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Theory of Macroscopic Excitations of Magnons*

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The excitation of ferromagnetic magnons by a macroscopic time-varying magnetic field is analyzed quantum mechanically. The quantization of the spin excitations is made by the method of Holstein and Primakoff, generalized for the case of a general nonuniform static field. Both linear and nonlinear excitation mechanisms are considered, with the driving field being either perpendicular or parallel to the static field. Particular attention is given to the coherence properties of the magnon states generated.

I. INTRODUCTION

It is well known that strongly magnetic systems have low-energy wavelike excitations called spin waves or magnons. The concept of spin waves was introduced by Bloch[1] in 1930 to explain the thermodynamic properties of ferromagnets at low temperatures. The interest in this field was renewed by the work of Suhl,[2] which explained the saturation effects observed in ferromagnetic resonance experiments as arising from the unstable growth of spin waves. Considerable progress in the subject was made after this work with the development of several methods for generating and detecting spin waves, measuring their properties, and studying their interactions with other elementary excitations.

Besides Suhl's nonlinear process, it was found by Morgenthaler[3] and Schlömann[4] that spin waves could be parametrically excited by an rf field applied parallel to the static field, instead of perpendicular. Schlömann[5] also first proposed a linear excitation mechanism, making use of an rf field perpendicular to a nonuniform static field. This excitation method proved to be very useful in low-magnetic-loss materials, such as yttrium iron garnet, providing a new possibility for delaying and processing information contained in electromagnetic signals. It has been used in several experimental investigations of properties of spin waves, such as their coupling to elastic waves in spatially[6,7] and time-varying[8] magnetic fields, their amplification by parametric pumping,[9] their interaction with light beams,[10] and others.

The macroscopic excitation of spin waves in ferromagnets has been discussed both semiclassically and quantum mechanically. The semiclassical treatments are mostly based on the magnetization equation-of-motion method introduced by Herring and Kittel.[11] In most of the previous quantum treatments one uses the equations of motion for the magnon creation and annihilation operators of the Holstein-Primakoff[12] formalism. The solutions of these equations are used to find the time dependence of the expectation values of the magnetization and other related operators. These results, however, do not contain all the information that can be obtained from the knowledge of the time-evolution operator.

Recently these authors[13,14] pointed out the importance of introducing coherent states[15] of a quantum oscillator in the description of spin-wave excitations. In the present paper we study the excitation of ferromagnetic magnons by a time-varying macroscopic magnetic field, which can be either parallel or perpendicular to the static field. In particular, we analyze the magnetization and the coherence properties of the system in each case from the corresponding evolution operator. The quantization of the spin excitations is made by the method of Holstein and Primakoff, generalized for the case of a nonuniform static field. In a first approximation of this method, magnons are considered as noninteracting bosons. This approximation is satisfactory for most of this work if we assume that the temperature of the ferromagnet is well below the Curie temperature.

In Sec. II we present the necessary background on the quantization of spin waves. In Sec. III the description of spin waves in terms of coherent states is studied. Section IV is devoted to the problem of linear excitation of spin waves by a spatial and time-varying macroscopic field perpendicular to the static field, assumed in general to be nonuniform. In Sec. V we consider the nonlinear perpendicular and parallel pumping mechanisms.

II. BACKGROUND: DIAGONALIZATION OF UNPERTURBED HAMILTONIAN

The analysis presented in this paper applies to a simple Heisenberg ferromagnet. The extension to ferri- and antiferromagnets should be straightforward. The Hamiltonian of the system under study is assumed to include Zeeman, exchange, and dipolar interactions[16,17]

$$\mathcal{H} = -2\mu_B \sum_i \vec{S}_i \cdot \vec{H}_i - \sum_{i\neq j} J_{ij}\vec{S}_i\cdot\vec{S}_j + \tfrac{1}{2}\sum_{i\neq j}(2\mu_B)^2\left(\frac{\vec{S}_i\cdot\vec{S}_j}{r_{ij}^3} - \frac{3(\vec{r}_{ij}\cdot\vec{S}_i)(\vec{r}_{ij}\cdot\vec{S}_j)}{r_{ij}^5}\right), \tag{2.1}$$

where μ_B is the Bohr magneton, $\vec{S}_i$ is the spin at the lattice site i (in units of $\hbar$), assumed to have a g factor of 2, J_{ij} is the exchange constant of the spins $\vec{S}_i$ and $\vec{S}_j$, and $\vec{r}_{ij}$ is their relative position vector. $\vec{H}_i$ is the applied field at site i and is the sum of a static component lying in the z direction and a small perturbating (excitation) component. The electron spin is taken parallel to its magnetic moment, as in most quantum treatments of spin waves. In our study we will neglect the effects of relaxation, magnetic anisotropy, magnet-elastic interaction, and boundary conditions.

The Hamiltonian (2.1) can be written as $\mathcal{H}(t)=\mathcal{H}_0+\mathcal{H}_1(t)$, where $\mathcal{H}_0$ comes from the static applied field and $\mathcal{H}_1(t)$ is due to the excitation field. In this section we investigate the eigenstates of the unperturbed term. These can be found by means of a diagonalization procedure, obtained by a series of transformations performed on the spin operators. These transformations were first introduced by Holstein and Primakoff[12] for the case where the static field is uniform. Here we discuss the more general case of nonuniform field. The first transformation is the same as that of Holstein and Primakoff[12]

$$S_i^+ = S_i^x + iS_i^y = (2S)^{1/2}(1 - a_i^\dagger a_i/2S)^{1/2} a_i\,, \tag{2.2}$$

$$S_i^- = S_i^x - iS_i^y = (2S)^{1/2}a_i^\dagger (1 - a_i^\dagger a_i/2S)^{1/2}\,, \tag{2.3}$$

