fine field which we have not considered here. The important ones may be magnon-magnon, magnon-phonon, and electron-phonon interactions. None of these is expected to be very effective for YIG in the temperature range of our experiment. A more comprehensive discussion of these different processes will be included in a future publication.

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Theory of Acoustic Paramagnetic Resonance in Dense Magnetic Insulators*

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Several mechanisms which describe the interaction between spins and phonons in a dense paramagnetic insulator are theoretically explored in order to estimate their resonant contribution to the ultrasonic attenuation at high temperatures. In particular, one mechanism provides a quasiresonant contribution to the attenuation which is roughly proportional to the square root of the difference of the frequency and certain multiples of the Larmor frequency. Other mechanisms may lead to the same type of line shape.

I. INTRODUCTION

In this paper we shall theoretically examine the question of acoustic paramagnetic resonance (APR) in dense magnetic insulators. The basic model used for the spin system is a Heisenberg paramagnet with an isotropic exchange energy much greater than the Zeeman energy due to an applied magnetic field or any anisotropic spin-dependent forces. For the most part, only temperatures much greater than the magnetic transition temperature of the spins are considered. For our purposes the "resonant" contribution to the acoustic attenuation will be taken to mean that contribution which depends on an externally applied magnetic field.

Electron paramagnetic resonance (EPR) experi-

ments in dense magnetic insulators show an intense, rather narrow line at the resonant frequency $\omega_0=\gamma H_0$, where H_0 is the applied magnetic field and γ is the electronic gyromagnetic ratio. One might naively expect similar results in an APR experiment as is the case with paramagnetic impurities in insulating dielectrics.[1] For example, an estimate[2] of the peak attenuation due to the acoustically modulated spin dipolar coupling for a typical dense magnetic insulator (MnF_2) at a temperature of 300 °K and a frequency of 1 GHz is about 3×10^{-2} cm^{-1}—if a linewidth of 0.3 kG (1 GHz) is assumed. Observation of such a change in attenuation is within the range of present-day experimental techniques. Unfortunately, estimates from moment calculations[2,3] suggest that a linewidth of 300 kG (10^3 GHz) is more appropriate. Not only would it reduce the peak attenuation by three orders of magnitude but it would make the "line" almost impossible to sweep.

The results[4] obtained from experiments and our analysis indicate that the situation is somewhere between these two extreme cases. In the high-temperature limit we obtain magnetic-field-dependent contributions to the ultrasonic attenuation which are roughly proportional to $-|\omega-m\omega_0|^{1/2}$, where m can take on the values ±1 and ±2. Other mechanisms which are weaker in the high-temperature limit,[5] but may still dominate in a temperature range well in excess of the magnetic transition temperature, may give similar contributions.

The reason why EPR and APR experiments yield different results is simply that, even at high temperatures, the Heisenberg paramagnet is a strongly interacting system and is not equivalent to an equal number of noninteracting spins. In fact, bandwidths due to spin-spin exchange interactions can be orders of magnitude larger than typical Zeeman energies. Since the coupling of an electromagnetic field to the spins is linear in the spin operators, since the wavelength of electromagnetic radiation is very large, and since the total spin of the system commutes with the exchange interaction, this bandwidth is relatively unimportant in EPR experiments. The isotropic exchange interaction does not directly contribute to the EPR line shape (or magnitude) but comes into play only through exchange narrowing of the relatively small anisotropic dipolar interaction. On the other hand, since spin-phonon couplings are quadratic in the spin, the complexities of the spin-spin exchange forces are of central importance in APR.

The rest of this section contains a description of the various interactions that are used in this paper. In Sec. II the formalism and approximations used in the calculations are developed. A discussion and tabulation of the results are contained in Sec. III. For readers not interested in technical details, Sec. II may be omitted.

The Hamiltonian for a magnetic insulator is written as a sum of spin, phonon, and spin-phonon interaction parts:

$$H=H_{\text{spin}}+H_{\text{phonon}}+H_{\text{int}}. \tag{1}$$

The spin part is taken to be

$$H_{\text{spin}}=-\hbar\gamma\sum_{\alpha}\vec{H}_0\cdot\vec{S}(\vec{\alpha},t)+\tfrac{1}{2}\sum_{\alpha,\alpha'}S_i(\vec{\alpha},t)J_{ij}(\vec{\alpha},\vec{\alpha}')S_j(\vec{\alpha}',t)+\sum_{\alpha}E_a[\vec{S}(\vec{\alpha},t)], \tag{2}$$

where $\vec{S}(\vec{\alpha},t)$ is the spin operator at the lattice site $\vec{\alpha}$ at time t in the Heisenberg representation. Latin subscripts refer to Cartesian directions, H_0 is an external magnetic field, and $J_{ij}(\vec{\alpha},\vec{\alpha}')$ describes the interaction between spins at different sites $\vec{\alpha}$ and $\vec{\alpha}'$. It is assumed that J_{ij} has an isotropic part (usual Heisenberg exchange) that is much larger than both its anisotropic (presumably dipolar) part and $\hbar\gamma H_0$. Any single-ion anisotropy energy (due to crystal-field and spin-orbit effects) is included in $E_a[\vec{S}]$ which is also much smaller than the isotropic part of J_{ij}. It will be our convention that all repeated Latin subscripts are summed over. The spin operators obey the usual commutation relations of

$$[S_x(\vec{\alpha},t),S_y(\vec{\alpha}',t)]=i\delta(\vec{\alpha},\vec{\alpha}')S_z(\vec{\alpha},t),\qquad [S_x(\vec{\alpha},t),S_x(\vec{\alpha}',t)]=0, \tag{3}$$

and cyclic permutations.

