temperature variation of the magneto-elastic energy is higher in the ferromagnetic state than in the helical state. Noteworthy is the minimum of the magneto-elastic energy in the helical state, at 95°K. At this temperature the volume compressibility, Fig. 4, is at its minimal value.

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Coupled Electronic Spins, Nuclear Spins, and Phonons in a Cubic Antiferromagnet*

PETER A. FEDDERS
Arthur Holly Compton Laboratory of Physics, Washington University, St. Louis, Missouri 63130
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The spin response functions for the electronic and nuclear spins in a cubic antiferromagnet are obtained in the random-phase approximation. These response functions are then used to investigate the coupled electronic-spin–nuclear-spin–phonon system. Predicted changes in phonon velocity and attenuation in $RbMnF_3$ due to interaction with electronic and nuclear spin waves are obtained. Some of these predictions agree with previous experiments and theory, and others can be checked by further experiments.

I. INTRODUCTION

THE coupled system of electronic and nuclear spins[1,2] and its interaction with acoustic phonons[3,4] in an antiferromagnet with cubic symmetry have been of some interest lately. It is the purpose of this paper to treat this coupled problem consistently from a unified point of view within the random-phase approximation (RPA) and, in particular, to obtain detailed predictions for $RbMnF_3$. Because of the complicated geometry of the equilibrium magnetization of a cubic antiferromagnet, this treatment is limited to configurations in which an external magnetic field lies in a restricted range of a {110} plane. As will be seen, because of the relatively strong coupling between the electronic and nuclear spins in $RbMnF_3$, the combined system must be taken into account in order to obtain correct results for the effects of the antiferromagnetism on the phonons.

The Hamiltonian used to describe the coupled electronic and nuclear spins is

$$H_{en}=\tfrac{1}{2}\sum_{\alpha,\alpha'}J(\alpha,\alpha')\mathbf{S}(\alpha)\cdot\mathbf{S}(\alpha')-\sum_{\alpha}A\mathbf{I}(\alpha)\cdot\mathbf{S}(\alpha)-\sum_{\alpha}\mathbf{H}_0\cdot[\mu_e\mathbf{S}(\alpha)+\mu_n\mathbf{I}(\alpha)]+H_a, \quad (1.1)$$

where $\mathbf{S}(\alpha)$ and $\mathbf{I}(\alpha)$ are the electronic- and nuclear-spin operators, respectively, at the magnetic site α. The exchange interaction is written so that $J(\alpha,\alpha')$ is positive for antiferromagnetic coupling, $\mathbf{H}_0$ is a uniform applied magnetic field and μ_e (μ_n) is the electronic (nuclear) magnetic moment. H_a is a term describing the electronic-spin anisotropy energy which, for a cubic system, is

$$E_a=\tfrac{1}{2}K(S_{x_c}{}^4+S_{y_c}{}^4+S_{z_c}{}^4) \quad (1.2)$$

per site. The subscript c denotes the usual coordinate system coincident with the cubic axes of the crystal.

The spins and phonons interact through the single-ion magnetostriction Hamiltonian H_{ep} which, for a lattice with cubic symmetry, takes the form

$$H_{ep}=G_{11}\sum_{\alpha}\{\epsilon_{xx}(\alpha)[S_x{}^2(\alpha)-\tfrac{1}{2}S_y{}^2(\alpha)-\tfrac{1}{2}S_z{}^2(\alpha)]+(\text{cyclic permutations})\}+G_{44}\sum_{\alpha}\{\epsilon_{xy}(\alpha)\times[S_x(\alpha)S_y(\alpha)+S_y(\alpha)S_x(\alpha)]+(\text{cyclic permutations})\}. \quad (1.3)$$

The strain componets $\epsilon_{ij}(\alpha)$ can be expressed in terms of the phonon displacement operation $U_i(\alpha)$ by the equation

$$\epsilon_{ij}(\alpha)=\frac{1}{2}\left[\frac{\partial U_i(\alpha)}{\partial\alpha_j}+\frac{\partial U_j(\alpha)}{\partial\alpha_i}\right]. \quad (1.4)$$

The method of attacking the problem is to first calculate the electronic-spin correlation function using thermodynamic Green's functions and then to use it to obtain predictions about various experiments. Section II contains a discussion of the model, the approximations employed, and the use of the spin correlation functions. In order (it is hoped) to make the paper more useful to a variety of readers, formulas for the spin correlation functions are derived in Appendix A, while

* Work supported in part by National Science Foundation under Grant No. GP-9573.
[1] W. J. Ince, Phys. Rev. **184**, 574 (1969).
[2] L. W. Hinderks and P. M. Richards, Phys. Rev. **183**, 575 (1969).
[3] R. L. Melcher and D. I. Bolef, Phys. Rev. **186**, 491 (1969). Hereafter referred to as M & B.
[4] A. Platzker and F. R. Morganthaler, Phys. Rev. Letters **22**, 1051 (1969).

the magnetoelastic coupling is treated in Appendix B. In Sec. III, the correlation functions are discussed for values of the parameters appropriate to $RbMnF_3$. Also, some of the effects of the antiferromagnetism on acoustic phonon velocity and attenuation are considered with a comparison to experiment.

In order to make contact with other work,[5] the following definitions are made:

$$\begin{gathered} S_0=\tfrac{1}{2}(S_1+S_2)\,, \quad I_0=\tfrac{1}{2}(I_1+I_2)\,, \\ \hbar\omega_A=\mu_e H_A=\tfrac{4}{3}kS_0{}^3\,, \quad \hbar\omega_E=\mu_e H_E=S_0J_0\,, \\ \hbar\omega_{NE}=\mu_e H_{NE}=AI_0\,, \quad \mu_e H_0=\hbar\omega_0\,, \\ \chi_\perp=\mu_e{}^2/J_0\,, \quad \hbar\omega_N=\mu_e H_N=AS_0\,, \end{gathered} \tag{1.5}$$

where S_1 (I_1) and S_2 (I_2) are the average electronic (nuclear) spins per site on the two magnetic sublattices and J_0 is the sum of $J(\alpha,\alpha')$ over all sites α' of sublattice 2 with α fixed at a site of sublattice 1. The magnetic sites form a simple cubic lattice with nearest neighbors belonging to different sublattices. The microscopic constants G_{ii} can be related to the phenomenological magneto-elastic coupling constants b_i of M&B through the equations

$$\begin{gathered} (\tfrac{3}{2}G_{11})^2S_0{}^3/M=b_1{}^2\mu_e/\rho M_0\,, \\ G_{44}{}^2S_0{}^3/M=b_2{}^2\mu_e/\rho M_0\,. \end{gathered} \tag{1.6}$$

Here, M is the mass of a magnetic cell, ρ is the density of the system, and M_0 is the magnetic moment per unit volume of the system.

