Thermodynamics of thin ferromagnetic films in the presence of anisotropy and dipolar coupling

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(Received 6 May 1991)

We discuss aspects of the thermodynamics of ultrathin ferromagnetic films, with dipolar couplings and anisotropy present. We present a formalism that allows one to form proper normal-mode spin-wave annihilation and creation operators, from the left and right eigenvectors generated by the non-Hermitian matrix encountered in the spin-wave equations of motion. We present numerical calculations that explore the dependence on magnetic field (parallel to the surface) of the magnetization of an ultrathin film with easy axis normal to the surface. The magnetization has a singularity at the critical field where the magnetization of the film just aligns parallel to the surface. We discuss the nature of the spin correlations near the critical field.

INTRODUCTION

Currently, there is great interest in the properties of ultrathin (few atomic layer) ferromagnetic films. These materials have very interesting physical properties. For example, their Curie temperature can be substantially depressed below that of the bulk material. When these films are modeled as two-dimensional Heisenberg ferromagnets, their transition temperature should in fact be strictly zero. It has been argued that the strong uniaxial anisotropy found often in these films is responsible for inducing long-range order.[1]

Well below the ordering temperature, spin-wave theory may be applied to the ultrathin films. This paper is concerned with aspects of their thermodynamics that we view as striking and unique. Consider a film with an easy axis that orients the magnetization normal to the film surface. Application of a magnetic field of magnitude H_{ex} parallel to the film surfaces cants the spins; at a certain critical field H_{ex}^c, the magnetization just aligns parallel to the surface and remains parallel thereafter for stronger fields. When $H_{ex}=H_{ex}^c$ we have a second-order phase transition. Within spin-wave theory, the magnetization $M(T)/M(0)$ of an N-layer film may be written $M(T)/M(0)=1-(1/S)\Delta_N(T)$, where S is the length of a spin. At the critical field H_{ex}^c, we show that $\Delta_N(T)$ diverges logarithmically as $H_{ex}\rightarrow H_{ex}^c$ from above or below. This is a consequence of the presence of a "soft mode" at H_{ex}^c, which in fact has been studied experimentally by the light-scattering method.[2]

While above remarks are directed to the case where the easy axis orients the magnetization normal to the surface in zero external field, we note that ultrathin Fe films grown on W(110) have an easy axis in the plane of the film.[3] In this latter case, a similar singular behavior for $\Delta_N(T)$ is predicted if the magnetization is reoriented perpendicular to the easy axis by a field applied parallel to the film surfaces and normal to the easy axis. Thus a singular behavior for $\Delta_N(T)$ is predicted if the magnetization is reoriented perpendicular to the easy axis, whether the easy axis is normal or parallel to the film surfaces. It should be noted that, in our discussion below, we also remark on the physical origin of the anisotropy, as inferred from the behavior of $\Delta_N(T)$ for the Fe films on W(110).

Some years ago rare-earth films of macroscopic thickness were fabricated, ultimately for use in magnetic-bubble technology.[4] These have an easy axis normal to the surface and will experience a spin-reorientation transition, similar to the one considered above, when a magnetic field is applied parallel to the film plane. There will be a soft mode in such systems, which has properties very similar to that discussed in Ref. 2. However, we argue below that the singularity in $\Delta_N(T)$ is in fact very modest in such macroscopic films. In a sense described below, the singular contribution vanishes in the thermodynamic limit. Thus the singular behavior of $\Delta_N(T)$ explored here is a feature of ultrathin films only, whose thermodynamic properties are quasi-two-dimensional.

This paper may be viewed as an extension of our earlier work,[5] which examined features of spin waves in very thin films, in the presence of spin canting, within a fully microscopic theory. Here we show how to use the information generated in Ref. 5 to study the thermodynamic properties of the film. The extension is in fact nontrivial and has led us to examine and solve an issue present for many years in the literature on spin waves. The resolution we describe will prove useful in other areas of condensed-matter theory, where a quantum theoretic description leads one to a Hamiltonian that is a Hermitian structure with certain "anomalous terms," as discussed many years ago by Bogoliubov,[6] who treated them by means of a transformation that bears his name.

The basic approach to the theory of spin waves in magnetically ordered materials was put forward many years ago by Holstein and Primakoff.[7] When this approach is applied to an infinitely extended lattice of ferromagnetically aligned, exchange-coupled spins, the Hamiltonian is diagonalized quite trivially in the spin-wave limit, as discussed in many textbooks. If dipolar interactions are present or if the magnetic ground-state spin arrangement consists of sublattices of noncollinear spins, the problem is a bit more complex. The Hamiltonian of the infinitely

extended lattice is a quadratic form in the spin-wave annihilation and creation operators $a_{\mathbf{k}}$ and $a_{\mathbf{k}}^{\dagger}$, but contains terms involving the combination $a_{\mathbf{k}}a_{-\mathbf{k}}$ and its Hermitian adjoint, in addition to the diagonal terms $a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}$. The terms in $a_{\mathbf{k}}a_{-\mathbf{k}}$ and $a_{\mathbf{k}}^{\dagger}a_{-\mathbf{k}}^{\dagger}$ are the anomalous terms encountered by Bogoliubov. The spin-wave Hamiltonian is then diagonalized by the transformation mentioned earlier. By now there are a large number of examples in the literature where this is done;[6–9] the original Holstein-Primakoff paper incorporated dipolar couplings that generate such terms in the ferromagnet, for example.

So far as we know, however, there has never been a careful and complete discussion of the method of diagonalizing the spin Hamiltonian when off-diagonal terms are encountered, especially within the context of a film geometry. As remarked above, there is in fact a tricky issue that is not revealed in examples that are dealt with in a few lines of algebra. We address this question in general terms, resolve it by constructing appropriate transformations, and then apply the results to the study of the thermodynamic properties of very thin ferromagnetic films, where the spins interact via exchange and dipole coupling and are also influenced by surface or interfacial anisotropy.

The issue is the following, expressed in the language of spin-wave theory. In our earlier paper,[5] we explored the theory of spin waves in a ferromagnetic film in which the spins interact by means of isotropic exchange couplings and dipolar interactions. Spin canting could be induced by the combination of surface or interfacial anisotropy and an externally applied magnetic field. The spin-wave frequencies, along with their eigenvectors, can be found by diagonalizing a certain $2N\times 2N$ matrix, with N the number of layers in the film. This matrix is in fact non-Hermitian, in the presence of either noncollinear spins (produced by layer-dependent canting) or dipolar coupling. There are then right eigenvectors, which are those produced by the conventional diagonalization procedure, and also a distinct set known as left eigenvectors,[10] which we show below can be derived from the right eigenvectors for a spin-wave Hamiltonian of general structure (in the spin- wave limit). The original Hamiltonian is itself a Hermitian form, of course.

So far as we know, there is no prescription in the literature for constructing the spin-wave annihilation and creation operators from the left and right eigenvectors that emerge from the analysis of the non-Hermitian form associated with the equations of motion. The difficulty is serious, because in the absence of a general prescription, it is not clear how to calculate various thermodynamic properties of the film, how to generate various spin-correlation functions, and so on, from information generated from the equations of motion. As remarked earlier, the only examples in the literature are handled rather trivially in a few lines of algebra. The extension to the more complex problems encountered in the theory of multilayer films leads us to interesting aspects of the linear algebra of non-Hermitian forms, as we shall see. The analysis presented here provides a general prescription for constructing the proper spin-wave annihilation and creation operators of boson character in this circumstance and casting the Hamiltonian into diagonal form. We can then proceed to study any desired thermodynamic property of the film, at least in the spin-wave regime.

In Sec. II we derive the explicit form of the canonical transformation that diagonalizes the Hamiltonian, and in Sec. III we present studies of the physics of ultrathin films using the formalism. Section IV contains some final remarks.

II. CANONICAL FORM OF THE SPIN-WAVE HAMILTONIAN

As in Ref. 5, we consider a ferromagnetic film, infinite in planar extent, with two (001) surfaces. The film consists of N layers stacked to form a cubic crystal (fcc or bcc), with one atom per unit cell, in the bulk realization of the structure. In our model we include nearest-neighbor Heisenberg exchange, dipole-dipole coupling, and a uniaxial surface or interface anisotropy, with the easy axis normal to the film surfaces. The anisotropy energy includes terms quadratic and quartic in the relevant spin components. We also include an external magnetic field $\mathbf{H}_{\rm ex}$ of arbitrary magnitude and direction.

