MASTER

Investigations of Nuclear and Electronic
Spin Waves by Parallel Pumping

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I. INTRODUCTION

A. Introduction

The concept of nuclear spinwaves is the result of attempts to explain seemingly anomalous nuclear magnetic resonance (NMR) data of Mn^{++} in antiferromagnetic $KMnF_3$. In a ferromagnet or antiferromagnet with a hyperfine interaction the MNR frequency which is measured is in disagreement with that predicted by the following simple argument.

One first represents the hyperfine interaction by

$$\mathcal{H}_{hf} = A \underline{I} \cdot \underline{S} \quad (1.1)$$

where A is the hyperfine constant, $\underline{I}$ and $\underline{S}$ are the nuclear and electronic spins, respectively. This energy may be thought of as arising from a nuclear spin in an average hyperfine field as follows

$$\mathcal{H}_{hf} = \underline{H}_{nn} \cdot \underline{I} \quad (1.2)$$

This implies a precessional frequency of the nuclei in this field of

$$\frac{H_{nn}}{\gamma} = \omega_n = A \langle S \rangle \quad (1.3)$$

where $\langle \rangle$ represents thermal average and γ is the nuclear gyromagnetic ratio. (we take $\hbar = 1$ throughout).

At low temperatures $\langle S \rangle \approx S$. The assumption has been made that H_{nn} is much greater than the external applied field H . It should be noted that one has tacitly assumed that there is no electronic-nuclear

correlation. In other words, the electronic spin is held rigid while the nuclear spin precesses in the hyperfine field caused by the electron.

Electron paramagnetic resonance studies of Mn^{++} in a diamagnetic host can yield a value for A .² This value of A predicts an NMR frequency much higher than that which is actually measured³ in $KMnF_3$. To account for this the coupled electron-nuclear motion must be considered. In a system in which the electronic spin system is tightly coupled, such as $KMnF_3$, the possibility immediately arises that one precessing nucleus can disturb the whole electronic spin system. If one nuclear spin disturbs a neighboring electronic spin then the neighboring nuclear spin is also disturbed because of the strong hyperfine coupling. The net effect is one in which nuclei can "communicate" with other nuclei via the electronic system. When such a collective effect is quantized the result is a so called nuclear magnon or nuclear spinwave.⁴ Nuclear spinwaves differ from electronic spinwaves in that the nuclear spin system is far from ordered, in fact at 4.2°K $\langle I_z \rangle / I$ is of the order of one percent; however, a well defined spectrum is proved to exist.⁵

The concept of nuclear spinwaves predicts accurately the value of the NMR ($k = 0$) frequency and provides answers to other problems encountered with high power nonlinear absorption in $KMnF_3$ as well as other related compounds, such as $RbMnF_3$ and $CsMnF_3$. Nuclear spinwaves, like electronic spinwaves, are characterized by a specific relationship between frequency ω and wavevector k . Conventional NMR can yield information only about the $k = 0$ spinwave while the parallel pumping⁶ experiment can be used to measure lifetimes of spinwaves of arbitrary wavevector.

The powerful tool of parallel pumping was discounted by previous

authors as a possibility for investigation of nuclear spinwave relaxation rates.¹ The process they envisioned is one in which one photon creates two nuclear magnons by direct coupling of the rf field to the nuclear spin system. Impossibly high rf powers are required for this excitation. In another attempt to show the feasibility of exciting nuclear spinwaves by parallel pumping Nino and Keffer⁷ considered the case of one photon exciting simultaneously one nuclear and one electronic spinwave. They predicted the threshold field for such a process to be just within experimental limits. Their theory, however, when applied to the antiferromagnet RbMnF_3 holds only for very low external fields in which case the magnetization vectors are approximately parallel and antiparallel to the external field, and they also neglected the possibility of exciting nuclear spinwaves alone. Hinderks and Richards^{8,9} considered the case of RbMnF_3 in an external field larger than the field required to "flop" the sublattice magnetization vectors approximately perpendicular to the external field. In this "flopped" configuration they assumed the parallel pumping rf to couple only to the electronic spin system. In the flopped configuration the spins are canted slightly toward the external field direction. This canting allows parallel pump processes in which one photon creates two electronic, one nuclear and one electronic and two nuclear spinwaves. Also, they showed experimental results of the parallel pumping of one nuclear and one electronic spinwave in RbMnF_3 . M.H. Seavey¹⁰ independently considered the possibility of parallel pumping two electronic spinwaves in the flopped antiferromagnet CsMnF_3 . Later, Adams, Hinderks, and Richards¹¹ reported excitation of two nuclear spinwaves by parallel pumping in CsMnF_3 . Morgenthaler and

Platzker¹² point out that one must include effects of Suhl first order processes because of the ability of canted antiferromagnets to absorb rf power linearly when the rf field is parallel to the external magnetic field.

In this work parallel pumping experiments on CsMnF_3 and RbMnF_3 are reported and relaxation rates inferred. We analyze the problem of the Suhl first order process as well as the parallel pumping process in the context of the Holstein-Primakoff¹³ formalism and arrive at generally the same results as Morgenthaler and Platzker.¹² One parallel pumping experiment is performed to confirm directly the necessity for including the Suhl first order process.

These experiments represent the first direct confirmation of nuclear spinwaves. The experimental methods used to excite two nuclear spinwaves by parallel pumping represent totally new techniques in microwave spectrometers at frequencies in the 1000 MHz range. This technique, which uses a resonant helix rather than the conventional coaxial cavity, permits high sensitivity because of the large ratio of sample volume to volume of the rf fields.

B. Mode Geometries

The basic mode patterns of RbMnF_3 and CsMnF_3 in the flopped state are shown in Fig. 1-1. The two magnetization vectors precess in opposite directions much like a simple antiferromagnet. In the simple antiferromagnet there exist two degenerate normal mode frequencies.¹⁴ However, because of the canting this degeneracy is broken up in a canted antiferromagnet and hence, there are two distinct normal mode frequencies corresponding to two nearly orthogonal motions. Also, in the flopped

Fig. 1-1 Elliptical precession of sublattice magnetizations about equilibrium directions, canted at an angle θ_c with respect to a plane perpendicular to H . The axes of ellipse are denoted by a and b .

state, the magnetization vectors precess in an ellipse rather than a circle. In the absence of any hyperfine interaction and for a substance with uniaxial anisotropy, the resonance frequencies for the two modes are approximately

$$\begin{aligned}\Omega_1 &= \gamma H \\ \Omega_2 &= \gamma (H_E H_A)^{1/2}\end{aligned}\quad (1.4)$$

where γ is the gyromagnetic ratio and H is the external field. H_E and H_A are the exchange and anisotropy fields. The assumption has been made that $H_E \gg H$. It is assumed that a plane perpendicular to the uniaxial axis, which we take as the y -direction, is an easy plane of anisotropy and that H_A stabilizes the electronic spins in this plane.

It is interesting to calculate with a simple method the actual degree of ellipticity, since it is this ellipticity which gives rise to parallel pumping.

The energy of the system is taken to be

$$E = \lambda \vec{M}_A \cdot \vec{M}_B - H \cdot (\vec{M}_A + \vec{M}_B) + \frac{H_A}{2M} (M_A^y{}^2 + M_B^y{}^2) \quad (1.5)$$

with the condition that $|\vec{M}_A| = M = |\vec{M}_B|$. The position of the magnetization vectors at various times are shown in Fig. 1-2 and 1-3 for the field dependent mode with frequency Ω_1 and the field independent mode with frequency Ω_2 respectively. Reference to Fig. 1-2a shows for the field dependent mode that

$$E(t=0) - E_0 = M H \alpha^2 \sin \theta_c \quad (1.6)$$

Fig. 1-2 Field dependent mode. (a) Magnetizations at time $t = 0$.
 (b) Magnetizations at time $t = \pi/2\Omega_1$.

Fig. 1-3 Field independent mode. (a) Magnetizations at time $t = 0$.
(b) Magnetizations at time $t = \pi/2\Omega_1$.

where E_0 is the energy in the equilibrium position. It is assumed that $\alpha \ll 1$. With the help of Fig. 1-2b, one has

$$E(t = \pi/2\Omega_1) - E_0 = 2M H_E \beta^2 \quad (1.7)$$

where $\lambda M = H_E$ and it is assumed $\beta \ll 1$ and $H_E \gg H_A$. Using the fact that the precession is one of constant energy one can equate Eqs. (1.6) and (1.7) yielding

$$\frac{\alpha^2}{\beta^2} = \frac{2 H_E}{H \sin \theta_c} \quad (1.8)$$

Elementary considerations show

$$\sin \theta_c = H/2H_E \quad (1.9)$$

Combining Eqs. (1.4), (1.8) and (1.9) yields

$$\frac{\alpha}{\beta} = \frac{2\Gamma H_E}{\Omega_1} \gg 1$$

Hence, there is large ellipticity in the motion of the sublattice magnetizations. Since the magnitude of M_A or M_B is constant and the motion is an ellipse of precessional frequency Ω_1 about the equilibrium direction, the projection of a sublattice magnetization vector on the equilibrium direction must have a $2\Omega_1$ time dependence. If this is now projected on to the z axis as shown in Fig. 1-4a, the z component of total magnetization M_T^z must also have a $2\Omega_1$ time dependence. The ratio

Fig. 1-4 Perspective view of total magnetization. (a) Field dependent mode. (b) Field independent mode.

of the maximum M_T^Z to the minimum M_T^Z is derived in the following way.

The minimum value of M_T^Z corresponds to the situation in Fig. 1-2a.

In this position it is clear that

$$\begin{aligned} M_T^Z(\min) &= M_A^Z + M_B^Z \\ &= M \sin(\theta_c - \alpha) + M \sin(\theta_c + \alpha) \end{aligned} \quad (1.10)$$

$$M_T^Z(\min) = 2M \sin \theta_c \cos \alpha$$

The maximum value of M_T^Z corresponds to Fig. 1-2b. In this position one has

$$M_T^Z(\max) = M_A^Z + M_B^Z \quad (1.11)$$

$$M_T^Z(\max) = 2M \sin \theta_c$$

Therefore, the ratio of $M_T^Z(\max)$ to $M_T^Z(\min)$ is given by

$$\frac{M_T^Z(\max)}{M_T^Z(\min)} = \frac{1}{\cos \alpha} \approx 1 + \frac{\alpha^2}{2} \quad (1.12)$$

Postulating that the motion of M_T is an ellipse, it is instructive to calculate the actual amount of ellipticity. M_T^X corresponding to the x component of M_T in Fig. 1-2a is

$$M_T^x = M_A^x + M_B^x$$

$$= M \cos(\theta_c - \alpha) - M \cos(\theta_c + \alpha)$$

$$M_T^x = 2M \sin \theta_c \sin \alpha \quad (1.13)$$

M_T^y , defined as the y component of $\vec{M}_T$ in Fig. 1-2b is

$$M_T^y = M_A^y + M_B^y$$

$$M_T^y = 2M \cos \theta_c \sin \beta \quad (1.14)$$

Hence, the ratio of the axes of the ellipse is

$$\frac{M_T^y}{M_T^x} = \tan \theta_c \frac{\sin \alpha}{\sin \beta} = 1 \quad (1.15)$$

where use has been made of Eqs. (1.8) and (1.9) and θ_c , α and β are assumed to be much less than unity. The motion is therefore circular. Eqs. (1.12) and (1.15) imply that the magnitude of $\vec{M}_T$ is not constant, as is commonly the case in ferromagnets. We restate that the magnitudes of the individual sublattice magnetizations $\vec{M}_A$ and $\vec{M}_B$ are of course constant.