Because only changes in the phonon spectrum due to the spin-phonon interactions are being considered, it is sufficient to use the harmonic approximation for H_{phonon}. In terms of the normal coordinates,[6]

$$H_{\text{phonon}}=\tfrac{1}{2}\sum_{\vec{q},\lambda}[\dot{Q}(\vec{q},\lambda,t)\dot{Q}(-\vec{q},\lambda,t)+\omega_0^2(\vec{q},\lambda)Q(\vec{q},\lambda,t)Q(-\vec{q},\lambda,t)], \tag{4a}$$

$$[\dot{Q}(\vec{q},\lambda,t),Q(\vec{q}',\lambda',t)]=-i\hbar\delta(\lambda,\lambda')\delta(\vec{q}+\vec{q}'),\qquad [\dot{Q}(\vec{q},\lambda,t),\dot{Q}(\vec{q}',\lambda',t)]=[Q(\vec{q},\lambda,t),Q(\vec{q}',\lambda',t)]=0. \tag{4b}$$

In these equations, $\vec{q}$ is a wave vector in the first Brillouin zone, λ specifies the branch or normal mode, and $\omega_0(\vec{q},\lambda)$ is the harmonic frequency of the mode $(\vec{q},\lambda)$. The displacement of atom b in the $\vec{\alpha}$th unit cell, $U_i(\vec{\alpha},b,t)$, is

$$U_i(\vec{\alpha},b,t)=\sum_{\vec{q},\lambda}\frac{1}{(NM_b)^{1/2}}e_i(\vec{q},\lambda,b)\,Q(\vec{q},\lambda,t)\,e^{i\vec{q}\cdot\vec{\alpha}}, \tag{5}$$

where there are N unit cells in the crystal, M_b is the mass of atom b, and $e_i(\vec{q},\lambda,b)$ is the appropriate unit polarization vector.

The spin-phonon interactions considered in this paper are linear in the strains or normal coordinates and quadratic in the spin operators. Interactions of higher order in the spin variables should be much smaller while interactions of higher order in the strains should contribute very little to any resonant process. Since the exchange interaction depends upon the distance between the spins, lattice vibrations modulate this coupling and give rise to the volume magnetostriction described by $H_{\rm int}^{(V)}$,

$$H_{\rm int}^{(V)}=\tfrac{1}{2}\sum_{\vec{\alpha},\vec{\alpha}',\lambda,\vec{q}}\frac{e_i(\vec{q},\lambda)}{(NM)^{1/2}}\,Q_{jk}^{i}(\vec{\alpha}-\vec{\alpha}')S_j(\vec{\alpha},t)\times S_k(\vec{\alpha}',t)[e^{i\vec{q}\cdot\vec{\alpha}}-e^{i\vec{q}\cdot\vec{\alpha}'}]Q(\vec{q},\lambda,t), \quad (6)$$

where

$$Q_{jk}^{i}(\vec{\alpha}-\vec{\alpha}')=(\nabla_\alpha)_i J_{jk}(\vec{\alpha}-\vec{\alpha}'). \quad (7)$$

Only the magnetic ion in a unit cell will enter any of our expressions and thus the label b is suppressed. The anisotropy energy of a spin depends on the positions of the neighboring atoms and is therefore also modulated by lattice vibrations. This gives rise to the single-ion magnetostriction described by $H_{\rm int}^{(s)}$, conveniently written as

$$H_{\rm int}^{(s)}=i\sum_{\vec{\alpha},\vec{q},\lambda}\frac{1}{(MN)^{1/2}}\,G_{ij}q_jF_{ij}[\vec{S}(\vec{\alpha},t)]e^{i\vec{q}\cdot\vec{\alpha}}\times e_i(\vec{q},\lambda)Q(\vec{q},\lambda,t). \quad (8)$$

The magneto-elastic constants G_{ij} and the form of F_{ij} [which is quadratic in $S(\vec{\alpha},t)$] depend on the crystal structure under consideration. Different structures are considered in Sec. III.