In the limit of small-amplitude spin waves and after a rotation to a set of layer-dependent local coordinates of the classical ground state,[5] we may write the Hamiltonian as

$$H=E_0-\frac{1}{4S}\sum_{l,l'}[S_-(l)\ \ S_+(l)]\times\begin{bmatrix}A(l,l') & B(l,l')^*\\ B(l,l') & A(l',l)\end{bmatrix}\begin{bmatrix}S_+(l')\\ S_-(l')\end{bmatrix}-\sum_l H_{\rm loc}(l_\perp)\Delta S_z(l)\ , \tag{1a}$$

where E_0 is the classical ground-state energy. The spins have length S, $S_\pm(l)$ are raising and lowering operators, and

$$S_z(l)=S+\Delta S_z(l)\ . \tag{1b}$$

The $+,-$ and z axes are reckoned with respect to the layer-dependent local coordinate system defined in Ref. 5; the local z axis, denoted by the unit vector $\hat{\mathbf{z}}(l_\perp)$, is aligned along the equilibrium orientation of the spins in layer $l_\perp$. At each site l, in film layer $l_\perp$, there is a local magnetic field, aligned parallel to this $\hat{\mathbf{z}}(l_\perp)$ axis of quantization, whose magnitude is given by

$$H_{\rm loc}(l_\perp)=\hat{\mathbf{z}}(l_\perp)\cdot\mathbf{H}_{\rm ex}+\sum_{l'}\Lambda_{zz}^{R}(l,l')+[h_{s_1}(l_\perp)+h_{s_2}(l_\perp)\cos^2\theta(l_\perp)]\times\cos^2\theta(l_\perp)\ , \tag{1c}$$

where $\theta(l_\perp)$ is the angle of canting, with respect to the film normal, of a spin in film layer $l_\perp$. In addition, the coefficients appearing in the 2×2 matrix of Eq. (1a) are defined as

$$A(\mathbf{l},\mathbf{l}')=\Lambda^R_{++}(\mathbf{l},\mathbf{l}')+\tfrac{1}{2}h_s(l_\perp)\sin^2\theta(l_\perp)\delta_{\mathbf{l},\mathbf{l}'}\ , \tag{1d}$$

$$B(\mathbf{l},\mathbf{l}')=\Lambda^R_{-+}(\mathbf{l},\mathbf{l}')+\tfrac{1}{2}h_s(l_\perp)\sin^2\theta(l_\perp)\delta_{\mathbf{l},\mathbf{l}'}\ , \tag{1e}$$

$$h_s(l_\perp)=h_{s_1}(l_\perp)+3h_{s_2}(l_\perp)\cos^2\theta(l_\perp)\ . \tag{1f}$$

Equations (1c–1e) contain coefficients of the form $\Lambda^R_{\alpha\beta}(\mathbf{l},\mathbf{l}')$, which are components of the exchange and dipole tensor Λ^R, rotated into the coordinates of the classical ground state, as discussed in Ref. 5. The surface or interface anisotropy dependence enters through the pinning fields $h_{s_1}(l_\perp)$ and $h_{s_2}(l_\perp)$, which represent the strength of terms quadratic and quartic in the spin components perpendicular to the film surfaces. A detailed discussion of the nature of the classical ground state under various conditions has been given by us earlier.[5]

We consider the second quantization of the Hamiltonian of Eq. (1a) via the Holstein-Primakoff transformation, retaining only the leading terms relevant to the spin-wave limit:

$$\begin{bmatrix}S_+(\mathbf{l})\\ S_-(\mathbf{l})\end{bmatrix}=\sqrt{2S}\,\sigma(\mathbf{l})\ , \tag{2a}$$

$$\Delta S_z(\mathbf{l})=-a^\dagger(\mathbf{l})a(\mathbf{l})\ , \tag{2b}$$

where $a(\mathbf{l})$ and $a^\dagger(\mathbf{l})$ are boson annihilation and creation operators, and the vector operator $\sigma(\mathbf{l})$ is defined as

$$\sigma(\mathbf{l})=\begin{bmatrix}a(\mathbf{l})\\ a^\dagger(\mathbf{l})\end{bmatrix}\ . \tag{2c}$$

If we apply Eqs. (2) to Eqs. (1), we find that we may write

$$H=E_0+E_H+H_{\rm SW}\ , \tag{3a}$$

where

$$E_H=-\tfrac{1}{2}\sum_{\mathbf{l}}H_{\rm loc}(l_\perp)\ , \tag{3b}$$

and, most importantly,

$$H_{\rm SW}=\tfrac{1}{2}\sum_{\mathbf{l},\mathbf{l}'}\sum_{\alpha,\beta=\pm}\sigma^\dagger_\alpha(\mathbf{l})\mathcal{H}_{\alpha\beta}(\mathbf{l},\mathbf{l}')\sigma_\beta(\mathbf{l}')\ , \tag{3c}$$

where we label the entries of two-dimensional vectors and matrices by $+,-$. The Hermitian tensor $\mathcal{H}$, whose components are $\mathcal{H}_{\alpha\beta}(\mathbf{l},\mathbf{l}')$, can be inferred from the two-dimensional matrix

$$\mathcal{H}(\mathbf{l},\mathbf{l}')=\begin{bmatrix}H_{\rm loc}(l_\perp)\delta_{\mathbf{l},\mathbf{l}'}-A(\mathbf{l},\mathbf{l}') & -B(\mathbf{l},\mathbf{l}')^*\\ -B(\mathbf{l},\mathbf{l}') & H_{\rm loc}(l_\perp)\delta_{\mathbf{l},\mathbf{l}'}-A(\mathbf{l}',\mathbf{l})\end{bmatrix}\ . \tag{3d}$$

We now focus our attention on the spin-wave term $H_{\rm SW}$. Consider a Fourier transform defined by

$$\sigma_\alpha(\mathbf{l})=\frac{1}{\sqrt{N_\parallel}}\sum_{\mathbf{k}_\parallel}\sigma_\alpha(\mathbf{k}_\parallel,l_\perp)e^{i\mathbf{k}_\parallel\cdot\mathbf{x}(\mathbf{l})}\ , \tag{4a}$$

where $N_\parallel$ is the number of atoms per film layer, $\mathbf{x}(\mathbf{l})$ is the position vector of lattice site $\mathbf{l}$, and $\mathbf{k}_\parallel$ is a wave vector whose direction is parallel to the film surfaces. Here we have defined the vector $\sigma(\mathbf{k}_\parallel,l_\perp)$, whose components are boson operators, by

$$\sigma(\mathbf{k}_\parallel,l_\perp)=\begin{bmatrix}a(\mathbf{k}_\parallel,l_\perp)\\ a^\dagger(-\mathbf{k}_\parallel,l_\perp)\end{bmatrix}\ . \tag{4b}$$

Therefore, substituting Eq. (4a) into $H_{\rm SW}$ of Eq. (3c), we find

$$H_{\rm SW}=\tfrac{1}{2}\sum_{\mathbf{k}_\parallel}\sum_{l_\perp,\alpha}\sum_{l'_\perp,\beta}\sigma^\dagger_\alpha(\mathbf{k}_\parallel,l_\perp)\mathcal{H}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l_\perp')\sigma_\beta(\mathbf{k}_\parallel,l'_\perp)\ , \tag{5a}$$

where the $2N$-dimensional Hermitian matrix $\mathcal{H}(\mathbf{k}_\parallel)$ has components

$$\mathcal{H}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l'_\perp)=\sum_{l'_\parallel}\mathcal{H}_{\alpha\beta}(\mathbf{l},\mathbf{l}')e^{-i\mathbf{k}_\parallel\cdot\mathbf{x}(\mathbf{l},\mathbf{l}')}\ , \tag{5b}$$

and $\mathbf{x}(\mathbf{l},\mathbf{l}')$ is the vector from site $\mathbf{l}$ to site $\mathbf{l}'$. Via Eqs. (3d) and (5b) and the properties of the tensor Λ^R, which can be inferred from the Hermiticity of the Hamiltonian, we may express the components $\mathcal{H}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l'_\perp)$ in terms of a 2×2 matrix. Thus we may write

$$\mathcal{H}(\mathbf{k}_\parallel;l_\perp,l'_\perp)=\begin{bmatrix}H_{\rm loc}(l_\perp)\delta_{l_\perp,l'_\perp}-A(\mathbf{k}_\parallel;l_\perp,l'_\perp) & -B(-\mathbf{k}_\parallel;l_\perp,l'_\perp)^*\\ -B(\mathbf{k}_\parallel;l_\perp,l'_\perp) & H_{\rm loc}(l_\perp)\delta_{l_\perp,l'_\perp}-A(-\mathbf{k}_\parallel;l_\perp,l'_\perp)^*\end{bmatrix}\ , \tag{6}$$

where

$$A(\mathbf{k}_\parallel;l_\perp',l_\perp)^*=A(\mathbf{k}_\parallel;l_\perp,l_\perp') , \tag{7a}$$

$$B(-\mathbf{k}_\parallel;l_\perp',l_\perp)=B(\mathbf{k}_\parallel;l_\perp,l_\perp') . \tag{7b}$$

The form of Eq. (6), and therefore Eq. (5a), results strictly from the hermiticity of $H_{\rm SW}$ and is independent of the particular rotational symmetries of a given infinitely extended film.