The rate at which energy is absorbed is given by

$$P_{abs} = \left(\int \hbar \cdot \frac{dM}{dt} dv \right)_{\text{time avg.}} \quad (1.16)$$

Only if h_x or h_y has an $e^{i\Omega_1 t}$ time dependence, will it survive the time average. Also h_z must have an $e^{2i\Omega_1 t}$ time dependence to survive the time average. These represent transverse or resonant pumping and parallel or parametric pumping respectively.

A similar analysis can be given for the field independent mode (see Fig. 1-3).

$$E(t=0) - E_0 = 2 M H_E \alpha^2 \quad (1.17)$$

$$E(t=\pi/2\Omega_2) - E_0 = M H_A \beta^2 \quad (1.18)$$

Equating (1.17) and (1.18) for the same reason discussed preceding Eq.

(1.8) yields

$$\frac{\alpha}{\beta} = \frac{(2 H_E H_A)^{1/2}}{2 H_E} = \frac{\Omega_2}{2 \Gamma H_E} \ll 1 \quad (1.19)$$

Again, the motion of an individual magnetization vector is highly elliptical, but careful inspection shows that the total of the two magnetization vectors does not precess at all, but only moves up and down the z axis as shown in Fig. 1-4b. Referring to Eq. (1.16), it is clear that rf power will be absorbed by this mode only if h_{rf} is in the

z direction. An $e^{i\Omega_2 t}$ time dependence for h_z will survive the time average and it is this absorption which gives rise to resonant pumping. If M_T^z has a Fourier component at $2\Omega_2$ then power can be absorbed by an h_z with an $e^{2i\Omega_2 t}$ time dependence and the parallel pumping type of parametric excitation can occur. It is the ability of the field independent mode to absorb both linearly and parametrically when the driving rf is applied parallel to the external field which complicates the parallel pumping problem. This problem is discussed in Chapter II when the parallel pumping thresholds are considered.

II. A DETAILED TREATMENT OF NUCLEAR AND ELECTRONIC SPINWAVES IN A CANTED ANTIFERROMAGNET

A. Normal Modes

The two antiferromagnets RbMnF_3 and CsMnF_3 have the common features of canting and low anisotropy. These lead to pronounced effects on the normal modes of their spin systems. The effects are, briefly, relatively low normal mode frequencies and high ellipticity of precession.¹⁸ Another interesting feature is the large hyperfine interaction which results in a high degree of coupling between the low and high frequency branches of their frequency spectrum.¹ This is the so called "pulling" effect. To explain these features and show the feasibility of nonlinear excitation in these substances, the normal modes of the spin systems must be calculated.

The ground state of the system is assumed to be an approximately antiparallel arrangement of the magnetic spins and small departures from this ground state can be approximated by a noninteracting spinwave formalism. The methods of Holstein and Primakoff are used to derive the normal modes.¹³

The Hamiltonian for the system is considered to be a sum of the exchange, Zeeman, anisotropy and hyperfine terms. For simplicity, only the "flopped" configuration of the spins is considered. The equilibrium position of the spins is shown in Fig. 2-1. This "flopped" position becomes energetically more favorable when the external applied field exceeds H_{SF} defined by

$$H_{SF} = \left(\frac{3}{2} H_E H_A \right)^{1/2} \quad (2.1)$$

where H_E and H_A are the exchange and anisotropy fields respectively. The conditions under which Eq. (2.1) may be expressed in this form are discussed in section B. Eq. (2.1) holds in RbMnF_3 when the external field is along the $\langle 100 \rangle$ crystal direction at low temperatures.¹⁵ In CsMnF_3 the in-plane anisotropy is approximately zero and, as a result, the sublattice magnetizations flop upon the application of any field in the basal plane.¹⁶ In both substances the canting angle θ_c is¹⁴

$$\theta_c = H / 2H_E \quad (2.2)$$

The large hyperfine interaction forces the nuclear spins to align parallel to the electronic spins and thus the nuclear Zeeman contribution to the Hamiltonian can be ignored, since laboratory applied fields are much weaker than the hyperfine fields seen by the nuclei. The additional canting of the nuclei due to their interaction with the external field is¹⁷

$$\theta \approx H / H_{nn}$$

where H_{nn} is the field seen at the nucleus and is typically 650×10^3 Oe. RbMnF_3 is adequately described by a two sublattice model¹⁸ while CsMnF_3 is most accurately described by a six sublattice model.¹⁹

The following sections are devoted to a derivation of the normal modes and instability thresholds for a two sublattice antiferromagnet with cubic or uniaxial anisotropy. In the cubic case, the external field is assumed to be along a $\langle 100 \rangle$ direction. For the uniaxial case, the field is applied in the basal plane which is considered an easy plane of anisotropy. The methods used to derive normal modes for the two sublattices can in principle be applied to the full six sublattice

system, but the results for the simpler two sublattice model show all the pertinent features of the canted antiferromagnet with a large hyperfine interaction.

It is important to be particularly careful with the three magnon terms which arise from the exchange interaction because these may lead to a nonlinear process in which a uniform precession magnon decays into two $k \neq 0$ spinwaves much like the Suhl first order process.²⁰ Platzker and Morganthaler¹² originated this concept, which has been ignored by previous authors.^{9,21} The following sections show that certain first order Suhl-like processes are indeed possible in the framework of the Holstein Primakoff formulation and cannot be neglected.

B. Approximating The Hamiltonian With Spinwave Operators

The Hamiltonian for the system is written in the form ($\hbar = 1$)

$$\mathcal{H} = \gamma H \left(\sum_i S_{iz} + \sum_j S_{jz} \right) + \sum_{ij} J(R_{ij}) \underline{S}_i \cdot \underline{S}_j \quad (2.3)$$

$$- A \left(\sum_i \underline{I}_i \cdot \underline{S}_i + \sum_j \underline{I}_j \cdot \underline{S}_j \right) + \mathcal{H}_{anis}$$

where the i and j refer to the i th and j th lattice site on different sublattices. H is the external field applied in the z direction as shown in Fig. 2-2 and γ is the electronic gyromagnetic ratio. γ is assumed to be positive and the sign is taken into account explicitly in Eq. (2.3). $\underline{S}$ and $\underline{I}$ are the electronic and nuclear spin operators respectively. The terms represent in order the Zeeman, exchange, hyperfine and anisotropy Hamiltonians.

Super-exchange acting through the F^- ions is thought to be the cause of the antiferromagnetic alignment. The magnitude of the exchange

Fig. 2-1 (a) Sublattice magnetization in normal or unflopped state.
 (b) Sublattice magnetizations in flopped state with magnetizations canted at an angle θ_c .

Fig. 2-2 Coordinate system used for normal mode calculation. z'' , z' , x'' and x' lie in the x - z plane.

is proportional to J . The J in Eq. (2.3) is taken as positive, and the sign of J is explicitly taken into account to make the electronic spin system favor antiparallel alignment. The hyperfine constant is also taken to be positive. The explicit sign in Eq. (2.3) is chosen to make the nuclear and electronic spins parallel as is the case in RbMnF_3 and CsMnF_3 . It is interesting to note that various authors assume the electronic spins to be parallel as well as antiparallel.^{7,22,23}

The relative direction of the spins is ultimately controlled by the sign of the hyperfine constant A . Such a difference will only manifest itself at high applied magnetic fields and does not affect the results to be presented here.

The anisotropy Hamiltonian takes the form of an appropriate power series expansion of the direction cosines compatible with crystal symmetry. For the cubic RbMnF_3 only the lowest order terms need be kept.

$$\mathcal{H}_{\text{anis}} = \frac{K_c}{S^4} \left[\sum_i (S_{ix}^2 S_{iy}^2 + S_{iy}^2 S_{iz}^2 + S_{iz}^2 S_{ix}^2) + \sum_j (S_{jx}^2 S_{jy}^2 + S_{jy}^2 S_{jz}^2 + S_{jz}^2 S_{jx}^2) \right] \quad (2.4)$$

where $2K_c/S = \Gamma H_A$. If the anisotropy field is described in this manner, then Eq. (2.1) will hold. The form in uniaxial CsMnF_3 is

$$\mathcal{H}_{\text{anis}} = \frac{K_u}{S^2} \left(\sum_i S_{iy}^2 + \sum_j S_{jy}^2 \right) \quad (2.5)$$

where $2K_u/S = \Gamma H_A$. For both types of anisotropy the coordinates are shown in Fig. 2-2.

The electronic part of the Hamiltonian can be expressed in terms of boson operators, using the coordinate system in Fig. 2-2 and defining

spin deviation operators as follows

$$\begin{aligned}
 S_i^+ &= S_{ix'} + i S_{iy'} = (2S)^{1/2} a_i \\
 S_i^- &= S_{ix'} - i S_{iy'} = (2S)^{1/2} a_i^+ \\
 S_j^+ &= S_{jx''} + i S_{jy''} = (2S)^{1/2} b_j \\
 S_j^- &= S_{jx''} - i S_{jy''} = (2S)^{1/2} b_j^+
 \end{aligned}
 \tag{2.6}$$

$$\begin{aligned}
 S_{iz'} &= S - a_i^+ a_i \\
 S_{jz''} &= S - b_j^+ b_j
 \end{aligned}
 \tag{2.7}$$

where the subscripts i and j refer to the A and B sublattices respectively. This approximation is valid at low temperatures. Further, only nearest neighbor exchange will be assumed. Defining the spatial Fourier transforms in the usual way

$$\begin{aligned}
 a_k &= N^{-1/2} \sum_i a_i e^{-i\mathbf{k} \cdot \mathbf{r}_i} \\
 b_k &= N^{-1/2} \sum_j b_j e^{-i\mathbf{k} \cdot \mathbf{r}_j}
 \end{aligned}
 \tag{2.8}$$

with N equal to the number of spins on a sublattice, allows one to write the exchange Hamiltonian

$$\mathcal{H}_{ex} = \sum_{i,j} J(R_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j
 \tag{2.9}$$

in the form keeping only two magnon terms

$$\mathcal{H}_{ex} = \sum_K \left\{ \Gamma H_E \cos 2\theta_c (a_K^\dagger a_K + b_K^\dagger b_K) \right. \\ \left. + \frac{1}{2}(1 - \cos 2\theta_c) \Gamma H_E \gamma_K (a_K b_K^\dagger + a_K^\dagger b_K) \right. \\ \left. - \frac{1}{2}(1 + \cos 2\theta_c) \Gamma H_E \gamma_K (a_K b_{-K} + a_K^\dagger b_{-K}^\dagger) \right\} \quad (2.10)$$

with

$$\Gamma H_E = S \sum_z J(R_z) \quad (2.11)$$

$$\gamma_K = \frac{\sum_z J(R_z) e^{-i\vec{K} \cdot \vec{R}_z}}{\sum_z J(R_z)} \quad (2.12)$$

where z represents the nearest neighbors. Thus, γ_K is a function of the geometry of the crystal.