$$S_i^z = S - a_i^\dagger a_i\,, \tag{2.4}$$

where $a_i^\dagger$ and a_i are creation and annihilation operators that satisfy the usual Bose commutation relations. Using (2.2)–(2.4) in (2.1) and neglecting products of three or more Bose operators, we obtain[17]

$$\mathcal{H}_0 = \sum_{i,j} R_{ij}a_i^\dagger a_j + \tfrac{1}{2}\sum_{i\neq j} S_{ij}a_i a_j + \tfrac{1}{2}\sum_{i\neq j} S_{ij}^* a_i^\dagger a_j^\dagger\,, \tag{2.5}$$

where

$$R_{ij} = \left[2\mu_B H(\vec{r}_i) + 2S\sum_{l\neq i} J_{il} - 4\mu_B^2 S\sum_{l\neq i}\frac{1}{r_{il}^3}\left(1-\frac{3z_{il}^2}{r_{il}^2}\right)\right]\delta_{ij} - 2SJ_{ij} - \frac{2\mu_B^2 S}{r_{ij}^3}\left(1-\frac{3z_{ij}^2}{r_{ij}^2}\right), \tag{2.6}$$

$$S_{ij} = -\frac{6\mu_B^2 S}{r_{ij}^3}\left(\frac{x_{ij}+iy_{ij}}{r_{ij}}\right)^2. \tag{2.7}$$

The Hamiltonian (2.5) cannot be diagonalized by the other Holstein-Primakoff transformations when H is a function of space. We use here the more general transformation of Bogoliubov and Tyablikov[18]

$$a_i = \sum_k [\psi_k(i)c_k + \xi_k^*(i)c_k^\dagger]\,, \tag{2.8}$$

where the functions $\psi_k(i)$ and $\xi_k(i)$ are the eigensolutions of the system of equations

$$\begin{aligned} E_k\psi_k(i) &= \textstyle\sum_j R_{ij}\psi_k(j) + \sum_j S_{ij}\xi_k(j)\,,\\ -E_k\xi_k(i) &= \textstyle\sum_j R_{ij}^*\xi_k(j) + \sum_j S_{ij}^*\psi_k(j)\,. \end{aligned} \tag{2.9}$$

The eigenfunctions form a complete set and satisfy the orthonormality conditions

$$\begin{aligned} &\textstyle\sum_i \psi_k(i)\psi_{k'}^*(i) - \sum_i \xi_k(i)\xi_{k'}^*(i) = \delta_{kk'}\,,\\ &\textstyle\sum_i \psi_k(i)\xi_{k'}(i) - \sum_i \psi_{k'}(i)\xi_k(i) = 0\,,\\ &\textstyle\sum_k \psi_k(i)\xi_k(j) - \sum_k \psi_k(j)\xi_k(i) = 0,\\ &\textstyle\sum_k \psi_k(i)\psi_k^*(j) - \sum_k \xi_k(j)\xi_k^*(i) = \delta_{ij}\,. \end{aligned} \tag{2.10}$$

With the transformation (2.8), the Hamiltonian takes the diagonal form

$$\mathcal{H}_0 = \sum_k E_k c_k^\dagger c_k\,. \tag{2.11}$$

The inverse transformation

$$c_k = \sum_i [\psi_k^*(i) a_i - \xi_k^*(i) a_i^\dagger] \tag{2.12}$$

defines operators $c_k^\dagger$ and c_k which satisfy the Bose commutation relations

$$[c_k, c_{k'}] = 0, \quad [c_k, c_{k'}^\dagger] = \delta_{kk'} . \tag{2.13}$$

The operator $c_k^\dagger$ creates a magnon of eigenvalue E_k, and c_k annihilates a magnon of eigenvalue E_k. The eigenfunctions $\psi_k(i)$ and $\xi_k(i)$ which characterize the spatial variation of the magnon excitation are given by (2.9). In the particular case of an uniform applied field, the eigenfunctions reduce to the usual plane-wave form

$$\psi_k(i) = N^{-1/2} u_k e^{i\vec{k}\cdot\vec{r}_i} , \quad \xi_k(i) = -N^{-1/2} v_k^* e^{i\vec{k}\cdot\vec{r}_i} , \tag{2.14}$$

where N is the total number of spins and $\vec{k}$ denotes the wave vector of the excitation. Here the index k is identified with a wave vector, although the vector sign is kept out for clarity of the notation. The parameters in (2.14) are

$$u_k = \cosh\mu_k , \quad v_k = e^{i2\phi_k} \sinh\mu_k , \tag{2.15}$$

where

$$\tanh 2\mu_k = |B_k| / A_k \tag{2.16}$$

and, for a simple cubic lattice, with exchange interaction only between nearest neighbors, in the long-wavelength limit, A_k and B_k are given by[16]

$$A_k = Dk^2 + 2\mu_B H + \mu_B 4\pi M \sin^2\theta_k , \qquad B_k = \mu_B 4\pi M \sin^2\theta_k e^{-i2\phi_k} , \tag{2.17}$$

where $D = 2SJa^2$, a is the lattice parameter, M is the saturation magnetization, and θ_k and ϕ_k are the polar and azymuthal angles of the wave vector. In this case the magnon energy also reduces to a simple form

$$E_k = \hbar\omega_k = (A_k^2 - |B_k|^2)^{1/2} . \tag{2.18}$$

A continuous magnetization operator $\vec{M}(\vec{r})$ can be introduced through the relation $\vec{M}(\vec{r}) = 2\mu_B \sum_i S_i / \delta V$, where the summation runs over the sites inside a small volume δV, around the point $\vec{r}$, which contains many sites. Using (2.2) and (2.3), we can obtain the components of $\vec{M}$ transverse to the static field. In the Heisenberg picture, to first order in the magnon variables, we have