A dynamical matrix $\mathcal{D}(\mathbf{k}_\parallel)$, from which spin-wave frequencies and eigenvectors may be generated, can be determined from the equations of motion of the annihilation and creation operators. In our notation these can be written (in units with $\hbar=1$)

$$i\frac{\partial}{\partial t}\sigma_\alpha(\mathbf{k}_\parallel,l_\perp;t)=[\sigma_\alpha(\mathbf{k}_\parallel,l_\perp;t),H_{\rm SW}] . \tag{8a}$$

Upon using Eq. (5a) for $H_{\rm SW}$, we have

$$i\frac{\partial}{\partial t}\sigma_\alpha(\mathbf{k}_\parallel,l_\perp;t)=\sum_{l_\perp',\beta}\mathcal{D}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l_\perp')\sigma_\beta(\mathbf{k}_\parallel,l_\perp';t) , \tag{8b}$$

where components of the dynamical matrix are given by

$$\mathcal{D}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l_\perp')=\alpha\mathcal{H}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l_\perp') . \tag{8c}$$

Here $\alpha,\beta=\pm$ and one can see that $\mathcal{D}(\mathbf{k}_\parallel)$ is in general a complex, non-Hermitian matrix of dimension $2N$. Explicitly, we have

$$\mathcal{D}(\mathbf{k}_\parallel;l_\perp,l_\perp')=\begin{bmatrix} H_{\rm loc}(l_\perp)\delta_{l_\perp,l_\perp'}-A(\mathbf{k}_\parallel;l_\perp,l_\perp') & -B(-\mathbf{k}_\parallel;l_\perp,l_\perp')^* \\ B(\mathbf{k}_\parallel;l_\perp,l_\perp') & -H_{\rm loc}(l_\perp)\delta_{l_\perp,l_\perp'}+A(-\mathbf{k}_\parallel;l_\perp,l_\perp')^* \end{bmatrix} . \tag{8d}$$

By seeking a solution of Eq. (8b) of the form

$$\sigma_\alpha(\mathbf{k}_\parallel,l_\perp;t)=\sigma_\alpha(\mathbf{k}_\parallel,l_\perp)e^{-i\Omega(\mathbf{k}_\parallel)t} , \tag{8e}$$

we can attempt to diagonalize $\mathcal{D}(\mathbf{k}_\parallel)$; however, it is easy to see that $\mathcal{D}(\mathbf{k}_\parallel)$ is not a normal matrix since $[\mathcal{D}(\mathbf{k}_\parallel),\mathcal{D}^\dagger(\mathbf{k}_\parallel)]\neq 0$. Hence a similarity transformation, if one exists, cannot be unitary.[11] It follows that, for $\mathcal{D}(\mathbf{k}_\parallel)$, we must introduce the concept of right and left eigenvectors, which we shall denote by the column vector $\mathbf{e}^{(s)}(\mathbf{k}_\parallel)$ and the row vector $\mathbf{u}^{(s)}(\mathbf{k}_\parallel)$, respectively.[10] Here the index s is defined to be an integer $|s|\leq N$, $s\neq 0$. The components of our eigenvectors satisfy the equations

$$\sum_{l_\perp',\beta}\mathcal{D}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l_\perp')e_\beta^{(s)}(\mathbf{k}_\parallel;l_\perp')=\Omega^{(s)}(\mathbf{k}_\parallel)e_\alpha^{(s)}(\mathbf{k}_\parallel;l_\perp) , \tag{9a}$$

$$\sum_{l_\perp',\beta}u_\beta^{(s)}(\mathbf{k}_\parallel;l_\perp')\mathcal{D}_{\beta\alpha}(\mathbf{k}_\parallel;l_\perp',l_\perp)=\Omega^{(s)}(\mathbf{k}_\parallel)u_\alpha^{(s)}(\mathbf{k}_\parallel;l_\perp) , \tag{9b}$$

where $\Omega^{(s)}(\mathbf{k}_\parallel)$ is the frequency, measured in magnetic-field units, of a spin-wave mode. A basic property of the right and left eigenvectors is the analog of the completeness and orthonormality relations of the theory of Hermitian matrices:

$$\sum_s e_\alpha^{(s)}(\mathbf{k}_\parallel;l_\perp)u_\beta^{(s)}(\mathbf{k}_\parallel;l_\perp')=\delta_{l_\perp,l_\perp'}\delta_{\alpha\beta} , \tag{10a}$$

$$\sum_{l_\perp\alpha}u_\alpha^{(s)}(\mathbf{k}_\parallel;l_\perp)e_\alpha^{(s')}(\mathbf{k}_\parallel;l_\perp)=\delta_{ss'} . \tag{10b}$$

If the classical ground state is stable (there is no guarantee of this), then the Hermitian matrix $\mathcal{H}(\mathbf{k}_\parallel)$, as it appears in Eq (5a), must be positive definite. In addition, in view of Eq. (8c), we may construct the $2N$-dimensional Hermitian matrix $\mathcal{I}$, defining its components by

$$\mathcal{I}_{\alpha\beta}(l_\perp,l_\perp')=\alpha\delta_{l_\perp,l_\perp'}\delta_{\alpha\beta} , \tag{11}$$

such that $\mathcal{D}(\mathbf{k}_\parallel)$ can be written as the matrix product $\mathcal{I}\mathcal{H}(\mathbf{k}_\parallel)$. Since $\mathcal{D}(\mathbf{k}_\parallel)$ can be written as the product of two Hermitian matrices, where $\mathcal{H}(\mathbf{k}_\parallel)$ is positive definite, then $\mathcal{D}(\mathbf{k}_\parallel)$ can be diagonalized, such that all eigenvalues $\{\Omega^{(s)}(\mathbf{k}_\parallel);\ |s|\leq N,\ s\neq 0\}$ are real numbers.[12]

From the form of the dynamical matrix, as given by Eqs. (8d) and (7), one can determine important relations among the eigenvalues and eigenvectors of $\mathcal{D}(\mathbf{k}_\parallel)$. These relations are easily verified; we state them below. (i) For every *right* eigenvector $\mathbf{e}^{(s)}(\mathbf{k}_\parallel)$, with eigenvalue $\Omega^{(s)}(\mathbf{k}_\parallel)$, there exists another *right* eigenvector, which we label by the index $-s$, with components that can be defined as

$$e_\alpha^{(-s)}(\mathbf{k}_\parallel;l_\perp)=[e_{-\alpha}^{(s)}(-\mathbf{k}_\parallel;l_\perp)]^* , \tag{12a}$$

and whose eigenvalue corresponds to

$$\Omega^{(-s)}(\mathbf{k}_\parallel)=-\Omega^{(s)}(-\mathbf{k}_\parallel) . \tag{12b}$$

(ii) For every *right* eigenvector $\mathbf{e}^{(s)}(\mathbf{k}_\parallel)$, there exists a *left* eigenvector, corresponding the the *same* eigenvalue, whose components can be defined as

$$u_\alpha^{(s)}(\mathbf{k}_\parallel;l_\perp)=\mathrm{sgn}(s)[\alpha e_\alpha^{(s)}(\mathbf{k}_\parallel;l_\perp)]^* , \tag{12c}$$

where $\mathrm{sgn}(s)$ denotes the sign value of s. The factor $\mathrm{sgn}(s)$ ensures that Eqs. (12a) and (12c) are consistent with the completeness and orthonormality relations of Eqs. (10), where Eqs. (10) must hold for $-\mathbf{k}_\parallel$ as well as $\mathbf{k}_\parallel$. (iii) Since $\mathcal{H}(\mathbf{k}_\parallel)$ is assumed positive definite, then $e^{(s)}(\mathbf{k}_\parallel)^\dagger\cdot\mathcal{H}(\mathbf{k}_\parallel)\cdot e^{(s)}(\mathbf{k}_\parallel)>0$ for any s. However, from this

statement and Eqs (8c) and (12c), it is easy to verify that

$$\Omega^{(s)}(\mathbf{k}_\parallel)=\begin{cases} u^{(s)}(\mathbf{k}_\parallel)\cdot\mathcal{D}(\mathbf{k}_\parallel)\cdot e^{(s)}(\mathbf{k}_\parallel)<0, & s<0 \\ u^{(s)}(\mathbf{k}_\parallel)\cdot\mathcal{D}(\mathbf{k}_\parallel)\cdot e^{(s)}(\mathbf{k}_\parallel)>0, & s>0 \end{cases} . \tag{12d}$$