II. FORMULATION

A. Ultrasonic Attenuation

For our purposes it is most convenient to discuss the acoustic properties of the system in terms of the phonon Green's function,

$$D(\vec{q},\lambda,t-t')=(i/\hbar)\langle(Q(\vec{q},\lambda,t)Q(-\vec{q},\lambda,t'))_+\rangle, \quad (9)$$

where the angular brackets $\langle X\rangle$ denote the average value of X in the canonical ensemble and $(\cdots)_+$ denotes the Wick time ordering operation. D is Fourier transformed in time according to the prescription[6]

$$D(\omega_\nu)=\int_0^{-i\hbar\beta}dt\,e^{i\omega_\nu(t-t')}\,D(t-t'), \quad (10)$$

where $\omega_\nu=(\pi\nu/-i\hbar\beta)$ and ν is an even integer. If we consider only single-ion magnetostriction, Eqs. (4) and the Heisenberg equation of motion,

$$i\hbar\frac{\partial A(t)}{\partial t}=[A(t),H(t)], \quad (11)$$

lead to an exact equation of motion for the phonon Green's function:

$$\left[\frac{\partial^2}{\partial t^2}+\omega_0^2(\vec{q},\lambda)\right]D(\vec{q},\lambda,t-t')=\delta(t-t')-\hbar^{-1}(MN)^{-1/2}\textstyle\sum_{\vec{\alpha}}G_{ij}q_j\,e^{-i\vec{q}\cdot\vec{\alpha}}\times e_i^*(\vec{q},\lambda)\langle(F_{ij}[S(\vec{\alpha},t)]Q(-\vec{q},\lambda,t'))_+\rangle. \quad (12)$$

Following Ref. 7, the term on the right-hand side of Eq. (12) is expanded to lowest order in $H_{\rm int}^{(s)}$ yielding

$$\left(\frac{\partial^2}{\partial t^2}+\omega_0^2(\vec{q},\lambda)\right)D(\vec{q},\lambda,t-t')=\delta(t-t')+\int_0^{-i\hbar\beta}d\bar{t}\,\Pi(\vec{q},\lambda,t-\bar{t})D(\vec{q},\lambda,\bar{t}-t'), \quad (13)$$

$$\Pi(\vec{q},\lambda,t-\bar{t})=\frac{i}{\hbar NM}\sum_{\alpha\alpha'}e^{-i\vec{q}\cdot(\vec{\alpha}-\vec{\alpha}')}\,\bar{G}_{ij}(\vec{q},\lambda)\bar{G}_{kl}(\vec{q},\lambda)\times\langle(S_i(\vec{\alpha},t)S_j(\vec{\alpha},t)S_k(\vec{\alpha}',\bar{t})S_l(\vec{\alpha}',\bar{t}))_+\rangle, \quad (14)$$

where the $\bar{G}_{ij}(\vec{q},\lambda)$ is defined by the equation

$$e_i(\vec{q},\lambda)G_{ij}q_jF_{ij}[\vec{S}(\vec{\alpha},t)]=S_i(\alpha,t)\bar{G}_{ij}(\vec{q},\lambda)S_j(\alpha,t). \quad (15)$$

This expansion is well justified because the spins and phonons are only weakly coupled. There are also terms contributing to Π from the volume magnetostriction and from interference terms between $H_{\rm int}^{(s)}$ and $H_{\rm int}^{(V)}$. These terms will be included later.

In order to proceed further the four-spin correlation function in Eq. (14) must be evaluated. Following other authors,[7,8] we shall approximate it by the sum of all possible products of two-spin correlation functions containing different times:

$$\langle([S_i(\vec{\alpha}_1,t)S_j(\vec{\alpha}_2,t)]_{\rm st}[S_k(\vec{\alpha}_3,\bar{t})S_l(\vec{\alpha}_4,\bar{t})]_{\rm st})_+\rangle=G_{ik}(\vec{\alpha}_1-\vec{\alpha}_3,t-\bar{t})G_{jl}(\vec{\alpha}_2-\vec{\alpha}_4,t-\bar{t})+G_{il}(\vec{\alpha}_1-\vec{\alpha}_4,t-\bar{t})G_{jk}(\vec{\alpha}_2-\vec{\alpha}_3,t-\bar{t}), \quad (16)$$

where $G_{ij}(\vec{\alpha}_1-\vec{\alpha}_3,t-\bar{t})$ is the usual two-spin correlation function,

$$G_{ij}(\vec{\alpha}-\vec{\alpha}',t-\bar{t})=\langle(S_i(\vec{\alpha},t)S_j(\vec{\alpha}',\bar{t}))_+\rangle. \quad (17)$$

The symbol $[\,]_{\rm st}$ denotes the symmetrized, traceless form of the operators enclosed,

$$[S_i(\vec{\alpha},t)S_j(\vec{\alpha}',t)]_{\rm st}=\tfrac{1}{2}[S_i(\vec{\alpha},t),S_j(\vec{\alpha}',t)]-\tfrac{1}{3}S(S+1)\delta_{ij}\delta(\vec{\alpha},\vec{\alpha}'). \quad (18)$$

Correlations that are independent of time will not contribute at finite frequencies and are neglected. Our calculation is primarily to be taken at temperatures much greater than the magnetic transition temperature where this decomposition is expected to be most valid.

