

Simultaneous Parallel Pumping of Nuclear and Electronic Spin Waves*

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In a magnetic medium in which the nuclei carry a magnetic moment, there exists a spectrum of nuclear spin waves in addition to the usual electronic spin waves, provided the temperature is sufficiently low. The possibility is investigated of joint excitation of nuclear and electronic magnons by parallel pumping; this process is analogous to "exchange pumping" of acoustic and exchange magnons in a ferrimagnet. First a simple ferromagnet is considered, then the ferrimagnet manganese ferrite, and finally the cubic antiferromagnet RbMnF_3 . It is suggested that the process should be feasible, and that it should be possible thereby to excite nuclear spin waves of arbitrary k . (In ordinary NMR only $k=0$ is excited.) The threshold pump field in a typical material is estimated to be of order 36 Oe in magnitude, but it can be smaller in carefully prepared samples.

I. INTRODUCTION

IT has been shown by de Gennes *et al.*¹ that, because of hyperfine interaction, a simple ferromagnet in which the magnetic ions also carry a nuclear moment has two spin-wave branches at sufficiently low temperatures. The upper branch relates essentially to the electronic system and deviates slightly from the "usual" electronic spin-wave spectrum. The lower branch relates essentially to the nuclear system but deviates to a relatively greater extent from the "usual" NMR frequency. These branches will be referred to as electronic and nuclear spin-wave branches, respectively.

In the same paper, DPHW¹ also considered the possibility of parallel pumping the nuclear spin waves, i.e., excitation by application of an oscillating magnetic field parallel to the direction of magnetization. The process envisaged was the absorption of one photon and the consequent creation of *two nuclear magnons*. They came to the conclusion that the condition for instability of the nuclear system would be difficult to satisfy. This, however, does not rule out parallel pumping, for there still remains the possibility of simultaneous excitation of nuclear and electronic spin waves: each photon would give rise to *one nuclear and one electronic magnon*. This process is analogous to "exchange pumping" in the two sublattice ferrimagnet with unequal g factors.^{2,3}

We first consider a simple ferromagnet and present an elementary analysis of the $k=0$ case (Sec. II). Then the nuclear and electronic spectra will be derived using the method of Holstein and Primakoff,⁴ and the threshold field worked out for parallel pumping insta-

bility using the method of transition probabilities outlined by Callen.⁵ In the present treatment, dipole-dipole interactions are ignored. This will mean that, with the pumping field on, the total angular momentum along the field is a constant of the motion. Thus if magnons are to be excited jointly from the nuclear and electronic branches, in such a way that their numbers increase exponentially with time, the nuclear and electronic spins will have to point in opposite directions, so that any gain in angular momentum by one mode is exactly balanced by loss in the other. We are therefore restricted to hyperfine interactions $A \sum_i \mathbf{L}_i \cdot \mathbf{S}_i$ in which A is positive. In most magnetic materials, however, A is negative.

In Sec. III, we outline a corresponding theory for manganese ferrite. Its unit cell consists of four Fe^{3+} ions and two Mn^{2+} ions, each with electron spin $S=\frac{5}{2}$, but with all Fe^{3+} pointing in opposite direction⁶ to the Mn^{2+} . In addition, NMR data by Heeger and Houston⁷ show that the nuclear angular momentum of the Mn, which also has $I=\frac{5}{2}$, is parallel to its electronic spin. Thus $A < 0$. However, any change in the angular momentum of the electron and nuclear spins of the Mn can be offset by a change in the Fe^{3+} electronic angular momentum, so that angular momentum is conserved. Of the eight spin-wave branches present here, two are nuclear branches of which only one is appreciably depressed by the hyperfine interaction. It will be shown that, as far as the pumping field is concerned, two of the branches can be ignored, while the other six can be divided into two equal lots—not connected by the rf field—with each containing a nuclear mode. The calculation shows that joint excitation of nuclear and electronic magnons within each of these two lots is theoretically possible.

Finally in Sec. IV, we consider RbMnF_3 , which is a cubic antiferromagnetic. This has low anisotropy, so that the electronic frequencies (in the absence of a

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¹ P. G. deGennes, P. A. Pincus, F. Hartmann-Boutron, and J. M. Winter, *Phys. Rev.* **129**, 1105 (1963). These authors will be referred to as DPHW.

² F. R. Morgenthaler, *Phys. Rev. Letters* **11**, 69 (1963). Also see: E. Schlomann, in *Proceedings of the International Conference on Solid State Physics in Electronics and Telecommunications, Brussels, 1958* (Academic Press Inc., New York, 1960), Vol. 3.

³ F. R. Morgenthaler, *J. Appl. Phys.* **36**, 3102 (1965).

⁴ T. Holstein and H. Primakoff, *Phys. Rev.* **58**, 1098 (1940).

⁵ H. Callen, in *Fluctuation, Relaxation and Resonance in Magnetic Systems*, edited by D. ter Haar (Oliver and Boyd Ltd., Edinburgh, 1962).

⁶ J. M. Hastings and L. M. Corliss, *Phys. Rev.* **104**, 328 (1956).

⁷ A. J. Heeger and T. W. Houston, *Phys. Rev.* **135**, A661 (1964).

field) are comparatively low—a condition favorable to the pumping process.⁸

II. FERROMAGNET

A. Simple Derivation, $\mathbf{k}=0$ Modes

We consider a system with Hamiltonian

$$\mathcal{H} = - \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \gamma_e \hbar H \sum_i S_i^z - \gamma_n \hbar H \sum_i I_i^z + A \sum_i \mathbf{I}_i \cdot \mathbf{S}_i, \quad (1)$$

where the terms, in order, represent the exchange energy between electronic spins, the electronic and the nuclear Zeeman energies, and the electron-nucleus hyperfine coupling. The magnetic field H is in the direction of the positive z axis, and γ_e is the (intrinsically negative) electronic magnetogyric ratio. In general the nuclear magnetogyric ratio γ_n and the hyperfine coupling constant A may each be either positive or negative; but as mentioned earlier and as explicit calculation shows, A must be positive for parallel pumping to be feasible. This is discussed in Appendix D using the formalism of Sec. II B below.

For the present case the $\mathbf{k}=0$ modes may be obtained from the two sublattice approximation

$$(\mathcal{H}/V) \simeq \lambda \mathbf{M}_e \cdot \mathbf{M}_e - H M_e^z - H M_n^z + (AV/N\hbar^2\gamma_e\gamma_n) \mathbf{M}_e \cdot \mathbf{M}_n, \quad (2)$$

where

$$\mathbf{M}_e = \gamma_e \hbar \sum_i \mathbf{S}_i / V, \quad \mathbf{M}_n = \gamma_n \hbar \sum_i \mathbf{I}_i / V \quad (3)$$

are the magnetizations, respectively, of the electronic and nuclear sublattices, λ is a molecular-field parameter, and N is the number of electronic or nuclear spins. The step from (1) to (2) involves the assumption that all \mathbf{I}_i and \mathbf{S}_i are independent of i , and that therefore the spin systems are translationally invariant. This correctly yields the $\mathbf{k}=0$ modes.

The Hamiltonian (2) gives rise to the equations of motion

$$d\mathbf{M}_e/dt = \gamma_e \mathbf{M}_e \times [H\hat{z} - (AV/N\hbar^2\gamma_e\gamma_n)\mathbf{M}_n], \quad (4a)$$

$$d\mathbf{M}_n/dt = \gamma_n \mathbf{M}_n \times [H\hat{z} - (AV/N\hbar^2\gamma_e\gamma_n)\mathbf{M}_e], \quad (4b)$$

with \hat{z} a unit vector along the positive z axis.

These equations may be written in the form

$$\begin{aligned} \dot{M}_e^\pm &= \mp i\gamma_e \\ &\times [M_e^\pm H - (AV/N\hbar^2\gamma_e\gamma_n)(M_e^\pm M_n^z - M_e^z M_n^\pm)], \end{aligned} \quad (5a)$$

$$\begin{aligned} \dot{M}_n^\pm &= \mp i\gamma_n \\ &\times [M_n^\pm H - (AV/N\hbar^2\gamma_e\gamma_n)(M_n^\pm M_e^z - M_n^z M_e^\pm)], \end{aligned} \quad (5b)$$

$$\begin{aligned} \dot{M}^z &= (\gamma_n - \gamma_e)(AV/N\hbar^2\gamma_e\gamma_n)(i/2) \\ &\times (M_e^+ M_n^- - M_e^- M_n^+). \end{aligned} \quad (5c)$$

Here $M^\pm = M^x \pm iM^y$, and M^z is the total magnetization along positive z , i.e., $M_e^z + M_n^z$. For small oscillations, both M_e^z and M_n^z will not change very much and we may, to a first approximation, consider these quantities constant when solving the equations in M^\pm . This is the *linearization approximation*, discussed below. We thus set, in Eqs. (5a) and (5b),

$$M_e^z = \gamma_e \hbar (N/V) \langle S^z \rangle; \quad M_n^z = \gamma_n \hbar (N/V) \langle I^z \rangle. \quad (6)$$

It is precisely the variation of M^z , however, which permits parallel pumping. Therefore it is assumed that this variation is both small enough not measurably to effect the M^\pm equations of motion, and yet large enough to allow a z -directed rf field to couple to the system.

With the above assumption, and with M_e^+ and M_n^+ taken proportional to $\exp(i\omega t)$, the secular equation becomes

$$\begin{vmatrix} \omega + \gamma_e [H - (A/\hbar\gamma_e) \langle I^z \rangle] & (A\gamma_e/\hbar\gamma_n) \langle S^z \rangle \\ (A\gamma_n/\hbar\gamma_e) \langle I^z \rangle & \omega + \gamma_n [H - (A/\hbar\gamma_n) \langle S^z \rangle] \end{vmatrix} = 0. \quad (7)$$

If the off-diagonal elements are treated as perturbations, the eigenfrequencies are

$$\omega_{0\alpha} = \omega_e + \frac{(A^2/\hbar^2) \langle I^z \rangle \langle S^z \rangle}{\omega_e - \omega_n}, \quad (8)$$

$$\omega_{0\beta} = \omega_n + \frac{(A^2/\hbar^2) \langle I^z \rangle \langle S^z \rangle}{\omega_n - \omega_e}, \quad (9)$$

where

$$\omega_e = -\gamma_e H + (A/\hbar) \langle I^z \rangle, \quad (10)$$

$$\omega_n = -\gamma_n H + (A/\hbar) \langle S^z \rangle, \quad (11)$$

are the unperturbed electronic and nuclear frequencies, respectively. In comparing these equations with the more general ones of the next section it is helpful to note that there $\langle S^z \rangle$ is taken as $-S$, and that the energy $\epsilon_{0\beta}$ is equal to $-\hbar\omega_{0\beta}$.

The above approximation is valid provided, of course, that the perturbation is small compared to $|\omega_e - \omega_n|$.

⁸ Such pumping in RbMnF_3 in the spin-flopped state has recently been observed experimentally by L. W. Hinderks and P. M. Richards, J. Appl. Phys. (to be published).

Under these conditions, the resonance (8) comprises a relatively small amplitude precession of the nuclear sublattice superposed onto a much larger amplitude precession of the electronic sublattice, whereas in the resonance (9) the nuclear sublattice has the larger amplitude of precession. In either resonance, as seen from inspection of Eq. (5c), \dot{M}^z is zero.

If, however, both modes of resonance are simultaneously present, a nonzero value of \dot{M}^z is achieved. In this case M_e^\pm is essentially proportional to $\exp(\pm i\omega_{0\alpha}t)$ and M_n^\pm is essentially proportional to $\exp(\pm i\omega_{0\beta}t)$ and

$$\dot{M}^z = (N/V)A(\gamma_e - \gamma_n)\langle S^\pm \rangle \langle I^\pm \rangle \sin(\omega_{0\alpha} - \omega_{0\beta})t. \quad (12)$$

Here $\langle S^\pm \rangle$ and $\langle I^\pm \rangle$ are average components normal to the z direction and represent the precessional amplitudes.

The conversion to spin-wave language is achieved by

$$n_{0\alpha} = N(S + \langle S^z \rangle), \quad (13)$$

$$n_{0\beta} = N[\langle I^z \rangle - \langle I^z(n) \rangle], \quad (14)$$

where $\langle I^z(n) \rangle$ is the expectation value of I_i^z when $n_{0\beta} = n$; and where $n_{0\alpha}$ and $n_{0\beta}$ are the number of $\mathbf{k}=0$ spin waves present in the electronic (α) and nuclear (β) spin-wave branches, respectively. The state from which spin waves are generated, i.e., the nonresonating state, is taken to have complete electronic alignment $-NS$ but only partial nuclear polarization $N\langle I^z \rangle$.