It follows that half the eigenvalues for a given wave vector $\mathbf{k}_\parallel$ are positive and half are negative. By our definition of Eq. (12c), we have ordered the eigenvalues and eigenvectors so that for positive integers s, $1\le s\le N$, we have $\Omega^{(s)}(\mathbf{k}_\parallel)>0$ and for negative integers s $-N\le s\le -1$, we have $\Omega^{(s)}(\mathbf{k}_\parallel)<0$. As one would expect, there are precisely N distinct physical frequencies that result from the diagonalization of $\mathcal{D}(\mathbf{k}_\parallel)$; in addition, one obtains, via Eq. (12b), the negative of the N physical frequencies for $-\mathbf{k}_\parallel$. We thus have arranged the eigenvalues so the following relation holds:

$$\Omega^{(s)}(\mathbf{k}_\parallel)=\mathrm{sgn}(s)\Omega^{(|s|)}(\mathrm{sgn}(s)\mathbf{k}_\parallel) , \tag{12e}$$

where once again $\mathrm{sgn}(s)$ denotes the sign value of s.

In view of Eq. (12e), we introduce the positive integer n, $1\le n\le N$, and define a canonical transformation by

$$a(\mathbf{k}_\parallel,l_\perp)-\sum_{n=1}^{N}[e_+^{(n)}(\mathbf{k}_\parallel;l_\perp)a(\mathbf{k}_\parallel,n) +e_+^{(-n)}(\mathbf{k}_\parallel;l_\perp)a^\dagger(-\mathbf{k}_\parallel,n)] \tag{13a}$$

and its inverse by

$$a(\mathbf{k}_\parallel,n)=\sum_{l_\perp=1}^{N}[u_+^{(n)}(\mathbf{k}_\parallel;l_\perp)a(\mathbf{k}_\parallel,l_\perp) +u_-^{(n)}(\mathbf{k}_\parallel;l_\perp)a^\dagger(-\mathbf{k}_\parallel,l_\perp)] , \tag{13b}$$

where, for example, $a(\mathbf{k}_\parallel,n)$ is a boson annihilation operator. One can easily verify, via Eqs. (10) and (12), that $a(\mathbf{k}_\parallel,n)$ and $a^\dagger(\mathbf{k}_\parallel,n)$ satisfy the commutation relations appropriate to boson operators. The relations in Eqs. (13) constitute the generalized Bogoliubov transformation discussed in Sec. I.

We can also express the above transformation in a more convenient form by defining the matrix $\Gamma(\mathbf{k}_\parallel)$ to represent the $2N$-dimensional similarity transformation implied by Eqs. (9). We therefore let

$$\Gamma_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,n)=e_\alpha^{(\beta n)}(\mathbf{k}_\parallel;l_\perp) , \tag{14a}$$

$$\Gamma^{-1}_{\alpha\beta}(\mathbf{k}_\parallel;n,l_\perp)=u_\beta^{(\alpha n)}(\mathbf{k}_\parallel;l_\perp) . \tag{14b}$$

We may then write

$$\sigma_\alpha(\mathbf{k}_\parallel,l_\perp)=\sum_{n,\beta}\Gamma_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,n)\sigma_\beta(\mathbf{k}_\parallel,n) , \tag{14c}$$

$$\sigma_\alpha(\mathbf{k}_\parallel,n)=\sum_{l_\perp,\beta}\Gamma^{-1}_{\alpha\beta}(\mathbf{k}_\parallel;n,l_\perp)\sigma_\beta(\mathbf{k}_\parallel,l_\perp) , \tag{14d}$$

where we have introduced the vector operator $\sigma(\mathbf{k}_\parallel,n)$, given by

$$\sigma(\mathbf{k}_\parallel,n)=\begin{bmatrix} a(\mathbf{k}_\parallel,n) \\ a^\dagger(-\mathbf{k}_\parallel,n) \end{bmatrix} . \tag{14e}$$

We can now demonstrate, with the aid of Eqs. (10) and (12), that H_{SW} is indeed diagonal in $a(\mathbf{k}_\parallel,n)$ and $a^\dagger(\mathbf{k}_\parallel,n)$. We first multiply $\mathcal{H}(\mathbf{k}_\parallel)$ by $\Gamma(\mathbf{k}_\parallel)$, as defined in Eq. (14a), to obtain, with the aid of Eqs. (8c) and (9a), the result

$$\sum_{l'_\perp,\beta}\mathcal{H}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l'_\perp)\Gamma_{\beta\gamma}(\mathbf{k}_\parallel;l'_\perp,n) =\alpha\Omega^{(\gamma n)}(\mathbf{k}_\parallel)e_\alpha^{(\gamma n)}(\mathbf{k}_\parallel;l_\perp) . \tag{15a}$$

Then, applying the relations of Eqs. (12c) and (12e) and using the definition for the inverse of the similarity transformation given by Eq. (14b), we find

$$\sum_{l'_\perp,\beta}\mathcal{H}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l'_\perp)\Gamma_{\beta\gamma}(\mathbf{k}_\parallel;l'_\perp,n) =\Omega^{(n)}(\gamma\mathbf{k}_\parallel)(\Gamma^\dagger)^{-1}_{\alpha\gamma}(\mathbf{k}_\parallel;l_\perp,n) . \tag{15b}$$

Rearranging this expression, we have

$$\sum_{l_\perp,\alpha}\sum_{l'_\perp,\beta}\Gamma^\dagger_{\gamma\alpha}(\mathbf{k}_\parallel;n,l_\perp)\mathcal{H}_{\alpha\beta}(\mathbf{k}_\parallel;l_\perp,l'_\perp) \Gamma_{\beta\gamma'}(\mathbf{k}_\parallel;l'_\perp,n')=\Omega^{(n)}(\gamma\mathbf{k}_\parallel)\delta_{nn'}\delta_{\gamma\gamma'} . \tag{15c}$$

It is now straightforward to write H_{SW} in diagonal form. Substituting Eq. (14c) into Eq. (5a) and utilizing Eq. (15c) above, we arrive at the result

$$H_{\mathrm{SW}}=\sum_{\mathbf{k}_\parallel,n}\Omega^{(n)}(\mathbf{k}_\parallel)[a^\dagger(\mathbf{k}_\parallel,n)a(\mathbf{k}_\parallel,n)+\tfrac{1}{2}] . \tag{16}$$

As we have shown, the frequency $\Omega^{(n)}(\mathbf{k}_\parallel)$ that appears in Eq. (16) is a positive number.

As a last remark in this section, we would like to comment on properties of the dispersion relations in the spin-wave limit, specifically, under what conditions $\Omega^{(n)}(\mathbf{k}_\parallel)$ is an even function of $\mathbf{k}_\parallel$. If the ground-state spin configuration is one with all spins parallel, whether aligned normal or tangent to the film surfaces (or canted out of the plane in the absence of dipole-dipole coupling), the dispersion relations are even functions of $\mathbf{k}_\parallel$. This is easy to see since, under these circumstances, one has $A(\mathbf{k}_\parallel;l_\perp,l'_\perp)=A(-\mathbf{k}_\parallel;l_\perp,l'_\perp)^*$ in Eq. (8d). Hence we find that $\Omega^{(n)}(\mathbf{k}_\parallel)$, which is an even function of $\mathbf{k}_\parallel$ within the context of a purely exchange-coupled spin model, is also an even function of $\mathbf{k}_\parallel$ even when dipole-dipole coupling is introduced, regardless of the symmetry of the infinitely extended film.

However, in the presence of a surface anisotropy with the easy axis normal to the film, where one finds the classical ground-state spin configuration to be one canted out of the film plane, the demagnetizing effect of dipole-dipole coupling renders a twist in the spin configuration; i.e., the angle of canting is a function of film layer. When such a deviation from "hard" ferromagnetic alignment occurs, the result is, quite generally, a nonreciprocal dispersion relation, i.e., $\Omega^{(n)}(-\mathbf{k}_\parallel)\neq\Omega^{(n)}(\mathbf{k}_\parallel)$. Under conditions where the exchange coupling is much stronger than dipole coupling, a feature found in very thin Fe films, the twist present in the ground-state spin arrangement is quite small. Our earlier calculations[5] show that this is less than 1° in Fe films if one compares the directions of spins in, say, a four- or five-layer film. Under

these conditions, to excellent approximation, $\Omega^{(n)}(\mathbf{k}_\parallel)$ is an even function of $\mathbf{k}_\parallel$.