II. MODEL

The electronic-spin correlation function or dynamic susceptibility can be defined as

$$\begin{gathered} D_{ij}(\alpha\alpha',\omega)=\frac{i}{\hbar}\int_0^\infty dt\, e^{i\omega t}\langle[\tilde{S}_i(\alpha,t),\tilde{S}_j(\alpha',0)]\rangle\,, \\ \tilde{S}_i(\alpha,t)=S_i(\alpha,t)-\langle S_i(\alpha)\rangle\,, \end{gathered} \tag{2.1}$$

where the angular brackets $\langle\ \rangle$ denote that the expectation value is to be taken over a thermal ensemble. This complex susceptibility can be thought of as giving the response of $S_i(\alpha)$ to a perturbation on $S_j(\alpha')$ with a frequency ω. It is directly related to the absorption of electromagnetic waves by the spin system, and it can be used in approximations to obtain the effects of the spins on the phonons.

It is also convenient to Fourier-transform $D_{ij}(\alpha\alpha',\omega)$ in space. Since there are two magnetic sublattices, a unit cell of the system contains two magnetic ions. Thus, the position index α is a combination of a vector index 1 denoting the unit cell and an index a denoting the position of the ion within the unit cell. The index a takes the values 1 and 2 corresponding to the two magnetic sublattices. Since the system possess translational symmetry, $D_{ij}(\alpha_1\alpha_2)$ can be Fourier-transformed as

$$D_{ij}(a_1a_2,\mathbf{l}_1-\mathbf{l}_2)=N^{-1}\sum_{\mathbf{q}} D_{ij}(a_1a_2,\mathbf{q})e^{i\mathbf{q}\cdot(\mathbf{l}_1-\mathbf{l}_2)}\,, \tag{2.2}$$

where N is the number of unit cells in the crystal and the summation is over all values of $\mathbf{q}$ in the first Brillouin zone.

This treatment of the spin correlation functions is restricted to cases where $\mathbf{H}_0$ lies in the $(\bar{1}\ 1\ 0)$ plane between the [001] and [110] axes. Further, ψ, the angle between $\mathbf{H}_0$ and the [001] axis, is restricted to the range from 0 to $\arcsin\frac{2}{3}$ and the point $\psi=\frac{1}{4}\pi$.

First consider the case where $0<\psi<\arcsin\frac{2}{3}$, which shall be called case A. It is convenient to work in the coordinate system defined by the equations

$$\begin{aligned} x&=(x_c+y_c)/\sqrt{2}\,, \quad z=z_c\,, \\ y&=(y_c-x_c)/\sqrt{2}\,. \end{aligned} \tag{2.3}$$

This is the coordinate system used in Appendix A in which both sublattice magnetizations lie in the x-z plane. The equilibrium configuration for the spins in this case has been worked out by Ince.[1] $\mathbf{H}_0$ makes an angle ψ with the z or [001] axis, and the sublattice spins $\mathbf{S}_1$ and $\mathbf{S}_2$ make angles of $\theta-t$ and $\theta+\pi+t$, respectively, with the z axis. The equilibrium values for θ and t are given by the formulas

$$\begin{gathered} H_0{}^2(1-\alpha)\sin2\phi+3H_EH_A\sin2\theta(1-\tfrac{3}{2}\sin^2\theta)\,, \\ t=H_0\sin\phi/2H_E\,, \quad \phi=\theta-\psi\,, \quad \alpha=\chi_{11}/\chi_\perp\,, \\ S_1-S_2=\chi_{11}H_0\cos\phi/\mu_e\,. \end{gathered} \tag{2.4}$$

The lowest-energy solution of the above equations is the one which minimizes $|\phi-\frac{1}{2}\pi|$.

It is also convenient to introduce the following coordinate systems, one for each magnetic sublattice, denoted by a bar:

$$\begin{aligned} \bar{x}(a)&=[S_z(a)x-S_x(a)z]/S(a)\,, \\ \bar{y}(a)&=y\,, \\ \bar{z}(a)&=[S_x(a)x+S_z(a)z]/S(a)\,, \end{aligned} \tag{2.5}$$

where x, y, and z refer to Eqs. (2.3).

In these coordinate systems, $\bar{z}(a)$ points along the direction of magnetization of the sublattice a, and the common $\bar{y}$ direction is perpendicular to both sublattice magnetizations. The utility of this transform is that $D_{ij}(aa',\mathbf{q},\omega)$ is zero if i is $\bar{z}(a)$ or if j is $\bar{z}(a')$.

Case B, where $\psi=\frac{1}{2}\pi$, is somewhat simpler because $|S_1|$ is equal to $|S_2|$ and $\mathbf{H}_0$ is perpendicular to $\mathbf{S}_1-\mathbf{S}_2$. In the coordinate system defined by

$$\begin{aligned} x&=(-\eta x_c+\eta y_c+z_c)/\sqrt{3}\,, \\ y&=(x_c-y_c+2\eta z_c)/\sqrt{6}\,, \\ z&=(x_c+y_c)/\sqrt{2}\,, \end{aligned} \tag{2.6}$$

where z is along the [110] axis and is parallel to $\mathbf{H}_0$, x is

[5] The notation in this paper, in so far as is possible, is like that of Refs. 1 and 3. Values for the parameters in $RbMnF_3$ are given in these references.

along the $[-\eta\ \eta\ 1]$ axis which is parallel to $\mathbf{S}_1-\mathbf{S}_2$, and y is along the $[1\ -1\ 2\eta]$ axis which is perpendicular to $\mathbf{S}_1$ and $\mathbf{S}_2$. The quantity η takes on the values $+1$ and -1. The sublattice spins $\mathbf{S}_1$ and $\mathbf{S}_2$ make angles of t and $\pi-t$, respectively, with the x axis, and the equilibrium value for t is

$$t=H_0/2H_E. \tag{2.7}$$

The energies associated with the two values of η are degenerate. Equations (2.5) can also be used to define the bar coordinate systems in case B, where the x, y, z system is now given by Eqs. (2.6).