From Eqs. (13) and (14) one finds

$$\langle S^\pm \rangle^2 = (2S/N)n_{0\alpha}[1 - (n_{0\alpha}/2SN)], \quad (15)$$

$$\langle I^\pm \rangle^2 = (2\langle I^z \rangle N)n_{0\beta}[1 - (n_{0\beta}/2\langle I^z \rangle N)], \quad (16)$$

and hence

$$\dot{M}^z \approx (2A/V)(\langle I^z \rangle S)^{1/2}(\gamma_e - \gamma_n)(n_{0\alpha}n_{0\beta})^{1/2} \sin(\omega_{0\alpha} - \omega_{0\beta})t. \quad (17)$$

If now a rf field of magnitude h and frequency $|\omega_e - \omega_n|$ is set into oscillation along z , the maximum steady power which can be delivered to the spin system (power factor = 1) is, per unit volume,

$$P_{\text{in}} \approx (hA/V)(\langle I^z \rangle S)^{1/2} |\gamma_e - \gamma_n| (n_{0\alpha}n_{0\beta})^{1/2}. \quad (18)$$

The total decay rates of electronic and nuclear $\mathbf{k}=0$ spin waves from all processes (spin-spin and spin-lattice) are taken as $\Gamma_{0\alpha}$ and $\Gamma_{0\beta}$, respectively. The power-out of the $\mathbf{k}=0$ systems is then, per unit volume,

$$P_{\text{out}} = (\Gamma_{0\alpha}/V)n_{0\alpha}\hbar\omega_{0\alpha} + (\Gamma_{0\beta}/V)n_{0\beta}\hbar|\omega_{0\beta}|. \quad (19)$$

Instability sets in, that is, power may be absorbed from parallel pumping, when h is sufficiently large so that (18) exceeds (19). Just before instability, $n_{0\alpha}$ and $n_{0\beta}$ are very small, and $\dot{n}_{0\alpha}$ and $\dot{n}_{0\beta}$ are zero. The power-in creates $n_{0\alpha}$ and $n_{0\beta}$ at equal rates R (otherwise angular momentum is not conserved), and

$$\dot{n}_{0\alpha} = R - \Gamma_{0\alpha}n_{0\alpha} = 0, \quad (20a)$$

$$\dot{n}_{0\beta} = R - \Gamma_{0\beta}n_{0\beta} = 0. \quad (20b)$$

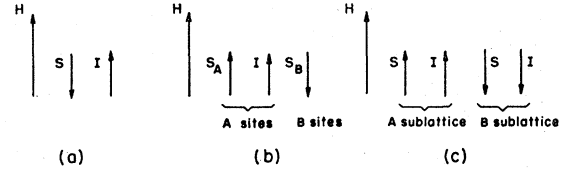


FIG. 1. (a) Ferromagnetic case with a positive hyperfine interaction. (b) Ferrimagnetic case with a negative hyperfine interaction. Note that the orientation of I with respect to the external and hyperfine fields is the same as in (a), resulting in the same "unperturbed" NMR frequency. (c) Antiferromagnetic case with negative hyperfine interaction.

Hence, at the instability point,

$$n_{0\alpha}/n_{0\beta} = \Gamma_{0\beta}/\Gamma_{0\alpha}. \quad (21)$$

On combining Eqs. (18), (19), and (21), one finds that the power-in exceeds the power-out when h exceeds a critical field h_c given by

$$h_c = \frac{(\Gamma_{0\alpha}\Gamma_{0\beta})^{1/2}\hbar(\omega_{0\alpha} + |\omega_{0\beta}|)}{|\gamma_e - \gamma_n| A(\langle I^z \rangle S)^{1/2}}. \quad (22)$$

This is a special case of *exchange-pumping*, for which the general formula has been given by Morgenthaler.³ His Eq. (35) yields the above h_c with the substitutions

$$|w_{12}| \rightarrow -AV/N\hbar^2\gamma_e\gamma_n, \quad (23a)$$

$$\alpha_i \rightarrow \frac{1}{2}\Gamma_i. \quad (23b)$$

In the present case, the hyperfine coupling between electronic and nuclear sublattices plays the role of an exchange field.

The above derivation yields a simple physical picture of exchange pumping, and in particular makes it clear why the two magnetogyric factors must differ. If $\gamma_e = \gamma_n$, then the exchange torques on the two sublattices, as given by Eqs. (5a) and (5b), are balanced; and it is possible for nuclear and electronic sublattices to precess without flexing against one another. With $\gamma_e \neq \gamma_n$, however, flexing takes place; and this gives rise to nonzero total \dot{M}^z .

The simple physical derivation is inadequate to handle complicated ferrimagnetic-nuclear and anti-ferromagnetic nuclear systems, with their many modes. We now turn to the more powerful operator technique, first making application to the ferromagnet.

B. Normal Modes, All \mathbf{k}

The normal modes for (1) at very low temperatures have been obtained by DPHW¹ from the equations of motion, by replacing S^z by S and $I_{\mathbf{a}}^z$ by its average $\langle I^z \rangle$. We proceed instead, by using conventional spin-wave theory⁴ through the substitutions [Fig. 1(a)]:

$$S_i^+ = (2S)^{1/2}a_i^\dagger,$$

$$S_i^- = (2S)^{1/2}a_i,$$

$$S_i^z = -S + a_i^\dagger a_i,$$

$$I_i^+ = (2\langle I^z \rangle)^{1/2}b_i,$$

$$I_i^- = (2\langle I^z \rangle)^{1/2}b_i^\dagger,$$

$$I_i^z = \langle I^z \rangle - b_i^\dagger b_i, \quad (24)$$

where $[a_i, a_i^\dagger] = [b_i, b_i^\dagger] = 1$ for all i , all other pairs of creation and annihilation operators commuting.

The linearization implied by the above use of $\langle I^z \rangle$ has been justified in detail by DPHW,¹ and it is here that the requirement of very low temperatures comes in. Inserting (24) in (1) and dropping quartic terms gives

$$\begin{aligned} \mathcal{H} = & C + \sum_{ij} 2SJ_{ij}(a_j^\dagger a_j - a_i^\dagger a_i) \\ & + \sum_i [(A \langle I^z \rangle - \gamma_e \hbar H) a_i^\dagger a_i \\ & + (AS + \gamma_n \hbar H) b_i^\dagger b_i + A (\langle I^z \rangle S)^{1/2} (a_i b_i + a_i^\dagger b_i^\dagger)], \end{aligned}$$

where

$$C = -S^2 \sum_{ij} J_{ij} - (\gamma_n \langle I^z \rangle - \gamma_e S) \hbar H N - A \langle I^z \rangle S N,$$

and N is the number of unit cells in the crystal.

Transforming now by

$$\begin{aligned} a_k &= N^{-1/2} \sum_j \exp(-i\mathbf{k} \cdot \mathbf{R}_j) a_j, \\ b_k &= N^{-1/2} \sum_j \exp(i\mathbf{k} \cdot \mathbf{R}_j) b_j \end{aligned} \quad (25)$$

and setting

$$J_k = 2S \sum_j J_{ij} \exp[i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)],$$

where \mathbf{k} belongs to the first Brillouin zone, we get

$$\mathcal{H} = C + \sum_k [A_k a_k^\dagger a_k + B b_k^\dagger b_k + F(a_k b_k + a_k^\dagger b_k^\dagger)], \quad (26)$$

where

$$\begin{aligned} A_k &= -\gamma_e \hbar H + A \langle I^z \rangle + J_0 - J_k, \\ B &= \gamma_n \hbar H + AS, \end{aligned}$$

and

$$F = A (\langle I^z \rangle S)^{1/2}. \quad (27)$$

\mathcal{H} may then be diagonalized by the canonical transformation

$$\begin{aligned} a_k &= \alpha_k \cosh \theta_k + \beta_k^\dagger \sinh \theta_k, \\ b_k &= \alpha_k^\dagger \sinh \theta_k + \beta_k \cosh \theta_k, \end{aligned} \quad (28)$$

where

$$\tanh 2\theta_k = -2F/(A_k + B) \quad (29)$$

and we assume that $|2F/(A_k + B)| < 1$. There results

$$\mathcal{H} = C - \frac{1}{2} \sum_k (A_k + B) + \sum_k (\epsilon_{k\alpha} \alpha_k^\dagger \alpha_k + \epsilon_{k\beta} \beta_k^\dagger \beta_k),$$

where

$$\begin{aligned} \epsilon_{k\alpha} &= \frac{1}{2} [(A_k - B) + (A_k + B)/\cosh 2\theta_k], \\ \epsilon_{k\beta} &= \frac{1}{2} [-(A_k - B) + (A_k + B)/\cosh 2\theta_k]. \end{aligned} \quad (30)$$

Since in fact $|2F/(A_k + B)| \ll 1$, one has

$$\begin{aligned} (\cosh 2\theta_k)^{-1} \\ = (1 - \tanh^2 2\theta_k)^{1/2} \approx 1 - \frac{1}{2} \tanh^2 2\theta_k = 1 - 2F^2/(A_k + B)^2 \end{aligned}$$

so that

$$\begin{aligned} \epsilon_{k\alpha} &\approx A_k - F^2/(A_k + B), \\ \epsilon_{k\beta} &\approx B - F^2/(A_k + B). \end{aligned}$$

Thus, referring to (27), it is seen that α and β are, respectively, the electronic and nuclear spin-wave branches. The corresponding frequencies of course agree with those obtained from DPHW,¹ when the appropriate signs for S , A , $\langle I^z \rangle$, and H are taken.

C. Parallel Pumping

We now apply a pumping field $h \sin \omega t$ along the z axis and treat the corresponding interaction

$$V(t) = -\hbar h \sin \omega t \sum_i (\gamma_e S_i^z + \gamma_n I_i^z)$$

as a perturbation on \mathcal{H} . After transforming to the normal modes α, β by inserting successively (24), (25), and (28), and ignoring terms diagonal in the α, β representation (since these cause no transitions), one gets

$$V(t) = \frac{1}{2} (\gamma_n - \gamma_e) \hbar h \sin \omega t \sum_k \sinh 2\theta_k (\alpha_k \beta_k + \alpha_k^\dagger \beta_k^\dagger). \quad (31)$$

The perturbation therefore excites and de-excites magnon pairs: one magnon from each branch. By ordinary time-dependent perturbation theory, the growth rate of occupation numbers is given by

$$\begin{aligned} \dot{n}_{k\alpha} = \dot{n}_{k\beta} = & \frac{1}{8} \pi (\gamma_e - \gamma_n)^2 h^2 \sinh^2 2\theta_k (n_{k\alpha} + n_{k\beta} + 1) \\ & \times \delta(\omega - \omega_{k\alpha} + \omega_{k\beta}), \end{aligned}$$

where

$$\hbar \omega_{k\alpha} = \epsilon_{k\alpha}; \quad \hbar \omega_{k\beta} = -\epsilon_{k\beta}. \quad (32)$$

Following Callen,⁵ magnon relaxation is then introduced phenomenologically by the addition of dissipative terms $-(n_{k\sigma} - \bar{n}_{k\sigma}) \Gamma_{k\sigma}$ where $\bar{n}_{k\sigma}$ is an equilibrium occupation number, together with the replacement $\delta(x) \rightarrow \frac{1}{2} \Gamma / [\pi(x^2 + \frac{1}{4} \Gamma^2)]$. Here $\frac{1}{2} \Gamma$ is the decay factor for the amplitude of the pair-magnon state and equals $\frac{1}{2} (\Gamma_{k\alpha} + \Gamma_{k\beta})$.

The net rate of change of occupation numbers then becomes

$$\begin{aligned} \dot{n}_{k\sigma} = & \frac{1}{8} \pi (\gamma_e - \gamma_n)^2 h^2 \sinh^2 2\theta_k (n_{k\alpha} + n_{k\beta} + 1) \\ & \times \frac{\Gamma_{k\alpha} + \Gamma_{k\beta}}{(\omega - \omega_{k\alpha} + \omega_{k\beta})^2 + \frac{1}{4} (\Gamma_{k\alpha} + \Gamma_{k\beta})^2} \\ & - (n_{k\sigma} - \bar{n}_{k\sigma}) \Gamma_{k\sigma}, \quad (\sigma = \alpha, \beta). \end{aligned}$$

This coupled system of equations will have a solution that increases exponentially with time when the rf field satisfies

$$h^2 > \frac{16 \Gamma_{k\alpha} \Gamma_{k\beta}}{(\Gamma_{k\alpha} + \Gamma_{k\beta})^2} \frac{[(\omega - \omega_{k\alpha} + \omega_{k\beta})^2 + \frac{1}{4} (\Gamma_{k\alpha} + \Gamma_{k\beta})^2]}{(\gamma_e - \gamma_n)^2 \sinh^2 2\theta_k}.$$

Therefore, denoting the right-hand side by h_{ck} , the first mode to go unstable will be when h exceeds a critical

field given by $h_c = \min h_{ck}$ where the minimum is to be sought over all k .

To estimate this expression, assume $\Gamma_{k\alpha}$, $\Gamma_{k\beta}$ are independent of k . Then since by (29), (30), and (32),

$$\omega_{k\alpha} - \omega_{k\beta} = -2F/(\hbar \sinh 2\theta_k),$$

the minimum occurs for $\omega_{k\alpha} - \omega_{k\beta} \simeq \omega$,

$$h_c = \hbar\omega(\Gamma_\alpha\Gamma_\beta)^{1/2}/|\gamma_e - \gamma_n| A(\langle I^z \rangle S)^{1/2}. \quad (33)$$

This formula is very similar to that obtained in the exchange pumping of a two-sublattice ferrimagnet.^{2,3,9} It may be noted that if the hyperfine interaction is anisotropic

$$A_t I^z S^z + A_t(I^x S^x + I^y S^y),$$

A in Eq. (33) gets replaced by the transverse component A_t .