The electronic Zeeman Hamiltonian

$$\mathcal{H}_z = \Gamma H (\sum_i S_{iz} + \sum_j S_{jz}) \quad (2.13)$$

with H in the z direction can be expressed as follows again keeping only two magnon terms

$$\mathcal{H}_z = \Gamma H \sin \theta_c \sum_K (a_K^\dagger a_K + b_K^\dagger b_K) \quad (2.14)$$

The anisotropy Hamiltonian given by either Eq. (2.4) or (2.5) can be expressed in the form

$$\mathcal{H}_{anis} = \sum_K \left\{ E (a_K^\dagger a_K + b_K^\dagger b_K) \right. \\ \left. + \frac{D}{2} (a_K a_{-K} + a_K^\dagger a_{-K}^\dagger + b_K b_{-K} + b_K^\dagger b_{-K}^\dagger) \right\} \quad (2.15)$$

In terms of Eq. (2.4), D and E are

$$D = \frac{9}{4} \frac{K_C}{S} = \frac{9}{8} \Gamma H_A \quad (2.16)$$

$$E = \frac{3}{4} \frac{K_C}{S} = \frac{3}{8} \Gamma H_A$$

For Eq. (2.5), D and E are

$$D = -\frac{K_U}{S} = -\frac{\Gamma H_A}{2} \quad (2.17)$$

$$E = \frac{K_U}{S} = \frac{\Gamma H_A}{2}$$

The hyperfine interaction is represented by the Hamiltonian³²

$$\mathcal{H}_{\text{hf}} = -A \left(\sum_i \vec{S}_i \cdot \vec{I}_i + \sum_j \vec{S}_j \cdot \vec{I}_j \right) \quad (2.18)$$

The electronic variables are converted to spinwave variables by Eq. (2.6) through (2.8). The nuclear variables are approximated by boson operators in the following way. Consider the vectors $\vec{J}_i$ and $\vec{J}_j$ defined by

$$|\vec{J}_i| = \langle I_{iz} \rangle = \langle I_{jz''} \rangle = |\vec{J}_j|$$

$\langle I_{iz} \rangle$ equals $\langle I_{jz''} \rangle$ because of the identical hyperfine coupling of each sublattice. The bars represent magnitude and the brackets mean thermal averages. In equilibrium $\vec{J}_i$ and $\vec{J}_j$ are in the z' and z'' directions respectively. Small departures from equilibrium of the vectors $\vec{J}_i$ and $\vec{J}_j$ are handled similarly to Eq. (2.6) through (2.8)

$$J_i^+ = J_{ix'} + i J_{iy'} = (2\langle I_z \rangle)^{1/2} \alpha_i$$

$$J_j^+ = J_{jx''} + i J_{jy''} = (2\langle I_z \rangle)^{1/2} \beta_j \quad (2.20)$$

$$J_{iz'} = \langle I_{iz'} \rangle - \alpha_i^+ \alpha_i \quad (2.21)$$

$$J_{jz''} = \langle I_{jz''} \rangle - \beta_j^+ \beta_j$$

where the new variables α and β satisfy boson commutation relations.

The Fourier transforms are given by

$$\alpha_k = N^{-1/2} \sum_i \alpha_i e^{-i\mathbf{k} \cdot \mathbf{r}_i} \quad (2.22)$$

$$\beta_k = N^{-1/2} \sum_j \beta_j e^{-i\mathbf{k} \cdot \mathbf{r}_j}$$

Replacing I_z by a vector which has magnitude $\langle I_z \rangle$ was first done by DeGennes, Pincus, Hartman-Boutron, and Winter. In this approximation the very sizeable fluctuations in I_z , as evidenced by a small degree (1%) of nuclear polarization, are ignored; so all that remains is a vector of length $\langle I_z \rangle \ll \langle I_z^2 \rangle^{1/2}$. A similar approach is to employ the random phase approximation (RPA). The RPA method leads to exactly the same results as the approach used here.

In the RPA the Fourier transforms of the following operators are considered

$$\begin{aligned} I_{KA}^+ &= N^{-1/2} \sum_i I_{Ai}^+ e^{-i\mathbf{k} \cdot \mathbf{r}_i} \\ I_{KB}^+ &= N^{-1/2} \sum_j I_{Bj}^+ e^{-i\mathbf{k} \cdot \mathbf{r}_j} \end{aligned} \quad (2.23)$$

$$\begin{aligned} I_{KA}^{z'} &= N^{-1/2} \sum_i I_{iz'} e^{-i\mathbf{k} \cdot \mathbf{r}_i} \\ I_{KB}^{z''} &= N^{-1/2} \sum_j I_{jz''} e^{-i\mathbf{k} \cdot \mathbf{r}_j} \end{aligned} \quad (2.24)$$

where the A and B refer to the A and B sublattices. Thus i and j refer

to lattice sites on sublattices A and B. In the RPA, $I_{kA}^{z'}$ and $I_{kB}^{z''}$ are replaced by their thermal averages thus

$$\begin{aligned} I_{kA}^{z'} &\rightarrow \langle I_{kA}^{z'} \rangle \\ I_{kB}^{z''} &\rightarrow \langle I_{kB}^{z''} \rangle \end{aligned} \quad (2.25)$$

$$\langle I_{kA}^{z'} \rangle = \langle I_{kB}^{z''} \rangle = N^{1/2} \langle I_z \rangle \delta_{k0}$$

with δ_{k0} being Kronecker delta function. The Fourier transforms of the usual nuclear spin commutation relations are

$$[I_{kA}^+, I_{qA}^-] = 2N^{-1/2} I_{(k-q)A}^{z'} \quad (2.26)$$

$$[I_{kA}^\pm, I_{qA}^{z'}] = \mp N^{-1/2} I_{(k\pm q)A}^\pm$$

These can satisfy boson like commutation relations only in the RPA. New boson operators α and β can be defined as before and small departures of these operators from $\langle I_z \rangle$ are considered.

The hyperfine Hamiltonian may now be written in the form

$$\begin{aligned} \mathcal{H}_f = \sum_k \{ & rH_n (a_k^+ a_k + b_k^+ b_k) + \omega_n (\alpha_k^+ \alpha_k + \beta_k^+ \beta_k) \\ & - (rH_n \omega_n)^{1/2} (\alpha_k^+ a_k + \alpha_k a_k^+ + \beta_k^+ b_k + \beta_k b_k^+) \} \end{aligned} \quad (2.27)$$

with $rH_n = A \langle I_z \rangle$ and $\omega_n = AS$. This assumes that the nuclear spin is aligned parallel with the electronic spin. The hyperfine frequency $\omega_n/2\pi$ is typically of the order of 600 MHz which implies that the field seen at the nucleus is of the order of 10^5 Oe as mentioned earlier. In light of this it is easily seen that the nuclear Zeeman term can be ignored.

The entire two magnon Hamiltonian takes the form

$$\mathcal{H} = \mathcal{H}_e + \mathcal{H}_{en} + \mathcal{H}_n \quad (2.28)$$

with

$$\mathcal{H}_e = \sum_k \{ A(a_k^\dagger a_k + b_k^\dagger b_k) + B(a_k^\dagger b_k + a_k b_k^\dagger) \quad (2.29)$$

$$+ C_k(a_k b_{-k} + a_k^\dagger b_{-k}^\dagger) + \frac{D}{2}(a_k^\dagger a_{-k}^\dagger + a_k a_{-k} + b_k^\dagger b_{-k}^\dagger + b_k b_{-k}) \} \quad (2.30)$$

$$\mathcal{H}_{en} = -(\Gamma H_N \omega_n)^{1/2} \sum_k (\alpha_k^\dagger a_k + \alpha_k a_k^\dagger + \beta_k^\dagger b_k + \beta_k b_k^\dagger) \quad (2.31)$$

$$\mathcal{H}_n = \omega_n \sum_k (\alpha_k^\dagger \alpha_k + \beta_k^\dagger \beta_k)$$

$$A = \Gamma H_E \cos 2\theta_c + \Gamma H \sin \theta_c + E + \Gamma H_N \quad (2.32)$$

$$B = \frac{1}{2} \gamma_k \Gamma H_E (1 - \cos 2\theta_c)$$

$$C = -\frac{1}{2} \gamma_k \Gamma H_E (1 + \cos 2\theta_c)$$

$$D = D$$

This Hamiltonian neglects linear terms because they are assumed to cancel if z' and z'' are equilibrium directions. Three magnon terms will be discussed in section D. It is easy to see from Eq. (2.41) that in the absence of any coupling of the nuclear and electronic variables the nuclei would have a normal mode frequency of ω_n . It is precisely H_{en} which is responsible for the coupling and hence the pulling of the nuclear normal mode frequencies from the unpulled frequency ω_n .

C. Diagonalization Of The Hamiltonian

The Hamiltonian containing only electronic variables, given by Eq. (2.29), is first considered. It may be expressed in matrix form as follows²⁴

$$\mathcal{H}_e = X^\dagger H_e X \quad (2.33)$$

$$X = \begin{pmatrix} a_k \\ b_k \\ a_{-k}^+ \\ b_{-k}^+ \end{pmatrix} \quad H_e = \begin{pmatrix} A & B & D & C \\ B & A & C & D \\ D & C & A & B \\ C & D & B & A \end{pmatrix} \quad (2.34)$$

where A, B, C and D are the same as in Eq. (2.32). In the above $X^\dagger$ is the transpose of the Hermitian adjoint of X. Knowing that the elements of X are boson operators allows one to write the following matrix equation

$$g H_e T = T g H' \quad (2.35)$$

where

$$g = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (2.36)$$

$$H' = \begin{pmatrix} \Omega_1 & 0 & 0 & 0 \\ 0 & \Omega_2 & 0 & 0 \\ 0 & 0 & \Omega_1 & 0 \\ 0 & 0 & 0 & \Omega_2 \end{pmatrix} \quad (2.37)$$

and T is the transformation matrix which diagonalizes H_e and defines the normal mode operators by

$$X = T Y \quad (2.38)$$

where

$$Y = \begin{pmatrix} c_k \\ d_k \\ c_{-k}^+ \\ d_{-k}^+ \end{pmatrix} \quad (2.39)$$