III. CALCULATION OF THE TEMPERATURE AND FIELD VARIATION OF THE MAGNETIZATION IN ULTRATHIN FILMS: THE MAGNETIC-FIELD VARIATION OF THE MAGNETIZATION NEAR THE SPIN-REORIENTATION TRANSITION

The discussion of the previous section provides us with the formal apparatus required to calculate thermodynamic properties and spin-correlation functions in the ultrathin film. We have seen that the eigenvalues of the $2N\times 2N$ non-Hermitian matrix $\mathcal{D}(\mathbf{k}_\parallel)$, associated with the equations of motion, quite generally yield N distinct positive frequencies $\Omega^{(n)}(\mathbf{k}_\parallel)$, which are, with $\hbar=1$, the various excitation energies of the spin waves. The boson annihilation and creation operators associated with these modes are $a(\mathbf{k}_\parallel,n)$ and $a^\dagger(\mathbf{k}_\parallel,n)$, and these are related, via the generalized Bogoliubov transformation of Eqs. (13), to the spin-deviation operators $a(\mathbf{k}_\parallel,l_\perp)$ and $a\dagger(\mathbf{k}_\parallel,l_\perp)$ defined in Eq. (4). Keep in mind that $l_\perp$ labels a particular plane in the film. Furthermore, if the spins in the ground state in plane $l_\perp$ are canted with respect to the film normal, then the z axis referred to in Eqs. (1b) and (2b) is a "local" z axis, aligned along the direction of the spins in layer $l_\perp$.

In this section we shall explore the behavior of

$$\langle S_z(l)\rangle = S - \langle a^\dagger(l)a(l)\rangle \equiv S - \Delta_N(l_\perp,T)\ , \tag{17}$$

where

$$\Delta_N(l_\perp,T)=\langle a^\dagger(l)a(l)\rangle = \frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\langle a^\dagger(\mathbf{k}_\parallel,l_\perp)a(\mathbf{k}_\parallel,l_\perp)\rangle$$

is the spin deviation in layer $l_\perp$ of an N-layer film.

A measurement of the magnitude and temperature variation of the hyperfine field, provided by Mössbauer experiments such as those reported in Ref. 3, gives access to $\Delta_N(l_\perp,T)$. If the ground-state spin arrangement is canted with respect to an applied magnetic field, the component of the total magnetization of the film along the direction of the applied field is given by

$$M_H(T)=\frac{M_0}{NS}\sum_{l_\perp}\langle S_z(1)\rangle\cos\Psi(l_\perp)\ , \tag{18}$$

where M_0 is the saturation magnetization and $\Psi(l_\perp)$ is the angle between the spins in layer $l_\perp$ and the applied field. Thus the measurement of the total magnetization of the film provides information on $\Delta_N(l_\perp,T)$ also.

The transformation in Eqs. (13) allow us to express $\Delta_N(l_\perp,T)$ in terms of the eigenvector components generated by the equation of motion analysis. We have

$$\Delta_N(l_\perp,T)=\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{n=1}^{N}\{|e_+^{(n)}(\mathbf{k}_\parallel,l_\perp)|^2 n_B(\Omega^{(n)}(\mathbf{k}_\parallel))+|e_-^{(n)}(\mathbf{k}_\parallel,l_\perp)|^2[1+n_B(\Omega^{(n)}(\mathbf{k}_\parallel))]\}\ , \tag{19}$$

where $n_B(\Omega)=(e^{\Omega/k_BT}-1)^{-1}$ is the Bose-Einstein function.

Note that $\Delta_N(l_\perp,T)$ remains finite as the temperature $T\to 0$ because of the presence of the $1+n_B$ term in Eq. (19). In general, there are zero-point oscillations in the spin system. In the absence of dipolar interactions and if there is no layer-dependent anisotropy (i.e., we have a perfectly aligned, ferromagnetic array of exchange-coupled spins, possibly in an external magnetic field), then $B(l,l')$ in Eq. (3d) vanishes, $|e_-^{(n)}(\mathbf{k}_\parallel,l_\perp)|^2$ vanishes, and $\Delta_N(l_\perp,T)\to 0$ as $T\to 0$. More generally, zero-point spin motions exist, an issue that may cause concern in thin films where the exchange and dipolar couplings are of comparable magnitude. The role of zero-point oscillations would be a concern in such a sample if one wishes to extract absolute values of the hyperfine coupling constants from the data.

We now turn to the results of our numerical calculations of $\Delta_N(l_\perp,T)$. We shall show results for $\Delta_N(l_\perp,T)$ for the monolayer and for a trilayer, with the parameters in the latter case chosen to coincide with those deduced from the soft-mode data of the authors of Ref. 2.

The films have nearest-neighbor exchange coupling J whose strength is such that, in the long-wavelength limit, the exchange stiffness constant D of bulk Fe is reproduced. The trilayer consists of three layers arranged in a fcc fashion, with an external field $\mathbf{H}_{ex}$ applied in the [100] direction. The structure of the monolayer is defined by removing two layers from the trilayer. The dipole moment per spin is adjusted so that, for bulk material formed from such spin arrays, $4\pi M_0=18$ kG. We apply easy-axis anisotropy to the two outer layers of spins of the trilayer and to the monolayer, with the parameters h_{s1} and h_{s2} in Eq. (1c) equal to 30.90 and 1.68 kG, respectively. These parameters nicely reproduce the field dependence reported for the $\mathbf{k}_\parallel=0$ spin-wave mode in the elegant Brillouin light-scattering experiment of Ref. 2. Our fit to the data is displayed in Fig. 5 of Ref. 5. The parameters are also compatible with those used in Ref. 2, in the theoretical analysis reported there.

In Fig. 1 we sketch the ground-state spin arrangement in the trilayer film as a function of external field. In zero field the easy-axis anisotropy orients all spins normal to the film surfaces, as illustrated in Fig. 1(a). Then application of the field parallel to the film surfaces leads to spin canting, as illustrated in Fig. 1(b). In our calculations we allow for layer-dependent spin canting although, in our model of very thin Fe film, the variation in canting angle from layer to layer is quite small, the order of one degree. If the field is less than the critical field H_{ex}^c

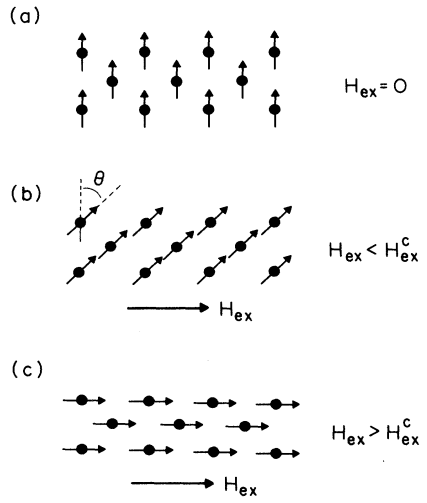

FIG. 1. Pictured is a schematic illustration of the ground-state spin arrangement for the model film explored in Sec. III. (a) When the external field is zero, the magnetization is normal to the film surfaces by virtue of the easy-axis anisotropy. (b) The external magnetic field H_{ex} applied parallel to the film surfaces is less than the critical field H_{ex}^{c} required to rotate the magnetization through 90°. (c) When $H_{ex} > H_{ex}^{c}$, the magnetization is strictly parallel to the film surfaces.

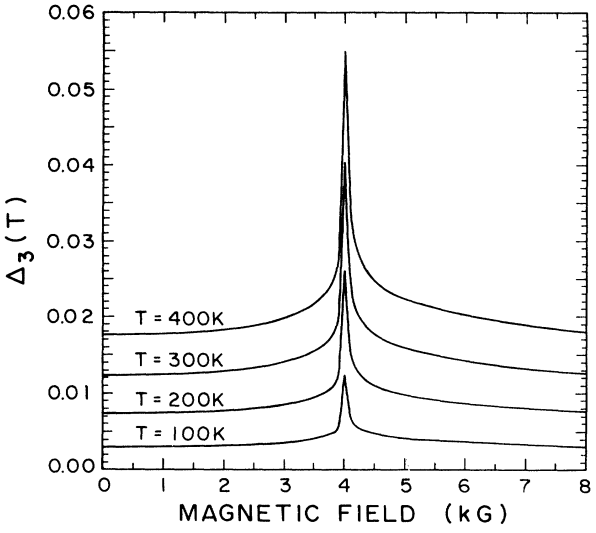

FIG. 2. Plotted is the dependence on magnetic field of the spin deviation $\Delta_3(T)$, corresponding to a layer in the trilayer film. We show calculations for various temperatures and for parameters chosen as described in the text. The spin deviation is essentially identical in all three layers of the trilayer.