If one takes the values for MnFe_2O_4 (for which admittedly A is large as well as negative), one has at liquid helium temperatures, $\Gamma_\alpha \sim 10^9 \text{ sec}^{-1}$,¹⁰ $\Gamma_\beta \sim 10^5 \text{ sec}^{-1}$, $A \sim 10^{-18} \text{ ergs}$,⁷ and say $\langle I^z \rangle \sim 2 \times 10^{-2}$, giving $h_c \sim 36 \text{ Oe}$ at $\omega = 10^{10} \text{ sec}^{-1}$. To achieve a pump frequency ω this small it may be necessary to take advantage of the effect of the demagnetizing field of a disk-shaped sample on the magnitude of $\omega_{k\alpha}$.

In arriving at the above estimate for $\Gamma_{k\beta}$ we have approximated $\Gamma_{k\beta} \sim \Gamma_{0\beta}$ and taken $\Gamma_{0\beta} \simeq (2/T_2)$, where T_2 is the nuclear spin-spin relaxation time. The lifetime of a nuclear spin wave is thus assumed to be governed by the dephasing of the nuclear spins; this is reasonable because the major portion of the nuclear spin system is unpolarized. Suhl¹¹ has estimated $(2/T_2)$ from the square root of the second moment of the nuclear resonance absorption, obtaining¹²

$$\Gamma_{0\beta} \simeq 2/T_2 \\ \simeq [I(I+1)/24\pi S^2]^{1/2} (\omega_n^2/\omega_{ex}^{3/4} \omega_e^{1/4}).$$

Here ω_{ex} is a measure of the exchange coupling between electronic spins. With $\omega_{ex} \sim 3 \times 10^{13}$, $\omega_e \sim 6 \times 10^9$, $\omega_n \sim 3 \times 10^9$, $S = I = \frac{5}{2}$, one obtains $T_2 \sim 10^{-5} \text{ sec}$. This is in agreement with a measurement by spin-echo techniques.¹³

In Table I are listed the values of h_c for a number of different processes. It is seen that, even in a powder sample, provided

$$\omega_{0\beta} < \omega = \omega_{k\alpha} - \omega_{k\beta} < 2\omega_{0\alpha}$$

⁹ F. Keffer, *Handbuch der Physik* (Springer-Verlag, Berlin, 1966), Vol. 18/2.

¹⁰ J. F. Dillon, Jr., S. Geschwind, and V. Jaccarino, *Phys. Rev.* **100**, 750 (1955). This is most probably an upper limit to the actual decay constant.

¹¹ H. Suhl, *Phys. Rev.* **109**, 606 (1958).

¹² This formula is incorrectly reproduced in DPHW (Ref. 1). Their equation (3.13) should be multiplied by $(1/4\pi^2)$ on the right-hand side. This correction improves their estimate of the validity of the linearization procedure, and allows reasonable definition of the nuclear spin-wave spectrum at temperatures as high as 4°K.

¹³ H. Yasuoka, H. Abe, M. Matsuura, and A. Hirai, *J. Phys. Soc. Japan* **18**, 1554 (1963).

TABLE I. Various critical fields, arranged in usual order of magnitude for same pump frequency ω . Note, however, that certain processes will require in general higher values of ω . In the formulas, ω_e and ω_n are given by (10) and (11), the quantity ω_m is $-4\pi\gamma_e M_e$, and Γ_1 is $1/T_1$, where T_1 is the nuclear spin-lattice relaxation time. The formula for the fifth listed process is from this paper, those of the second and fourth from Morgenthaler (Ref. 3) and the remainder from DeGennes *et al.* (Ref. 1).

Process	Pump frequency ω	Formula for h_c	Approximate ratio of h_c to that of parallel pump of a nuclear plus an electronic magnon for same value of ω	Typical value of previous column
Nuclear resonance saturation in perpendicular pump	$\omega_{0\beta}$	$(\Gamma_{0\beta}\Gamma_1)^{1/2}\hbar\omega_e/ \gamma_e AS$	$(\Gamma_1/\Gamma_{k\alpha})^{1/2}(\omega_e/\omega_e + \omega_m) \langle I^z \rangle / S)^{1/2}$	10^{-3}
Parallel pump of two electronic magnons in different spin-wave branches (exchange pumping)	$\omega_{k\alpha} - \omega_{k\beta}$	$(\Gamma_{k\alpha}\Gamma_{k\beta})^{1/2}\hbar\omega_e/ \gamma_e - \gamma_e 2J_z (SS)^{1/2}$	$(\Gamma_{k\alpha}/\Gamma_{k\beta})^{1/2}(\gamma_e - \gamma_n / \gamma_e - \gamma_e)(\omega_n/\omega_{ex}) \langle I^z \rangle / S)^{1/2}$	10^{-3}
Perpendicular pump of two nuclear magnons (Suhl instability)	$\omega_{0\beta}$	$\Gamma_{k\beta}\hbar\omega_e/ \gamma_e AS$	$(\Gamma_{k\beta}/\Gamma_{k\alpha})^{1/2}(\omega_e/\omega_e + \omega_m) \langle I^z \rangle / S)^{1/2}$	10^{-2}
Parallel pump of two electronic magnons in acoustic branch	$2\omega_{k\alpha}$	$\Gamma_{k\alpha}\omega_e/\omega_m \sin^2\theta_k$	$(\Gamma_{k\alpha}/\Gamma_{k\beta})^{1/2}(\omega_m/\omega_e) \langle I^z \rangle / S)^{1/2}$	10^{-1}
Parallel pump of a nuclear plus an electronic magnon	$\omega_{k\alpha} - \omega_{k\beta}$	$(\Gamma_{k\alpha}\Gamma_{k\beta})^{1/2}\hbar\omega_e/ \gamma_e - \gamma_n A \langle I^z \rangle S)^{1/2}$	1	1
Parallel pump of two nuclear magnons	$2\omega_{k\beta}$	$(\Gamma_{k\beta}\hbar^2\omega_e\omega_e/ \gamma_n A^2 \langle I^z \rangle S) \omega_e/\omega_m \sin^2\theta_k$	$(\Gamma_{k\beta}/\Gamma_{k\alpha})^{1/2}(\gamma_e / \gamma_n)(\omega_e/\omega_m) \langle I^z \rangle / S)^{1/2}$	10^2

the instability envisioned in this paper should be the first to go.

III. MANGANESE FERRITE MODEL

Manganese ferrite (MnFe_2O_4) has the spinel crystal structure with 80% of the Mn^{2+} ions on the tetrahedral (A) sites and 90% of the Fe^{3+} ions on the octahedral (B) sites.⁶ The Mn^{2+} ions have electronic spin $S=\frac{5}{2}$ and the nuclear spin $I=\frac{5}{2}$ directed parallel to the electronic spin⁷; the nuclear gyromagnetic ratio is $\gamma_n \approx 7 \times 10^3$ (sec G)⁻¹. The Fe^{3+} have $S=\frac{5}{2}$ and, except for Fe^{57} , which is of low abundance, $I=0$. Hence for the present purposes, we assume all the Mn^{2+} are on A sites and all the Fe^{3+} on B sites, and further that only the Mn nuclei carry a magnetic moment. MnFe_2O_4 is ferrimagnetic with the Mn^{2+} and Fe^{3+} electronic spins oppositely directed.

A detailed analysis of the electronic spin-wave modes of a normal spinel was first given by Kaplan.¹⁴ It will be convenient in what follows to use some of his notation. The spinel structure may be defined (following Kaplan), by the primitive translation vectors

$$a_1 = \frac{1}{2}a(1, 1, 0), \quad a_2 = \frac{1}{2}a(0, 1, 1), \quad a_3 = \frac{1}{2}a(1, 0, 1) \quad (34)$$

together with the basis

$$\begin{aligned} \mathbf{e}_1^A &= 0, & \mathbf{e}_2^A &= \frac{1}{4}a(1, 1, 1), \\ \mathbf{e}_1^B &= \frac{1}{8}a(1, 5, 1), & \mathbf{e}_2^B &= \frac{1}{8}a(3, 5, 3), \\ \mathbf{e}_3^B &= \frac{1}{8}a(3, 7, 1), & \mathbf{e}_4^B &= \frac{1}{8}a(1, 7, 3), \end{aligned} \quad (35)$$

where $[a_{l\alpha}, a_{l\alpha}^\dagger] = [b_{m\beta}, b_{m\beta}^\dagger] = [c_{l\alpha}, c_{l\alpha}^\dagger] = 1$ (all l, m, α, β), all other pairs of creation and annihilation operators commuting. The above choice of operators is in accordance with the data given earlier as to orientation of the spins [Fig. 1(b)]. Substitution of Eq. (37) into Eq. (36), with neglect of quartic terms, followed by the canonical transformations

$$\begin{aligned} a_{k\alpha} &= N^{-1/2} \sum_l \exp(-i\mathbf{k} \cdot \mathbf{R}_{l\alpha}) a_{l\alpha}, \\ b_{k\beta} &= N^{-1/2} \sum_l \exp(i\mathbf{k} \cdot \mathbf{R}_{l\beta}) b_{l\beta}, \end{aligned}$$

and

$$c_{k\alpha} = N^{-1/2} \sum_l \exp(i\mathbf{k} \cdot \mathbf{R}_{l\alpha}) c_{l\alpha}, \quad (38)$$

where N is the number of unit cells in the crystal,

the numbers in parentheses being the components with respect to some rectangular coordinate system. Here a^3 is four times the volume of the primitive unit cell.

A. Hamiltonian

The Hamiltonian for the nuclear electronic system is taken to be

$$\begin{aligned} \mathcal{H} &= 2J \sum_{lm\alpha\beta} \mathbf{S}_{l\alpha} \cdot \mathbf{S}_{m\beta} - \gamma_e \hbar H_a \sum_{l\alpha} S_{l\alpha}^{z'} - \gamma_e \hbar H_b \\ &\times \sum_{m\beta} S_{m\beta}^{z'} - A \sum_{l\alpha} \mathbf{I}_{l\alpha} \cdot \mathbf{S}_{l\alpha} - \gamma_n \hbar H \sum_{l\alpha} I_{l\alpha}^{z'}. \end{aligned} \quad (36)$$

Here l, m label the unit cell; $\alpha=1, 2$ labels the A sites and $\beta=1, 2, 3, 4$ the B sites in a unit cell. The terms, in order, are: antiferromagnetic exchange between A and B spins ($J>0$); combined electronic Zeeman and anisotropy energy for A spins and for B spins; hyperfine interaction between A electronic and A nuclear spins¹⁵; nuclear Zeeman energy. The external field H is along an easy direction z' and we have written $H_a = H - H_A$, $H_b = H + H_B$ where H_A, H_B are effective anisotropy fields on the A and B sublattices, respectively. The summation over exchange terms will be taken only over nearest A - B pairs. From the paper of Heeger and Houston cited earlier,⁷ $J/k \approx 22.7^\circ\text{K}$. Also, as may be seen from Eq. (49) below, *their* effective ferromagnetic anisotropy field H_A must be identified with our $(2H_B - H_A)$. This is about 800 Oe. at liquid-helium temperatures.

Following standard spin-wave theory, we first set

$$\begin{aligned} S_{l\alpha}^+ &= (2S)^{1/2} a_{l\alpha}, & S_{l\alpha}^- &= (2S)^{1/2} a_{l\alpha}^\dagger, & S_{l\alpha}^{z'} &= S - a_{l\alpha}^\dagger a_{l\alpha}, \\ S_{m\beta}^+ &= (2S)^{1/2} b_{m\beta}^\dagger, & S_{m\beta}^- &= (2S)^{1/2} b_{m\beta}, & S_{m\beta}^{z'} &= -S + b_{m\beta}^\dagger b_{m\beta}, \\ I_{l\alpha}^+ &= (2\langle I^z \rangle)^{1/2} c_{l\alpha}, & I_{l\alpha}^- &= (2\langle I^z \rangle)^{1/2} c_{l\alpha}^\dagger, & I_{l\alpha}^{z'} &= \langle I^z \rangle - c_{l\alpha}^\dagger c_{l\alpha}, \end{aligned} \quad (37)$$

yields

$$\begin{aligned} \mathcal{H} &= \text{constant} + \hbar \sum_k \{ \tilde{A} \sum_\alpha a_{k\alpha}^\dagger a_{k\alpha} + B \sum_\beta b_{k\beta}^\dagger b_{k\beta} \\ &+ \gamma \sum_{\alpha\beta} [\zeta_{\alpha\beta}(-\mathbf{k}) a_{k\alpha} b_{k\beta} + \zeta_{\alpha\beta}(\mathbf{k}) a_{k\alpha}^\dagger b_{k\beta}^\dagger] \\ &- F \sum_\alpha (a_{k\alpha}^\dagger c_{k\alpha} + a_{k\alpha} c_{k\alpha}^\dagger) + D \sum_\alpha c_{k\alpha}^\dagger c_{k\alpha} \}, \end{aligned} \quad (39)$$

where \mathbf{k} runs over the first Brillouin zone, and

$$\begin{aligned} \hbar \tilde{A} &= A \langle I^z \rangle - \gamma_e \hbar H_a + 24SJ, \\ \hbar B &= -\gamma_e \hbar H_b + 12SJ, \\ \hbar \gamma &= 2SJ, \\ \hbar F &= A (S \langle I^z \rangle)^{1/2}, \\ \hbar D &= \gamma_n \hbar H + AS, \\ \zeta_{\alpha\beta}(\mathbf{k}) &= \sum_m \exp[i\mathbf{k} \cdot (\mathbf{R}_{m\beta} - \mathbf{R}_{l\alpha})], \end{aligned} \quad (40)$$

¹⁴ T. A. Kaplan, Phys. Rev. **109**, 782 (1958).