$$\begin{aligned} \vec{m}(\vec{r}, t) &= \vec{m}^{(+)}(\vec{r}, t) + \vec{m}^{(-)}(\vec{r}, t), \\ m_x^{(+)}(\vec{r}, t) &= [m_x^{(-)}(\vec{r}, t)]^\dagger \\ &= (\mu_B MN/V)^{1/2} \sum_k e^{-i\omega_k t} [\psi_k(\vec{r}) + \xi_k(\vec{r})] c_k , \\ m_y^{(+)}(\vec{r}, t) &= [m_y^{(-)}(\vec{r}, t)]^\dagger \\ &= -i(\mu_B MN/V)^{1/2} \sum_k e^{-i\omega_k t} [\psi_k(\vec{r}) - \xi_k(\vec{r})] c_k , \end{aligned} \tag{2.19}$$

where V is the volume of the crystal. The longitudinal component of $\vec{M}$ is $M_z = M - m_z$, where $m_z \simeq (m_x^2 + m_y^2)/2M$. In the continuum approximation, the unperturbed part of the Hamiltonian can also be written as[19]

$$\mathcal{H}_0 = \int dr^3 : \left(H m_z + \frac{D}{4\mu_B M} \frac{\partial m_i}{\partial x_j} \frac{\partial m_i}{\partial x_j} + \frac{|\vec{h}|^2}{8\pi} \right) : = \sum_k \hbar\omega_k c_k^\dagger c_k , \tag{2.20}$$

where $\vec{h}$ is the volume dipolar field operator. We have used the normal product, denoted by $:\cdots:$, defined taking all creation operators to the left of all annhilation operators in order to eliminate infinite zero-point energies. The repeated indices in (2.20) indicate summation.

Another operator of interest in the continuum approach of spin waves is the linear-momentum density. We construct this operator by analogy to the magnon momentum suggested by Morgenthaler.[20] Its component g_i becomes

$$g_i = \frac{\hbar}{4\mu_B M} \left(\vec{m} \times \frac{\partial \vec{m}}{\partial x_i} \right) \cdot \hat{z} . \tag{2.21}$$

In the case of an uniform static field, using (2.13) and (2.21), one can show that total momentum is given by the expected form

$$\vec{P} = \int d^3 r \vec{g}(\vec{r}) = \sum_k \hbar \vec{k}\, c_k^\dagger c_k , \tag{2.22}$$

provided that terms with three or more magnon operators are discarded.

III. COHERENT MAGNON STATES

The unperturbed Hamiltonian $\mathcal{H}_0$ of the Heisenberg ferromagnet, in its quadratic approximation (2.11), characterizes a system of noninteracting magnons. Its eigenstates can be written in the N representation as

$$|n_k\rangle = [(c_k^\dagger)^{n_k} / (n_k!)^{1/2}] |0\rangle , \tag{3.1}$$

where the vacuum state is defined by the condition $c_k |0\rangle = 0$. These stationary states describe systems with a well-defined number of magnons and uncertain phase. They have been used in nearly all quantum treatments of thermodynamic properties, relaxation mechanisms, and magnon interaction processes in ferromagnets. On the other hand, they do not correspond to the Herring-Kittel spin waves used in the semiclassical treatments.[13] This is clear from the fact that the first-order components of the transverse magnetization (2.19) have zero expectation values in the stationary states. In addition, a system that behaves nearly classically should involve a large and uncertain number of magnons, with well-defined phases. The present authors have indicated[13,14] that in order to establish a correspondence between classical and quantum spin waves one should use the concept of coherent magnon states. These are defined by analogy to the coherent photon states introduced by Glauber.[15]

A coherent magnon state is defined as the simultaneous eigenket of the positive frequency parts of $m_x(\vec{r}, t)$ and $m_y(\vec{r}, t)$,

$$m_i^{(+)}(\vec{r}, t)\,|\,\rangle = m_i(\vec{r}, t)\,|\,\rangle\ , \tag{3.2}$$

where the eigenvalues $m_i(\vec{r}, t)$ are complex numbers, which obey the semiclassical equations of motion for the magnetization. The dual of $|\,\rangle$ is also an eigenbra of the negative frequency parts of $m_i(\vec{r}, t)$. The vector $|\,\rangle$ can be written as a direct product of single-mode coherent magnon states, defined by

$$c_k\,|\,\alpha_k\rangle = \alpha_k\,|\,\alpha_k\rangle\ , \tag{3.3}$$

where the complex eigenvalue α_k is directly related to the eigenvalue $m_i(\vec{r}, t)$. The states defined in (3.3) can be expanded in terms of the eigenstates of the Hamiltonian[15]:

$$|\,\alpha_k\rangle = e^{-1/2|\alpha_k|^2} \sum_{n_k} \frac{(\alpha_k)^{n_k}}{(n_k!)^{1/2}}\,|\,n_k\rangle\ . \tag{3.4}$$

The properties of the states $|\,\alpha_k\rangle$ have been extensively studied by Glauber.[15] The probability of finding n_k magnons in the coherent state $|\,\alpha_k\rangle$ is given by a Poisson distribution

$$|\langle n_k\,|\,\alpha_k\rangle|^2 = (|\,\alpha_k\,|^{2n_k}/n_k!)\,e^{-|\alpha_k|^2}\ . \tag{3.5}$$

The mean value $|\,\alpha_k\,|^2$ is the expectation value of the occupation number $n_k = c_k^\dagger c_k$ in the coherent state.