($H_{ex}^{c}=4$ kG for the trilayer and the above parameters), the canting angle is less than $\pi/2$. When $H_{ex}=H_{ex}^{c}$ the canting angle is $\pi/2$; this is the field at which the $\mathbf{k}_{\parallel}=\mathbf{0}$ low-lying spin wave mode "goes soft." For $H_{ex}>H_{ex}^{c}$, of course, the canting angle remains pinned at the value $\pi/2$, and as discussed in Ref. 2, the $\mathbf{k}_{\parallel}=\mathbf{0}$ spin-wave mode has a finite frequency that increases linearly with external field.

We found $\Delta_N(l_\perp,T)$ to be essentially identical in each layer of the trilayer for the values of the parameters we considered. Hence, suppressing the index $l_\perp$, we show in Fig. 2 the magnetic-field dependence of $\Delta_3(T)$ for various temperatures. There is clearly a divergence at the critical field H_{ex}^{c} for all temperatures. We presume that, in fact, right at H_{ex}^{c} where the low-lying spin-wave branch has zero frequency at $\mathbf{k}_{\parallel}=\mathbf{0}$, spin-wave theory must break down. True long-range order should disappear at this critical field.

The calculations in Fig. 2 include the contribution to $\Delta_3(T)$ from each of the three spin-wave branches associated with each wave vector $\mathbf{k}_{\parallel}$. In fact, the low-lying branch provides the dominant contribution. We have tested this by calculating $\Delta_3(l_\perp,T)$ for each of the three layers, to find, as we have mentioned, only very small differences for the temperatures considered in Fig. 2. The low-lying branch is an acoustical branch, wherein the spins in each of the three layers precess very nearly in phase and with equal amplitude for all values of $\mathbf{k}_{\parallel}$ significant in the thermodynamics. The very small dependence of $\Delta_3(l_\perp,T)$ on $l_\perp$ thus shows that this one branch dominates the thermodynamics, even for temperatures well above room temperature. Hence, in the regime of temperatures covered in Fig. 2, the statistical mechanics of the film is indeed two dimensional to a very high degree of accuracy.

We wish to comment on the relationship between our results and those reported by the authors of Ref 3, who measured, by means of Mössbauer spectroscopy, the magnitude and temperature variation of $\Delta_N(T)$ for Fe films on W(110). The structure of these films (unknown in detail) is different from the model film used to generate Fig. 2. For example, the magnetization lies in plane in the samples used in Ref 3, and surely the two-dimensional unit cell of these ultrathin absorbed Fe films is rectangular and the same as that of the W(110) substrate upon which the films are deposited.

The authors of Ref. 3 fit the data to a power law $\Delta_N(T)=bT^n$ to find that n lies in the range of 1.5 ± 0.2. The zero-field results displayed in Fig. 2 can be fitted with a power law also, where $n\cong1.3$, the lower end of the range quoted in Ref. 3. There is no theoretical justification for the power-law behavior of $\Delta_N(T)$ in such films, so far as we know; it is often the case that information which covers a limited temperature range can be reproduced by a power law. The effective exponent n will surely be affected by details of the anisotropy, which in part controls the gap at $\mathbf{k}_{\parallel}=0$ in the low-lying acoustic spin-wave mode that we have seen controls the thermodynamic properties of the ultrathin film. Nonetheless, our calculations show clearly that $\Delta_N(T)$ displays a temperature dependence stronger than linear and not so far off the well-known $T^{3/2}$ law provided by bulk spin-wave theory.

There is a most remarkable feature in the data reported in Ref. 3, which is noted and discussed by these authors. If we write $\Delta_N(T)=bT^n$, then for films that range in thickness from one to four monolayers the coefficient b scales *inversely* with film thickness; to within experimental uncertainty the value of b for a four-layer film is smaller than that of a monolayer by a factor of 4.

The authors of Ref 3 argue that if the dispersion relation of the spin-wave branch that controls the thermodynamics of the film (our low-lying acoustic spin-wave branch) is independent of film thickness, then this result follows. One may see this scaling behavior from our Eq. (10b), noting the relation between the left and right eigenvectors. For the low-lying acoustic spin-wave branch, $e_\alpha^{(s)}(\mathbf{k}_\parallel,l_\perp)$ will be independent of $l_\perp$ to a very good approximation, and so as layers are added, $|e_\alpha^{(s)}(\mathbf{k}_\parallel,l_\perp)|^2$ will scale inversely with film thickness [note the sum on $l_\perp$ in Eq. (10b)]. From Eq. (19) it follows that $\Delta_N(l_\perp,T)$ does also, provided the dispersion relation is independent of film thickness.

One must then inquire under what conditions the dispersion relation is independent of film thickness. If the spins in the film surface or at an interface experience anisotropy substantially different from those in the film interior (of a multilayer structure), then the gap in the acoustic spin-wave branch, and consequently the dispersion relation of the acoustic branch, will *not* be independent of film thickness. Under these conditions the scaling relation evident in the data of Ref. 3 will *not* be obeyed. We illustrate this in Fig. 3. Our model calculations for the trilayer (Fig. 2) assume that the spins in only the outermost layers experience the single-site anisotropy. while those in the interior layer experience no anisotropy.

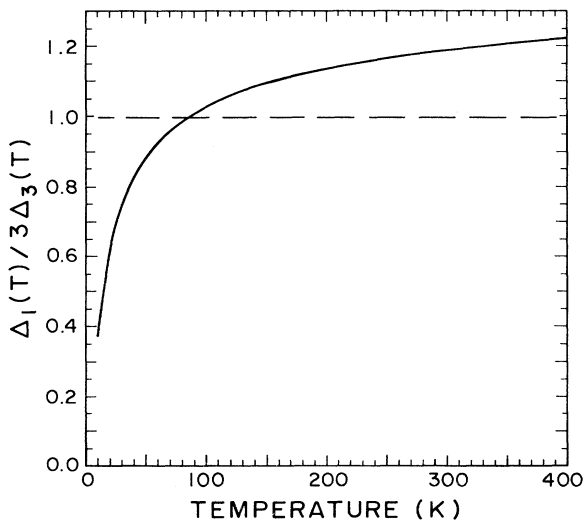

FIG. 3. In zero magnetic field we plot the temperature variation of $\Delta_1(T)/3\Delta_3(T)$, where $\Delta_1(T)$ is the spin deviation in the monolayer and $\Delta_3(T)$ is the spin deviation in a layer of the trilayer. The spin deviation is nearly identical in all three layers of the trilayer.

In Fig. 3 we calculate the ratio $\Delta_1(T)/3\Delta_3(T)$, where $\Delta_3(T)$ is the spin deviation for the model trilayer in zero field (the zero-field results of Fig. 2) and $\Delta_1(T)$ is that for a monolayer in which all spins "feel" anisotropy equal to that in the outer layers of the trilayer. The ratio is clearly not unity.

The differences are influenced both by the gap at $\mathbf{k}_\parallel=0$ and by the fact that the curvature of the dispersion relations at large $\mathbf{k}_\parallel$ differs for the two cases. In the monolayer a given spin is exchange coupled to fewer neighbors than, on the average, a spin in the trilayer. The curvature in the monolayer dispersion relation is thus smaller than in the trilayer at large $\mathbf{k}_\parallel$; the large-$\mathbf{k}_\parallel$ spin waves are "softer" in the monolayer than in the trilayer, and so $\Delta_1(T)/3\Delta_3(T)$ rises above unity at higher temperatures.[13] The gap at $\mathbf{k}_\parallel$ in the monolayer is larger than that in the trilayer, and so $\Delta_1(T)/3\Delta_3(T)$ plunges below unity at the lower temperatures.

If, in a multilayer film, spins in all layers experience identical anisotropy, then within the framework of a macroscopic theory, such as that presented in Ref. 2, the gap in the low-lying spin-wave branch will be nearly independent of film thickness, provided the anisotropy is much larger than the maximum dipole field of $4\pi M_0$. Under these conditions one may expect that the scaling with film thickness, evident in the data, may be realized more closely in theory. At least the sharp drop in the ratio of $\Delta_1(T)/3\Delta_3(T)$ at lower temperatures (see Fig. 3) will be absent.