In Appendix A, equations for the electronic-spin correlation functions $D_{ij}(a\,a',\mathbf{q},\omega)$ are obtained and solved. The Hamiltonian given by Eq. (1.1) is used together with the RPA for all terms except the H_a term, which is treated semiclassically. For small values of $\mathbf{q}$, the magnetoelastic coupling given by Eq. (1.3) will have a negligible effect on $D_{ij}(a\ a',\mathbf{q},\omega)$ because the coupling is a deformation potential. On the other hand, this coupling is responsible for the changes in the phonon modes due to the antiferromagnetism, as is derived in Appendix B. The reason why the nuclear-spin modes couple to the phonons in this scheme is that they appear in the electronic-spion response function. This is further discussed in Sec. III.

III. RESULTS

Although the Appendixes contain expressions for the spin correlation functions at arbitrary temperatures, the results in this section are restricted to situations in which χ_{11} is unimportant (that is, to low enough temperatures so that (χ_{11}/χ_1) is nearly zero or to large enough H_0, so that $\cos\phi$ is nearly zero). Neglecting terms of order $(\omega_0/\omega_E)^2$, $(\omega_{NE}+\omega_A)/\omega_E$, and $\omega_N^2/\omega_E(\omega_A+\omega_{NE})$ compared to 1, the electronic-spin correlation fundtions for case A can be written as

$$\begin{aligned}
D_{\bar{x}\bar{x}}(aa,\mathbf{q},\omega)&=D_{\bar{x}\bar{x}}(a\tilde{a},\mathbf{q},\omega)=(\omega_{e+}^2-\Omega_-^2)F_+(\omega)\\
&\quad+(\Omega_-^2-\omega_{e-}^2)F_-(\omega),\\
D_{\bar{y}\bar{y}}(aa,\mathbf{q},\omega)&=-D_{\bar{y}\bar{y}}(a\tilde{a},\mathbf{q},\omega)=(\omega_{e+}^2-\Omega_+^2)F_+(\omega)\\
&\quad+(\Omega_+^2-\omega_{e-}^2)F_-(\omega),\\
D_{\bar{x}\bar{y}}(aa,\mathbf{q},\omega)&=D_{\bar{y}\bar{x}}(a\tilde{a},\mathbf{q},\omega)=-D_{\bar{x}\bar{y}}(a\tilde{a},\mathbf{q},\omega)\\
&=-D_{\bar{y}\bar{x}}(aa,\mathbf{q},\omega)\\
&=2i\omega\omega_0\cos\phi[F_+(\omega)-F_-(\omega)],\\
F_\pm(\omega)&=[S_0\omega_E/\hbar(\omega_{e+}^2-\omega_{e-}^2)][(\omega^2-\omega_{e\pm}^2)^{-1}\\
&\quad+(2\omega_E\omega_N^2\omega_{NE}/\omega_{e\pm}^4)(\omega^2-\omega_{n\pm}^2)^{-1}],
\end{aligned} \tag{3.1}$$

where

$$\begin{aligned}
\Omega_+^2&=\omega_0^2(2\sin^2\phi-1)+2\omega_E[\omega_{NE}+\tfrac{3}{2}\omega_A f(\theta)+\omega_E Q(\mathbf{q})],\\
\Omega_-^2&=\omega_0^2(\sin^2\phi-1)+2\omega_E[\omega_{NE}+\tfrac{3}{2}\omega_A g(\theta)+\omega_E Q(\mathbf{q})],\\
\omega_{e\pm}^2&=\tfrac{1}{2}\{\Omega_+^2+\Omega_-^2+4\omega_0^2\cos^2\phi\\
&\quad\pm[(\Omega_+^2+\Omega_-^2+4\omega_0^2\cos^2\phi)^2-4\Omega_+^2\Omega_-^2]^{1/2}\},\\
\omega_{n\pm}^2&=\omega_N^2[1-(2\omega_E\omega_{NE}/\omega_{e\pm}^2)].
\end{aligned} \tag{3.2}$$

The $\mathbf{q}$ dependence of these quantities is contained in $Q(\mathbf{q})$ which, with the quantities $f(\theta)$ and $g(\theta)$ are defined by Eq. (A22). As is expressed by Eq. (A14), $D_{\bar{i}\bar{j}}(a\ a',\mathbf{q},\omega)$ means that $\bar{\imath}$ is $\bar{\imath}(a)$ and $\bar{\jmath}$ is $\bar{\jmath}(a')$. The equations for the spin correlation function in case B are also given by Eqs. (3.1) and (3.2), but with $\theta=\phi=\frac{1}{2}\pi$.