It can be shown that coherent states are not orthogonal to one another, but they form a complete set. This property allows the expansion of an arbitrary state in terms of coherent states.[15] Another convenient property is that a coherent state can be generated by the application of a displacement operator to the vacuum,

$$|\,\alpha_k\rangle = D(\alpha_k)\,|\,0\rangle\ , \tag{3.6}$$

where[15]

$$D(\alpha_k) = \exp(\alpha_k c_k^\dagger - \alpha_k^* c_k)\ . \tag{3.7}$$

The coherent states are not eigenstates of the unperturbed Hamiltonian. On the other hand, as opposed to the stationary states, they have nonzero expectation values for the magnetization. With (2.19) and (3.3) we have for a state $|\,\alpha_k\rangle$

$$\langle m_x(\vec{r}, t)\rangle = (\mu_B MN/V)^{1/2}\left[(\psi_k + \xi_k)\alpha_k e^{-i\omega_k t} + \text{c. c.}\right],$$
$$\langle m_y(\vec{r}, t)\rangle = -(\mu_B MN/V)^{1/2}\left[i(\psi_k - \xi_k)\alpha_k e^{-i\omega_k t} + \text{c. c.}\right]. \tag{3.8}$$

As $\psi_k(\vec{r})$ and $\xi_k(\vec{r})$ are simply functions of the position, (3.8) represents at each point a magnetization vector precessing elliptically around the static magnetic field, which agrees with the macroscopic picture of a spin wave. In the particular case of an uniform static field, Eq. (2.14) shows that the phase of the precession of the magnetization varies harmonically in space, so that one can write for a single-mode coherent state $|\,\alpha_k\rangle$

$$\langle \vec{m}(\vec{r}, t)\rangle = \hat{x}\, a_k \cos(\vec{k}\cdot\vec{r} - \omega_k t + \beta_k) - \hat{y}\, b_k \sin(\vec{k}\cdot\vec{r} - \omega_k t + \beta_k), \tag{3.9}$$

where

$$a_k = (4\mu_B M/V)^{1/2}(u_k - v_k)\,|\,\alpha_k\,|\ ,$$
$$b_k = (4\mu_B M/V)^{1/2}(u_k + v_k)\,|\,\alpha_k\,|\ , \tag{3.10}$$

and β_k is the argument of α_k. In order to write the magnetization in the form (3.9) we assume, without loss of generality, that the mode considered has azymuthal angle $\phi_k = 0$. The ellipticity of the spin wave is

$$e_k = \frac{a_k}{b_k} = \left(\frac{A_k - |\,B_k\,|}{A_k + |\,B_k\,|}\right)^{1/2}\ , \tag{3.11}$$

like the semiclassical result.[16,17] From (3.10) one can show that the mean number of magnons in a coherent state is related to the product of the major and minor axes of the ellipse by

$$\langle n_k\rangle = |\,\alpha_k\,|^2 = a_k b_k\, V/4\mu M\ , \tag{3.12}$$

which is also in agreement with the semiclassical value.[20] These considerations about the expectation values of the magnetization do not give a complete view of the relation between a classical system and a system in a coherent state. In fact, there are an infinite number of linear superpositions of the eigenstates of the Hamiltonian which satisfy (3.9) and (3.11). To understand better the meaning of the coherent states, let us calculate the variances of the components of the magnetization

$$[\Delta m_x(\vec{r}, t)]^2 = \langle m_x^2\rangle - \langle m_x\rangle^2\ ,$$
$$[\Delta m_y(\vec{r}, t)]^2 = \langle m_y^2\rangle - \langle m_y\rangle^2\ . \tag{3.13}$$

For a single-mode coherent state in an uniform field, we find

$$[\Delta m_x(\vec{r}, t)]^2 = (\mu_B M/V)(u_k - v_k)^2\ ,$$
$$[\Delta m_y(\vec{r}, t)]^2 = (\mu_B M/V)(u_k + v_k)^2\ , \tag{3.14}$$

and, for the product of the two normalized uncertainties, we find

$$(\Delta m_x/a_k)(\Delta m_y/b_k) = \mu_B M/(a_k b_k V) = (4\langle n_k\rangle)^{-1}\ . \tag{3.15}$$

This result shows that a coherent state propagates with no spread in the variances of the components of the magnetization. Furthermore, the relative variances go to zero as the number of magnons approaches infinite, as required in a classical system.

To conclude this section let us investigate the properties of coherent states related to energy and momentum. Due to the definition (3. 3), the expectation value of an operator written as products of magnon operators can readily be found for coherent states. This is the case of the Hamiltonian (2. 20) and the linear momentum (2. 21), which have in a general coherent state $|\,\rangle = \Pi_k |\,\alpha_k\rangle$,

$$\langle \mathcal{H}_0\rangle = \int d^3r \left(H\langle m_z\rangle + \frac{D}{4\mu_B M}\,\frac{\partial\langle m_i\rangle}{\partial x_j}\,\frac{\partial\langle m_i\rangle}{\partial x_j} + \frac{1}{8\pi}\,|\langle \vec{h}\rangle|^2 \right), \quad (3.16)$$

$$\langle P_i\rangle = \int d^3r\, \frac{\hbar}{4\mu_B M}\left(\langle \vec{m}\rangle \times \frac{\partial\langle\vec{m}\rangle}{\partial x_i}\right)\cdot \hat{z}\,. \quad (3.17)$$

Also

$$\langle \mathcal{H}_0\rangle = \textstyle\sum_k \hbar\omega_k\, |\,\alpha_k|^2\,, \quad (3.18)$$

and, in the case of an uniform static field,

$$\langle \vec{P}\rangle = \textstyle\sum_k \hbar\vec{k}\,|\,\alpha_k|^2\,. \quad (3.19)$$

Equations (3. 16) and (3. 17) show that the average values of the energy and linear momentum of a coherent state can be factorized as products of functions of the mean values of the magnetization components, in the same way as the classical quantities. One can prove, with arguments analogous to those used by Kroll[21] in the photon case, that the state which satisfies (3. 8) and (3. 18) is uniquely determined, and therefore it is a coherent state. One can also show that for a coherent state

$$\frac{(\Delta\mathcal{H})^2}{\langle\mathcal{H}\rangle^2} = \frac{\sum\hbar^2\omega_k^2\langle n_k\rangle}{(\sum\hbar\omega_k\langle n_k\rangle)^2}\,, \quad \frac{(\Delta P_i)^2}{\langle P_i\rangle^2} = \frac{\sum\hbar^2 k_i^2\langle n_k\rangle}{(\sum\hbar k_i\langle n_k\rangle)^2}\,, \quad (3.20)$$

where the second relation is valid only for an uniform field. These equations show that in a coherent state the relative variances of the energy and the momentum also vanish as the mean number of magnons increases.