Performing the algebra yields

$$\begin{aligned} \Omega_1^2 &= (A+B)^2 - (C+D)^2 \\ \Omega_2^2 &= (A-B)^2 - (C-D)^2 \end{aligned} \quad (2.40)$$

$$T = \begin{pmatrix} R & -S & T & U \\ R & S & T & -U \\ T & U & R & -S \\ T & -U & R & S \end{pmatrix}$$

$$R = \frac{C+D}{[4\Omega_1(A+B-\Omega_1)]^{1/2}}$$

$$T = \frac{C+D}{[4\Omega_1(A+B+\Omega_1)]^{1/2}}$$

$$S = \frac{C-D}{[4\Omega_2(A-B-\Omega_2)]^{1/2}}$$

$$U = \frac{C-D}{[4\Omega_2(A-B+\Omega_2)]^{1/2}}$$

It is interesting to note that

$$R \approx T \approx - \left(\frac{\Gamma H_E}{4\Omega_1} \right)^{1/2}$$

(2.43)

$$S \approx U \approx - \left(\frac{\Gamma H_E}{4\Omega_2} \right)^{1/2}$$

The total Hamiltonian for the electronic and nuclear system in terms of the normal mode electronic variables becomes in matrix form

$$\mathcal{H} = W^\dagger H W$$

(2.44)

where

$$W = \begin{pmatrix} c_k \\ d_k \\ \alpha_k \\ \beta_k \\ c_k^+ \\ d_k^+ \\ \alpha_{-k}^+ \\ \beta_{-k}^+ \end{pmatrix}$$

(2.45)

$$H = \left(\begin{array}{cccc|cccc} \Omega_1 & 0 & G & G & 0 & 0 & K & K \\ 0 & \Omega_2 & H & -H & 0 & 0 & -L & L \\ G & H & \omega_n & 0 & K & -L & 0 & 0 \\ G & -H & 0 & \omega_n & K & L & 0 & 0 \\ \hline 0 & 0 & K & K & \Omega_1 & 0 & G & G \\ 0 & 0 & -L & L & 0 & \Omega_2 & H & -H \\ K & -L & 0 & 0 & G & H & \omega_n & 0 \\ K & L & 0 & 0 & G & -H & 0 & \omega_n \end{array} \right)$$

(2.46)

with

$$G = -\omega_n \Gamma H_n T$$

$$H = -\omega_n \Gamma H_n U$$

$$K = -\omega_n \Gamma H_n R$$

$$L = -\omega_n \Gamma H_n S$$

(2.47)

The Hamiltonian is diagonalized by the following transformation

$$W = U Z$$

(2.48)

with

$$Z = \begin{pmatrix} f_{1k} \\ f_{2k} \\ f_{3k} \\ f_{4k} \\ f_{1-k}^+ \\ f_{2-k}^+ \\ f_{3-k}^+ \\ f_{4-k}^+ \end{pmatrix}$$

(2.49)

and U defined by the following equation

$$gH(U = U gH')$$

(2.50)

with

$$H' = \left(\begin{array}{cccc|cccc} \omega_1 & 0 & 0 & 0 & & & & \\ 0 & \omega_2 & 0 & 0 & & & & \\ 0 & 0 & \omega_3 & 0 & & & & \\ 0 & 0 & 0 & \omega_4 & & & & \\ \hline & & & & \omega_1 & 0 & 0 & 0 \\ & & & & 0 & \omega_2 & 0 & 0 \\ & & & & 0 & 0 & \omega_3 & 0 \\ & & & & 0 & 0 & 0 & \omega_4 \end{array} \right)$$

(2.51)

$$\omega_1^2 = \frac{\alpha + \sqrt{\alpha^2 - 4\delta}}{2}$$

$$\omega_2^2 = \frac{\beta + \sqrt{\beta^2 - 4\gamma}}{2}$$

(2.52)

$$\omega_3^2 = \frac{\alpha - \sqrt{\alpha^2 - 4\delta}}{2} = \frac{\delta}{\omega_1^2} \quad (2.53)$$

$$\omega_4^2 = \frac{\beta - \sqrt{\beta^2 - 4\gamma}}{2} = \frac{\gamma}{\omega_2^2}$$

$$\begin{aligned} \alpha &= \Omega_1^2 + 4(G^2 - K^2) + \omega_n^2 \\ \beta &= \Omega_2^2 + 4(H^2 - L^2) + \omega_n^2 \end{aligned} \quad (2.54)$$

$$\begin{aligned} \delta &= \Omega_1^2 \omega_n^2 - 4\Omega_1 \omega_n (G^2 + K^2) + 4(G^2 - K^2)^2 \\ \gamma &= \Omega_2^2 \omega_n^2 - 4\Omega_2 \omega_n (H^2 + L^2) + 4(H^2 - L^2)^2 \end{aligned}$$

$$\omega_1 \approx \Omega_1 + \frac{2\omega_n}{\Omega_1^2} (G^2 + K^2) + \frac{2}{\Omega_1} (G^2 - K^2) \quad (2.55a)$$

$$\omega_1 \approx \Omega_1 + \frac{\omega_n \Gamma H_N}{\Omega_1} \left(1 + \frac{\omega_n \Gamma H_E}{\Omega_1^2} \right)$$

$$\omega_2 \approx \Omega_2 + \frac{2\omega_n}{\Omega_2^2} (H^2 + L^2) + \frac{2}{\Omega_2} (H^2 - L^2) \quad (2.55b)$$

$$\omega_2 \approx \Omega_2 + \frac{\omega_n \Gamma H_N}{\Omega_2} \left(1 + \frac{\omega_n \Gamma H_E}{\Omega_2^2} \right)$$

$$\omega_3 \approx \omega_n \left[1 - \frac{4(G^2 + K^2)}{\Omega_1 \omega_n} \right]^{1/2} \approx \omega_n \left[1 - \frac{2H_E H_N}{\Omega_1^2} \right]^{1/2} \quad (2.56a)$$

$$\omega_4 \approx \omega_n \left[1 - \frac{4(H^2 + L^2)}{\Omega_2 \omega_n} \right]^{1/2} \approx \omega_n \left[1 - \frac{2H_E H_N}{\Omega_2^2} \right]^{1/2} \quad (2.56b)$$

In a recent paper⁹ the assumption is made that G equals K [see Eq. (2.47)] and thus the term $2(G^2 - K^2)/\Omega_1$ can be neglected. A more careful analysis reveals that such a term must be kept. It turns out, however, that the more exact expression used here yields the same results as before⁹ because of a fortunate cancellation. Using Eq. (2.51), (2.50), and (2.46) yields the result

$$U = \left(\begin{array}{cccc|cccc} 1 & 0 & j_1 & 0 & 0 & 0 & m_1 & 0 \\ 0 & 1 & 0 & -j_2 & 0 & 0 & 0 & m_2 \\ h_1 & -h_2 & p_1^+ & -p_2^+ & -h_1 & -h_2 & -p_1^- & -p_2^- \\ h_1 & h_2 & p_1^+ & p_2^+ & -h_1 & h_2 & -p_1^- & p_2^- \\ \hline 0 & 0 & m_1 & 0 & 1 & 0 & j_1 & 0 \\ 0 & 0 & 0 & m_2 & 0 & 1 & 0 & -j_2 \\ -h_1 & -h_2 & -p_1^- & -p_2^- & h_1 & -h_2 & p_1^+ & -p_2^+ \\ -h_1 & 0 & -p_1^- & p_2^- & h_1 & h_2 & p_1^+ & p_2^+ \end{array} \right) \quad (2.57)$$

$$h_i = \left(\frac{\omega_n}{8\Omega_{ik}} \right)^{1/2} \left(\frac{2\Gamma^2 H_E H_N}{\Omega_{ik}^2} \right)^{1/2}$$

$$j_i = h_i \left(\frac{2\omega_n}{\omega_{ik}} \right) \quad i = 1, 2 \quad (2.58)$$

$$p_i^\pm = \frac{\omega_n \pm \omega_{ik}}{2(2\omega_n \omega_{ik})^{1/2}}$$

$$m_i = j_i \left(1 - \frac{2\omega_{ik}}{\Omega_{ik}} \right)$$

$$\omega_{ik} = \omega_n \left[1 - \frac{2\Gamma^2 H_E H_N}{\Omega_{ik}^2} \right]^{1/2}$$

D. Parallel Pumping

It is well known that two magnons with a frequency $\omega_p/2$ can be excited parametrically with a pump frequency of ω_p . This technique,^{25,26} known as parallel pumping, has been used extensively on ferrimagnets, such as yttrium iron garnet, to measure magnon relaxation rates. Ninio and Keffer⁷ presented the first realistic possibility of parallel pumping nuclear spin waves in RbMnF_3 . They did not, however, consider the flopped state, but rather limited themselves to small external fields. Recently the parallel pumping of a flopped antiferromagnet was considered. In the two canted antiferromagnets studied here, there exists a linear susceptibility when the rf and external fields are parallel. This susceptibility is due to the field-independent mode. Morgenthaler and Platzker¹² point out that there exist other first order nonlinear processes similar to the Suhl first order process due to this linear susceptibility. This possibility is examined in the context of the Holstein-Primakoff formulation in this section. The words "parallel pumping" are taken to mean that the external and rf fields are parallel regardless of what processes are excited.

The oscillating field couples much more strongly to the electronic spins than to the nuclear spins and therefore the rf Hamiltonian is

$$\mathcal{H} = P \cos \omega_p t \left[\sum_i \vec{S}_i \cdot \vec{h} + \sum_j \vec{S}_j \cdot \vec{h} \right] \quad (2.61)$$

The terms in Eq.(2.61) which couple two magnon terms to $\vec{h}$ clearly must arise from S_{iz} and S_{jz} . At first glance it would seem possible to couple with $\vec{h}$ parallel or perpendicular to $\vec{H}$, because of the canting.

Analysis reveals⁹

$$\mathcal{H}_{rf \parallel} (2mag) = \Gamma h \cos(\omega_p t) \sin \theta_c \times \sum_K (a_K^\dagger a_K + b_K^\dagger b_K) \quad (2.62)$$

where $\parallel$ indicates that the rf field is parallel to H_0 and in the plane formed by H_0 and the sublattice magnetizations. For the perpendicular direction one has¹⁰

$$\mathcal{H}_{rf \perp} (2mag.) = \Gamma h \cos(\omega_p t) \cos \theta_c \times \sum_K (a_K^\dagger a_K - b_K^\dagger b_K) \quad (2.63)$$

where the $\perp$ symbol means that the rf field is perpendicular to H_0 and in the same plane as above. The angle θ_c is of the order of H/H_E and thus much less than one. This might lead one erroneously to conclude that Eq.(2.63) leads to the most efficient means of coupling to the two magnon terms.

In actuality, if one examines closely the excitation of any pair f_{ik}, f_{j-k} with wavevectors k and $-k$, it is easy, with the help of Eq. (2.59), to show that $a_{k,k}^\dagger a_k = b_{k,k}^\dagger b_k$. This can also be arrived at by considering the symmetry of the two sublattices. Thus the most efficient means of two magnon excitation is with the rf applied parallel to the external field. If one expresses Eq. (2.62) in terms of normal mode operators then he has

$$\sum_K (a_K^\dagger a_K + b_K^\dagger b_K) = \sum_{i \leq j} B_{ij} \sum_K (f_{ik} f_{j-k}^\dagger + Hc) \quad (2.64)$$

where Hc refers to the Hermitian conjugate of all the preceding terms.