The two-spin correlation function (or Green's function) is conveniently transformed according to the prescription

$$G_{ij}(\vec{\alpha}-\vec{\alpha}', t-t') = \frac{1}{N}\sum_{\vec{q}}\frac{1}{\beta}\sum_{\nu} e^{i\vec{q}\cdot(\vec{\alpha}-\vec{\alpha}')} e^{-i\omega_\nu(t-t')} G_{ij}(\vec{q}, \omega_\nu), \quad (19)$$

where $G_{ij}(\vec{q}, \omega_\nu)$ is expressed in terms of its spectral weight function[8] $\chi''(\vec{q}, \omega)$,

$$G_{ij}(\vec{q}, \omega_\nu) = \int_{-\infty}^{\infty} \frac{d\omega}{\pi}\frac{\chi''_{ij}(\vec{q}, \omega)}{\omega-\omega_\nu}. \quad (20)$$

From Eqs. (10), (14), (16), and (19), it is easily seen that

$$\Pi(\vec{q}, \lambda, \omega_\nu) = (\beta MN)^{-1}\sum_{\vec{q}', \nu'} \bar{G}_{ij}(\vec{q}, \lambda)\bar{G}_{kl}(\vec{q}, \lambda) \times [G_{ik}(\vec{q}'_+, \omega_{\nu'})G_{jl}(-\vec{q}'_-, \omega_\nu-\omega_{\nu'}) + G_{il}(\vec{q}'_+, \omega_{\nu'})G_{jk}(-\vec{q}'_-, \omega_\nu-\omega_{\nu'})], \quad \vec{q}'_\pm = \vec{q}' \pm \tfrac{1}{2}\vec{q}. \quad (21)$$

Now, if the anharmonic coupling is small, the ultrasonic dispersion and absorption are obtained by the prescription[6]

$$\Pi(\vec{q}, \lambda, \omega) = \Pi'(\vec{q}, \lambda, \omega) + i\Pi''(\vec{q}, \lambda, \omega) = \lim_{\epsilon\to 0^+}\Pi(\vec{q}, \lambda, \omega_\nu \to \omega + i\epsilon),$$
$$\Delta\omega(\vec{q}, \lambda) = -\Pi'(\vec{q}, \lambda, \omega_0(\vec{q}, \lambda))/2\omega_0(\vec{q}, \lambda),$$
$$\gamma(\vec{q}, \lambda) = \Pi''(\vec{q}, \lambda, \omega_0(\vec{q}, \lambda))/2\omega_0(\vec{q}, \lambda). \quad (22)$$

Using Eq. (20) the frequency sum in Eq. (21) is performed using the identity

$$\frac{1}{\beta\hbar}\sum_{\nu}\frac{1}{\omega-\omega_\nu} = \tfrac{1}{2}\coth\tfrac{1}{2}\beta\hbar\omega. \quad (23)$$

After letting $\omega_\nu \to \omega + i\epsilon$ and using the fact that $\beta\hbar\omega \ll 1$ for all relevant frequencies at temperatures much greater than the magnetic ordering temperature, one obtains

$$\Pi(\vec{q}, \lambda, \omega) = \frac{1}{\beta MN}\sum_{\vec{q}'}\int\frac{d\omega_1}{\pi}\int\frac{d\omega_2}{\pi}\frac{\gamma_{ij}(\vec{q}, \vec{q}')\gamma_{kl}(\vec{q}, \vec{q}')}{\omega_1\omega_2} \times \frac{\omega_1+\omega_2}{\omega_1+\omega_2-\omega-i\epsilon}[\chi''_{il}(\vec{q}'_+, \omega_1)\chi''_{jk}(-\vec{q}'_-, \omega_2) + \chi''_{ik}(\vec{q}'_+, \omega_1)\chi''_{jl}(-\vec{q}'_-, \omega_2)]. \quad (24)$$

When both the single-ion magnetostriction and the volume magnetostriction are included,

$$\gamma_{ij}(\vec{q}, \vec{q}') = [\bar{G}_{ij}(\vec{q}, \lambda) - \tfrac{1}{2}i(Q_{ij}(\vec{q}'_-) - Q_{ij}(\vec{q}'_+))],$$
$$Q_{jk}(\vec{q}) = e_i(\vec{q}, \lambda)\, Q^i_{jk}(\vec{q}), \quad (25)$$

where $Q^i_{jk}(\vec{q})$ is the Fourier transform of $Q^i_{jk}(\vec{\alpha}-\vec{\alpha}')$,

$$Q(\vec{q}) = N^{-1}\sum_{\vec{\alpha}} e^{i\vec{q}\cdot\vec{\alpha}} Q(\vec{\alpha}). \quad (26)$$