IV. LINEAR EXCITATION OF MAGNONS

Spin waves can be macroscopically generated in a ferromagnetic crystal by means of a microwave magnetic field conveniently oriented. The application of the rf field transverse to the static field provides a linear coupling between the electromagnetic field and the spin waves. In actual experiments this coupling is increased by the use of nonuniform static fields, which normally exist in nonellipsoidal samples, according to a mechanism first suggested by Schlömann.[5] This method of generating spin waves has been studied semiclassically by Schlömann[5,22] and Lüthi.[23] In this section we present a quantum formulation of the theory of this excitation mechanism. We assume that the role played by magnetostatic modes in the coupling mechanism is negligible.

Let us consider a ferromagnetic medium magnetized in the z direction by a static nonuniform field $H(\vec{r})$. At $t=0$ a transverse driving field $\hat{x}h(\vec{r},\,t)$ is turned on. The Hamiltonian of the system can be written as $\mathcal{H}(t)=\mathcal{H}_0+\mathcal{H}_1(t)$, where the static term is given by (2. 11). The perturbing term, which arises from the Zeeman contribution, is given by

$$\mathcal{H}_1(t) = -\mu_B \textstyle\sum_i h(\vec{r}_i,\,t)(S_i^+ + S_i^-)\,. \quad (4.1)$$

Considering only the first-order terms in the expansions (2. 2) and (2. 3) and using the transformation (2. 8), the Hamiltonian $\mathcal{H}_1(t)$ becomes

$$\mathcal{H}_1(t) = \theta(t)\textstyle\sum_k [\, g_k(t)c_k + \text{H. c.}\,]\,, \quad (4.2)$$

where $\theta(t)$ is the Heaviside function and

$$g_k(t) = -\mu_B(2S)^{1/2}\textstyle\sum_i [\,\psi_k(i)+\xi_k(i)\,]\,h(\vec{r}_i,\,t)\,. \quad (4.3)$$

In the Schrödinger picture the state at an instant t is related to its value at $t=0$ through the time-evolution operator

$$|\,t\rangle = U(t,\,0)\,|\,t=0\rangle\,. \quad (4.4)$$

To find $U(t,\,0)$ it is convenient to use the interaction picture.[24] Let $U=U^0U^I$, where $U^0(t,\,0) = e^{-i\mathcal{H}_0 t/\hbar}$. The intermediate evolution operator is governed by

$$i\hbar\,\frac{d}{dt}\,U^I = \mathcal{H}^I U^I\,, \quad (4.5)$$

where

$$\mathcal{H}^I(t) = \theta(t)\textstyle\sum_k [\, g_k(t)\,e^{-i\omega_k t}c_k + \text{H. c.}\,]\,. \quad (4.6)$$

As the commutator of $\mathcal{H}^I$ with itself at different times is a c-number function, an explicit solution for (4. 5) can be found immediately,[25] leading to

$$U(t,\,0) = \exp[-i\mathcal{H}_0 t/\hbar + i\beta(t)\,]\exp[\textstyle\sum_k(\gamma_k c_k^\dagger - \gamma_k^* c_k)\,]\,, \quad (4.7)$$

where $\beta(t)$ is an unimportant phase and $\gamma_k(t)$ is a c-number function given by

$$\gamma_k(t) = (i\hbar)^{-1}\int_0^t g_k^*(t')\,e^{i\omega_k t'}\,dt'\,. \quad (4.8)$$

The last exponential operator in (4. 7) is the product of displacement operators for coherent states defined by (3. 6) and (3. 7). Therefore, if prior to $t=0$ the state of the system is the vacuum, the transverse driving field excites coherent magnon states given by

$$|\,t\rangle = U\,|\,0\rangle = \exp[-i\mathcal{H}_0 t/\hbar + i\beta(t)\,]\textstyle\prod_k |\,\gamma_k(t)\rangle = e^{i\beta(t)}\prod_k |\,\gamma_k(t)e^{-i\omega_k t}\rangle\,. \quad (4.9)$$

This result is valid under general conditions. The spatial variation of the static field is taken into account in the generalized magnon operators. The external field creates coherent magnon states even with no preexisting spin fluctuations. The expectation values of the transverse components of the magnetization are

$$\langle m_x\rangle = (\mu_B MN/V)^{1/2} \textstyle\sum_k \{ \gamma_k(t) [\psi_k(\vec{r}) + \xi_k(\vec{r})] e^{-i\omega_k t} + \text{c. c.} \},$$

$$\langle m_y\rangle = -i(\mu_B MN/V)^{1/2} \textstyle\sum_k \{ \gamma_k(t) [\psi_k(\vec{r}) - \xi_k(\vec{r})] e^{-i\omega_k t} - \text{c. c.} \}, \tag{4.10}$$

which describe macroscopic spin waves propagating in space according to the variation of ψ_k and ξ_k.