Fig. 2-3 (a) Parallel pumping interaction. (b) Suhl first order process. represents a photon. represents a magnon.

We have

$$\begin{aligned}
 B_{11} &= 2RT & B_{13} &= 2j_1(R+T)^2 \\
 B_{22} &= -2US & B_{24} &= 2j_2(U+S)^2 \\
 B_{33} &= j_1^2(R+T)^2 & & \\
 B_{44} &= -j_2^2(U+S)^2 & &
 \end{aligned} \tag{2.65}$$

with all other B_{ij} 's = 0. These B_{ij} 's which are zero correspond to coupling to two magnons from different branches. The approximation has been made that $j_i = m_i$ which is justified because the exact expressions involve only the sum $m_i + j_i$, and $m_i - j_i \ll m_i + j_i$, as seen from Eq. (2.58).

Because of the existence of three magnon terms in the exchange Hamiltonian, and because of the large linear susceptibility of the field independent mode when the external field and the rf field are parallel, one should also look for a process in which one photon creates a field independent uniform mode magnon f_{20} , which in turn decays into two $k \neq 0$ magnons. Such a process is shown schematically in Fig. 2-3b. One does not consider a process in which the rf field couples linearly to the f_{10} magnon because the rf susceptibility for that mode in the parallel geometry is zero. The exchange Hamiltonian contains the following three magnon terms

$$\begin{aligned}
 \mathcal{H}_{ex}(3 \text{ mag}) &= \left(\frac{S}{2}\right)^{1/2} \sin 2\theta_c \times \\
 &\sum_{i,j} J_{ij} (a_i^+ a_i b_j^+ - b_j^+ b_j a_i^+ + Hc)
 \end{aligned} \tag{2.66}$$

and the process just mentioned may arise from these terms. This expression is now Fourier transformed into

$$\begin{aligned} \mathcal{H}_{ex}(3mag) = & \frac{\Gamma H_E \sin 2\theta_c}{\sqrt{2NS}} \times \\ & \sum_k \left\{ \gamma_k [a_0(a_k^+ b_{-k}^+ + a_k^+ b_k) \right. \\ & \left. - b_0(b_k^+ a_{-k}^+ + b_k^+ a_k)] \right. \\ & \left. + \gamma_0(b_0 a_k^+ a_k - a_0 b_k^+ b_k) + H_c \right\} \end{aligned} \quad (2.67)$$

Only terms in which two $k \neq 0$ magnons are excited by one with zero wavevector are kept since we are interested only in the process of Fig. 2-3b. With the help of Eq. (2.59), Eq. (2.67) may be written

$$\begin{aligned} \mathcal{H}_{ex}(3mag) = & \frac{\Gamma H_E \sin 2\theta_c}{\sqrt{2NS}} \times \\ & [f_{20}^+ \sum_{\substack{i \leq j \\ k}} (C_{ij} f_{ik} f_{j-k} \\ & + E_{ij} f_{ik}^+ f_{j-k}^+ + H_c)] \end{aligned} \quad (2.68)$$

The reason for not including the f_{40} term is discussed at the end of this section. The C's and the E's of Eq. (2.68) are defined as follows where the approximation has been made that $j_i = m_i$ ³⁴

$$\begin{aligned}
C_{11} &= 2\gamma_k (T^2 U_0 - R^2 S_0) + 2RT(U_0 - S_0)(\gamma_k - \gamma_0) \\
C_{22} &= 2\gamma_k [S^2 S_0 - U^2 U_0 + 2SU(U_0 - S_0)] - 2US(U_0 - S_0)(\gamma_k - \gamma_0) \\
C_{33} &= 2\gamma_k j_1^2 (R+T)^2 (U_0 - S_0) + 2j_1^2 (R+T)^2 (U_0 - S_0)(\gamma_k - \gamma_0) \\
C_{44} &= 2\gamma_0 j_2^2 (U_0 - S_0)(U+S)^2 \quad (2.69) \\
C_{13} &= 4\gamma_k j_1 (R+T)(TU_0 - RS_0) + 2j_1 (R+T)(RU_0 - ST_0)(\gamma_k - \gamma_0) \\
C_{24} &= 2\gamma_k (UU_0 - SS_0 + US_0 - SU_0)(\gamma_k - \gamma_0) \\
&\quad + (4\omega_{2k}/\Omega_{2k})(U-S)(SS_0 + 2UU_0 - SU_0)
\end{aligned}$$

$$\begin{aligned}
E_{11} &= 2\gamma_k (U_0 R^2 - S_0 T^2) + 2RT(U_0 - S_0)(\gamma_k - \gamma_0) \\
E_{22} &= 2\gamma_k [U^2 S_0 - S^2 U_0 + 2SU(U_0 - S_0)] - 2US(U_0 - S_0)(\gamma_k - \gamma_0) \\
E_{33} &= 2\gamma_k j_1^2 (R+T)^2 (U_0 - S_0) + 2j_1^2 (R+T)^2 (U_0 - S_0)(\gamma_k - \gamma_0) \\
E_{44} &= 2\gamma_0 j_2^2 (U-S)^2 (U_0 - S_0) \quad (2.70) \\
E_{13} &= 4\gamma_k j_1 (R+T)(RU_0 - TS_0) + 2j_1 (R+T)^2 (U_0 - S_0)(\gamma_k - \gamma_0) \\
E_{24} &= 4\gamma_k j_2 (U+S)(SS_0 - UU_0) + 2j_2 (U+S)^2 (U_0 - S_0)(\gamma_k - \gamma_0)
\end{aligned}$$

with all other C_{ij} 's and E_{ij} 's, corresponding to excitation of magnons from different branches, being zero. The subscript on the transformation coefficients indicates they are to be evaluated for $k = 0$.

These expressions may be evaluated by using the following approximations keeping the lowest nonvanishing order in $\Omega_k/\Gamma H_E$

$$R \approx - \left(\frac{\Gamma H_E}{4\Omega_{1K}} \right)^{1/2} \left(1 + \frac{\Omega_{1K}}{2\Gamma H_E} \right)$$

$$S \approx - \left(\frac{\Gamma H_E}{4\Omega_{2K}} \right)^{1/2} \left(1 + \frac{\Omega_{2K}}{2\Gamma H_E} \right)$$

$$T \approx - \left(\frac{\Gamma H_E}{4\Omega_{2K}} \right)^{1/2} \left(1 - \frac{\Omega_{1K}}{2\Gamma H_E} \right)$$

$$U \approx - \left(\frac{\Gamma H_E}{4\Omega_{2K}} \right)^{1/2} \left(1 - \frac{\Omega_{2K}}{2\Gamma H_E} \right)$$

At first glance it might appear that the Zeeman and exchange^{17,27} contribute other three magnon terms formed by expanding the one magnon terms to a higher order. The one magnon terms originally arose from the operators S^+ and S^- , which convert to spinwave variables a and $a^\dagger$ by the following method

$$S^+ = (2S)^{1/2} \left(1 - \frac{a^\dagger a}{2S} \right)^{1/2} a \quad (2.71)$$

$$S^- = (2S)^{1/2} a^\dagger \left(1 - \frac{a^\dagger a}{2S} \right)^{1/2}$$

It is the expansion of the square root which leads to higher order terms. It must be remembered that the one magnon terms all cancel because they essentially define the equilibrium direction of the sublattice magnetizations; hence, the three magnon terms arising from these one magnon terms must also cancel.

The relationship between f_{20}^+ and the rf field h , when the rf field is parallel to the external field H , is found by considering Eq. (2.61) and keeping only the terms containing f_{20}^+ and f_{20} .

$$\mathcal{H}_{rf}(1mag) = \Gamma h (2NS)^{1/2} (f_{20}^+ + f_{20}) \quad (2.72)$$

The subscripts on U and S mean that they are to be evaluated for $k = 0$. The f_{40} terms are not considered for reasons discussed later.

If one assumes a linearly polarized field

$$h = h_0 \cos \omega_p t \quad (2.73)$$

and then decomposes this into rotating and counterrotating circularly polarized fields he finds

$$h = \frac{h_0}{2} e^{i\omega_p t} + \frac{h_0}{2} e^{-i\omega_p t} \quad (2.74)$$

Both senses of rotation must be kept because of the possibility of driving the field independent mode far off resonance. The Hamiltonian in the region where f_i , $k \neq 0$ is small, which is the case before the instability sets in, is

$$\mathcal{H} = \Omega_{20} f_{20}^+ f_{20}^+ - T_2 (e^{i\omega_p t} + e^{-i\omega_p t}) \times (f_{20}^+ + f_{20}) \quad (2.75)$$

$$\text{where } T_2 = \frac{\Gamma h_0}{2} (2NS)^{1/2} (U_0 - S_0) \quad (2.76)$$

The equations of motion can be formed as follows

$$\dot{f}_{20}^+ = i[\mathcal{H}, f_{20}^+] \quad (2.77)$$

$$\dot{f}_{20}^+ = i\Omega_{20} f_{20}^+ - iT_2(e^{i\omega_p t} + e^{-i\omega_p t}) \quad (2.78)$$

Assuming a solution

$$f_{20}^+ = ae^{-i\omega_p t} + be^{i\omega_p t} \quad (2.79)$$

requires

$$a = \frac{T_2}{\Omega_{20} + \omega_p - i\eta_{20}}$$

$$b = \frac{T_2}{\Omega_{20} - \omega_p - i\eta_{20}} \quad (2.80)$$

In arriving at Eq. (2.80) Ω_{20} is replaced by $\Omega_{20} - i\eta_{20}$, where η_{20} is a phenomenological damping parameter of the field independent mode.

The three magnon Hamiltonian is then of the form

$$\mathcal{H}_{ex}(3 \text{ mag}) = \frac{\rho h_0}{2} \sin \theta_c \times$$

$$\sum_{\substack{i \neq j \\ k}} \left\{ \left[\frac{D_{ij} e^{i\omega_p t}}{\Omega_{20} - \omega_p + i\eta_{20}} + \frac{F_{ij} e^{i\omega_p t}}{\Omega_{20} + \omega_p - i\eta_{20}} \right] f_{ik} f_{j-k} \right. \quad (2.81)$$

$$\left. + \left[\frac{D_{ij} e^{-i\omega_p t}}{\Omega_{20} + \omega_p - i\eta_{20}} + \frac{F_{ij} e^{-i\omega_p t}}{\Omega_{20} - \omega_p + i\eta_{20}} \right] f_{ik} f_{j-k} + H_c \right\}$$

where

$$D_{ij} = (\Omega_{20} \Gamma H_E)^{1/2} C_{ij} \quad (2.82)$$

$$F_{ij} = (\Omega_{20} \Gamma H_E)^{1/2} E_{ij} \quad (2.83)$$

and we have used $U_0 - S_0 \approx \left(\frac{\Omega_{20}}{4\Gamma H_E}\right)^{1/2}$.