¹⁵ The hyperfine constant has been taken to be $-A$, so that $A > 0$ in Eq. (36).

the last sum being over m for which $m\beta$ is nearest neighbor to $l\alpha$. $\mathbf{R}_{l\alpha(\beta)}$ is the position vector of $A(B)$ site $\alpha(\beta)$ in the unit cell l .

From Eqs. (34) and (35), one may verify that

$$\zeta_{1\beta}(\mathbf{k}) = \zeta_{2\beta}(-\mathbf{k}). \quad (41)$$

It will be convenient to let $\zeta_{\alpha\beta}^e, \zeta_{\alpha\beta}^o$ be, respectively, the real and imaginary parts of $\zeta_{\alpha\beta}$, and to introduce the quantities

$$\xi_e = \sum_{\beta=1}^4 (\zeta_{1\beta}^e)^2, \quad \xi_o = \sum_{\beta=1}^4 (\zeta_{1\beta}^o)^2, \quad \xi' = \sum_{\beta=1}^4 \zeta_{1\beta}^e \zeta_{1\beta}^o. \quad (42)$$

B. Normal Modes

To diagonalize \mathcal{H} , it suffices to solve the equation of motion¹⁶

$$i(d\mathbf{X}/dt) = \mathbf{M}\mathbf{X}, \quad (43)$$

where \mathbf{X} is the transpose of $(a_{k1}, a_{k2}, c_{k1}, c_{k2}, b_{k1}^\dagger, b_{k2}^\dagger, b_{k3}^\dagger, b_{k4}^\dagger)$ and

$$\mathbf{M} = \begin{pmatrix} \tilde{A} & 0 & F & 0 & \gamma\zeta_{11} & \gamma\zeta_{12} & \gamma\zeta_{13} & \gamma\zeta_{14} \\ 0 & \tilde{A} & 0 & F & \gamma\zeta_{21} & \gamma\zeta_{22} & \gamma\zeta_{23} & \gamma\zeta_{24} \\ F & 0 & D & 0 & 0 & 0 & 0 & 0 \\ 0 & F & 0 & D & 0 & 0 & 0 & 0 \\ -\gamma\zeta_{21} & -\gamma\zeta_{11} & 0 & 0 & -B & 0 & 0 & 0 \\ -\gamma\zeta_{22} & -\gamma\zeta_{12} & 0 & 0 & 0 & -B & 0 & 0 \\ -\gamma\zeta_{23} & -\gamma\zeta_{13} & 0 & 0 & 0 & 0 & -B & 0 \\ -\gamma\zeta_{24} & -\gamma\zeta_{14} & 0 & 0 & 0 & 0 & 0 & -B \end{pmatrix}. \quad (44)$$

Equation (41) has been used to ensure that in \mathbf{M} all ζ have argument k . In Appendix A, it is shown that the eigenvalues of \mathbf{M} are given by

$$\omega = -B \quad (2\text{-fold}), \quad (45)$$

and by the roots of the two cubics

$$(\omega - D)[(\omega - \tilde{A})(\omega + B) + \gamma^2 \xi] - F^2(\omega + B) = 0, \quad (46)$$

where

$$\xi = \xi_e + \xi_o \pm [(\xi_e - \xi_o)^2 + 4\xi'^2]^{1/2}. \quad (47)$$

The roots referring to the + sign will be labeled $\omega_1, \omega_2,$ and ω_3 ; those to the - sign $\omega_4, \omega_5,$ and ω_6 . We also set $\omega_7 = \omega_8 = -B$.

Since $F^2 = A^2 \langle I^z \rangle S$ is small, for orientation, we may neglect it in (46). This gives

$$\omega \approx D; \quad \omega \approx \frac{1}{2}(\tilde{A} - B) \pm \left[\frac{1}{4}(\tilde{A} - B)^2 + (\tilde{A}B - \gamma^2 \xi) \right]^{1/2}. \quad (48)$$

¹⁶ A more extensive discussion of the diagonalization procedure, relevant to Eqs. (43), (52), (53), and (54), will be found in R. M. White, M. Sparks, and I. Ortenburger, Phys. Rev. 139, A450 (1965).

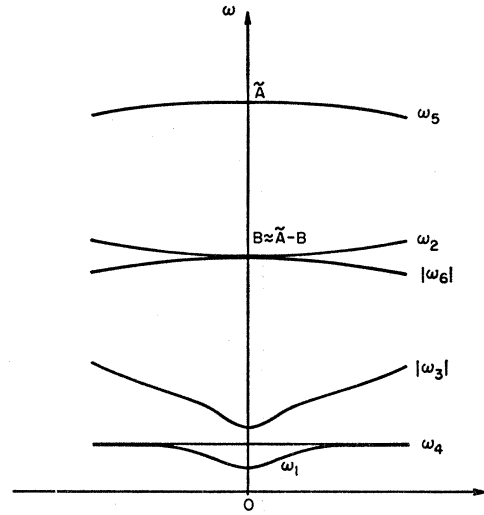


FIG. 2. Spectrum of manganese ferrite for branches 1-6 near $k=0$. The other two modes have $|\omega_7| = |\omega_8| = B$.

Thus each cubic has a nuclear frequency $\approx D$. Let the exact values be ω_1 and ω_4 .

Next, using in Eq. (48) the definitions (40) with the data given earlier for MnFe_2O_4 , shows that each cubic has a negative root. Let these be ω_3 and ω_6 . The roots ω_2 and $\omega_5, \dots, \omega_8$ are of order 10^{14} cps and are of no interest for the present purposes. In Appendix B, it is shown that for the nuclear branches

$$\hbar\omega_{1,4} \approx AS + \gamma_n \hbar H - \frac{A^2 S \langle I^z \rangle}{AS - \gamma_e \hbar (H_a + 2H_b) + 24SJ[1 - \xi(\mathbf{k})/72]}; \quad (49)$$

and for small ka ,

$$-\hbar\omega_3 \approx -\gamma_e \hbar (H_a + 2H_b) + A \langle I^z \rangle + 24SJ[1 - \xi(\mathbf{k})/72] - \frac{A^2 S \langle I^z \rangle}{AS - \gamma_e \hbar (H_a + 2H_b) + 24SJ[1 - \xi(\mathbf{k})/72]}. \quad (50)$$

From Eqs. (34), (35), and (40)-(42), it follows that to order k^2 ,¹⁴

$$\xi_o = k^2 a^2 / 16, \quad \xi_e = 36 - 33k^2 a^2 / 16, \quad \xi' = 0;$$

hence

$$\xi = \xi_e + \xi_o \pm |\xi_e - \xi_o| = 2\xi_e = 72 - 33k^2 a^2 / 8 \quad \text{in } \omega_1, \omega_3 \\ = 2\xi_o = k^2 a^2 / 8 \quad \text{in } \omega_4. \quad (51)$$

Thus of the two nuclear branches 1 and 4, only ω_1 is depressed appreciably near $k=0$. Further, $|\omega_3|$, which is the electronic acoustic mode, is depressed by the same amount as ω_1 , and is of the order of 10^{10} cps near $k=0$. The branches are sketched in Fig. 2.

The normal modes of the system will be given by

$$(\alpha_{k1}, \alpha_{k2}, \alpha_{k3}^\dagger, \alpha_{k4}, \alpha_{k5}, \alpha_{k6}^\dagger, \alpha_{k7}^\dagger, \alpha_{k8}^\dagger)^T = \mathbf{S}\mathbf{X}, \quad (52)$$

where T means transpose, and where \mathbf{S} is to be chosen so that

$$\mathbf{S}\mathbf{M}\mathbf{S}^{-1} = \text{diag}(\omega_1, \dots, \omega_8) \quad (53)$$

and

$$\mathbf{S} \text{diag}(1, 1, 1, 1, -1, -1, -1, -1) \mathbf{S}^* = \text{diag}(1, 1, -1, 1, 1, -1, -1, -1). \quad (54)$$

Equations (52) and (53) decouple the equations of motion, while Eq. (54) ensures the α 's satisfy the Bose commutation rules.¹⁶ The notation * means Hermitian conjugate.

In terms of the eigenvalues,

$$\mathbf{S} = \begin{pmatrix} \tau_{1\lambda_1} & \lambda_1 & \frac{-F\tau_{1\lambda_1}}{D-\omega_1} & \frac{-F\lambda_1}{D-\omega_1} & \frac{\tau_{21\lambda_1}}{B+\omega_1} & \frac{\tau_{22\lambda_1}}{B+\omega_1} & \frac{\tau_{23\lambda_1}}{B+\omega_1} & \frac{\tau_{24\lambda_1}}{B+\omega_1} \\ \tau_{1\lambda_2} & \lambda_2 & \frac{-F\tau_{1\lambda_2}}{D-\omega_2} & \frac{-F\lambda_2}{D-\omega_2} & \frac{\tau_{21\lambda_2}}{B+\omega_2} & \frac{\tau_{22\lambda_2}}{B+\omega_2} & \frac{\tau_{23\lambda_2}}{B+\omega_2} & \frac{\tau_{24\lambda_2}}{B+\omega_2} \\ \tau_{1\lambda_3} & \lambda_3 & \frac{-F\tau_{1\lambda_3}}{D-\omega_3} & \frac{-F\lambda_3}{D-\omega_3} & \frac{\tau_{21\lambda_3}}{B+\omega_3} & \frac{\tau_{22\lambda_3}}{B+\omega_3} & \frac{\tau_{23\lambda_3}}{B+\omega_3} & \frac{\tau_{24\lambda_3}}{B+\omega_3} \\ -\tau_{1\lambda_4} & \lambda_4 & \frac{F\tau_{1\lambda_4}}{D-\omega_4} & \frac{-F\lambda_4}{D-\omega_4} & \frac{\tau_{31\lambda_4}}{B+\omega_4} & \frac{\tau_{32\lambda_4}}{B+\omega_4} & \frac{\tau_{33\lambda_4}}{B+\omega_4} & \frac{\tau_{34\lambda_4}}{B+\omega_4} \\ -\tau_{1\lambda_5} & \lambda_5 & \frac{F\tau_{1\lambda_5}}{D-\omega_5} & \frac{-F\lambda_5}{D-\omega_5} & \frac{\tau_{31\lambda_5}}{B+\omega_5} & \frac{\tau_{32\lambda_5}}{B+\omega_5} & \frac{\tau_{33\lambda_5}}{B+\omega_5} & \frac{\tau_{34\lambda_5}}{B+\omega_5} \\ -\tau_{1\lambda_6} & \lambda_6 & \frac{F\tau_{1\lambda_6}}{D-\omega_6} & \frac{-F\lambda_6}{D-\omega_6} & \frac{\tau_{31\lambda_6}}{B+\omega_6} & \frac{\tau_{32\lambda_6}}{B+\omega_6} & \frac{\tau_{33\lambda_6}}{B+\omega_6} & \frac{\tau_{34\lambda_6}}{B+\omega_6} \\ 0 & 0 & 0 & 0 & u_{11}^* & u_{12}^* & u_{13}^* & u_{14}^* \\ 0 & 0 & 0 & 0 & u_{21}^* & u_{22}^* & u_{23}^* & u_{24}^* \end{pmatrix}, \quad (55)$$

where

$$\begin{aligned} \tau_1 &= (\xi_e - \xi_0 - 2i\xi') / [(\xi_e - \xi_0)^2 + 4\xi'^2]^{1/2}, \\ \tau_{\alpha\beta} &= \gamma \{ \zeta_{2\beta} + (-1)^{\alpha} \zeta_{1\beta} (\xi_e - \xi_0 - 2i\xi') / [(\xi_e - \xi_0)^2 + 4\xi'^2]^{1/2} \}, \quad (\alpha = 1, 2). \\ \sum_{\beta=1}^4 u_{\alpha\beta}^* u_{\alpha'\beta} &= \delta_{\alpha\alpha'}, \quad \sum_{\beta=1}^4 \zeta_{\alpha\beta}^e u_{\alpha'\beta} = \sum_{\beta=1}^4 \zeta_{\alpha\beta}^0 u_{\alpha'\beta} = 0, \quad (\alpha, \alpha' = 1, 2) \end{aligned}$$

and

$$2 |\lambda_j|^2 \left(1 + \frac{F^2}{(D-\omega_j)^2} - \frac{\gamma^2}{(B+\omega_j)^2} \{ \xi_e + \xi_0 \pm [(\xi_e - \xi_0)^2 + 4\xi'^2]^{1/2} \} \right) = \pm 1, \quad (56)$$

where on the left, the + goes with $j=1, 2, 3$ and the - with $j=4, 5, 6$; and on the right the + goes with $j=1, 2, 4, 5$ and the - with $j=3, 6$.