If prior to $t=0$ the system is in thermal equilibrium at a finite temperature, i.e., it can be described by a chaotic mixture of eigenstates of $\mathcal{H}_0$, the application of $h(\vec{r}, t)$ will result in the excitation of a state with the expectation value of the magnetization still given by (4.10). The value of $\langle m_x^2\rangle$ ($\langle m_y^2\rangle$) will be given by the sum of two separate contributions: One is its initial value, and the other is the value of $\langle m_x\rangle^2$ ($\langle m_y\rangle^2$) obtained squaring (4.10).

In the case that at $t=0$ the system is in a general coherent state, the excitation mechanism does not destroy the coherence properties of the system.[26] The resulting state is a new coherent state in which the eigenvalues of c_k are displaced by $\gamma_k(t)\, e^{-i\omega_k t}$.

In order to understand the physical meaning of the eigenvalues $\gamma_k(t)$ of the coherent states generated by the transverse field, let us consider two particular cases. In the first we assume that the static field is uniform. Using the plane-wave forms (2.14) and replacing the summation over the lattice sites in (4.8) by an integral, we obtain

$$\gamma_k(t) = (i\hbar)^{-1}(\mu_B M/V)^{1/2}(v_k - u_k) \times \int d^3r \int_0^t dt'\, e^{i(\omega_k t' - \vec{k}\cdot\vec{r})}\, h(\vec{r}, t'). \tag{4.11}$$

The eigenvalue of the generated $\vec{k}$-mode magnon state, as well as the transverse magnetization, is proportional to the corresponding Fourier transform of the driving field. This means that in order to excite a spin wave with given frequency one needs an rf field with the same frequency and a spatial variation in some region comparable to the variation of the mode. This is the main result of the work of Lüthi.[23] Note that Eq. (4.10) with the value of γ_k given by (4.11) is essentially his Eq. (2).

The second case of interest is that of the excitation of spin waves in a nonuniform static field by a transverse driving field which is essentially uniform. This is the case considered by Schlömann. For simplicity we neglect the dipolar terms. In this approximation the coefficients S_{ij} in (2.7) vanish and R_{ij} in (2.6) becomes

$$R_{ij} = [2\mu_B H(\vec{r}_i) + 2S\textstyle\sum_l J_{il}]\,\delta_{ij} - 2SJ_{ij}\ . \tag{4.12}$$

In the long-wavelength limit, with the nearest-neighbor exchange approximation, the eigenvalue system (2.9) becomes

$$[2\mu_B H(\vec{r}_i) - D\nabla^2]\psi_k(i) = \hbar\omega_k\, \psi_k(i)\ . \tag{4.13}$$

From (4.10) we see that this equation governs the dependence of the magnetization in space. The similarity between (4.13) and the Schrödinger equation is the source of a well-known and fruitful analogy between the behavior of magnons in a nonuniform field and mass particles in a space-dependent potential. Using this analogy we can readily understand the result (4.9) for this case. Assuming that the driving field is harmonic, $h(\vec{r}, t) = h(\vec{r})\sin\omega t$, the expectation value of the magnetization operator $m^+ = (2\mu S^+/V)$, in the state $|\,t\rangle$, is in this case

$$m^+(\vec{r}, t) \simeq i\left(\frac{\mu_B MN}{\hbar V}\right)\sum_k e^{-i\omega_k t}\ \frac{e^{i(\omega_k-\omega)t}-1}{\omega_k-\omega}\ \psi_k(\vec{r})\int d^3r'\,\psi_k^*(\vec{r}\,')h(\vec{r}\,')\ , \tag{4.14}$$

where we have neglected the nonresonant terms and replaced the sum over the lattice sites by the integral. This equation shows that after a few periods of oscillation only the modes with eigenvalues $\omega_k \simeq \omega$ will be excited. From the Schrödinger-like equation (4.13) one can see that at points where $H \simeq \hbar\omega/2\mu_B$, the magnetization of these modes is a slowly varying function of space. At larger values of the static field the magnetization is a rapidly decaying function of space, whereas at smaller fields it is an oscillating function. One can conclude that if the driving field is essentially uniform, almost all the contribution to the integral in (4.14) arises from the region where $H(\vec{r}) \simeq \hbar\omega/2\mu_B$. Therefore, in order to excite spin waves with frequency ω, it is necessary that the driving field extends through the region where $H(r) \simeq \hbar\omega/2\mu_B$.

V. NONLINEAR GENERATION OF MAGNONS

Spin waves can be nonlinearly excited in strongly magnetic systems by means of a microwave magnetic field applied either perpendicular or parallel to the static field. The excitation is due to the oscillation of the coupling parameter between two or more magnon modes, and for this reason the process is called parametric. As in other nonlinear processes, the excitation is very intense when the driving field exceeds a threshold value. In the perpendicular pumping case the coupling between the magnon modes is made through the uniform precession mode, which is excited by the external field.[2] In the parallel pumping case there is a direct coupling between magnon pairs and the parallel field.[3,4] The previous theoretical discussions of these processes have been concerned with the macroscopic aspects of the excitations, such as the threshold fields and the susceptibilities.[2,3,4,27,28] Here we discuss the statistical properties of the state generated under parametric excitation.