Also $\sin 2\theta_c$ is replaced by $2 \sin \theta_c$ in arriving at Eq. (2.81), which is valid for $\theta_c \ll 1$. This may be combined with Eq. (2.62) and (2.64) to yield the total effective Hamiltonian used for two magnon excitation which is

$$\begin{aligned} \mathcal{H}_{\text{eff}} = & \frac{\Gamma h_0}{2} \sin \theta_c \sum_{i \leq j} \left\{ [B_{ij} \right. \\ & + \frac{D_{ij}}{\Omega_{20} - \omega_p + i\eta_{20}} + \frac{F_{ij}}{\Omega_{20} + \omega_p - i\eta_{20}}] e^{i\omega_p t} f_{ik} f_{j-k} \\ & \left. + H.c. \right\} \end{aligned} \quad (2.84)$$

The term involving $e^{i\omega t} f_{ik} f_{j-k}$ is not considered because it does not contribute to the instability.

To find the critical fields consider

$$\dot{f}_{ik}^+ = i [\mathcal{H}_{\text{eff}}, f_{ik}] \quad (2.85)$$

Thus one can write

$$\dot{f}_{ik} = -i\omega_{ik} f_{ik} + i G_{ij} (1 + \delta_{ij}) \times \frac{\Gamma h_0}{2} \sin \theta_c e^{i\omega_p t} f_{j-k}^+ \quad (2.86)$$

$$\dot{f}_{j-k}^+ = i\omega_{ik} f_{j-k}^+ - i G_{ij} (1 + \delta_{ij}) \times \frac{\Gamma h_0}{2} \sin \theta_c e^{-i\omega_p t}$$

where ω_{ik} is the normal mode frequency. Assuming a phenomenological relaxation rate η implies that ω_{ik} should be replaced by

$$\omega_{ik} = \omega_{ik} - i\eta_{ik} \quad (2.87)$$

Solving Eqs. (2.86) with (2.87) inserted, it can be shown that the pair $f_{ik}f_{j-k}$ become excited to a very large amplitude when h exceeds h_c , with h_c defined as^{26,28}

$$\Gamma h_c = \frac{2(\eta_{ik}\eta_{jk})}{|G_{ij}|(1+\delta_{ij})\sin\theta_c} \quad (2.88)$$

where

$$G_{ij} = B_{ij} + \frac{D_{ij}}{\Omega_{20} - \omega_p - i\eta_{20}} + \frac{F_{ij}}{\Omega_{20} + \omega_p - i\eta_{20}}$$

with the condition

$$\omega_p = \omega_{ik} + \omega_{jk} \quad (2.89)$$

The B_{ij} 's, D_{ij} 's and F_{ij} 's are given by Eqs. (2.65), (2.69), (2.70), (2.82) and (2.83) respectively, and it is obvious by looking at them that one can only excite magnons from the same branch. Thus, there are three types of parallel pumping experiments for each branch: excitation of two electronic spinwaves, excitation of one nuclear and one electronic spinwave, and excitation of two nuclear spinwaves. A detailed examination of G_{22} , G_{24} and G_{44} reveals that they are very small for all k . Thus, the critical fields corresponding to excitation of any spinwaves from the field independent branches are very large.

Specific numerical examples are given in Chapter IV (page 93). Table II-1 lists the critical field relationships for the field independent modes. In evaluating the critical fields the approximations that $\Omega_{20} \gg \omega_n$ and $\sin \theta_c = H/2H_E$ are made and we assume $|\Omega_{20} - \omega_p| \gg \eta_{20}$ for all processes except those where ω_p can be near Ω_{20} .

The reason for neglecting a process such as $f_{40}^\dagger f_{ik} f_{j-k}$, where the $f_{40}^\dagger$ refers to the field independent nuclear branch is two fold.

One must consider the rf susceptibility and the coefficient of such a term in H_{ex} (3 mag.). The new quantities J and M are defined as follows

$$\begin{aligned} J &= S j_2 + U m_2 \\ M &= U j_2 + S m_2 \end{aligned} \quad (2.90)$$

Using these expressions it is easily shown that in the expressions for the rf susceptibility U and S are replaced by M and J respectively, when considering f_{40} ; thus

$$\begin{aligned} \frac{\chi(f_{40})}{\chi(f_{20})} &= \frac{(J-M)/(\omega_{20}-\omega_p-i\eta_{40})}{(U-S)/(\Omega_{20}-\omega_p-i\eta_{20})} \\ \frac{\chi(f_{40})}{\chi(f_{20})} &= 2 \frac{\omega_{20}}{\Omega_{20}} j_2 \left| \frac{\Omega_{20}-\omega_p-i\eta_{20}}{\omega_{20}-\omega_p-i\eta_{40}} \right| \end{aligned} \quad (2.91)$$

which is clearly much less than unity for all acceptable pumping frequencies since j_2 is equal to $\frac{\omega_n}{\Omega_{2k}} \frac{2\Gamma^2 H_E H_N}{\Omega_{2k}^2} \ll 1$.

The coefficients corresponding to the D_{ij} and F_{ij} must now be examined. Such a term must arise from a term similar to $a_0 a_k^\dagger b_{-k}^\dagger$. The a_0 part is replaced by Jf_{40} rather than Uf_{20} . With this in mind, the coefficients of the f_{40} terms must be the corresponding D_{ij} and F_{ij} multiplied by $\frac{J-M}{U-S}$ which equals $2j_2 \frac{\omega_{20}}{\Omega_{20}}$. Thus combining this with Eq. (2.89) yields

$$\frac{\mathcal{H}(f_{40})}{\mathcal{H}(f_{20})} = 4 j_2^2 \left(\frac{\omega_{20}}{\Omega_{20}} \right)^2 \left| \frac{\Omega_{20} - \omega_p - i\gamma_{10}}{\omega_{20} - \omega_p - i\gamma_{40}} \right| \ll 1 \quad (2.92)$$

and therefore $H_{f_{40}}$ is neglected as long as $\omega_p \gtrsim 2\omega_{20}$, which is the case for two magnon pumping.

E. Spinwave Spectrum

It is possible to obtain the spinwave dispersion relation for the uniaxial and cubic anisotropy case. Substituting the values for A, B, C and D yields the two normal mode frequencies, one of which is independent of the external field.

The frequencies are

$$\begin{aligned} \left(\frac{\Omega_{1K}}{\Gamma} \right)^2 &= H^2 - \frac{3}{2} H_E H_A + 2 H_E H_N + \frac{1}{3} K^2 a^2 H_E^2 \\ \left(\frac{\Omega_{2K}}{\Gamma} \right)^2 &= 3 H_E H_A + 2 H_E H_N + \frac{1}{3} K^2 a^2 H_E^2 \end{aligned} \quad (2.93)$$

for RbMnF_3 , with a being the lattice spacing, and H applied in the $\langle 100 \rangle$ direction. For CsMnF_3 , with H in the basal plane

$$\left(\frac{\Omega_{1K}}{\Gamma}\right)^2 = H^2 + 2H_E H_N + \alpha k_t^2 + \beta k_z^2$$

$$\left(\frac{\Omega_{2K}}{\Gamma}\right)^2 = 2H_E H_N + 2H_E H_A + \alpha k_t^2 + \beta k_z^2 \quad (2.94)$$

$$\alpha = a^2 H_E^2 / 9 \quad \beta = c^2 H_E^2 / 36$$

where a and c are the dimensions of the unit cell and k_t is the component of k in the basal plane and k_z the component of k along the "c" axis.

A more exact discussion of CsMnF_3 is presented elsewhere.¹⁷ The more exact expressions take into account that CsMnF_3 is a six sublattice system and hence has six normal mode electronic frequencies. The other four normal modes not considered here are strictly optical.

The quantity α takes on a slightly different form when the more exact six sublattice model is used. Also, in the more rigorous six sublattice case there are six normal mode nuclear frequencies. The other four nuclear modes, corresponding to the optical electronic modes, must all have a frequency of ω_n .

F. Conclusion

In the previous sections it is seen that care must be taken to evaluate the transformation matrices to the order $(\Omega_k / \Gamma H_E)$ to allow for the possible contribution of three magnon terms in the exchange Hamiltonian which otherwise would cancel if the transformation matrices

are evaluated to a lower order. It is shown that these three magnon terms give rise to a first order Suhl-like process as predicted by Platzker and Morgenthaler. The Holstein-Primakoff (HP) method used here yields the same results for pumping of field dependent magnons as Platzker et. al. when the following approximations in their expressions are made

$$\frac{\omega_p^2 - \omega_n^2}{\omega_p^2 - \omega_{20}^2} \approx 1 \quad (2.95)$$

and $\omega_{20} \gg \omega_n$. Both approximations are well satisfied. Threshold fields, for parallel pumping magnons from the field independent branch, in general do not approach infinity as Morgenthaler and Platzker predict. This work predicts threshold fields approaching infinity only when one is exciting $k = 0$ field independent magnons.

The HP method has the advantage of physical clarity because it utilizes the creation and annihilation operators and yields essentially the same results as Platzker et. al. This is reasonable because both methods contain the same physical content. There are two striking predictions which the preceding theory makes: First, the impossibility of exciting two field independent nuclear spinwaves by parallel pumping and second, extremely low rf fields necessary for the excitation of the parallel pumping instability when the pump frequency approaches the frequency of the field independent mode.

TABLE II-1

PROCESS	PUMP FREQUENCY ω_p	THRESHOLD FIELD h_c
Two electronic spinwaves with frequency Ω_{1k}	$2\Omega_{1k}$	$\frac{2\eta_{1k}^e \Omega_{1k}}{\Gamma^2 H} \left \frac{\Omega_{20}^2 - \omega_p^2 + 2i\eta_{20}\omega_p}{\Omega_{20}^2} \right $
Two nuclear spinwaves with frequency ω_{1k}	$2\omega_{1k}$	$\frac{2\eta_{1k}^n \Omega_{1k}^2 \omega_{1k}}{\Gamma^2 H (\omega_n^2 - \omega_{1k}^2)}$
One nuclear spinwave with frequency ω_{1k} and one electronic spinwave with frequency Ω_{1k}	$\omega_{1k} + \Omega_{1k}$	$\frac{2(\eta_{1k}^e \eta_{1k}^n)^{1/2} \Omega_{1k}}{\Gamma^2 H} \left(\frac{\Omega_{1k} \omega_{1k}}{\omega_n^2 - \omega_{1k}^2} \right)^{1/2} \times \left \frac{\Omega_{20}^2 - \omega_p^2 + 2i\eta_{20}\omega_p}{\Omega_{20}^2} \right $
Two electronic spinwaves with frequency Ω_{2k}	$2\Omega_{2k}$	$\frac{4\eta_{2k}^e \Omega_{2k}}{\Gamma^2 H} \left \frac{4\Omega_{2k}^2 - \Omega_{20}^2}{(3\Omega_{20}^2 - 2\Omega_{2k})(\Omega_{20}^2 - \Omega_{2k})} \right $
Two nuclear spinwaves with frequency ω_{2k}	$2\omega_{2k}$	$\frac{\eta_{2k}^n \Omega_{2k}^2 \omega_{2k}}{(\omega_n^2 - \omega_{2k}^2) \Gamma^2 H} \frac{\Omega_{20}^2}{2\omega_{2k}^2}$
One nuclear spinwave with frequency ω_{2k} and one electronic spinwave with frequency Ω_{2k}	$\omega_{2k} + \Omega_{2k}$	$\frac{8(\eta_{2k}^e \eta_{2k}^n)^{1/2} \Omega_{2k}}{\Gamma^2 H} \left(\frac{\Omega_{2k} \omega_{2k}}{\omega_n^2 - \omega_{2k}^2} \right)^{1/2}$ where $D = \frac{(\Omega_{20}^2 - \Omega_{2k}^2) - 2\omega_{2k}(\Omega_{2k} - \Omega_{20} - i\eta_{20}) - 2\omega_{2k}^2 + 2i\eta_{20}\Omega_{2k}}{(\Omega_{20}^2 - \Omega_{2k}^2) - \omega_{2k}(2\Omega_{2k} + \omega_n)}$

III. PARALLEL PUMPING OF TWO NUCLEAR SPINWAVES

A. Introduction

Chapter II predicts that it is possible by parallel pumping to excite simultaneously two nuclear spinwaves belonging to the same branch. The relation of the critical field to the relaxation rate of the spinwaves is given in Table 2-1. Analysis of critical field data requires accurate knowledge of ω_n , H_E , H_N , H_A . A variety of normal resonance techniques are needed to ascertain these parameters.