C. Parallel Pumping

With a pumping field $h \sin\omega t$ along Oz' , the interaction is

$$V(t) = -\hbar h \sin\omega t [\gamma_e \sum_{l\alpha} S_{l\alpha} z' + \gamma_o \sum_{m\beta} S_{m\beta} z' + \gamma_n \sum_{l\alpha} I_{l\alpha} z']$$

If we now make the approximation (37) and apply successively the transformations (38) and (52), using (55) and (42), we get

$$V(t) = \hbar h \sin\omega t \sum_k \left\{ \begin{aligned} & [\alpha_{k1}^\dagger, \alpha_{k2}^\dagger, -\alpha_{k3}] \begin{bmatrix} 0 & W_{12}^{(k)} & W_{13}^{(k)} \\ W_{21}^{(k)} & 0 & W_{23}^{(k)} \\ W_{31}^{(k)} & W_{32}^{(k)} & 0 \end{bmatrix} \begin{bmatrix} \alpha_{k1} \\ \alpha_{k2} \\ -\alpha_{k3}^\dagger \end{bmatrix} \\ & + [\alpha_{k4}^\dagger, \alpha_{k5}^\dagger, -\alpha_{k6}] \begin{bmatrix} 0 & W_{45}^{(k)} & W_{46}^{(k)} \\ W_{54}^{(k)} & 0 & W_{56}^{(k)} \\ W_{64}^{(k)} & W_{65}^{(k)} & 0 \end{bmatrix} \begin{bmatrix} \alpha_{k4} \\ \alpha_{k5} \\ -\alpha_{k6}^\dagger \end{bmatrix} \end{aligned} \right\} \quad (57)$$

in which terms diagonal in the α 's have been ignored, and where, omitting the index k ,

$$W_{ij} = W_{ji}^* = 2\lambda_i \lambda_j^* \left(\gamma_e + \frac{\gamma_n F^2}{(D - \omega_i)(D - \omega_j)} - \frac{\gamma_e}{(B + \omega_i)(B + \omega_j)} \gamma^2 \{ \xi_e + \xi_0 \pm [(\xi_e - \xi_0)^2 + 4\xi^2]^{1/2} \} \right), \quad (58)$$

where $+$ is for $(ij) = (12), (13), (23)$ and $-$ for $(ij) = (45), (46), (56)$.

Thus the perturbation does not mix different k 's, nor does it mix any of 1, 2, 3 with any of 4, 5, 6 (or 7, 8); but it does contain a term $\alpha_{k1}^\dagger \alpha_{k3}^\dagger$ which indicates the possibility of simultaneous pumping in the 1 and 3 branches. It is apparent since branches 1 and 3 are well removed from the remainder, that if the pumping frequency is in the neighborhood of $\omega_1 - \omega_3$ (note: $\omega_3 < 0$), we need only consider transitions of the 1 and 3 branches. Then, as in the case of a ferromagnet, the rate equations with damping are

$$\dot{n}_{kj} = \frac{1}{2} \hbar^2 |W_{13}^{(k)}|^2 (n_{k1} + n_{k3} + 1) \frac{\Gamma_{k1} + \Gamma_{k3}}{(\omega - \omega_{k1} + \omega_{k3})^2 + \frac{1}{4}(\Gamma_{k1} + \Gamma_{k3})^2} - (n_{kj} - \bar{n}_{kj}) \Gamma_{kj} \quad (j=1, 3),$$

where Γ_{k1}, Γ_{k3} are linewidths appropriate to the 1, 3 branches, respectively. The threshold for instability of the 1-3 system is therefore

$$h_c = \left\{ \frac{4\Gamma_{k1}\Gamma_{k3}[(\omega - \omega_{k1} + \omega_{k3})^2 + \frac{1}{4}(\Gamma_{k1} + \Gamma_{k3})^2]}{(\Gamma_{k1} + \Gamma_{k3})^2 |W_{13}^{(k)}|^2} \right\}_{\min k}^{1/2}. \quad (59)$$

It will now be shown that as long as $|\omega_3| \ll B \sim 12SJ/\hbar$ and $ka \ll 1$, the above critical field is essentially the same as for the ferromagnetic case. This is not surprising, since it is known that low-frequency modes in the acoustic branch generally are "unaware" of the crystal structure. First, from Eqs. (56) and (58),

$$|W_{13}|^2 = \frac{([\gamma_n F^2 / (D - \omega_1)(D - \omega_3)] + \gamma_e \{ [\gamma^2 \xi / (B + \omega_1)(B + \omega_3)] - 1 \})^2}{\{1 + [F^2 / (D - \omega_1)^2] - [\gamma^2 \xi / (B + \omega_1)^2]\} \{[\gamma^2 \xi / (B + \omega_3)^2] - [F^2 / (D - \omega_3)^2] - 1\}}. \quad (60)$$

Next, from Eq. (B4), and the fact that $B \approx \bar{A} - B$ [Eq. (40)], it follows that

$$\begin{aligned} \omega_1 - D &= - \frac{BF^2}{D(\bar{A} - B) + \bar{A}B - \gamma^2 \xi} \approx - \frac{F^2}{D + [(\bar{A}B - \gamma^2 \xi) / (\bar{A} - B)]} \\ &\approx -F^2 / (\omega_1 - \omega_3), \end{aligned} \quad (61)$$

the last step coming from Eqs. (B4) and (B6). Hence

$$F^2 / (D - \omega_1)(D - \omega_3) \approx (\omega_1 - \omega_3) / (D - \omega_3) \approx 1$$

and

$$F^2 / (D - \omega_1)^2 \approx (\omega_1 - \omega_3)^2 / F^2 \gg 1.$$

Also, assuming $|\omega_3| \ll B \sim 12SJ/\hbar$, we have by Eqs. (40) and (41),

$$\gamma^2 \xi / B^2 \approx 2 - 11k^2 a^2 / 96.$$

Hence to a good approximation,

$$|W_{13}|^2 \approx \frac{\{\gamma_n - \gamma_e [(\gamma^2 \xi / B^2) - 1]\}^2}{(\omega_1 - \omega_3)^2 / F^2 [(\gamma^2 \xi / B^2) - 1]} \approx \frac{\gamma_e^2 F^2 [1 - (11k^2 a^2 / 96)]}{(\omega_1 - \omega_3)^2} \approx \frac{\gamma_e^2 F^2}{(\omega_1 - \omega_3)^2}, \quad (62)$$

the \mathbf{k} variation being determined essentially by the denominator. Introducing Eq. (62) into Eq. (59) then gives

$$\begin{aligned} h_c &= \left\{ \frac{4\Gamma_{k1}\Gamma_{k3}(\omega_{k1} - \omega_{k3})^2 [(\omega - \omega_{k1} + \omega_{k3})^2 + \frac{1}{4}(\Gamma_{k1} + \Gamma_{k3})^2]}{\gamma_e^2 F^2 (\Gamma_{k1} + \Gamma_{k3})^2} \right\}_{\min k}^{1/2}, \\ &= \frac{\hbar \omega (\Gamma_1 \Gamma_3)^{1/2}}{\gamma_e A (\langle I^2 \rangle S)^{1/2}}, \end{aligned} \quad (63)$$

where $\omega = \omega_{k1} + |\omega_{k3}|$ and we assume Γ_{k1}, Γ_{k3} are independent of \mathbf{k} . This field is the same as in Eq. (33) for the ferromagnetic case.

In the same way, the presence of a term $\alpha_{k4}^\dagger \alpha_{k5}^\dagger$ in Eq. (57), suggests the possibility of simultaneous pumping in the 4 and 5 branches. It is likely however, that the high frequency of the 5 branch ($\sim 10^{14}$ cps) would require high pump powers. But it should be noted that in the conventional NMR experiment, in which the static and rf fields are perpendicular to each other, it is the depressed nuclear mode ω_1 which is excited: Coupling to the other nuclear mode ω_4 is negligible, essentially because the two nuclear moments in a unit cell precess 180° out of phase (at $k=0$). Thus in principle, parallel pumping is a means of exciting the ω_4 branch.

IV. ANTIFERROMAGNET WITH APPLICATION TO RbMnF₃

We now consider the cubic antiferromagnet and specialize to RbMnF₃. RbMnF₃ is a cubic antiferromagnet with very low anisotropy, the magnetic ions

being Mn²⁺. As in the case of MnFe₂O₄, it will be assumed that for a given Mn ion, the nuclear and electronic spins point in the same direction.

A. Hamiltonian

An appropriate Hamiltonian is

$$\begin{aligned} \mathcal{H} = & 2J \sum_{\alpha\beta} \mathbf{S}_\alpha \cdot \mathbf{S}_\beta + \gamma_e \hbar (H_A - H) \\ & \times \sum_{\alpha} S_{\alpha^z} - \gamma_e \hbar (H_A + H) \sum_{\beta} S_{\beta^z} + \gamma_n \hbar H \\ & \times [\sum_{\alpha} I_{\alpha^z} + \sum_{\beta} I_{\beta^z}] - A [\sum_{\alpha} \mathbf{I}_\alpha \cdot \mathbf{S}_\alpha + \sum_{\beta} \mathbf{I}_\beta \cdot \mathbf{S}_\beta], \end{aligned} \quad (64)$$

where α and β refer to the A and B sublattices, respectively. The terms in order are: antiferromagnetic exchange ($J > 0$); combined electronic Zeeman and anisotropy energy for the A sublattice and for the B sublattice; nuclear Zeeman energy; hyperfine interaction ($A > 0$). The external field is along an easy direction z (and is assumed to be less than the critical flop field). The summation over exchange terms will be taken only over nearest A - B pairs.

By the usual spin-wave theory, we first set

$$\begin{aligned} S_{\alpha^+} &= (2S)^{1/2} a_{\alpha}, & S_{\alpha^-} &= (2S)^{1/2} a_{\alpha}^\dagger, & S_{\alpha^z} &= S - a_{\alpha}^\dagger a_{\alpha}, \\ S_{\beta^+} &= (2S)^{1/2} b_{\beta}^\dagger, & S_{\beta^-} &= (2S)^{1/2} b_{\beta}, & S_{\beta^z} &= -S + b_{\beta}^\dagger b_{\beta}, \\ I_{\alpha^+} &= (2\langle I_A^z \rangle)^{1/2} c_{\alpha}, & I_{\alpha^-} &= (2\langle I_A^z \rangle)^{1/2} c_{\alpha}^\dagger, & I_{\alpha^z} &= \langle I_A^z \rangle - c_{\alpha}^\dagger c_{\alpha}, \\ I_{\beta^+} &= (2|\langle I_B^z \rangle|)^{1/2} d_{\beta}^\dagger, & I_{\beta^-} &= (2|\langle I_B^z \rangle|)^{1/2} d_{\beta}, & I_{\beta^z} &= \langle I_B^z \rangle + d_{\beta}^\dagger d_{\beta}, \end{aligned} \quad (65)$$

where $[a_{\alpha}, a_{\alpha}^\dagger] = [b_{\beta}, b_{\beta}^\dagger] = [c_{\alpha}, c_{\alpha}^\dagger] = [d_{\beta}, d_{\beta}^\dagger] = 1$ (all α, β), all other pairs of creation and annihilation operators commuting. The choice of operators is in accordance with Fig. 1(c). $\langle I_A^z \rangle, \langle I_B^z \rangle$ are the average nuclear angular momenta of the A and B sublattices, respectively.