B. Correlation Functions

In the long-wavelength limit, $qa \ll 1$ (where a is typical lattice spacing), $\gamma_{ij}(\vec{q}, \vec{q}')$ is linear in q. Furthermore, if $q'a \ll 1$, $\gamma_{ij}(\vec{q}, \vec{q}')$ is also independent $\vec{q}'$ and thus we write

$$\gamma_{ij}(\vec{q}, \vec{q}') \to q\lambda_{ij}. \quad (27)$$

In order to evaluate the sum and integrals in Eq. (24) we shall use the low-frequency, small wave vector form of the spectral weight function. In a coordinate system where a uniform external field points along the z axis, and if the interaction Hamiltonian commutes with the spin, hydrodynamical arguments[9] demand that

$$\chi''_{\mu\nu}(\vec{q}, \omega) = \chi_{\mu\nu}(\vec{q})\omega Dq^2[(\omega-\omega_\mu)^2 + D^2q^4]^{-1}\delta_{\mu\tilde{\nu}}. \quad (28)$$

The spherical vector components, denoted by Greek subscripts $\tilde{\mu} = (+, -, 0)$ and $\tilde{\mu} = (-, +, 0)$, are used where

$$A_\pm = A_x \pm iA_y, \quad A_0 = A_z. \quad (29)$$

The resonant frequencies associated with the three spherical components are

$$\omega_\pm = \pm\gamma H_0 \text{ and } \omega_0 = 0. \quad (30)$$

D is the spin diffusion coefficient and $\chi_{\mu\nu}(\vec{q})$ is the wave-vector-dependent susceptibility.

The effects of the terms in the spin Hamiltonian that do not commute with the total spin {the anisotropic part of $J_{ij}(\vec{\alpha}-\vec{\alpha}')$ and $E_a[\vec{S}(\vec{\alpha}, t)]$} are taken into account phenomenologically. At high enough temperatures the primary effect is a correction to the linewidth or decay rate of the spins due to the exchanged-narrowed anisotropic interactions. This is included by replacing[10] Dq^2 by $\Gamma(\vec{q})$ where

$$\Gamma(\vec{q}) = \Gamma_0 + Dq^2. \quad (31)$$

The quantity Γ_0 is the linewidth observed in EPR experiments. Since at temperatures much greater than the magnetic transition temperature,

$$\chi_{+-}(\vec{q}) = \chi_{-+}(\vec{q}) = 2\chi_{00}(\vec{q}) = 2\chi_0 = \tfrac{2}{3}\beta S(S+1), \quad (32)$$

it is convenient to write

$$\chi''_{ij}(\vec{q}, \omega) = \sum_\alpha \chi^\alpha_{ij}\omega\Gamma(\vec{q})[(\omega-\omega_\alpha)^2 + \Gamma^2(\vec{q})]^{-1}, \quad (33)$$

$$\chi^\alpha_{ij} = \gamma_{i\alpha}\gamma_{j\tilde{\alpha}}\chi_0(2-\delta_{\alpha,0}). \quad (34)$$

The quantities $\gamma_{i\alpha}$ are defined by the transformation of the Cartesian coordinate system of the crystal to the spherical coordinate system in which $\vec{H}_0$ points along the $\alpha = 0$ axis,

$$A_i = \sum_\alpha \gamma_{i\alpha}A_\alpha. \quad (35)$$

The sum and integrals in Eq. (24) can now be

performed. Let $T_{\alpha\beta}$ be defined as

$$T_{\alpha\beta}=\frac{1}{N}\sum_{\vec{q}'}\int\frac{d\omega_1}{\pi}\int\frac{d\omega_2}{\pi}\frac{\Gamma(\vec{q}')}{(\omega_1-\omega_\alpha)^2+\Gamma^2(\vec{q}'_+)}\times\frac{\Gamma(\vec{q}-\vec{q}')}{(\omega_2-\omega_\beta)^2+\Gamma^2(\vec{q}'_-)}\frac{\omega_1+\omega_2}{\omega_1+\omega_2-\omega-i\epsilon}. \quad (36)$$

The two frequency integrals are easily performed, yielding

$$T_{\alpha\beta}=1+\frac{1}{N}\sum_{\vec{q}'}\frac{\omega}{\omega_\alpha+\omega_\beta-i\Gamma(\vec{q}'_+)-i\Gamma(\vec{q}'_-)}. \quad (37)$$

The summation of $\vec{q}'$ over the first Brillouin zone is replaced by an integral over the Debye zone:

$$\frac{1}{N}\sum_q\rightarrow\frac{v_a}{(2\pi)^3}\int_0^{q_d}q^2dq\int_0^{2\pi}d\phi\int_0^{\pi}\sin\theta\,d\theta, \quad (38)$$

where v_a is the volume of a unit cell and $q_d=(6\pi^2/v_a)^{1/3}$. In the limit $|\omega_\alpha+\omega_\beta-\omega-2i\Gamma_0|\ll Dq_d^2$,

$$T_{\alpha\beta}=1+\frac{\omega v_a q_d^3}{4\pi^2\omega_E}[i-\tfrac{1}{2}\pi f_{\alpha\beta}(\omega)], \quad (39)$$

where

$$\omega_E=2Dq_d^2 \quad (40)$$

and

$$f_{\alpha\beta}(\omega)=i\{\tfrac{1}{2}[Y+(X^2+Y^2)^{1/2}]\}^{1/2}-(X/|X|)\{\tfrac{1}{2}[-Y+(X^2+Y^2)^{1/2}]\}^{1/2},$$
$$Y=(2\Gamma_0+\tfrac{1}{2}Dq^2)/\omega_E,\quad X=(\omega_\alpha+\omega_\beta-\omega)/\omega_E. \quad (41)$$