Until recently all measurements in the 500 to 1,000 MHz range were done using either wavelength coaxial cavities or placing the sample at the shorted end of a coaxial cable.^{3,22} These cavities have the disadvantage of low filling factor. A more sensitive method is to use a resonant helix. With such a system high filling factors and high Q's are possible making it easy to obtain rf fields of the order of several gauss from an input power of 1 watt. Such a system allows more sensitive measurements than permitted by coaxial cavity.

B. Experimental Method

H_E and H_A , the resonance parameters for RbMnF_3 , are measured at X band frequencies and compared to previously published values.^{17,18,22,29} The sample is placed in a special X band cavity shown in Fig. 3-1. This cavity has the feature that the sample is mounted on a side wall on a rotateable platform. In this arrangement the microwave rf field is always at right angles to the external field H regardless of the angle θ . When H is parallel to the $\langle 100 \rangle$ crystallographic direction the field H for resonance is largest. One places the magnetic field at a small angle ϵ in the x-y plane, and then assumes that the $\langle 100 \rangle$ direction is initially at some small angle δ , as shown in Fig. 3-2a. It is easy to see that

Fig. 3-1 X band and cavity with sample on rotating platform. h_{rf} is parallel to z and H lies in the x - y plane.

upon rotating the sample about the x axis, the $\langle 100 \rangle$ direction will, at some angle θ , lie in the x-y plane and the magnetic field for resonance H_{res} will be maximum because H and the $\langle 100 \rangle$ direction are more nearly parallel. This corresponds to the situation in Fig. 3-2b. The external field is now rotated until H_{res} is further maximized. At this point, H and $\langle 100 \rangle$ are parallel.

To obtain H_E and H_A , one measures the resonance frequency and magnetic field at two temperatures with H along the $\langle 100 \rangle$ direction. Using the fact that $H_N = 9.43/T$ for RbMnF_3 , one can determine H_E and H_A uniquely [see Eq. (2.93)]. In this measurement the temperatures used are below 4.2°K, so that H_E and H_A are essentially temperature independent. Other methods of obtaining H_E and H_A for RbMnF_3 are discussed elsewhere.²²

For CsMnF_3 , a single measurement of the H_{res} with H in the basal plane is enough to yield H_E because of zero in plane anisotropy. We take $H_N = 9.15/T$ for this measurement.¹⁶

The measurements in the 1000 MHz range are made with the use of a resonant helix. The helix consisted of simply a spiral wound length of wire shown in Fig. 3-3. Such a structure can propagate an electromagnetic wave with a wave length along the helix of λ_h .²⁴ The lowest order mode has no θ variation. All modes have a Bessel function radial dependence and sinusoidal z dependence. One useful feature of the helix is the relatively uniform cross sectional fields. This allows the sample to occupy the entire helix cross section and still have all parts see essentially the same intensity rf field. A half wave length resonator is made by using a section of helix $\lambda_h/2$ in length. Coupling can be any of the three standard methods: tap, probe, or loop. All three methods have been tried successfully.

The actual helix arrangement is shown in Fig. 3-3. The helicies

Fig. 3-2 (a) Initial orientation of RbMnF_3 with H in the x-y plane.
 (b) Orientation after $\langle 100 \rangle$ is rotated about x axis until the $\langle 100 \rangle$ direction lies in the x-y plane.

Fig. 3-3 Properties and construction of helix.

used typically resonate at a frequency of about 1000 MHz and consist of 5 turns of #18 copper wire wound on a polystyrene tube with interwinding spacing equal to the wire diameter and with an inside diameter of .375 inch. The coupling loop consists of one and one half turns of #18 wire. The coupling loop is connected to the coaxial transmission line. The entire cavity can be enclosed by a stainless steel can to prevent helium from entering the helix.

The microwave spectrometer used for these measurements is shown in Fig. 3-4. The Hewlett Packard 612-A UHF signal generator used for the microwave source has provisions for external amplitude modulation. The generator is pulsed on by a General Radio 1217-C pulse generator. Pulse widths can be varied from one microsecond to greater than one millisecond. Pulse repetition rates are typically less than thirty pulses per second. Heating effects due to high repetition rates are encountered¹¹ when the can is used because of the poor thermal contact of the sample with the reservoir, the only method of heat transfer being through the helium exchange gas.

The decay of the spinwaves after the removal of the rf pulse is observed by closely monitoring the tail as shown in Fig. 3-5a. This straightforward procedure is complicated by the fact that the decay of the helix fields usually swamps out the spinwave decay. Therefore the actual signal observed is almost unresolvable. The high spike at the end of the pulse saturates the amplifying system. To be sure that one is actually observing the decay of the spinwaves and not the response of the amplifiers, an easy check is performed. The width of the applied rf pulse is first adjusted such that the instability amplitude reaches essentially the steady state value and the decay is noted. Then the pulse width is decreased to a value such that the

Fig. 3-4 Pulse spectrometer used for measurements in the 1GHz range.

Fig. 3-5 (a) Transient decay of nuclear spinwaves. (b) Pulse width too narrow to permit spinwave buildup and hence, no decay.

instability has not begun, as shown in Fig. 3-5b. The decay is again noted. Any amplifier saturation will remain in the case of the short pulse width and any effects due to spinwave decay will disappear.

The rf fields inside the helix are much more difficult to determine than those inside the usual waveguide cavity. A rough guess can be made using the following arguments. The energy stored in the helix fields is related to the power absorbed in the helix by the following well known relation defining the intrinsic Q_0 of a system

$$Q_0 = f \frac{\text{Energy stored}}{\text{Power loss}} \quad (3.2)$$

where f is the resonant frequency of the cavity. Using the fact that

$$U = \frac{1}{8\pi} \int h^2 dV \quad (3.3)$$

and assuming the fields are constant inside the helix and zero outside, one obtains

$$h^2 = \frac{8\pi Q P_{abs}}{f V} \quad (3.4)$$

where P_{abs} is the power absorbed by the helix and V is the volume of the helix. This is clearly an over simplification of the problem, but does provide one with an idea of the rf fields to expect. This expression is used to test the reasonableness of the answer obtained experimentally.

The actual rf fields are obtained by cw saturation of diphenylpicrylhydrazyl (DPPH). The observed susceptibility as a function of rf driving field in a paramagnet is

$$\chi'' = \frac{\chi_0''}{1 + \left(\frac{h}{\Delta H}\right)^2} \quad (3.5)$$

where the assumption is made that $T_1 = T_2$, where T_1 and T_2 are the longitudinal and transverse relaxation times respectively. The rf field h , is assumed to be linearly polarized and ΔH is the full width at half maximum power. Care must be taken to assure that χ is being measured. This is accomplished by making sure that the reflection coefficient on resonance Γ_{on} is very close to that off resonance Γ_{off} . The criteria used is that $(\Gamma_{on} - \Gamma_{off})/\Gamma_{off} \ll 1$.

The external field is adjusted to the value necessary for resonance and then the external field is modulated at a rate much less than T_1^{-1} . Modulating the signal permits the signal to be amplified by AC coupled amplifiers rather than the more drift prone DC amplifiers. The signal height as measured from the base line to the peak is considered proportional to the susceptibility. This method provides more sensitivity than a method in which the rf is pulsed. This is easily seen by remembering that in the sweep type cw measurement both on and off resonance signals are measured essentially simultaneously; whereas the pulse method requires that two separate measurements be made: one on resonance and one off resonance. The difference is then taken. Usually this difference is quite small; hence, small errors in either measurement appear magnified when the difference is taken.

In making the actual measurements, the apparatus shown in Fig. 3-6 is used with the DPPH surrounding the sample. The input power is adjusted by attenuator R1, while the off resonance power at the detector is maintained constant by adjusting precision attenuator R2 and monitoring the crystal voltage by a digital volt meter. In this method the crystal is always operated in the same region eliminating crystal response law correction. The inverse susceptibility is plotted as a

Fig. 3-6 Spectrometer used to determine rf fields in helix by saturating DPPH.

function of h^2 and fitted to a straight line. In this manner, the attenuator settings in dB can be converted to rf field values.

C. Discussion Of Results

A typical example of the saturation of DPPH with $\Delta H = 2.5$ Oe is shown in Fig. 3-7. From this graph it can be inferred that for a power of 0.128 watts an rf field of 2.5 Oe is produced. Using Eq. (3.4) with the following values: $Q_0 = 1000$, $V = 2\text{cm}^3$ and $P_{\text{abs}} = 0.128$ watts, one can infer that h would be 3.7 Oe. This is in qualitative agreement with that obtained by the saturation measurement.

NMR measurements on CsMnF_3 are shown in Fig. 3-8. If $1/\omega_{10}^2$ is plotted versus $1/H^2$, the result is a straight line [see Eq. (2.56)] which extrapolated to infinite field yields $\omega_n = 668 \pm 1.2$ MHz. Measurements of H_E at a pump frequency of 8.759 GHz and $T = 4.20^\circ\text{K}$ yields $H_E = 2.95 \times 10^5$ Oe for CsMnF_3 . H_E and H_A for RbMnF_3 at 4.2°K and a pump frequency of 8.491 GHz are 8.1×10^5 and 4.5 Oe respectively. Again, these are in complete agreement with published values. RbMnF_3 has a full width at half maximum of approximately 200 Oe, but its line shape is far from symmetrical as shown in Fig. 3-9. This asymmetry is thought to be caused by strains and may be accounted for by variations in the anisotropy field throughout the sample. This variation in the anisotropy field should also be reflected in the field independent mode so as to make the tail on the high frequency side. The frequency of this mode can not be measured directly but is inferred to be 10.7 GHz by measuring the resonance frequency as a function of angle and extrapolating to the $\langle 100 \rangle$ direction. This measurement is made by working a fixed frequency and rotating the sample until resonance is observed. Direct measurement of the field independent mode is extremely difficult because such a resonance is 200 Oe

Fig. 3-7 Typical data for saturation of DPPH when $x/x_0 = 2$,
then $h_{rf} = \Delta H$.