Substitution of Eq. (65) in Eq. (64), with neglect of quartic terms, followed by the canonical transformations

$$\begin{aligned} a_k &= N^{-1/2} \sum_{\alpha} \exp(i\mathbf{k} \cdot \mathbf{R}_{\alpha}) a_{\alpha}, \\ b_k &= N^{-1/2} \sum_{\beta} \exp(-i\mathbf{k} \cdot \mathbf{R}_{\beta}) b_{\beta}, \\ c_k &= N^{-1/2} \sum_{\alpha} \exp(i\mathbf{k} \cdot \mathbf{R}_{\alpha}) c_{\alpha}, \\ d_k &= N^{-1/2} \sum_{\beta} \exp(-i\mathbf{k} \cdot \mathbf{R}_{\beta}) d_{\beta}, \end{aligned} \quad (66)$$

where N is the number of unit cells in the crystal, yields

$\mathcal{H} = \text{const}$

$$\begin{aligned} & + \hbar \sum_k \{ \tilde{A} a_k^\dagger a_k + D_1 c_k^\dagger c_k - F_A (a_k^\dagger c_k + a_k c_k^\dagger) + B b_k^\dagger b_k \\ & + D_2 d_k^\dagger d_k - F_B (b_k^\dagger d_k + b_k d_k^\dagger) + \omega_{\text{ex}} \gamma_k (a_k b_k + a_k^\dagger b_k^\dagger) \}. \end{aligned} \quad (67)$$

Here

$$\begin{aligned} \tilde{A} &= \omega_{\text{ex}} - \gamma_e (H_A - H + H_{nA}), & H_{nA} &= -A \langle I_A^z \rangle / (\gamma_e \hbar), \\ B &= \omega_{\text{ex}} - \gamma_e (H_A + H + H_{nB}), & H_{nB} &= -A |\langle I_B^z \rangle| / (\gamma_e \hbar), \\ D_1 &= \omega_n + \gamma_n H, & \omega_n &= AS / \hbar, \\ D_2 &= \omega_n - \gamma_n H, \\ F_A &= (-\gamma_e \omega_n H_{nA})^{1/2}, \\ F_B &= (-\gamma_e \omega_n H_{nB})^{1/2}, \\ \omega_{\text{ex}} &= 2S_z J / \hbar, \\ \gamma_k &= z^{-1} \sum \exp(i\mathbf{k} \cdot \mathbf{R}), \end{aligned} \quad (68)$$

the last sum being over the z nearest neighbors of a given site. For RbMnF₃, it will be assumed that $\langle I_A^z \rangle = |\langle I_B^z \rangle| = \langle I^z \rangle$, which is adequate providing the external field is small compared to hyperfine field on the nuclei (~ 600 kOe). Then

$$H_{nA} = H_{nB} = H_n \equiv -A \langle I^z \rangle / (\gamma_e \hbar),$$

and

$$F_A = F_B = F \equiv (-\gamma_e \omega_n H_n)^{1/2}. \quad (69)$$

For later reference, we list some data obtained from

¹⁷ 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.* **10**, 293 (1963); H. Montgomery, D. T. Teaney and W. M. Walsh, Jr., Phys. Rev. **128**, 80 (1962).

the papers in Ref. 17. For RbMnF₃ at 4.2°K,

$$\begin{aligned} H_A &= 4.5 \text{ Oe}; & \omega_{\text{ex}} &= 1.6 \times 10^{13} \text{ cps}; \\ \omega_n &= 4.3 \times 10^9 \text{ cps}; & H_n &= 2.2 \text{ Oe}. \end{aligned} \quad (70)$$

B. Normal Modes

From Eq. (67) we get the equations of motion

$$i(d\mathbf{X}/dt) = \mathbf{M}\mathbf{X}, \quad (71)$$

where X is the transpose of $(a_k, c_k, b_k^\dagger, d_k^\dagger)$ and

$$\mathbf{M} = \begin{vmatrix} \tilde{A} & -F_A & \gamma_k \omega_{\text{ex}} & 0 \\ -F_A & D_1 & 0 & 0 \\ -\gamma_k \omega_{\text{ex}} & 0 & -B & F_B \\ 0 & 0 & F_B & -D_2 \end{vmatrix}. \quad (72)$$

The eigenvalues of \mathbf{M} are given by

$$\begin{aligned} & [(\tilde{A} - \omega)(B + \omega) - \gamma_k^2 \omega_{\text{ex}}^2] \\ & \times (D_1 - \omega)(D_2 + \omega) - F_A^2(B + \omega)(D_2 + \omega) \\ & - F_B^2(\tilde{A} - \omega)(D_1 - \omega) + F_A^2 F_B^2 = 0. \end{aligned} \quad (73)$$

With $F_A = F_B = 0$, Eq. (73) has the roots

$$\omega = x_1, D_1, -x_2, -D_2, \quad (74)$$

where

$$\begin{aligned} x_1 &= [\frac{1}{4}(\tilde{A} - B)^2 + \tilde{A}B - \omega_{\text{ex}}^2 \gamma_k^2]^{1/2} + \frac{1}{2}(\tilde{A} - B), \\ x_2 &= [\frac{1}{4}(\tilde{A} - B)^2 + \tilde{A}B - \omega_{\text{ex}}^2 \gamma_k^2]^{1/2} - \frac{1}{2}(\tilde{A} - B), \end{aligned} \quad (75)$$

with x_1 and x_2 the unperturbed electronic frequencies.

$$\mathbf{S} = \begin{vmatrix} \frac{\gamma_k \omega_{\text{ex}}(D_1 - \omega_1)\mu_1}{(\tilde{A} - \omega_1)(D_1 - \omega_1) - F_A^2} & \frac{\gamma_k \omega_{\text{ex}} F_A \mu_1}{(\tilde{A} - \omega_1)(D_1 - \omega_1) - F_A^2} & \mu_1 & \frac{F_B \mu_1}{D_2 + \omega_1} \\ \mu_2 & \frac{F_A \mu_2}{D_1 - \omega_2} & \frac{\gamma_k \omega_{\text{ex}}(D_2 + \omega_2)\mu_2}{(B + \omega_2)(D_2 + \omega_2) - F_B^2} & \frac{\gamma_k \omega_{\text{ex}} F_B \mu_2}{(B + \omega_2)(D_2 + \omega_2) - F_B^2} \\ \mu_3 & \frac{F_A \mu_3}{D_1 - \omega_3} & \frac{\gamma_k \omega_{\text{ex}}(D_2 + \omega_3)\mu_3}{(B + \omega_3)(D_2 + \omega_3) - F_B^2} & \frac{\gamma_k \omega_{\text{ex}} F_B \mu_3}{(B + \omega_3)(D_2 + \omega_3) - F_B^2} \\ \frac{\gamma_k \omega_{\text{ex}}(D_1 - \omega_4)\mu_4}{(\tilde{A} - \omega_4)(D_1 - \omega_4) - F_A^2} & \frac{\gamma_k \omega_{\text{ex}} F_A \mu_4}{(\tilde{A} - \omega_4)(D_1 - \omega_4) - F_A^2} & \mu_4 & \frac{F_B \mu_4}{D_2 + \omega_4} \end{vmatrix}, \quad (80)$$

where the μ 's satisfy

$$\begin{aligned} |\mu_j|^2 \left\{ 1 + \frac{F_A^2}{(D_1 - \omega_j)^2} - \frac{\gamma_k^2 \omega_{\text{ex}}^2 [(D_2 + \omega_j)^2 + F_B^2]}{[(B + \omega_j)(D_2 + \omega_j) - F_B^2]^2} \right\} &= +1, & j=2 \\ &= -1, & j=3 \\ |\mu_j|^2 \left\{ \frac{\gamma_k^2 \omega_{\text{ex}}^2 [(D_1 - \omega_j)^2 + F_A^2]}{[(\tilde{A} - \omega_j)(D_1 - \omega_j) - F_A^2]^2} - 1 - \frac{F_B^2}{(D_2 + \omega_j)^2} \right\} &= +1, & j=1 \\ &= -1, & j=4. \end{aligned}$$

C. Parallel Pumping

With a pumping field $h \sin \omega t$ along Oz, the interaction is

$$V(t) = -\hbar h \sin \omega t \left[\sum_{\alpha} (\gamma_e S_{\alpha}^z + \gamma_n I_{\alpha}^z) + \sum_{\beta} (\gamma_e S_{\beta}^z + \gamma_n I_{\beta}^z) \right].$$

For RbMnF₃, from Eq. (70)

$$\begin{aligned} D_1 &\simeq D_2 \simeq 4.3 \times 10^9 \text{ cps}, \\ x_1 x_2 &= \tilde{A}B - \omega_{\text{ex}}^2 \gamma_k^2 \simeq 3.9 \times 10^{21} \text{ cps} \quad \text{at } k=0, \end{aligned} \quad (76)$$

and we also have $x_1 \simeq x_2$.

Suppose with F_A and $F_B \neq 0$, the roots (74) go over into $\omega_1, \omega_2, \omega_3, \omega_4$, respectively. The corrections $(\omega_1 - x_1), (\omega_2 - D_1)$, etc., for the general case are given approximately in Appendix C.

For RbMnF₃, we may simplify further to obtain

$$\begin{aligned} \omega_1 &\simeq x_1 - \gamma_e \omega_n H_n (x_1^2 + \omega_{\text{ex}} \omega_n) / x_1^3, \\ \omega_2 &\simeq D_1 - \omega_n [1 - (1 + 2\gamma_e H_n \omega_{\text{ex}} / x_1 x_2)^{1/2}], \\ \omega_3 &\simeq -x_2 + \gamma_e \omega_n H_n (x_2^2 + \omega_{\text{ex}} \omega_n) / x_2^3, \\ \omega_4 &\simeq -D_2 + \omega_n [1 - (1 + 2\gamma_e H_n \omega_{\text{ex}} / x_1 x_2)^{1/2}]. \end{aligned} \quad (77)$$

Inserting the values in Eq. (70) it is seen that the fractional increase in the electronic frequencies (ω_1 and ω_3) is small ($\sim 7 \times 10^{-4}$) but the fractional decrease in the nuclear frequencies (ω_2 and ω_4) is substantial (~ 0.18).

The normal modes of the system will be given by

$$(\alpha_{k1}, \alpha_{k2}, \alpha_{k3}^\dagger, \alpha_{k4}^\dagger)^T = \mathbf{S}\mathbf{X}, \quad (78)$$

where \mathbf{S} is chosen so that¹⁶

$$\mathbf{S}\mathbf{M}\mathbf{S}^{-1} = \text{diag}(\omega_1, \omega_2, \omega_3, \omega_4),$$

and

$$\mathbf{S} \text{diag}(1, 1, -1, -1) \mathbf{S}^* = \text{diag}(1, 1, -1, -1), \quad (79)$$

(* denotes Hermitian conjugate). In terms of the eigenvalues,

By successive use of Eqs. (65), (66), (78), and (79), an expression of the form

$$V(t) = -\hbar h \sin \omega t \sum_k (\alpha_{k1}, \alpha_{k2}, -\alpha_{k3}^\dagger, -\alpha_{k4}^\dagger) [W_{ij}^{(k)}] (\alpha_{k1}^\dagger, \alpha_{k2}^\dagger, -\alpha_{k3}, -\alpha_{k4})^T$$

is obtained where terms diagonal in the α 's are omitted. Only the coefficients of $\alpha_{k1}^\dagger \alpha_{k4}^\dagger$ and $\alpha_{k2}^\dagger \alpha_{k3}^\dagger$ are of interest, corresponding to the joint excitation of nuclear and electronic magnons. Omitting the index k , one finds, for $ij=14$ or 23 ,

$$|W_{ij}|^2 = \frac{\{-\gamma_e [1 - (\omega_{ex} \gamma_k^2 G_i G_j / E_i E_j)] + \gamma_n [(\omega_{ex} \gamma_k^2 F_i^2 / E_i E_j) - (F_j^2 / H_i H_j)]\}^2}{\{[\omega_{ex}^2 \gamma_k^2 (G_i^2 + F_i^2) / E_i^2] - (F_j^2 / H_i^2) - 1\} \{1 + (F_j^2 / H_j^2) - [\omega_{ex}^2 \gamma_k^2 (G_j^2 + F_j^2) / E_j^2]\}},$$

where

$$\begin{aligned} E_{1,4} &\equiv (\tilde{A} - \omega_{1,4}) (D_1 - \omega_{1,4}) - F_A^2, \\ E_{2,3} &\equiv (B + \omega_{2,3}) (D_2 + \omega_{2,3}) - F_B^2, \\ G_{1,4} &\equiv D_1 - \omega_{1,4}; & G_{2,3} &\equiv D_2 + \omega_{2,3}, \\ H_{1,4} &\equiv D_2 + \omega_{1,4}; & H_{2,3} &\equiv D_1 - \omega_{2,3}, \\ F_1 &\equiv F_3 \equiv F_A; & F_2 &\equiv F_4 \equiv F_B. \end{aligned}$$

As for the ferromagnetic case, with $\omega \simeq \omega_1 + |\omega_4|$, we excite magnons in branches 1 and 4, and the rate equations are¹⁸

$$\dot{n}_{kj} = \frac{1}{4} h^2 |W_{14}|^2 (n_{k1} + n_{k4} + 1) \frac{(\Gamma_{k1} + \Gamma_{k4})}{(\omega - \omega_{k1} + \omega_{k4})^2 + \frac{1}{4} (\Gamma_{k1} + \Gamma_{k4})^2} - (n_{kj} - \bar{n}_{kj}) \Gamma_{kj}, \quad (j=1, 4)$$

where Γ_{kj} is the "linewidth" of branch j ; and the critical field is given by

$$h_c = \left\{ \frac{4\Gamma_{k1}\Gamma_{k4} [(\omega - \omega_{k1} + \omega_{k4})^2 + \frac{1}{4} (\Gamma_{k1} + \Gamma_{k4})^2]^{1/2}}{(\Gamma_{k1} + \Gamma_{k4})^2 |W_{14}^{(k)}|^2} \right\}_{\min k}.$$

With the data for RbMnF₃ [cf. (70)] as well as Eq. (77), $|W_{14}|^2$ can be simplified somewhat to

$$|W_{14}|^2 \simeq \frac{\gamma_e^2 \{ (1 - \gamma_k^2) - \gamma_k^2 [(\omega_{k1} + \omega_{k4}) / \omega_{ex}] \}^2}{[2\gamma_k^2 (\omega_{k1} / \omega_{ex}) - (1 - \gamma_k^2)] \{ (1 - \gamma_k^2) + [F^2 / (D_2 + \omega_{k4})^2] \}} \quad (81)$$

which is not as simple in its \mathbf{k} dependence as the corresponding expression for the ferromagnet.

