma_0^{(ba)}(-\Omega) =-(1/2L)(\omega_E/\omega_A-1)\delta(\Omega-(\omega_E\omega_A)^{1/2}),$$

so that

$$dE_s/dt=\pi g^2\mu_B^2h^2S(\omega_E\omega_A)^{1/2}(N/L)\delta(\Omega-(\omega_E\omega_A)^{1/2}).$$

This leads to the simple result mentioned in the Introduction:

$$\frac{\text{integrated strength of surface mode line}}{\text{integrated strength of main AFMR line}} =\tfrac{1}{2}(aS_A/V)(\omega_E/\omega_A)^{1/2},$$

where S_A is the total surface area of the sample and a is the lattice constant.

This result has a simple physical interpretation. When the surface mode is excited, we have seen that the spin deviation decays exponentially with distance from the surface. The mode penetrates a distance proportional to $a(\omega_E/\omega_A)^{1/2}$ into the medium. Then if the mode is excited by external radiation, a volume proportional to $aS_A(\omega_E/\omega_A)^{1/2}$ is responsible for the energy absorption. In bulk AFMR, energy is absorbed throughout the crystal volume. The result exhibited in this last equation is the ratio of the two volumes just mentioned, to within a factor of order unity.

We also remark that, in the present model, absorption of energy by the surface mode occurs for both senses of circular polarization. The integrated strength of the line is the same for both polarizations. This is an artificial consequence of the bond-cutting procedure, which produces one surface layer of A spins, and one surface layer of B spins. If radiation incident on a (100) surface of a semi-infinite medium is considered, absorption occurs only for one sense of circular polarization.

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