Except for the $\mathbf{q}$ dependence of the modes, Eqs. (3.2) are the same as derived by others.[6] All these modes have also been observed[7] electromagnetically in $RbMnF_3$. On the other hand, the present treatment also yields the spectral weights of the modes in all directions. Thus, one can see that when ϕ is $\frac{1}{2}\pi$, $\omega_{e\pm}=\Omega_\pm$ and that the electronic and nuclear modes labeled plus depend on H_0 and can be excited only by a spin perturbation parallel to $\mathbf{H}_0$, while the modes labeled minus do not depend on H_0 and can only be excited by spin perturbation perpendicular to $\mathbf{H}_0$, $\mathbf{S}_1$, and $\mathbf{S}_2$.[8] One can also calculate S_0 and χ_{11} within the RPA by using these functions.[8]

In Appendix B, some of the effects of the antiferromagnetism on the phonon spectrum are derived. A convenient way of expressing these results is through the equation

$$\begin{aligned}
\omega^2=\omega_0^2(\mathbf{q}\lambda)+M^{-1}S_0^2\sum_{a,a'}\{&C_{\bar{x}}(a)D_{\bar{x}\bar{x}}(aa',\mathbf{q},\omega)C_{\bar{x}}(a')\\
&+C_{\bar{y}}(a)D_{\bar{y}\bar{y}}(aa',\mathbf{q},\omega)C_{\bar{y}}(a')\},
\end{aligned} \tag{3.3}$$

where ω and $\mathbf{q}$ are the frequency and wave vector of the phonons, M is the mass of a magnetic cell, and $\omega_0(\mathbf{q},\lambda)$ is the frequency of the phonons in the branch λ in the absence of the antiferromagnetism. The C's for cases A and B are given by the equations

$$\begin{aligned}
C_{\bar{x}}(a)&=\tfrac{3}{2}G_{11}[S_x(a)S_z(a)/S^2(a)](e_xq_x+e_yq_y-2e_zq_z)\\
&\quad+G_{44}\{[S_x(a)S_z(a)/S^2(a)](e_xq_y+e_yq_x)\\
&\quad+[S_z^2(a)-S_x^2(a)]/\sqrt{2}S^2(a)\\
&\quad\times(q_ze_y+q_ye_z+q_ze_x+q_xe_z)\},\\
C_{\bar{y}}(a)&=\tfrac{3}{2}G_{11}[S_x(a)/S(a)](q_ye_y-q_xe_x)\\
&\quad+G_{44}[S_z(a)/\sqrt{2}S(a)](q_ze_y+q_ye_z-q_ze_x-q_xe_y),
\end{aligned} \tag{3.4a}$$

for case A and

$$\begin{aligned}
C_{\bar{x}}(a)&=\tfrac{3}{2}G_{11}\eta\tfrac{1}{3}(6^{1/2})(q_xe_x-q_ye_y)\\
&\quad+(G_{44}/6^{1/2})(-q_ze_y-q_ye_z-q_ze_x-q_xe_z),\\
C_{\bar{y}}(a)&=\tfrac{3}{2}G_{11}\eta(2^{1/2}/9^{1/2})(2q_ze_z-q_xe_x-q_ye_y)\\
&\quad+[G_{44}/(18)^{1/2}][2\eta(q_ye_x+q_xe_y)\\
&\quad+(q_ze_y+q_ye_z-q_ze_x-q_xe_z)]
\end{aligned} \tag{3.4b}$$

for case B. In these equations, e_i and q_i refer to the phonon polarization and wave vector along the cubic x, y, and z axes of the crystal, and $S_x(a)$ and $S_z(a)$ refer

[6] See Ref. 1 and references therein.

[7] D. T. Teaney, M. J. Freiser, and R. W. H. Stevenson, Phys. Rev. Letters **9**, 212 (1962); M. J. Freiser, P. E. Seiden, and D. T. Teaney, *ibid.* **7**, 293 (1963); A. J. Heeger and D. T. Teaney, J. Appl. Phys. **35**, 846 (1964).

[8] This differs, however, from notation used in Ref. 7.

TABLE I. Coefficients C_x^2 and C_y^2 to be used for cases A and B in Eq. (3.5). The vector $\mathbf{q}$ is the phonon wave vector and $\mathbf{e}$ is the polarization vector of the mode.

Elastic Mode	C_x^2 for case A	C_y^2 for case A	C_x^2 for case B	C_y^2 for case B
$\mathbf{q}\|\|\mathbf{e}\|\|[001]$	$9G_{11}^2\sin^2\theta\cos^2\theta$	0	0	$2G_{11}^2$
$\mathbf{q}\|\|\mathbf{e}\|\|[100]$	$(9/4)G_{11}^2\sin^2\theta\cos^2\theta$	$(9/4)G_{11}^2\sin^2\theta$	$\frac{3}{2}G_{11}^2$	$\frac{1}{2}G_{11}^2$
$\mathbf{q}\|\|\mathbf{e}\|\|[110]$	$(\frac{3}{2}G_{11}+G_{44})^2\sin^2\theta\cos^2\theta$	0	0	$(2/9)(G_{11}-G_{44})^2$
$\mathbf{q}\|\|\mathbf{e}\|\|[1\bar{1}0]$	$\frac{3}{2}(G_{11}-G_{44})^2\sin^2\theta\cos^2\theta$	0	0	$(2/9)(\frac{3}{2}G_{11}+G_{44})^2$
$\mathbf{q}\|\|[110],\mathbf{e}\|\|[1\bar{1}0]$	0	$(9/4)G_{11}^2\sin^2\theta$	$\frac{3}{2}G_{11}^2$	0
$\mathbf{q}\|\|[110],\mathbf{e}\|\|[001]$	$G_{44}^2(\cos^2\theta-\sin^2\theta)^2$	0	$\frac{1}{3}G_{44}^2$	0

to the components of sublattice spin in the coordinate systems given by Eqs. (2.3) and (2.6). Since these equations are somewhat ponderous, the results are tabulated in Table I in the form

$$\omega^2=\omega_0^2(\mathbf{q}\lambda)+4M^{-1}q^2S_0 \times[C_x^2D_{\bar{x}\bar{x}}(aa,\mathbf{q},\omega)+C_y^2D_{\bar{y}\bar{y}}(aa,\mathbf{q},\omega)] \quad (3.5)$$

for a few elastic modes. For acoustic phonons in $RbMnF_3$ and frequencies less than or of the order of $\omega_{n\pm}$ (less than 10^{10} sec^{-1}), the q dependence of the D's is negligible and is henceforth neglected.

If $D_{ij}(aa',0,\omega)$ in Eq. (3.3) is approximated by its zero frequency limit, our results are equivalent to the theoretically results in M&B[3] and in M[9] for case A. Their results were derived by neglecting the dynamic coupling between the electronic and nuclear spins (the $\bar{D}$ term in Eq. (A9) which is correct (except for very small correction terms) for $\omega=0$.