For a rough order of magnitude to h_c , we have

$$h_c \sim (\Gamma_1 \Gamma_4)^{1/2} / |W_{14}^{(0)}|. \quad (82)$$

Taking $\Gamma_1 \sim 5 \times 10^8 \text{ sec}^{-1}$, $\Gamma_4 \sim 10^5 \text{ sec}^{-1}$, and from Eq. (81), $|W_{14}^{(0)}| \sim 4 \times 10^5 \text{ (G sec)}^{-1}$, we find $h_c \sim 25 \text{ Oe}$.

All of the numerical values of h_c given in this paper can be made smaller by reduction of the magnitude of the decay rate of the electronic magnon, a reduction which can be achieved by increase of sample perfection. In very good single crystals of yttrium iron garnet this decay rate approaches (as $k \rightarrow 0$) the value¹⁹ $1.5 \times 10^7 \text{ sec}^{-1}$. Use of this value would reduce our estimates of h_c to $\sim 5 \text{ Oe}$.

Even the larger critical fields, however, can readily be achieved with use of pulsing techniques. Provided the pulse time exceeds T_2 , critical absorption should be observed; and it should thus be possible to excite nuclear magnons with fairly substantial values of k .

APPENDIX A

We derive here the cubics [Eq. (46)] satisfied by the eigenvalues of \mathbf{M} .

¹⁸ With RbMnF₃, branches 2, 3 may probably also be excited because $\omega_1 + |\omega_4| \simeq |\omega_2| + \omega_3$.

¹⁹ T. Kasuya and R. C. LeCraw, Phys. Rev. Letters **6**, 223 (1961).

By slight rearrangement, the characteristic equation of \mathbf{M} may be written

$$\begin{vmatrix} \Gamma & \zeta \\ \zeta^T & \Lambda \end{vmatrix} = 0, \quad (A1)$$

where

$$\mathbf{\Gamma} = \begin{vmatrix} 0 & \tilde{A} - \omega & F & 0 \\ \tilde{A} - \omega & 0 & 0 & F \\ 0 & F & D - \omega & 0 \\ F & 0 & 0 & D - \omega \end{vmatrix},$$

$$\mathbf{\Lambda} = (\mathbf{B} + \omega) \mathbf{I},$$

$$\zeta = \gamma \begin{vmatrix} \zeta_{11} & \zeta_{12} & \zeta_{13} & \zeta_{14} \\ \zeta_{21} & \zeta_{22} & \zeta_{23} & \zeta_{24} \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{vmatrix},$$

and T means transpose. It is an exercise in Ref. 20, p. 102, that from the nature of the above matrices, Eq. (A1) gives²⁰

$$\det(\mathbf{\Gamma} \mathbf{\Lambda} - \zeta \zeta^T) = 0, \quad (A2)$$

²⁰ P. R. Halmos, *Finite-Dimensional Vector Spaces* (D. Van Nostrand Company, Princeton, New Jersey, 1958), 2nd ed.

provided $\omega \neq -B$. After performing the matrix multiplications, using definitions (42), Eq. (A2) becomes

$$\begin{vmatrix} -\gamma^2(\xi_e - \xi_0 + 2i\xi') & (\tilde{A} - \omega)(\omega + B) - \gamma^2(\xi_e + \xi_0) & F(\omega + B) & 0 \\ (\tilde{A} - \omega)(\omega + B) - \gamma^2(\xi_e + \xi_0) & -\gamma^2(\xi_e - \xi_0 - 2i\xi') & 0 & F(\omega + B) \\ 0 & F(\omega + B) & (D - \omega)(\omega + B) & 0 \\ F(\omega + B) & 0 & 0 & (D - \omega)(\omega + B) \end{vmatrix} = 0.$$

In turn this may be partitioned into 2×2 matrices and, by use of the same device as above, reduced to a 2×2 determinant from which we get

$$(D - \omega)[(\tilde{A} - \omega)(\omega + B) - \gamma^2(\xi_e + \xi_0)] - F^2(\omega + B) = \pm \gamma^2(D - \omega)[(\xi_e - \xi_0)^2 + 4\xi'^2]^{1/2},$$

hence Eq. (46). The remaining two roots must be $\omega = -B$.

APPENDIX B

We estimate the two nuclear frequencies for MnFe_2O_4 , as well as the electronic frequency ω_s near $k=0$. It is convenient to write

$$(\omega - \tilde{A})(\omega + B) + \gamma^2 = (\omega - x_1)(\omega - x_2),$$

where

$$x_1 = \frac{1}{2}(\tilde{A} - B) + [\frac{1}{4}(\tilde{A} - B)^2 + \tilde{A}B - \gamma^2\xi]^{1/2},$$

and

$$x_2 = \frac{1}{2}(\tilde{A} - B) - [\frac{1}{4}(\tilde{A} - B)^2 + \tilde{A}B - \gamma^2\xi]^{1/2}. \quad (\text{B1})$$

Setting $\omega = D + \eta$ in Eq. (46), we find that η satisfies

$$\eta^2[\eta + 2D - x_1 - x_2] + \eta[(D - x_1)(D - x_2) - F^2] - (D + B)F^2 = 0,$$

so that

$$\eta = \frac{-[(D - x_1)(D - x_2) - F^2]}{2(\eta + 2D - x_1 - x_2)} \left[1 \pm \left(1 + \frac{4(\eta + 2D - x_1 - x_2)(D + B)F^2}{[(D - x_1)(D - x_2) - F^2]^2} \right)^{1/2} \right]. \quad (\text{B2})$$

As long as $|\eta| \ll \tilde{A} - B \sim 12SJ/\hbar$, it can be neglected on the right-hand side. Now, using the fact that the greatest value of ξ is 72, we have

$$\frac{4(x_1 + x_2 - 2D)(D + B)F^2}{[(x_1 - D)(D - x_2) + F^2]^2} < \frac{4(\tilde{A} - B - 2D)(D + B)F^2}{[-D^2 + D(\tilde{A} - B) + \tilde{A}B - 72\gamma^2]^2} \approx \frac{4BF^2}{(\tilde{A} - B)[D + (\tilde{A}B - 72\gamma^2)/(\tilde{A} - B)]^2}. \quad (\text{B3})$$

Substitution of Eq. (40) in Eq. (B3) and use of the data given earlier for MnFe_2O_4 , together with $\langle I^z \rangle \sim 2 \times 10^{-2}$ and $(H_a + 2H_b) \sim 10^8$ Oe., shows that

$$\text{Eq. (B3)} \sim \frac{4A^2S\langle I^z \rangle}{[\hbar D - \gamma_e \hbar(H_a + 2H_b)]^2} \ll 1.$$

The square root in Eq. (B2) may therefore be expanded, giving the two solutions

$$\eta \approx \frac{(D + B)F^2}{(D - x_1)(D - x_2) - F^2}, \frac{(D - x_1)(D - x_2) - F^2}{2D - x_1 - x_2} - \frac{(D + B)F^2}{(D - x_1)(D - x_2) - F^2},$$

and hence, on further neglecting small terms and using (B1),

$$\omega \approx D - BF^2/[D(\tilde{A} - B) + \tilde{A}B - \gamma^2\xi] \quad (\text{B4})$$

or

$$\omega \approx \frac{1}{2}(\tilde{A} - B) - [\frac{1}{4}(\tilde{A} - B)^2 + \tilde{A}B - \gamma^2\xi]^{1/2} + \frac{BF^2}{D(\tilde{A} - B) + \tilde{A}B - \gamma^2\xi}. \quad (\text{B5})$$

The first solution (B4) represents the nuclear modes and is certainly consistent with the assumption $\eta \ll \tilde{A} - B$. The second solution (B5) need not be. It will however be consistent if $\xi \approx 72$ in which case $(\tilde{A}B - \gamma^2\xi) \ll (\tilde{A} - B)^2$ and ω will be small. The usual approximation for the square root is then possible:

$$\omega \approx -\frac{\tilde{A}B - \gamma^2\xi}{\tilde{A} - B} + \frac{BF^2}{D(\tilde{A} - B) + \tilde{A}B - \gamma^2\xi}. \quad (\text{B6})$$

This solution is negative. Since $\xi(k=0) = 72$ for the + sign in Eq. (47), the frequency (B6) is identified with ω_3 , and the approximation will be valid for small k . On substituting Eq. (40) in Eqs. (B4) and (B5) and neglecting small terms, we get Eqs. (49) and (50).

APPENDIX C

We estimate the electronic and nuclear frequencies for an antiferromagnet.

To obtain ω_1 , we put $\omega = x_1 + \eta_1$ in Eq. (73). Then

$$\begin{aligned} \eta_1^2 & [\eta_1^2 + (x_1 - D_1)(x_1 + D_2) + (2x_1 - D_1 + D_2)(x_1 + x_2) - (F_A^2 + F_B^2)] \\ & + \eta_1 [\eta_1^2(3x_1 + x_2 - D_1 + D_2) + (x_1 - D_1)(x_1 + D_2)(x_1 + x_2) - F_A^2(2x_1 + B + D_2) - F_B^2(2x_1 - \tilde{A} - D_1)] \\ & - [F_A^2(x_1 + B)(x_1 + D_2) + F_B^2(x_1 - \tilde{A})(x_1 - D_1) - F_A^2 F_B^2] = 0. \end{aligned} \quad (C1)$$

If we assume $\eta_1 \lesssim 10^8$ and neglect small terms in Eq. (C1), we get

$$\begin{aligned} \eta_1^2 & [(x_1 - D_1)(x_1 + D_2) + (2x_1 - D_1 + D_2)(x_1 + x_2)] \\ & + \eta_1(x_1 - D_1)(x_1 + D_2)(x_1 + x_2) - [F_A^2(x_1 + B)(x_1 + D_2) + F_B^2(x_1 - \tilde{A})(x_1 - D_1)] = 0. \end{aligned}$$

Hence

$$\begin{aligned} \eta_1 &= \frac{-(x_1 - D_1)(x_1 - D_2)(x_1 + x_2)}{2[(x_1 - D_1)(x_1 + D_2) + (2x_1 - D_1 + D_2)(x_1 + x_2)]} \\ & \times \left\{ 1 \pm \left[1 + \frac{4[(x_1 - D_1)(x_1 + D_2) + (2x_1 - D_1 + D_2)(x_1 + x_2)][F_A^2(x_1 + B)(x_1 + D_2) + F_B^2(x_1 - \tilde{A})(x_1 - D_1)]}{[(x_1 - D_1)(x_1 + D_2)(x_1 + x_2)]^2} \right]^{1/2} \right\} \end{aligned}$$

and one must usually take only the - sign to be consistent with $\eta_1 \lesssim 10^8$. This yields $\omega_1 = x_1 + \eta_1$.

In the same way, putting $\omega = D_1 + \eta_2$, $\omega = -x_2 + \eta_3$, and $\omega = -D_2 + \eta_4$ in Eq. (73), yields ω_2 , ω_3 , and ω_4 , where

$$\begin{aligned} \eta_2 &= \frac{-\omega_n(D_1 - x_1)(D_1 + x_2)}{[(D_1 - x_1)(D_1 + x_2) + 2\omega_n(2D_1 - x_1 + x_2)]} \left\{ 1 - \left[1 + \frac{2[(D_1 - x_1)(D_1 + x_2) + 2\omega_n(2D_1 - x_1 + x_2)]F_A^2(D_1 + B)}{\omega_n(D_1 - x_1)^2(D_1 + x_2)^2} \right]^{1/2} \right\}, \\ \eta_3 &= \frac{(x_2 + D_1)(x_2 - D_2)(x_1 + x_2)}{2[(x_2 + D_1)(x_2 - D_2) + (2x_2 + D_1 - D_2)(x_1 + x_2)]} \\ & \times \left\{ 1 - \left[1 + \frac{4[(x_2 + D_1)(x_2 - D_2) + (2x_2 + D_1 - D_2)(x_1 + x_2)][F_A^2(x_2 - B)(x_2 - D_2) + F_B^2(x_2 - \tilde{A})(x_2 + D_1)]}{[(x_2 + D_1)(x_2 - D_2)(x_1 + x_2)]^2} \right]^{1/2} \right\}, \\ \eta_4 &= \frac{\omega_n(D_2 + x_1)(D_2 - x_2)}{(D_2 + x_1)(D_2 - x_2) + 2\omega_n(2D_2 + x_1 - x_2)} \left\{ 1 - \left[1 + \frac{2[(D_2 + x_1)(D_2 - x_2) + 2\omega_n(2D_2 + x_1 - x_2)]F_B^2(\tilde{A} + D_2)}{\omega_n(D_2 + x_1)^2(D_2 - x_2)^2} \right]^{1/2} \right\}. \end{aligned}$$

APPENDIX D

In Sec. II, it was assumed that $\gamma_n > 0$ and that the hyperfine coupling constant A was large and positive. It is of interest to look at some other possibilities:

- $\gamma_n > 0$ and A "small", i.e., $\gamma_n \hbar H \gg |A| > 0$.
- $\gamma_n > 0$ and A large and negative [written $-A$, with $A > 0$].
- $\gamma_n < 0$, A large, positive.
- $\gamma_n < 0$, A large, negative.
- $\gamma_n < 0$, A "small."