It should be noted that the part of $T_{\alpha\beta}$ which depends on H_0 (the part proportional to $f_{\alpha\beta}$) comes from the low-frequency and small-wave-vector parts of the frequency and wave-vector integrals. That is, the resonant or field-dependent part of $T_{\alpha\beta}$ comes from that region of phase space where the hydrodynamic approximation is valid.

By combining Eqs. (24), (27), (33), (36), and (39), one obtains the final result

$$\Pi(\vec{q},\lambda,\omega)=-(3\pi q^2\omega/2\beta M\omega_E)\lambda_{ij}(\lambda_{kl}+\lambda_{lk})\chi_{il}^{\alpha}\chi_{jk}^{\beta}f_{\alpha\beta}(\omega), \quad (42)$$

where all indices, Latin and Greek, are summed over.

III. RESULTS AND CONCLUSIONS

In Sec. II we have derived a formula that includes the resonant (magnetic-field-dependent) ultrasonic dispersion and absorption due to single-ion and volume magnetostriction. Since the infinite-temperature form of the magnetic susceptibility has been used, Eq. (42) is valid only at temperatures much greater than the magnetic transition temperature. The treatment could possibly be extended to lower temperatures but some of the other approximations may be far less valid there. In this section we shall apply the results to different types of crystals and acoustic modes in the high-temperature limit. We shall also comment briefly on other possible mechanisms with different temperature and angular dependences.

Our discussion will be limited to crystal structures that possess uniaxial (tetragonal) symmetry of which cubic crystals are a special case. It is convenient to define two coordinate systems; one (denoted by the subscript c) which coincides with the crystal axes and one (denoted by the subscript h) whose z axis points along the direction of the external magnetic field. If the direction of the magnetic field is given by the spherical angles (θ,ϕ) in the crystal coordinate system, a convenient transformation between the systems is

$$x_c=\cos\phi\ (x_h\cos\theta+z_h\sin\theta)-y_h\sin\phi,$$
$$y_c=\sin\phi\ (x_h\cos\theta+z_h\sin\theta)+y_h\sin\phi, \quad (43)$$
$$z_c=z_h\cos\theta-x_h\sin\theta.$$

For the spins it is also more convenient to work with the spherical vectors

$$A_\pm=A_{x_h}\pm iA_{y_h},\quad A_0=A_{z_h}, \quad (44)$$

than the Cartesian vectors. Any vector in the crystal coordinate system can be related to spherical vectors in the spins coordinate system through the transformation

$$A_i=\sum_\mu\gamma_{i\mu}A_\mu, \quad (45)$$

where $i=(x,y,z)$, $\mu=(+,-,0)$, and the complex direction cosines, $\gamma_{i\mu}$, are

$$\gamma_{x+}=\gamma_{x-}^*=\tfrac{1}{2}(\cos\theta\cos\phi+i\sin\phi),$$
$$\gamma_{y+}=\gamma_{y-}^*=\tfrac{1}{2}(\cos\theta\sin\phi-i\sin\phi),$$
$$\gamma_{z+}=\gamma_{z-}=-\tfrac{1}{2}\sin\theta, \quad (46)$$
$$\gamma_{z0}=\cos\theta,\quad \gamma_{x0}=\sin\theta\cos\phi,\quad \gamma_{y0}=\sin\theta\sin\phi.$$

All of these indices are somewhat overwhelming, but are necessary if we wish to consider arbitrary magnetic field angles and phonon modes. From Eqs. (34), (41), (42), and (45), one obtains the resonant part of the ultrasonic attenuation coefficient[11] α for acoustic phonons with velocity v,

$$\alpha(\lambda)=-(6\pi\omega^2\beta/Mv^3\omega_E)[S(S+1)/3]^2\times\{|\lambda_{++}|^2[f^{(2)}(\omega)+f^{(-2)}(\omega)]+|\lambda_{+0}|^2[f^{(1)}(\omega)+f^{(-1)}(\omega)]\}, \quad (47)$$

where

$$\lambda_{\alpha\beta}=\gamma_{i\alpha}\bar{G}_{ij}(\vec{q},\lambda)\gamma_{j\beta}/q \quad (48)$$

and

$$f^{(m)}(\omega)=\{2\Gamma_0+[(\omega-m\omega_0)^2+4\Gamma_0^2]^{1/2}\}^{1/2}/(2\omega_E)^{1/2}, \quad (49)$$

where $\omega=vq$ and λ specifies the phonon branch. For a tetragonal crystal with its c axis along the

TABLE I. Coupling constants λ_{+-} and γ_{+0} for various acoustic phonon modes which result from Eq. (48). The c axis of the crystal is in the z direction, the direction of the magnetic field is given by the spherical angles (θ, ϕ), $\vec{q}$ is the phonon wave vector, and $\vec{e}$ is the phonon polarization vector.