In spite of the fact that our results are essentially equivalent in case A, a couple of points are worth mentioning. First, when $\mathbf{H}_0$ is parallel to [001], $\cos\phi=\cos\theta$ approaches zero sharply but continuously as H_0 approaches the spin-flop field, $H_c=(3H_EH_A)^{1/2}$, from below. Since ω_{n+} is equal to zero when $H=H_c$, the zero-frequency limit is not appropriate near this point. Because of this, the fact that higher-order terms have been neglected, and the fact that a small error in ψ can have a large effect, the equations are somewhat unreliable for $\psi=0$ in small neighborhood of H_0 near H_c. Second, the theory and experiments agree only for angles ψ up to about 40° instead of $\arcsin\frac{2}{3}\approx 54.7°$. The reason for this is presumably that at least part of the sublattice magnetization is somewhat different from what has been used here.

Our case B is in agreement with the experimental findings of M but is in disagreement with his theoretical findings. The reason for this is almost certainly an error in the equilibrium magnetization used in M.

In addition to being in essential agreement with low-frequency ($\omega\ll\omega_{n\pm}$) measurements, Eq. (3.3) can also be checked against acoustic measurements at ω near ω_n[10,11] or $\omega_{e\pm}$. Since the theory used includes no mechanism for damping or line broadening, the phonon absorption derived from the imaginary part of Eq. (3.3) is a sum of δ functions. This is clearly unrealistic and may be remedied phenomenologically by replacing $\omega+i\tau^{-1}$, where τ is a phenomenological relaxation time. On the other hand, the change in phonon velocity near, but not on, resonance is relatively insensitive to τ if $\omega\tau\gg 1$.

Preliminary investigations[12] indicate that the changes in the phonon velocity near the nuclear resonance frequencies predicted by Eq. (3.3) are correct to within experimental error. A value of $b_1\approx 2.5\times 10^6$ erg/cm and the values used by Ince[1] for the quantities in Eqs. (1.5) where used in this comparison. These values of the parameters in Eq. (3.3) also give good agreement with the work of M&B and M. The reason for the difference in the value for b_1 used here and in the value used in M&B is that we have used Ince's value for H_A and they used a somewhat lower value. Except for H_0 near H_c, as discussed above, their measurements[13] depend only on the ratio b_1^2/H_A.

Finally, a brief summary of the approximations used in Appendix B to obtain Eq. (3.3) from the electronic-spin correlation functions is in order. These approximations are essentially the ones used by Bennett and Pytte[14] in a higher-temperature regime. First, only the lowest-order perturbation theory correction to the phonons through the magneto-elastic coupling is used. In view of the smallness of the coupling involved, this should be a good approximation. Next, a four-spin correlation function is approximated by the sum of all possible factorizations in terms of second-order (or less) correlation functions. This is the most shaky approximation used in the Appendix. Finally, only those terms that involved only one time-dependent correlation function were kept in obtaining Eq. (3.3). Clearly, only those terms contribute significantly for frequencies near $\omega_{e\pm}$ and $\omega_{n\pm}$. The contribution from the remaining terms at low frequencies and at the sums and differences

[9] R. L. Melcher, Ph.D. thesis, Washington University, 1968 (unpublished). Hereafter referred to as M.

[10] The nuclear acoustic resonance for Mn in $RbMnF_3$ has been reported by J. B. Merry and D. I. Bolef, Phys. Rev. Letters **23**, 126 (1969).

[11] The mechanism coupling the nuclear spins and phonons used here is essentially that used for a uniaxial antiferromagnet by S. D. Silverstein, Phys. Rev. **132**, 997 (1963).

[12] J. B. Merry and D. I. Bolef (private communication).

[13] The differences in the values for the anisotropy field is presumably due to differences in the crystals.

[14] H. S. Bennett and E. Pytte, Phys. Rev. **155**, 553 (1967).

of $\omega_{n+}(\mathbf{q})$, $\omega_{n-}(\mathbf{q})$, $\omega_{e+}(\mathbf{q})$, and $\omega_{e-}(\mathbf{q})$ are now under investigation.

APPENDIX A

In this Appendix, the equations of motion for the electronic-spin correlations are obtained and solved. Units where $\hbar=1$ are used, but $\hbar$ is reinserted when the formulas are used in Sec. II.

The contribution to $(\partial S_i/\partial t)$ from the anisotropy energy is treated semiclassically by quantizing the torque term

$$\left(\frac{\partial S_i}{\partial t}\right)_a=-\delta\left[S\frac{\partial E_a}{\partial S}\right]_i, \tag{A1}$$

where the Latin subscripts denote Cartesian directions. The operation δ denotes that S goes into $S+\delta S$ and that only first-order terms in δS are kept. The result is conveniently written in the form of a magnetic field contribution as

$$\left(\frac{\partial S_i}{\partial t}\right)_a=-\sum_{j,k}\epsilon_{ijk}\mu_e H_{ji}'\delta S_k, \tag{A2}$$

where ϵ_{ijk} is the usual antisymmetric tensor. The term is quantized by replacing δS with the operator S and replacing the rest of the S's by their thermal averages. By including the rest of the Hamiltonian given by Eq. (1.1), one finds that the operator equations of motion for S and I are

$$\begin{aligned}i\frac{\partial S_i(\alpha)}{\partial t}=&i\sum_{j,k}\epsilon_{ijk}\{-\mu_e[H_{0j}+H_{ji}'(\alpha)]S_k(\alpha)\\&+\sum_{\bar\alpha}J(\alpha,\bar\alpha)S_j(\alpha)S_k(\bar\alpha)-AI_j(\alpha)S_k(\alpha)\},\end{aligned} \tag{A3}$$

$$i\frac{\partial I_i(\alpha)}{\partial t}=i\sum_{j,k}\epsilon_{ijk}[-\mu_n H_{0j}I_k(\alpha)-AS_j(\alpha)I_k(\alpha)],$$

where H_{0j} is the jth component of $\mathbf{H}_0$.