These remarks have a bearing on the physical origin of the anisotropy in the films studied in Ref. 3; it is very likely that the anisotropy is strain induced, associated with the rectangular distortion required for these films to grow epitaxially on the W(110) substrate. If this were the dominant source of anisotropy, then all spins in the film would experience uniaxial anisotropy of the same magnitude. Such strain-induced anisotropy would also produce an easy axis that lies within the film plane, rather than perpendicular to its surface.

IV. FURTHER CONSIDERATIONS

We conclude with some final remarks. We have seen that for the ultrathin film, the magnetization as measured by $\Delta_N(l_\perp,T)$ is singular at the critical value of the applied field at which the magnetization just becomes parallel to the surface, [see Fig. 1(c)]. It is interesting to inquire how this singularity depends on film thickness. Unfortunately, it becomes prohibitive to carry out full and complete numerical calculations for films thick enough for the higher-lying spin-wave modes to influence the thermodynamics of the film importantly.

Nonetheless, some general remarks can be made. The physical situation with the trilayer is illustrated in Fig. 4(a). For each value of the wave vector $\mathbf{k}_\parallel$ parallel to the surface, we have three spin-wave branches, and the direction of spin-wave propagation is essentially isotropic (see Fig. 6 of Ref. 5 and the associated discussion). The lowest-frequency branch, the acoustical branch we have seen that dominates the thermodynamics, has a gap ω_0 at $\mathbf{k}_\parallel=\mathbf{0}$ controlled by the anisotropy, critical field, and di-

pole coupling. As $H_{ex} \to H_{ex}^{c}$ from either above or below, $\omega_0 \to 0$. The dispersion relation of this lowest branch is represented reasonably well, near $H_{ex}=0$, by the form

$$\Omega(\mathbf{k}_\parallel)=\omega_0+\tilde{D}k_\parallel^2 \ . \tag{20}$$

Again, we may suppose $\Delta_3(l_\perp,T)$ independent of $l_\perp$ to an excellent approximation, given the number of thermally excited spin waves present. Near $H_{ex}=0$ we have $B(\mathbf{k}_\parallel;l_\perp,l_\perp')\cong 0$, so that $|e_-(\mathbf{k}_\parallel,l_\perp)|^2\cong 0$. Ignoring the two higher branches in Fig. 4(a) gives, with the factor of 3 included from the normalization of the eigenvectors,

$$\Delta_3(T)\cong \frac{1}{3N_\parallel}\sum_{\mathbf{k}_\parallel} n_B(\Omega(\mathbf{k}_\parallel 0)) \cong \frac{A_0 k_B T}{12\pi\tilde{D}} \ln\left[\frac{k_B T}{\omega_0}\right] . \tag{21}$$

Here A is the surface area of the film, and so $A_0 = A/N_\parallel$ is the area of the two-dimensional unit cell.

Now suppose we add additional layers to the film, where we have N layers with $N \gg 1$. The behavior of the spin-wave spectrum then depends on the nature of the anisotropy. We may consider a thick film of material similar to that used in the magnetic-bubble samples. Then, to excellent approximation, overlooking details of surface effects, we have spin-wave branches with dispersion relations

$$\Omega^{(n)}(\mathbf{k}_\parallel)=\omega_0+\frac{1}{2}D\left[k_\parallel^2+\left[\frac{n\pi}{2Nd}\right]^2\right] , \tag{22}$$

where various subbands are labeled by integers $n=0,1,2,\ldots$. Here d is the spacing between film layers. If the $\mathbf{k}_\parallel=\mathbf{0}$, $n=0$ frequency is calculated, assuming the anisotropy field to be much larger than the dipole field $4\pi M_0$ and uniformly distributed across the thickness of the film, then ω_0 is independent of N.

The situation for large N is sketched in Fig. 4(b). For large N we may have many "minibands" that lie below $k_B T$ in excitation energy. Then letting ω_0 be driven to zero by an external field applied normal to the easy axis produces a modest singularity in the spin deviation $\Delta_N(T)$.

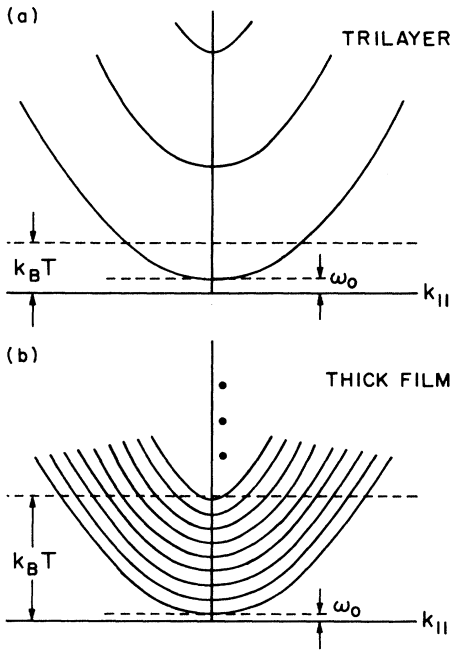

FIG. 4. Depicted are (a) the spin-wave normal modes of a trilayer, sketched at room temperature, and (b) the spin-wave "minibands" in a thick layer.

To see this, as a measure of the spin deviation, we simply use the number of thermally excited spin waves. Thus, with normalization factor $1/2N$ for standing cosine waves in a slab of width Nd,

$$\Delta_N(T)=\frac{1}{2NN_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{n=0}^{\infty} n_B(\Omega^{(n)}(\mathbf{k}_\parallel)) \ . \tag{23}$$

For large N we may replace the sum on n by an integral. We do this keeping the first correction provided by the Euler-MacLaurin formula:[14]

$$\sum_{n=n_m}^{n_M} f(n)=\int_{n_m}^{n_M} dn\, f(n)+\tfrac{1}{2}[f(n_m)+f(n_M)]+\cdots \ . \tag{24}$$

Hence, at the same time replacing the sum on $\mathbf{k}_\parallel$ by an integral,

$$\Delta_N(T)=\frac{A_0}{8\pi^2 N}\int d^2k_\parallel \int_0^\infty dn\, n_B(\Omega^{(n)}(\mathbf{k}_\parallel))+\frac{A_0}{2N}\int d^2k_\parallel n_B(\Omega^{(0)}(\mathbf{k}_\parallel))+\cdots \tag{25}$$

or

$$\Delta_N(T)=\frac{V_0}{2\pi^3}\int d^3k\, n_B\left[\omega_0+\frac{1}{2}Dk^2\right]+\frac{1}{N}\frac{\pi A_0 k_B T}{D}\ln\left[\frac{k_B T}{\omega_0}\right]+\cdots \ . \tag{26}$$

Here V_0 is the volume of the basic unit cell in the material.

The first term in Eq. (26) is well behaved and nonsingular in the limit $\omega_0 \to 0$. This is, in fact, the well-known formula for the mean spin deviation in a ferromagnet. As $\omega_0 \to 0$, we have the famous Bloch $T^{3/2}$ law.

The second term has a singularity identical in form to that in Eq. (21). However, it is proportional to $1/N$ and vanishes in the limit $N \to \infty$.

The singularity in the mean spin deviation at the critical field H_{ex}^c displayed in Fig. 2 is thus present only for the ultrathin films that are quasi-two-dimensional. A criterion for the crossover from quasi-two- to three-dimensional behavior is derived by requiring the separation between low-lying spin-wave branches to be small compared to $k_B T$. Thus we require $\tilde{D}(\pi/N_{a_0})^2 \ll k_B T$. This gives $N \gg N_c \equiv (\pi/\sqrt{2})\sqrt{\tilde{D}/A_0 k_B T}$ as the criterion for three-dimensional behavior. Numbers for Fe give $N_c \cong 2$ near room temperature, not far from unity, and so only ultrathin films exhibit the singularity.