In all cases we suppose there exists partial nuclear polarization as in Sec. IIB and that a linearization

procedure is permissible. However, the spin-wave approximation [of the type Eq. (24)] will have to be consistent with the equilibrium orientation of the electronic and nuclear spins as determined by (a)-(e). In any case, with H along the positive Z axis, the electronic spins will point down. Hence in (b), (d), and (e) the average nuclear spin also points down, and in (a) and (c) it points up.

Case (a). The large external field means that in the Hamiltonian equation (1), we still use the substitution of Eq. (24). Proceeding then as in Sec. IIB, one finds that all the results in that section hold except that now A may be positive or negative.

Case (b). The Hamiltonian is the same as Eq. (1) with A replaced by $-A$. The appropriate spin-wave

substitution is

$$S_i^+ = (2S)^{1/2} a_i^\dagger,$$

$$S_i^- = (2S)^{1/2} a_i,$$

$$S_i^z = -S + a_i^\dagger a_i;$$

$$I_i^+ = |2\langle I^z \rangle|^{1/2} b_i^\dagger,$$

$$I_i^- = |2\langle I^z \rangle|^{1/2} b_i,$$

$$I_i^z = \langle I^z \rangle + b_i^\dagger b_i;$$

yielding

$$\begin{aligned} \mathcal{H} = & C + \sum_{ij} 2SJ_{ij}(a_j^\dagger a_j - a_i^\dagger a_i) + (A | \langle I^z \rangle | - \gamma_e \hbar H) \\ & \times \sum_i a_i^\dagger a_i + (AS - \gamma_n \hbar H) \sum_i b_i^\dagger b_i - A | \langle I^z \rangle S |^{1/2} \\ & \times \sum_i (a_i b_i^\dagger + a_i^\dagger b_i), \end{aligned}$$

where

$$C = -S^2 \sum_{ij} J_{ij} - (\gamma_n \langle I^z \rangle - \gamma_e S) \hbar H N + AN \langle I^z \rangle S$$

and we retain only quadratic terms.

Applying next the Fourier transformations

$$a_k = N^{-1/2} \sum_i \exp(-i\mathbf{k} \cdot \mathbf{R}_i) a_i;$$

$$b_k = N^{-1/2} \sum_i \exp(-i\mathbf{k} \cdot \mathbf{R}_i) b_i$$

gives

$$\mathcal{H} = C + \sum_k [A_k a_k^\dagger a_k + B b_k^\dagger b_k - F(a_k b_k^\dagger + a_k^\dagger b_k)],$$

where

$$A_k = -\gamma_e \hbar H + A | \langle I^z \rangle | + J_0 - J_k,$$

$$B = AS - \gamma_n \hbar H,$$

$$F = A | \langle I^z \rangle S |^{1/2}.$$

\mathcal{H} may then be diagonalized by the canonical transformation

$$a_k = \alpha_k \cos \theta_k + \beta_k \sin \theta_k,$$

$$b_k = -\alpha_k \sin \theta_k + \beta_k \cos \theta_k,$$

where $\tan 2\theta_k = 2F/(A_k - B)$, and α and β are commuting Bose operators.

Finally, there results

$$\mathcal{H} = C + \sum_k (\epsilon_{k\alpha} \alpha_k^\dagger \alpha_k + \epsilon_{k\beta} \beta_k^\dagger \beta_k),$$

where

$$\epsilon_{k\alpha} = \frac{1}{2} \{ A_k + B + [(A_k - B)^2 + 4F^2]^{1/2} \},$$

$$\epsilon_{k\beta} = \frac{1}{2} \{ A_k + B - [(A_k - B)^2 + 4F^2]^{1/2} \}.$$

With $F/(A_k - B) \ll 1$, these become

$$\epsilon_{k\alpha} \simeq A_k + F^2/(A_k - B),$$

$$\epsilon_{k\beta} \simeq B_k - F^2/(A_k - B),$$

so that α and β are, respectively, the electronic and nuclear spin-wave branches.

If we now apply a pump field $\hbar \sin \omega t$ along the Z axis and proceed as in Sec. IIC we get the analog of Eq. (31):

$$V(t) = \frac{1}{2} (\gamma_n - \gamma_e) \hbar \sin \omega t \sum_k \sin 2\theta_k (\alpha_k^\dagger \beta_k + \alpha_k \beta_k^\dagger).$$

Hence by the perturbation theory

$$\begin{aligned} \dot{n}_{k\alpha} = -\dot{n}_{k\beta} \simeq & \frac{1}{3} \pi (\gamma_e - \gamma_n)^2 \hbar^2 \sin^2 2\theta_k (n_{k\beta} - n_{k\alpha}) \\ & \times \delta(\omega + \omega_{k\alpha} - \omega_{k\beta}), \end{aligned}$$

where $\hbar \omega_{k\alpha} = -\epsilon_{k\alpha}$; $\hbar \omega_{k\beta} = -\epsilon_{k\beta}$ [to preserve analogy with Eq. (32)]. With the modification for linewidth and dissipation, these become

$$\begin{aligned} \dot{n}_{k\alpha} = & \frac{1}{16} (\gamma_e - \gamma_n)^2 \hbar^2 \sin^2 2\theta_k (n_{k\beta} - n_{k\alpha}) \\ & \times \frac{\Gamma_{k\alpha} + \Gamma_{k\beta}}{(\omega + \omega_{k\alpha} - \omega_{k\beta})^2 + \frac{1}{4} (\Gamma_{k\alpha} + \Gamma_{k\beta})^2} - (n_{k\alpha} - \bar{n}_{k\alpha}) \Gamma_{k\alpha}, \\ \dot{n}_{k\beta} = & \frac{1}{16} (\gamma_e - \gamma_n)^2 \hbar^2 \sin^2 2\theta_k (n_{k\alpha} - n_{k\beta}) \\ & \times \frac{\Gamma_{k\alpha} + \Gamma_{k\beta}}{(\omega + \omega_{k\alpha} - \omega_{k\beta})^2 + \frac{1}{4} (\Gamma_{k\alpha} + \Gamma_{k\beta})^2} - (n_{k\beta} - \bar{n}_{k\beta}) \Gamma_{k\beta}, \end{aligned} \quad (D1)$$

which are of the form

$$\frac{d}{dt} \begin{pmatrix} n_\alpha \\ n_\beta \end{pmatrix} = \begin{pmatrix} -a - \Gamma_\alpha & a \\ a & -a - \Gamma_\beta \end{pmatrix} \begin{pmatrix} n_\alpha \\ n_\beta \end{pmatrix} + \begin{pmatrix} \bar{n}_\alpha \Gamma_\alpha \\ \bar{n}_\beta \Gamma_\beta \end{pmatrix},$$

where a is positive. For the square matrix on the right, the product of the eigenvalues is $(a + \Gamma_\alpha)(a + \Gamma_\beta) - a^2 > 0$, and the sum is $(-2a - \Gamma_\alpha - \Gamma_\beta) < 0$. The eigenvalues are negative and there are no unstable solutions. In fact the solutions decay exponentially with time, attaining equilibrium values \hat{n}_α , \hat{n}_β given by

$$\begin{pmatrix} \hat{n}_\alpha \\ \hat{n}_\beta \end{pmatrix} = \begin{pmatrix} a + \Gamma_\alpha & -a \\ -a & a + \Gamma_\beta \end{pmatrix}^{-1} \begin{pmatrix} \bar{n}_\alpha \Gamma_\alpha \\ \bar{n}_\beta \Gamma_\beta \end{pmatrix}.$$

One can see from the Eqs. (D1) that there is a kind of positive feedback: if $n_{k\alpha}$ increases, then $n_{k\beta}$ must decrease and the result is to reduce the rate of increase of $n_{k\alpha}$. This effect is due exclusively (if one ignores

damping) to conservation of angular momentum, and the assumption of the initial orientation of the nuclear spins. The presence of dissipation only makes it "more impossible" to obtain an instability.

Further because the solutions decay, there is no average power absorption.

Case (c) This goes through exactly as the problem considered in Sec. II, with the same conclusion.

Cases (d) and (e): These go through exactly as *Case (b)* and with the same conclusion.

Thus summarizing, pumping is possible for A large and positive, regardless of the sign of γ_n , or for $|A|$ small and $\gamma_n > 0$.²¹

²¹ We are indebted to Dr. E. Schlomann for pointing out this possibility (private communication).

Errata

Channeling in Diamond-Type and Zinc-Blende Lattices: Comparative Effects in Channeling of Protons and Deuterons in Ge, GaAs, and Si, A. R. SATTLER AND G. DEARNALEY [Phys. Rev. **161**, 244 (1967)]. The equation in Fig. 12 is incorrect. C should be replaced by $C' = C/A$. Table IV contains a tabulation of C' values (not C values).

Dynamical Spin Correlations in Many-Spin Systems. I. The Ferromagnetic Case, RAZA A. TAHIR-KHELIL [Phys. Rev. **159**, 439 (1967)]. In Eq. (C5) the first term on the left side of the second equality should be $\mathbf{k}^2 \mathcal{D}_{\mathbf{k}}^{(2)}$ rather than $\mathbf{k}^4 \mathcal{D}_{\mathbf{k}}^{(2)}$. The second RPA(II) expressions for the longitudinal Green's function, and consequently those of the longitudinal correlation function, should be reinterpreted as being the principal-value limits obtained when in the Green's function $\langle\langle S^+(1) S^-(1') \tilde{S}^z(3) \rangle\rangle$ the time τ_1 approaches the time τ_1' from below and from above. In other words, Eq. (B1) should read

$$M_{\mathbf{k}}^{(1)}(\nu) Z_{\nu} = \lim_{\epsilon \rightarrow -i\beta\Delta, \Delta=0} (-1/2\beta N) \sum_{\lambda}' \sum_{\rho} J_{++}(\lambda, k-\lambda) \\ \times [\exp(+iZ_{\rho}\epsilon) + \exp(-iZ_{\rho}\epsilon)] [Z_{\nu} + E_{k-\lambda} - E_{\lambda}]^{-1} \\ \times \{2M_{\mathbf{k}}^{(1)}(\nu) [G_{\lambda-k}(\rho-\nu) J_{0+}(\mathbf{k}, \mathbf{k}-\lambda) \\ - G_{\lambda}(\rho) J_{0+}(\mathbf{k}, \lambda)] + G_{\lambda}(\rho) - G_{\lambda-k}(\rho-\nu)\}.$$

Similarly, Eq. (B6) should read

$$[M_{\mathbf{k}}^{(1)}(\nu)]_{\text{kinematical sum rule}^{2\text{nd RPA (II)}}} \\ = \lim_{S=1/2, \epsilon \rightarrow -i\beta\Delta, \Delta=0} (-1/2\beta N) \sum_{\lambda}' \sum_{\rho} G_{\mathbf{k}-\lambda, \lambda}^{(1)}(\nu-\rho, \rho) \\ \times [\exp(+iZ_{\rho}\epsilon) - \exp(-iZ_{\rho}\epsilon)].$$

These prescriptions lead to the following unique results

$$[M_{\mathbf{k}}^{(1)}(\nu)]_{\text{dynamical sum rule}^{2\text{nd RPA (II)}}} = A_{\mathbf{k}}(\nu)/B_{\mathbf{k}}'(\nu),$$

$$[M_{\mathbf{k}}^{(1)}(\nu)]_{\text{kinematical sum rule}^{2\text{nd RPA (II)}}} = e_{\mathbf{k}}(\nu)/h_{\mathbf{k}}'(\nu),$$

where $A_{\mathbf{k}}(\nu)$ and $e_{\mathbf{k}}(\nu)$ are the same as given in Eqs. (B3) and (B7b) and where $B_{\mathbf{k}}'(\nu)$ is obtained from (B4a) by the relation

$$B_{\mathbf{k}}'(\nu) = \frac{1}{2} \sum_{j=1}^2 B_{\mathbf{k}}^{(j)}(\nu).$$

Similarly, $h_{\mathbf{k}}'(\nu)$ is obtained from Eqs. (B7c) and (B8b) by the relation

$$h_{\mathbf{k}}'(\nu) = \frac{1}{2} [h_{\mathbf{k}}^{(+)}(\nu) + h_{\mathbf{k}}^{(-)}(\nu)]$$