In order to obtain the electronic-spin correlation functions defined by Eq. (1.3), the following definitions are made:

$$\begin{aligned}D_{ij}(\alpha t,\alpha' t')&=-i\langle T(\tilde S_i(\alpha t)\tilde S_j(\alpha' t'))\rangle,\\ \bar D_{ij}(\alpha t,\alpha' t')&=-i\langle T(\tilde I_i(\alpha t)\tilde S_j(\alpha' t'))\rangle,\end{aligned} \tag{A4}$$

where T denotes that the bracketed operators are time ordered, $\tilde S$ is defined in Eq. (2.1), and $\tilde I$ is $I-\langle I\rangle$. In the RPA, expectation values of a product of three operators are approximated as follows:

$$\begin{aligned}\langle A\ B\ C\rangle=&\langle A\ B\rangle\langle C\rangle+\langle A\ C\rangle\langle B\rangle\\&+\langle B\ C\rangle\langle A\rangle+\langle A\rangle\langle B\rangle\langle C\rangle.\end{aligned}$$

One thus obtains

$$\begin{aligned}i\frac{\partial D_{il}(\alpha t,\alpha' t')}{\partial t}&+i\sum_{j,k}\epsilon_{ijk}\{\mu_e[H_{0j}+H_{ji}'(\alpha)]D_{kl}(\alpha t,\alpha' t')\\&+\sum_{\bar\alpha}J(\alpha,\bar\alpha)[S_j(\bar\alpha)D_{kl}(\alpha t,\alpha' t')-S_j(\alpha)D_{kl}(\bar\alpha t,\alpha' t')]\\&+A[I_j(\alpha)D_{kl}(\alpha t,\alpha' t')-S_j(\alpha)\bar D_{kl}(\alpha t,\alpha' t')]\}\\&=i\sum_j\epsilon_{ilj}S_j(\alpha)\delta(\alpha,\alpha')\delta(t-t')\end{aligned} \tag{A5}$$

and

$$\begin{aligned}i\frac{\partial\bar D_{il}(\alpha t,\alpha' t')}{\partial t}&+i\sum_{j,k}\epsilon_{ijk}\{[\mu_n H_{0j}+AS_j(\alpha)]\bar D_{kl}(\alpha t,\alpha' t')\\&-I_j(\alpha)D_{kl}(\alpha t,\alpha' t')\}=0.\end{aligned} \tag{A6}$$

It is convenient to Fourier-transform these equations in both space and time. The time transform is the usual one employed with thermodynamic Green's functions:

$$D(\omega_\nu)=\int D(t)e^{i\omega_\nu t}dt, \tag{A7}$$

where ω_ν is $(\pi\nu/-i\beta)$ and the integral runs from 0 to $-i\beta$. Here, β is $1/kT$, and ν is an even integer. The retarded Green's function may be obtained by the prescription

$$D(\omega)=\lim_{\epsilon\to0^+}D(\omega_\nu=\omega+i\epsilon). \tag{A8}$$

Using this and the spatial transform defined by Eq. (2.2), one obtains the equations

$$\begin{aligned}\omega D_{il}(aa',\mathbf{q},\omega)&+i\sum_{j,k}\epsilon_{ijk}[\bar H_{ji}(a,\mathbf{q})D_{kl}(aa',\mathbf{q},\omega)\\&+J(a\tilde a,\mathbf{q})S_j(a)D_{kl}(\tilde a a',\mathbf{q},\omega)-AS^j(a)\bar D_{kl}(aa',\mathbf{q},\omega)]\\&=i\sum_j\epsilon_{ilj}S_j(a)\delta(a,a'),\end{aligned} \tag{A9}$$

$$\begin{aligned}\omega\bar D_{il}(aa',\mathbf{q},\omega)&+i\sum_{j,k}\epsilon_{ijk}[B_j(a)\bar D_{kl}(aa',\mathbf{q},\omega)\\&-AI_j(a)D_{kl}(aa',\mathbf{q},\omega)]=0,\end{aligned} \tag{A10}$$

where $\tilde a$ is 1 (or 2) if a is 2 (or 1), $\mathbf{S}(a)$ and $\mathbf{I}(a)$ are the thermal averages of the electronic and nuclear spins on the sublattice a, and

$$\begin{aligned}\bar H_{ji}(a,\mathbf{q})=&\mu_e[H_{ji}'(a)+H_{0j}]-J(a\tilde a,0)S_j(a)\\&-[J(aa,0)-J(aa,\mathbf{q})]S_j(a)+AI_j(a),\end{aligned} \tag{A11}$$

$$B_j(a)=\mu_n H_{0j}+AS_j(a). \tag{A12}$$

It is convenient to first solve these equation by neglecting the $\bar D$ term in Eq. (A9). Using the easily verified facts that

$$\sum_i S_i(a)D_{ij}(aa')=\sum_j D_{ij}(aa')S_j(a')=0$$

and that $S_y(a)$ and $\bar{H}_{jy}(a)$ vanish in the coordinate systems defined by Eq. (2.3) for case A and by Eqs. (2.6) for case B, one can straightforwardly obtain the solution $D_{ij}(aa',\mathbf{q},\omega)$. The solution is most conveniently expressed in terms of the spherical components

$$D_{\mu\nu}(aa')=D_{\tilde{x}\tilde{x}}(aa')-\mu\nu D_{\tilde{y}\tilde{y}}(aa') +in D_{\tilde{y}\tilde{x}}(aa')-i\nu D_{\tilde{x}\tilde{y}}(aa'), \quad \text{(A13)}$$

where the $\mathbf{q}$ and ω dependence of D has been suppressed and $D_{\tilde{i}\tilde{j}}(aa')$ is a shorthand notation for

$$D_{\tilde{i}\tilde{j}}(aa')=D_{\tilde{i}(a),\tilde{j}(a')}(aa'). \quad \text{(A14)}$$

The indexes μ and ν take on the values $+1$ and -1.