Let us consider, for simplicity, that both the static and the pumping fields are uniform, and that the latter has a harmonic time dependence with frequency ω. As in Sec. IV, the total Hamiltonian of the system can be written as $\mathcal{H}(t)=\mathcal{H}_0+\mathcal{H}_1(t)$, where $\mathcal{H}_0$ is the same unperturbed term. The time-dependent term for both transverse and parallel processes are approximately given by[27]

$$\mathcal{H}_1(t)=\theta(t)\sum_k \tfrac{1}{2}\hbar\,(\rho_k e^{i\omega t}\,c_k^\dagger c_{-k}^\dagger+\text{H. c.}). \qquad (5.1)$$

For the parallel pumping process Eq. (5.1) is derived directly from the Zeeman contribution. The approximations, in this case, consist of neglecting higher-order terms in the magnon operators and a time-varying modulating term. In this case[27,28]

$$\rho_k=-\frac{2\mu_B}{\hbar}\,u_k v_k h=-\left(\frac{2\mu_B}{\hbar}\right)^2\frac{\pi M h}{\omega_k}\sin^2\theta_k\,e^{2i\phi_k}, \qquad (5.2)$$

where h is the amplitude of the pumping field. In the perpendicular process the term (5.1) arises from higher-order terms previously neglected in (2.2) and (2.3). The lowest-order process involves one uniform precession magnon ($k=0$) and a pair of $k\neq 0$ magnons, and can be represented by (5.1), with[2,27]

$$\rho_k=\left(\frac{\mu_B^3 M}{V}\right)^{1/2}\frac{4\pi}{\hbar^2\omega_k}\,(2\mu_B H+Dk^2+\hbar\omega_k)\times\sin 2\theta_k\,e^{-i\phi_k}\,c_0, \qquad (5.3)$$

where c_0 is the field amplitude of the uniform mode, assumed to be determined only by the external field[27]

$$c_0=h\,(\mu_B M V)^{1/2}\,[(\omega-\omega_0)^2+\eta_0^2)\,\hbar^2]^{-1/2}, \qquad (5.4)$$

where ω_0 and η_0 are the uniform-mode resonance and relaxation frequencies. In this case, $k=0$ is excluded in the summation of (5.1).

The perturbation Hamiltonian (5.1) has the same form as that generally used to describe parametric amplifier systems. These systems have been discussed by several authors, both with classical[29] and quantum[30] theories. The quantum theory developed by Mollow and Glauber[30] is based extensively on the P representation of the density operator, which provides a description of the fields closely resembling their classical description. We shall discuss here the properties of this nonlinear excitation process in the general nonresonant case, based on the time-evolution operator of the system.

In order to solve the equation of motion for the evolution operator, let us introduce an intermediate representation defined by $U(t,0)=A(t)U^I(t,0)$, where

$$A(t)=\exp(-i\tfrac{1}{2}\omega\textstyle\sum_k c_k^\dagger c_k\,t). \qquad (5.5)$$

From the equation for $U(t,0)$ one can show that

$$i\hbar\,\frac{dU^I}{dt}=(\mathcal{H}^I-\hbar\textstyle\sum_k\tfrac{1}{2}\omega\,c_k^\dagger c_k)U^I, \qquad (5.6)$$

where $\mathcal{H}^I=A^\dagger\mathcal{H}(t)A$ is given by

$$\mathcal{H}^I=\textstyle\sum_k[\hbar\omega_k\,c_k^\dagger c_k+\tfrac{1}{2}\hbar\,(\rho_k c_k^\dagger c_{-k}^\dagger+\rho_k^* c_k c_{-k})]. \qquad (5.7)$$

As $\mathcal{H}^I$ does not depend on t, (5.6) can be readily solved to give

$$U(t,0)=\exp(-i\tfrac{1}{2}\omega\textstyle\sum_k c_k^\dagger c_k\,t)\times\exp[-i\sum_k(\omega_k-\tfrac{1}{2}\omega)c_k^\dagger c_k\,t-\tfrac{1}{2}i\sum_k(\rho_k c_k^\dagger c_{-k}^\dagger+\rho_k^* c_k c_{-k})t]. \qquad (5.8)$$

This evolution operator can be used to calculate the state of the system at any instant t, for an arbitrary initial state. In the particular case of an initial vacuum state, (5.8) leads to the simple form

$$|\,t\rangle=Z\exp\Big(\sum_{k>0}-i\lambda_k\,c_k^\dagger c_{-k}^\dagger\Big)|\,0\rangle, \qquad (5.9)$$

where

$$Z=\prod_{k>0}(1-|\lambda_k|^2)^{1/2},$$
$$\lambda_k=\rho_k e^{-i\omega t}\sinh\kappa_k t\,[\kappa_k\cosh\kappa_k t+i(\omega_k-\tfrac{1}{2}\omega)\sinh\kappa_k t]^{-1},$$
$$\kappa_k^2=|\rho_k|^2-(\omega_k-\tfrac{1}{2}\omega)^2. \qquad (5.10)$$

This equation reveals that magnon pairs are emitted in all directions. However, as can be seen from (5.2) and (5.3), the most favored directions are $\theta=\tfrac{1}{2}\pi$ in the parallel pumping case and $\theta=\tfrac{1}{4}\pi$ in the perpendicular case. As it is well known, some pair modes are very strongly excited when the field intensity exceeds the threshold value for the given modes. In this nonlinear process, relaxation plays a very important role, and it can be taken into account by assigning an imaginary part to the magnon frequency ω_k. When this is done in (5.10), the threshold for instability is obtained equating the real part of κ_k to zero. This leads to results which agree with the known values.[2,3,4,27]

To find the properties of the system of nonlinearly excited magnons, we begin by considering the behavior of the expectation values of the magnon operators in the state $|\,t\rangle$. This is most easily done in the Heisenberg picture. From (5.8) we have

$$c_k^\dagger(t) = U^\dagger c_k^\dagger U = a_k(t) c_k^\dagger + b_k(t) c_{-k}\,, \tag{5.11}$$

where[31]

$$a_k(t) = e^{i(\omega/2)t}[\cosh\kappa_k t + i(\omega_k - \tfrac{1}{2}\omega)(\sinh\kappa_k t)/\kappa_k]\,,$$
$$b_k(t) = -ie^{i(\omega/2)t}\rho_k^*(\sinh\kappa_k t)/\kappa_k\,. \tag{5.12}$$