It is also of interest to examine the nature of spin correlations in the near vicinity of the critical field, in the ultrathin film limit. Consider, for example, the behavior of a correlation function such as $\langle S_-(l)S_+(l')\rangle$, which describes the amplitude and spatial variation of spin fluctuations in the plane perpendicular to the magnetization of the film. If we decompose l into a piece $l_\parallel$ parallel to the film surfaces and a piece $l_\perp \hat{\mathbf{n}}_\perp$ normal to the film surfaces, then our formalism gives, in the spin-wave limit,

$$\langle S_-(l)S_+(l')\rangle = \frac{2S}{N_\parallel}\sum_{n=1}^{N}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot\mathbf{x}(l_\parallel - l'_\parallel)}\{e_+^{(n)}(\mathbf{k}_\parallel;l_\perp)^* e_+^{(n)}(\mathbf{k}_\parallel;l'_\perp) n_B(\Omega^{(n)}(\mathbf{k}_\parallel)) + e_-^{(n)}(\mathbf{k}_\parallel;l_\perp) e_-^{(n)}(\mathbf{k}_\parallel;l'_\perp)^*[1+n_B(\Omega^{(n)}(\mathbf{k}_\parallel))]\} . \tag{27}$$

We assume that very near the critical field and particularly at large lateral separations $l_\parallel = l'_\parallel$, where small values of $\mathbf{k}_\parallel$ are emphasized, the soft spin-wave branch $n=1$ provides the dominant contribution. In this limit,

$$n_B(\Omega^{(1)}(\mathbf{k}_\parallel)) \cong 1 + n_B(\Omega^{(1)}(\mathbf{k}_\parallel)) \cong \frac{k_B T}{\Omega^{(1)}(\mathbf{k}_\parallel)} .$$

For simplicity we confine our attention to the monolayer; the results will be very similar in form for any ultrathin film or which the low-lying acoustical spin-wave mode dominates the thermodynamics. For the monolayer we may suppress reference to $l_\perp$ and $l'_\perp$ and to the mode index n, such that

$$\langle S_-(l)S_+(l')\rangle \cong \frac{2Sk_B T}{N_\parallel}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot\mathbf{x}(l-l')}\frac{1}{\Omega(\mathbf{k}_\parallel)}[|e_+(\mathbf{k}_\parallel)|^2 + |e_-(\mathbf{k}_\parallel)|^2] . \tag{28}$$

For the monolayer and for spins canted at a general angle θ relative to the film normal, one has[6]

$$\Omega(\mathbf{k}_\parallel) = \{[H_{\rm loc} - A(\mathbf{k}_\parallel)]^2 - |B(\mathbf{k}_\parallel)|^2\}^{1/2} , \tag{29}$$

where we may define $\cosh^2[\phi(\mathbf{k}_\parallel)] = |e_+(\mathbf{k}_\parallel)|^2$ and $\sinh^2[\phi(\mathbf{k}_\parallel)] = |e_-(\mathbf{k}_\parallel)|^2$, with

$$\tanh[2\phi(\mathbf{k}_\parallel)] = \frac{|B(\mathbf{k}_\parallel)|}{H_{\rm loc} - A(\mathbf{k}_\parallel)} . \tag{30}$$

The definitions of $H_{\rm loc}$, $A(\mathbf{k}_\parallel)$, and $B(\mathbf{k}_\parallel)$ are given by Eqs. (1c)–(1f), (3d), and (5b). In these definitions we encounter certain dipole sums denoted earlier[5] as $d_{11}(\mathbf{k}_\parallel)$, $d_{22}(\mathbf{k}_\parallel)$, $d_{\perp\perp}(\mathbf{k}_\parallel)$, and $d_{12}(\mathbf{k}_\parallel)$. We replace these by their limiting forms as $\mathbf{k}_\parallel \to 0$, noting $d_{12}(0)=0$, $d_{11}(0)=d_{22}(0)=(4\pi/3)\eta$, and $d_{\perp\perp}(0)=(8\pi/3)\eta$, where

$$\eta = \frac{3}{4\pi}\frac{1}{\sqrt{2}}\sum_{l,m'}\frac{l^2}{(l^2+m^2)^{5/2}} = 0.762\ldots . \tag{31}$$

The sum in Eq. (31) is evaluated quickly and efficiently using methods discussed a number of years ago.[15]

In the interest of simplicity, we ignore h_{s2}, since it has little influence on the qualitative structure of the spin-correlation function. The critical field is then $H_{ex}^c = h_{s1} - 4\pi\eta M_0$, and for external fields $H_{ex} < H_{ex}^c$,

$$H_{\rm loc} - A(\mathbf{k}_\parallel) \cong H_{ex}^c(1 - \tfrac{1}{2}\sin^2\theta) + \tfrac{1}{2}Dk_\parallel^2 \tag{32}$$

and

$$B(\mathbf{k}_\parallel) \cong \tfrac{1}{2}H_{ex}^c \sin^2\theta . \tag{33}$$

After some algebra, we find for the monolayer

$$\langle S_-(l)S_+(l')\rangle \cong \frac{2Sk_B T}{N_\parallel}\sum_{\mathbf{k}_\parallel}\left[\frac{H_{ex}^c(1-\frac{1}{2}\sin^2\theta)+\frac{1}{2}Dk_\parallel^2}{(H_{ex}^c+\frac{1}{2}Dk_\parallel^2)(H_{ex}^c\cos^2\theta+\frac{1}{2}Dk_\parallel^2)}\right] e^{i\mathbf{k}_\parallel\cdot\mathbf{x}(1-l')} . \tag{34}$$

For θ near $\pi/2 (H_{ex} \cong H_{ex}^c)$, the combination

$$\frac{H_{ex}^c(1-\frac{1}{2}\sin^2\theta)+\frac{1}{2}Dk_\parallel^2}{H_{ex}^c+\frac{1}{2}Dk_\parallel^2}$$

approaches $\frac{1}{2}$ as $\mathbf{k}_\parallel \to 0$ and unity as $\mathbf{k}_\parallel \to \infty$. Thus the ratio of these two factors has rather little influence on the result. As $|\mathbf{x}(l-l')| \to \infty$, it is the small values of $\mathbf{k}_\parallel$ that matter most, and so we replace this factor by $\frac{1}{2}$. Therefore, we have

$$\langle S_-(l)S_+(l')\rangle \cong \frac{Sk_BT}{N_\parallel}\sum_{\mathbf{k}_\parallel}\frac{e^{i\mathbf{k}_\parallel\cdot\mathbf{x}(l-l')}}{H_{ex}^c\cos^2\theta+\frac{1}{2}Dk_\parallel^2} . \quad (35)$$

If A_0 is the area of the unit cell, then, evaluating the sum on $\mathbf{k}_\parallel$,

$$\langle S_-(l)S_+(l')\rangle \cong \frac{A_0Sk_BT}{\pi D}K_0\left[\frac{|\mathbf{x}(l-l')|}{\xi_<}\right], \quad H_{ex}<H_{ex}^c \quad (36)$$

where the correlation length $\xi_<$ is given by

$$\xi_< = \left[\frac{D}{2H_{ex}^c}\right]^{1/2}\frac{1}{\cos\theta} . \quad (37)$$

The canting angle θ is determined from $\sin\theta = H_{ex}/H_{ex}^c$, so that Eq. (37) may be rewritten in the form, where $H_{ex} \approx H_{ex}^c$ implies $H_{ex}^c/(H_{ex}^c+H_{ex}) \approx \frac{1}{2}$,

$$\xi_< = \frac{\sqrt{D}}{2}\frac{1}{(H_{ex}^c-H_{ex})^{1/2}}, \quad H_{ex}<H_{ex}^c . \quad (38a)$$

For fields just above the critical field, the spin-correlation function is still described by Eq. (36), except the correlation length $\xi_>$ is replaced by

$$\xi_> = \left[\frac{D}{2}\right]^{1/2}\frac{1}{(H_{ex}-H_{ex}^c)^{1/2}}, \quad H_{ex}>H_{ex}^c . \quad (38b)$$

Thus, as the critical angle of $\pi/2$ is approached, by varying the external field through the critical field H_{ex}^c from either above or below, the spin correlations within the film become very long ranged. While we have worked out their form only for the monolayer, as remarked earlier, the structure of the spin-correlation function will be the same as displayed in Eq. (36) for any ultrathin film for which the thermodynamics is dominated by the single soft mode. Right at H_{ex}^c, the correlation length diverges and, as we have seen, so does $\Delta_N(l_\perp, T)$. This suggests that true long-range order is absent right at the critical field.

We hope that these remarks and the results displayed earlier will stimulate further study of this intriguing field-induced phase transition. The authors of Ref. 2, in their elegant Brillouin study, have established the presence of the soft mode and also the fact that the Brillouin intensity becomes very large at fields near H_{ex}^c. These data, however, do not allow one to draw quantitative conclusions about the nature of the spin fluctuations near T_c.

ACKNOWLEDGMENT

This research was supported by the U.S. Army Research Office, through Contract No. CS0011028.

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[11]See theorem 3.1.12 on p. 114 of J. M. Ortega, *Matrix Theory: A Second Course* (Plenum, New York, 1987) for a formal statement and proof.

[12]See theorem 6.2.3 on p. 232 of Ref. 11 for a formal statement and proof.

[13]We assume all exchange couplings in the monolayer and trilayer have the same strength. This may not be the case in real samples, of course.

[14]See p. 806 of *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (Dover, New York, 1965).

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