After some straightforward algebra, one obtains the following formulas:

$$\begin{aligned}
D_{\mu\nu}(aa)=&\mathfrak{D}^{-1}\{-\nu S(a)\mathfrak{D}/\omega+[\mu\omega S(a)+\mathfrak{K}_{\tilde{y}}(a)\cdot\mathbf{S}(a)]\\
&\times[C_1(\tilde{a})(\omega S(a)-\nu\mathfrak{K}_y(a)\cdot\mathbf{S}(a))\\
&-C_2(a)\nu J(a\tilde{a},\mathbf{q})\mathbf{S}(a)\cdot\mathbf{S}(\tilde{a})]/S(a)\omega\\
&+[J(a\tilde{a},\mathbf{q})S(a)/\omega]\\
&\times[C_2(\tilde{a})(\omega S(a)-\nu\mathfrak{K}_y(a)\cdot\mathbf{S}(a))\\
&-C_1(a)\nu J(\tilde{a}a,q)\mathbf{S}(a)\cdot\mathbf{S}(\tilde{a})]\},\\
D_{\mu\nu}(\tilde{a}a)=&\mathfrak{D}^{-1}\{[\mu\omega S(\tilde{a})+\mathfrak{K}_{\tilde{y}}(\tilde{a})\cdot\mathbf{S}(\tilde{a})]\\
&\times[C_2(\tilde{a})(\omega S(a)-\nu\mathfrak{K}_y(a)\cdot\mathbf{S}(a))\\
&-\nu C_1(a)J(\tilde{a}a,\mathbf{q})\mathbf{S}(a)\cdot\mathbf{S}(\tilde{a})]/\omega S(\tilde{a})\\
&+[J(\tilde{a}a,\mathbf{q})S(\tilde{a})/\omega]\\
&\times[C_1(\tilde{a})(\omega S(a)-\nu\mathfrak{K}_y(a)\cdot\mathbf{S}(a)\\
&-\nu C_2(a)J(\tilde{a}a,\mathbf{q})\mathbf{S}(a)\cdot\mathbf{S}(\tilde{a}))],
\end{aligned} \quad \text{(A15)}$$

where

$$\begin{aligned}
&\mathfrak{D}=C_1(1)C_1(2)-C_2(1)C_2(2),\\
&C_1(a)=\omega^2-\mathfrak{K}_y(a)\cdot\mathfrak{K}_{\tilde{y}}(a)-J(12,\mathbf{q})J(21,\mathbf{q})\mathbf{S}(1)\cdot\mathbf{S}(2),\\
&C_2(a)=J(a\tilde{a},\mathbf{q})\mathbf{S}(a)\cdot(\mathfrak{K}_{\tilde{y}}(\tilde{a})+\mathfrak{K}_y(a)),\\
&\mathfrak{K}_y(a)=(H_{xy}(a),0,H_{zy}(a)),\\
&\mathfrak{K}_{\tilde{y}}(a)=(H_{xz}(a),0,H_{zx}(a)).
\end{aligned} \quad \text{(A16)}$$

In order to obtain the electronic-spin correlation function for the system of couple electronic and nuclear spins, the coupled equations (A9) and (A10) must be solved. Towards this end, the further restriction that $|S(1)|$ equal $|S(2)|$ is imposed. Further, it is assumed that H_0 can be neglected with respect to $AS(a)$ in Eq. (A12), so that $\mathbf{S}(a)$ and $\mathbf{I}(a)$ are parallel. These restrictions are discussed in Sec. III.

In contrast to Eq. (A9), Eq. (A10) is a vector equation where $\bar{D}_{il}$ is considered as a vector in the index i. Schematically the equation reads

$$\omega\bar{\mathbf{D}}+i\mathbf{B}\times\bar{\mathbf{D}}-iA\mathbf{I}\times\bar{\mathbf{D}}=0.$$

Using the fact that $\mathbf{S}\cdot\mathbf{D}$ and $\mathbf{S}\cdot\bar{\mathbf{D}}$ are zero, one easily obtains the equation

$$\mathbf{S}\times\bar{\mathbf{D}}=A[-\mathbf{S}\times\mathbf{D}(\mathbf{B}\cdot\mathbf{I})-i\omega\mathbf{D}(\mathbf{S}\cdot\mathbf{I})]/(\omega^2-B^2).$$

Using this in Eq. (A10), it is easily seen that by making the substitutions

$$\begin{aligned}
&\omega\rightarrow\omega(1-A^2IS/(\omega^2-B^2)),\\
&\mathfrak{K}_{ji}(a)\rightarrow\mathfrak{K}_{ji}(a)+A^2BIS_j(a)/(\omega^2-B^2)
\end{aligned} \quad \text{(A17)}$$

that Eqs. (A15) and (A16) are again valid.

Using the prescription found in Eqs. (A1) and (A2) nonzero components of the "anisotropy fields" H' in case A are easily found to be

$$\begin{aligned}
&H_{zx}'(1)=H_{zy}'(1)=KS_1^3\cos\theta_-(5\sin^2\theta_--2),\\
&H_{xy}'(1)=KS_1^3\sin\theta_-(6-7\sin^2\theta_-),\\
&H_{xz}'(1)=2KS_1^3\sin^3\theta_-,\\
&\theta_-=\theta-t,
\end{aligned} \quad \text{(A18)}$$

for the sublattice 1. Those for the sublattice 2 are obtained by letting S_1 go into $-S_2$ and $\theta-t$ into $\theta+t$. In what follows, it will be assumed that $\omega_E\omega_A$ and $\omega_E\omega_{NE}$ are of the same order of magnitude and that ω_0^2 is, at most, of this order of magnitude. Further, ω_E is much greater than ω_0, ω_A or ω_{NE}. These assumption are valid for $RbMnF_3$ for magnetic fields of the order of a kG or less.