Equation (5.11) shows clearly that in the state (5.9) the expectation values of $c_k^\dagger(t)$ and $c_k(t)$, and consequently of $m_x(t)$ and $m_y(t)$, vanish at all times. The expectation value of n_k is

$$\langle n_k\rangle = \langle 0 \mid c_k^\dagger(t)\, c_k(t) \mid 0\rangle = (\,|\,\rho_k\,|^2/\,|\,\kappa_k\,|^2)\sinh^2\kappa_k t\,. \tag{5.13}$$

It is clear that the magnon occupation number grows exponentially with time when the driving field is above the threshold. The variances of m_x and m_y behave in the same way. This result reveals that the state (5.9) is not coherent, as opposed to the state generated by linear excitation. Mollow and Glauber[30] have shown that even when the initial state is coherent, an excitation of the type we are considering quickly destroys their properties of coherence. It is important to note that this generation process does not require the existence of magnons in the system before the commencement of the excitation.

The physical meaning of the state (5.9) can be clarified further by some of its statistical properties. We shall devote the rest of this section to a discussion of the behavior of the time-dependent density operator of the system. Soon after the generation of pairs, the correlation between the magnons in a pair can be neglected. Therefore we can base our study on the reduced-density operator for a mode k. Using the expansion of (5.9) in terms of the states $|\, n_k\rangle$ and the orthonormality between these states, we obtain for the reduced-density operator

$$\rho_k = \mathrm{tr}_{k'} \mid t\rangle\langle t \mid = \textstyle\sum_{m_k}(1 - |\,\lambda_k\,|^2)\,|\,\lambda_k\,|^{2m_k} \mid m_k\rangle\langle m_k \mid\,. \tag{5.14}$$

With (5.13), this can be rewritten as

$$\rho_k = \frac{1}{1+\langle n_k\rangle}\sum_{m_k}\left(\frac{\langle n_k\rangle}{1+\langle n_k\rangle}\right)^{m_k} \mid m_k\rangle\langle m_k \mid\,. \tag{5.15}$$

The distribution of the system in the number of magnons is of the same type as the Planck distribution for a system in thermal equilibrium. This behavior is characteristic of systems with identical oscillators which are statistically independent of one another. This result means that for each $\vec{k}$ mode, if we neglect the correlation with the $-\vec{k}$ mode, the magnon distribution of the system under nonlinear excitation is similar to the distribution of the system in thermal equilibrium. One difference to be noted is that in the former case the number of magnons can be several orders of magnitude higher than the latter. If we consider the system as a whole, the distributions are not the same in the two cases because in the situation of thermal equilibrium there is no preferential direction of excitation, whereas in the nonlinear excitation this occurs markedly.

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Nonlinear Electrical Conductivity of Superconducting Films below the Transition Temperature

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The Langevin equation for the superconducting order parameter above the transition temperature T_c proposed by Schmid is modified to calculate the nonlinear excess conductivity $\sigma'(T,E)$ slightly below T_c. The electric field dependence of $\sigma'(T,E)$ is described approximately by the same function of $E/E_c(T)$ as Schmid's function above T_c, with a newly defined characteristic field $E_c(T)$ below T_c. The experimental results of the nonlinear electrical conductivity of aluminum films below T_c are in fairly good agreement with the theory.

I. INTRODUCTION

The excess conductivity due to the thermodynamic fluctuations of the superconducting order parameter has been extensively investigated in the temperature region above and below the transition temperature T_c. Smith *et al.*[1] found that the excess current in thin films above T_c shows a nonlinear dependence on the electric field when the velocity v_s of the fluctuation Cooper pairs exceeds $\hbar/m\xi(T)$, where $\xi(T)$ is the temperature-dependent Ginzburg-Landau (GL) coherence length. Since then, the nonlinearity has been studied above T_c: theoretically by Hurault,[2] Schmid,[3] Tsuzuki,[4] and Gor'kov,[5] and experimentally by Thomas and Parks,[6] Klenin and Jensen,[7] and Kajimura and Mikoshiba[8] on thin aluminum films. The experimental results are in qualitative agreement with the theories.

According to the theories,[2-5] the characteristic field $E_c(T)$, at which the nonlinearity becomes appreciable, is proportional to $(T-T_c)^{3/2}$ in the case of thin films above T_c, while the zero-field excess conductivity is proportional to $(T-T_c)^{-1}$. However, the zero-field excess conductivity is observed to be continuous at T_c, and to increase exponentially as the temperature is lowered slightly below T_c. This behavior was successfully explained by Marčelja's theory.[9,10] In this temperature region the nonlinearity is expected to be greatly enhanced, but the theories of Refs. 2–4 cannot be applied to the immediate vicinity of T_c and below T_c. In this paper we present the result of a theoretical and experimental study on the nonlinear excess conductivity of thin films in this temperature region.

In Sec. II, we propose a Langevin equation appropriate for the temperature region slightly below T_c, and calculate the nonlinear excess conductivity. It is shown that the excess conductivity in the zero-electric-field limit reduces to the result given by Masker *et al.*,[9] and the electric field dependence of the excess conductivity is almost the same as that above T_c. The experimental procedure is given in Sec. III. In Sec. IV, we present the experimental results on the excess conductivity as a function of temperature and electric field, and compare them with the present calculation. The agreement between theory and experiment is fairly good. In Sec. V, discussions are given of the validity of the theory in terms of both a phenomenological treatment and a microscopic theory.[11]

II. THEORY

Schmid[3] proposed a Langevin equation for the superconducting order parameter $\Psi(\vec{r},t)$ above T_c and calculated the nonlinear excess conductivity. Tsuzuki[4] and Gor'kov[5] gave a support to this Langevin equation method by deriving the same result for the excess conductivity using the micro-