$\lambda_{+-}\{\vec{q} \parallel \vec{e} \parallel [001]\}$	$9G_{33}^2 \sin^4\theta/16$
$\lambda_{+0}\{\vec{q} \parallel \vec{e} \parallel [001]\}$	$9G_{33}^2 \sin^2 2\theta/4$
$\lambda_{+-}\{\vec{q} \parallel \vec{e} \parallel [110]\}$ (cubic crystal only)	$[\frac{3}{4}G_{11} \sin^2\theta + G_{44}(\sin^2\theta - 2) \sin\phi \cos\phi]^2/4 + G_{44}^2 \cos^2\theta(\sin^2\phi - \cos^2\phi)^2/4$
$\lambda_{+0}\{\vec{q} \parallel \vec{e} \parallel [110]\}$ (cubic crystal only)	$\sin^2 2\theta(\frac{3}{2}G_{11} + G_{44} \sin\phi \cos\phi)^2 + G_{44}^2 \sin^2\theta(\sin^2\phi - \cos^2\phi)^2/16$
$\lambda_{+-}\{\vec{q} \parallel [001], \vec{e} \parallel [\cos\psi, \sin\psi, 0]\}$	$G_{44}^2 \sin^2\theta[\sin^2(\phi - \psi) + \cos^2\theta \cos^2(\phi - \psi)]/16$
$\lambda_{+0}\{\vec{q} \parallel [001], \vec{e} \parallel [\cos\psi, \sin\psi, 0]\}$	$G_{44}^2[\cos^2\theta \sin^2(\phi - \psi) + (1 - 2\sin^2\theta)^2 \cos^2(\phi - \psi)]/16$

z direction, $\bar{G}_{ij}(\vec{q}, \lambda)$ can take the general form

$$\begin{aligned}
\bar{G}_{xx}(\vec{q}, \lambda) &= (G_{11} - G_{31})\epsilon_{xx} + (-G_{11} - 2G_{31})\epsilon_{yy}, \\
\bar{G}_{yy}(\vec{q}, \lambda) &= (G_{11} - G_{31})\epsilon_{yy} + (-G_{11} - 2G_{31})\epsilon_{xx}, \\
\bar{G}_{zz}(\vec{q}, \lambda) &= \tfrac{3}{2} G_{33}\epsilon_{zz}, \\
\bar{G}_{xy}(\vec{q}, \lambda) &= \bar{G}_{yx}(\vec{q}, \lambda) = G_{66}\epsilon_{xy}, \\
\bar{G}_{xz}(\vec{q}, \lambda) &= \bar{G}_{zx}(\vec{q}, \lambda) = G_{44}\epsilon_{zx}, \\
\bar{G}_{yz}(\vec{q}, \lambda) &= \bar{G}_{zy}(\vec{q}, \lambda) = G_{44}\epsilon_{yz},
\end{aligned} \tag{50}$$

where

$$\epsilon_{ij} = \tfrac{1}{2}[q_i e_j(\vec{q}, \lambda) + q_j e_i(\vec{q}, \lambda)] \tag{51}$$

and $e_i(\vec{q}, \lambda)$ is the usual phonon polarization vector. For a cubic crystal, $G_{33} = G_{11}$, $G_{31} = -\frac{1}{2} G_{11}$, and $G_{66} = G_{44}$.

The contributions to the G's come from both the single-ion magnetostriction and the anisotropic part of the volume magnetostriction. Since we have taken the spins spectral function to be diagonal in the spin [see Eq. (28) or (32)], the part of the volume magnetostriction due to modulation of the isotropic Heisenberg exchange does not contribute. Similarly, for our purposes, the diagonal part of the single-ion magnetostriction does not matter and Eq. (50) has been adjusted to our convenience with this in mind. From Eqs. (46), (48), (50), and (51), the values of $\lambda_{\alpha\beta}$ can be straightforwardly worked out for various phonon modes and magnetic field configurations. In addition, several simple cases are included in Table I. The "exchange frequency" ω_E is given by Eq. (40), and Γ_0 is the intrinsic linewidth such as is observed in EPR experiments [see discussion after Eq. (31)].