The electronic-spin wave-resonant frequencies are obtained by finding the zeroes of the function given in Eqs. (A16). With the definitions

$$\begin{aligned}
&C_1\binom{1}{2}=\omega^2-(L_1\pm L_2)_1,\\
&C_2\binom{1}{2}=L_3\pm L_4,
\end{aligned} \quad \text{(A19)}$$

$\mathfrak{D}$ can be written as

$$\begin{aligned}
&\mathfrak{D}=(\omega^2-\omega_{e+}^2)(\omega^2-\omega_{e-}^2),\\
&\omega_{e+}^2=L_1\pm[L_3^2+(L_4-L_2)(L_4+L_2)]^{1/2}.
\end{aligned} \quad \text{(A20)}$$

Neglecting terms of order ω_N/ω_E compared to 1, one obtains

$$\begin{aligned}
L_1=&\omega_0^2(1-\tfrac{1}{2}\sin^2\phi)-\omega_0^2\cos^2\phi[1-\tfrac{1}{2}\alpha]\alpha+2\omega_E\omega_{NE}\\
&+\tfrac{3}{2}\omega_E\omega_A[f(\theta)+g(\theta)]+2\omega_E^2Q(\mathbf{q}),\\
L_2+L_4=&4\omega_0\omega_E\cos\phi[1-\tfrac{1}{2}\alpha],\\
L_3=&\tfrac{1}{2}\omega_0^2\sin^2\phi+\omega_0^2\cos^2\phi\,[1-\tfrac{1}{2}\alpha]\alpha\\
&+\tfrac{3}{2}\omega_E\omega_A[f(\theta)-g(\theta)],\\
L_4-L_2=&\omega_0\cos\phi\{2\omega_{NE}(1-\tfrac{1}{2}\alpha)+2\omega_E(1-\tfrac{1}{2}\alpha)Q(\mathbf{q})\\
&+\tfrac{3}{2}\omega_A[f(\theta)+g(\theta)]+\tfrac{3}{8}\omega_A\alpha\\
&\times[4-26\sin^2\theta+24\sin^4\theta]\}+\tfrac{3}{8}\omega_A\omega_0\\
&\times\cos\theta\sin\phi\sin\theta[-4+6\sin^2\theta],
\end{aligned} \quad \text{(A21)}$$

where

$$\begin{aligned}
&f(\theta)=-\tfrac{1}{2}(2-13\sin^2\theta+12\sin^4\theta),\\
&g(\theta)=-\tfrac{1}{2}(2-7\sin^2\theta+3\sin^4\theta),\\
&Q(\mathbf{q})=[(J^2(12,0)-J(12,\mathbf{q})J(21,\mathbf{q}))-2J(12,0)\\
&\qquad\times(J(11,0)-J(11,\mathbf{q}))]/J^2(12,0).
\end{aligned} \quad \text{(A22)}$$

These equations are valid for values of q such that $(qa)^2$ is less than or of order ω_A/ω_E and $(\omega_0/\omega_E)^2$.

The effect of the nuclear modes on the electronic-spin correlation functions is evaluated only when χ_{11} can be neglected. This restricts the treatment to either low temperatures or magnetic fields many times $(H_E H_A)^2$. Under these conditions and the assumption that ω_N^2 is much less than $\omega_A\omega_E$, the nuclear modes are obtained from Eqs. (A18) and (A19) yielding

$$\omega_{n\pm}{}^2=\omega_N{}^2(1-2\omega_E\omega_{NE}/\omega_{e\pm}{}^2)\,. \tag{A23}$$

As well as yielding the normal modes of the system, the spin-correlation approach yields the spectral eight of the modes. As a function of $\mathbf{q}$ and ω, one obtains Eqs. (3.1) and (3.2).

The case where ψ is $\frac{1}{2}\pi$, case B, is also given by Eqs. (3.1) and (3.3) in the text with $\phi=\theta=\frac{1}{2}\pi$.

APPENDIX B

In this Appendix, equations are derived describing the effect of the antiferromagnetism on the phonons. Using the magneto-elastic Hamiltonian given by Eq. (1.3), one can derive the change in the phonon dispersion relation due to the electronic spin to lowest order in the magneto-elastic coupling[14]

$$\omega^2=\omega_0{}^2(\mathbf{q}\lambda)+(NM)^{-1}\sum e_i(\mathbf{q}\lambda)e_j(\mathbf{q}\lambda)G_{il}q_lG_{jm}q_m \times(-i)(-i\beta)^{-1}\int dt\,\langle T(F_{il}(\mathbf{l}\partial t)F_{jm}(\mathfrak{g}'a't'))\rangle \times\exp[i\beta\omega_\nu(t-t')-i\mathbf{q}\cdot(\mathbf{l}-\mathbf{l}')]\,, \tag{B1}$$

where the $e_i(\mathbf{q}\lambda)$ are the polarization vectors of the phonon modes, M is the mass of a magnetic ion, and the first summation is over $\mathbf{q}$, a, a', $\mathbf{l}$, $\mathbf{l}'$, i, j, l, and m. The nonzero G's are

$$\begin{aligned}&G_{33}=G_{22}=G_{11}\,,\\&G_{12}=G_{23}=G_{31}=G_{21}=G_{32}=G_{13}=G_{44}\,,\end{aligned} \tag{B2}$$

and the nonzero F's are

$$\begin{aligned}&F_{11}=S_x{}^2-\tfrac{1}{2}(S_y{}^2+S_z{}^2)\quad\text{and cyclic permutations,}\\&F_{12}=F_{21}=\tfrac{1}{2}(S_xS_y+S_yS_x)\quad\text{and cyclic permutations.}\end{aligned} \tag{B3}$$

The x, y, z here correspond to the cubic axes.

The approximation use here for the average of four spin operators is the sum of all possible factorizations in terms of lower-order correlation functions. Terms index of t do not contribute, and one is left with

$$\begin{aligned}\langle T(S_i(1)S_j(1)S_k(2)S_p(2))\rangle\rightarrow{}&(i)^2[D_{ik}(12)D_{jl}(12)\\&+D_{il}(12)D_{jk}(12)]+(i)[S_j(1)S_l(2)D_{ik}(12)\\&+S_j(1)S_k(2)D_{il}(12)+S_i(1)S_l(2)D_{jk}(12)\\&+S_i(1)S_k(2)D_{jl}(12)]\,,\\D_{ik}(12)\equiv{}&D_{ik}(\alpha_1l_1,\alpha_2l_2)\,.\end{aligned} \tag{B4}$$

In this paper, the terms in the first set of brackets are neglected. They do not contribute significantly to the phonon behavior near the spin wave-resonant frequencies.

After some lengthy but straightforward algebra, one arrives at Eqs. (3.3) and (3.4).