The distinctive feature of the resonant attenuation is the shape functions $f^{(m)}(\omega)$ which, in the absence of any intrinsic linewidth (Γ_0), are proportional to the square root of $|\omega - m\omega_0|$. The total field-dependent ultrasonic absorption is proportional to a sum of four terms with m equal to both ± 1 and ± 2. The minus sign in Eq. (47) means that the contribution of each $f^{(m)}(\omega)$ to α decreases as $|\omega - m\omega_0|$ increases. There is, of course, a field-independent contribution to α from the same mechanism which is larger in magnitude. In addition, α is proportional to the usual factors of ω^2, T^{-1}, and $(MV^3)^{-1}$. This line shape fits well with existing experimental results.[4]

This mechanism, which we have considered in detail, should dominate at sufficiently high temperatures. However, other mechanisms could also produce a line shape similar to that described by Eq. (49). For example, besides giving an intrinsic linewidth Γ_0, the anisotropic spin exchange and anisotropy energy will lead to off-diagonal elements of the spectral function [e.g., $\chi''_{+z}(\vec{q}, \omega)$]. The magnitude of this effect can be estimated from an expansion of the wave-vector-dependent susceptibility in powers of interaction energies divided by $k_b T$. This would lead to additional factors in the attenuation of order

$$[\tfrac{1}{3}\beta S(S+1)E_{\text{anis}}]^2, \tag{52}$$

where E_{anis} is due to anisotropic exchange and the anisotropy energy. Even though this factor is very small at temperatures above the magnetic transition temperature, the net effect of this "off-diagonal" process could be larger than the "diagonal" process already considered because the modulation of the isotropic exchange now enters. This modulated isotropic exchange can be several orders of magnitude larger than the modulated anisotropic exchange or single-ion magnetostriction.

These "off-diagonal" processes would be proportional to T^{-3} instead of T^{-1} and would have a different angular dependence. For example, in a cubic crystal, the anisotropic dipolar interaction leads to an angular-independent term. Other mechanisms involving more spin pairs may also be important which could give contributions proportional to $f^{(m)}(\omega)$ with $|m|$ greater than 2.

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Pseudohelicoidal Surface Spin Waves

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A simple theoretical semiclassical calculation of the surface ferromagnetic equilibrium is given. The equilibrium orientation of the magnetization in the layers which are near the surface is tilted from the direction of the magnetization in the bulk. This rotation depends on both surface and bulk anisotropies. Thus, some instabilities in the spectrum disappear, and optical surface spin waves are found to be less energetic than what is usually calculated. Moreover, a simple interpretation of a possible origin of the pinning of surface spins is given.

There have been recently a number of theoretical investigations of surface spin waves in Heisenberg ferromagnets,[1-7] where the existence of surface modes is related, in the case of nearest-neighbor exchange, to the variation of the bulk exchange parameter at the surface. Nevertheless, an important question which remains open is that of the direction of the magnetization at the surface. In this paper a simple theoretical calculation of the surface equilibrium configuration is given. The most interesting result is that when the spin layer draws near to the surface, the spin magnetization rotates, and a so-called "pseudohelicoidal" structure is found in the vicinity of the surface. Consequently, a simple interpretation of the origin of pinning effects is given.

Moreover, the surface spin-wave spectrum is found to be perturbed by the existence of surface anisotropy. A treatment dealing with the existence of optical surface spin waves has already been given in the special case where the bulk and the surface magnetizations are parallel.[8] However, the aim of this work is not to demonstrate that the surface and the bulk spectra are model dependent, but to lay emphasis on the fact that the magnetic structure of the surface often differs qualitatively from that of the bulk.

The direction of the bulk magnetization $\vec{M}$ is assumed to be determined by a total energy balance (bulk anisotropy, sample configuration, $\cdots$) and thus it is independent of the surface anisotropy parameters. Let α be its angle with the axis Oz, which is perpendicular to the surface. In order to describe the surface effects we introduce the following Hamiltonian[9,10]:

$$H=-\tfrac{1}{2}\sum_{f_1,f_2,\alpha} J(f_1-f_2)S^{\alpha}_{f_1}S^{\alpha}_{f_2}-\tfrac{1}{2}\sum_{f_1,f_2} I(f_1-f_2)S^{z}_{f_1}S^{z}_{f_2}\,. \tag{1}$$

The bulk anisotropy is not included in H ($\alpha=x,y,z$). The magnetocrystalline anisotropy integrals $I(f_1-f_2)$ differ from zero near the surface only, where we make the assumption that $I(f_1-f_2)$ is of the same order of magnitude as $J(f_1-f_2)$. The isotropic exchange $J(f_1-f_2)$ will also be perturbed. The penetration of the perturbation, i.e., the range of the helicoidal structure is given by the range where the I factors are nonzero, which is different from the surface spin-wave penetration. In order to simplify the calculation, we assume that only the first surface layer is perturbed. We define thus the following parameters:

$$J_{\parallel}=J(f_1-f_2)\,,\quad I_{\parallel}=I(f_1-f_2)\,,$$

where f_1 and f_2 are two nearest neighbors located on the surface layer and

$$J_{\perp}=J(f_1-f_2)\,,\quad I_{\perp}=I(f_1-f_2)\,,$$

where f_1 and f_2 are two nearest neighbors located on the first and second layers. Let us assume that