Fig. 3-8 The frequency of the uniform precession ($k = 0$) field dependent nuclear spinwave in CsMnF_3 as a function of external field. ω_h is that value of frequency corresponding to $H = \infty$.

Fig. 3-9 AFMR line shape of RbMnF_3 .

or approximately 560 MHz wide. With the klystron signal source used, it is possible to sweep over only a 75 MHz width. This value of Ω_{20} is also consistent with the measured values of H_E and H_A since

$$\begin{aligned}\Omega_{20}^2 &= \Gamma^2 (3 H_E H_A + 2 H_E H_N) \\ \Omega_{20}^2 &= (2\pi \times 10.75 \times 10^9)^2\end{aligned}\quad (3.6)$$

for the measured H_E and H_A .

Figs. 3-10 and 3-11 show the critical field data for CsMnF_3 and RbMnF_3 respectively. It is not possible with the present apparatus to excite two nuclear spinwaves in RbMnF_3 at 4.2°K because of insufficient pumping power. As is the case with the other pumping processes the nonlinear susceptibility for RbMnF_3 is considerably less than CsMnF_3 . This made it difficult to determine the exact value of critical field. The difference in nonlinear susceptibility may be explained by assuming that there exists a distribution of regions in the sample with different anisotropy fields which broadens the high frequency mode. This in turn broadens the low frequency line width which can be seen by differentiating Eq. (2.56) seen below

$$\delta \omega_k = \frac{2 \Gamma^2 H_E H_N \omega_n^2}{\omega_k \Omega_k^3} \delta \Omega_k \quad (3.7)$$

where $\delta \Omega_k$ is the width of the electronic mode. Thus, for a given frequency, not all the spins have the same k value [see Eq. (2.93)]. If one assumes that the relaxation rate depends on k then different values of k will become unstable at different powers. For RbMnF_3 and CsMnF_3 the AFMR line widths are typically 200 Oe and 20 Oe respectively. In the parallel pumping of two nuclear spinwaves the quantity Ω_k must remain constant even when the field is changed. This is easily seen by examining the following equation for the nuclear spinwave frequency

Fig. 3-10 Critical fields h versus external magnetic field H for CsMnF_3 .

Fig. 3-11 Critical fields h versus external magnetic field H for RbMnF_3 . Predicted field for $k = 0$ is shown.

$$\omega_k^2 = \left(\frac{\omega_p}{2}\right)^2 = \omega_n^2 \left[1 - \frac{2\Gamma^2 H_E H_N}{\Omega_k^2}\right] \quad (3.8)$$

Since the pump frequency ω_p is fixed in the parallel pumping experiment ω_k must also be fixed. This is depicted in Fig. 3-12. From this figure it is easy to see how various magnons with wavevector k and a frequency $\frac{\omega_p}{2}$ are selected by changing the external field.

The critical fields are converted to relaxation rates using Table II-1 and the external field values are converted into k values by Eqs. (2.93) and (2.94). The relaxation rates versus wavevector k are shown in Fig. 3-13 and 3-14 for CsMnF_3 and RbMnF_3 , respectively. It is impossible to relate exactly the external field H to the wavevector magnitude k in CsMnF_3 because of its hexagonal structure but since α approximates β in Eq. (2.94) one makes at most an eight percent error using a value midway between α and β . It is interesting to note the large difference in the relaxation rates of CsMnF_3 and RbMnF_3 . The measured nuclear relaxation rates for RbMnF_3 and CsMnF_3 at $k = 5 \times 10^4 \text{ cm}^{-1}$ and $T = 1.2^\circ\text{K}$ are 12×10^5 and 2×10^4 respectively. Such a difference is not predicted by Richards' theory and is not understood in view of existing theories. If the relaxation rate at various temperatures for $k = 0$ is examined the temperature does not follow exactly the expected linear temperature dependence as predicted by Richards' theory, but is found experimentally to have a $T^{1.3}$ temperature dependence. This is shown in Fig. 3-15. The relaxation rates inferred from transient decay are included in this graph. The temperature dependence is approximately the same for all measured values of k .

Fig. 3-12 Ability of external magnetic field to select various wavevectors when nuclear spinwave frequency ω_k is fixed. Wavevectors k_1 and k_2 are selected by external magnetic fields H_1 and H_2 respectively.

Fig. 3-13 Relaxation rates versus wavevector in CsMnF_3 . The solid line represents Richards' theory. The dash-dot line represents the inclusion of inhomogeneities. ● this work $T = 4.2^\circ\text{K}$. ▲ this work $T = 1.2^\circ\text{K}$. ■ Seavey's inferred nuclear relaxation rates at $T = 4.2^\circ\text{K}$.

Fig. 3-14 Relaxation rates versus wavevector in RbMnF_3 . The solid line represents Richards' theory. The dash-dot line represents the inclusion of inhomogeneities.

Fig. 3-15 Temperature dependence of nuclear spinwave relaxation rates in CsMnF_3 . ● Critical field data. △ Transient decay.

The transient decay is related to a relaxation rate by the following

$$\tau^{-1} = 4\eta_k$$

where τ is the characteristic decay time of the reflected power. The factor of four arises from the fact that power is proportional to the fourth power of the spinwave amplitude for two magnon processes. Typical transient decay data for CsMnF_3 at 1.2°K is shown in Fig. 3-16. The nuclear relaxation rates inferred from transient decay agree well with the nuclear relaxation rates inferred from critical field measurements as shown in Fig. 3-13. At the higher temperatures it is impossible to measure the transient decay.

Although the nuclear spinwave relaxation rates inferred from critical field measurements are independent of k , it becomes difficult to resolve the transient decay from the cavity ringing at higher k values because of the increased incident power (see Fig. 3-10).

D. Relaxation Mechanisms

We now attempt to understand the large discrepancy in the RbMnF_3 relaxation rates measured and the nuclear relaxation rates predicted by Richards' theory involving two magnon scattering.³⁰ This theory arrives at the expression

$$\eta_k = \frac{\pi}{6} \frac{I(I+1)}{N} \sum_q \left(\frac{\Delta\omega_q}{\langle I_z \rangle} \right)^2 \delta(\omega_k - \omega_q) \quad (3.9)$$

where I is the nuclear spin, $\langle I_z \rangle$ the thermal average of the z component of nuclear spin, N is the number of spins in a sublattice, hence half the total number, and $\Delta\omega_q$ is the difference between ω_n and ω_q . The approximation is made that the pulling is small. Therefore, the square

Fig. 3-16 (a) Observed transient decay of nuclear spinwaves in CsMnF_3 corresponding to situation in Fig. 3-5a. (b) Transient response corresponding to Fig. 3-5b.

root in the expression for the nuclear spinwave frequency can be expanded as shown below

$$\omega_K = \omega_n \sqrt{1 - \frac{2H_E H_N}{\Omega_K^2}} \approx \omega_n - \frac{\omega_n H_E H_N}{\Omega_K^2} \quad (3.10)$$

The above quantities are defined in Chapter II. This relaxation rate can be derived in a straightforward manner by use of the "Golden Rule" of time dependent perturbation theory. In the time dependent perturbation calculation it is the Suhl-Nakamura Hamiltonian^{4,5} appropriate to canted antiferromagnets which is used to compute matrix elements. Richards points out that when pulling exists the number of states on speaking terms with the excited state is greatly reduced. The narrowing due to the pulling is analogous to "motional" narrowing of paramagnetic resonance lines.

Consider now the case in which there exists a spread in the electronic frequencies due to sample inhomogenities. The electronic frequency is given by

$$\Omega_K^2 = \Omega_{10}^2 + \alpha K^2 \quad (3.11)$$

where $\Omega_{10}^2 = \Gamma^2(H^2 + 2H_E H_N + \frac{3}{2} H_E H_A)$ and $\alpha = \frac{1}{3} a^2 H_E^2$ for RbMnF_3 . These can be similarly defined for CsMnF_3 by reference to Eq. (2.94).

There is a spread $\Delta\Omega$ in electronic frequencies caused by the inhomogenities and is of the order of $\Gamma\Delta H$, where Γ is the electronic gyromagnetic ratio and ΔH is the full width at half maximum power of the antiferromagnetic resonance (AFMR). Immediately it can be seen that the density of nuclear spinwave states is increased near $k = 0$ as shown in

Fig. 3.17. Also, the ΔH for RbMnF_3 is typically 200 Oe, while corresponding for CsMnF_3 , it is 20 Oe. This appears to be a possible mechanism to explain the difference in the relaxation rates of CsMnF_3 and RbMnF_3 . The CsMnF_3 rates agree somewhat with Richards' theory, and with the above in mind, this is reasonable since the small AFMR width for CsMnF_3 contributes only a small addition to the density of states. On the other hand, the order of magnitude larger AFMR widths in RbMnF_3 leads to the possibility of a much larger density of states for small values of k . Further conjecture can only be substantiated by actually calculating the nuclear relaxation rates from Eq. (3.9) using the broadened AFMR spectrum given by Eq. (3.11).

For simplicity, it is assumed that there exists an equal population of spins of frequency between $\omega_k + \Delta\omega/2$ and $\omega_k - \Delta\omega/2$, and the frequency of any spin must lie within these limits. The sum is replaced in the usual manner by an integral

$$\sum_{\underline{g}} \rightarrow \frac{V}{8\pi^3} \int d\underline{g} \rightarrow \frac{V}{2\pi^2} \int dg \quad (3.12)$$

Eq. (3.11) is rewritten in the following manner

$$\Omega_k^2 = \Omega_{10}^2 + \Omega_{10} \Gamma \Delta H \epsilon_k + \alpha k^2 \quad (3.13)$$

where Ω_{10} is now taken to be the central frequency and ϵ_k is a dimensionless parameter which varies between ± 1 throughout the sample. We have assumed $\Gamma \Delta H \ll \Omega_{10}$. Using Eqs. (3.13) and (3.10) allows the delta function to be written

$$\delta(\omega_k - \omega_g) = \delta \left[B \left(\frac{\Omega_k^2 - \Omega_g^2}{\Omega_g^2} \right) \right] = \delta [f(g, \epsilon_g)]$$

with $B = \frac{\omega_n \Gamma^2 H_E H_N}{\Omega_K^2}$ and $f(q, \epsilon_q) = B \left(\frac{\Omega_K^2 - \Omega_q^2}{\Omega_q^2} \right)$

The relaxation rate of Eq. (3.9) may now be written

$$\eta_K = \frac{1}{2} \left(\frac{\Omega_K^2 B}{\langle I_z \rangle} \right) \frac{I(I+1)}{12\pi} \times \int_{K_1}^{K_2} q^2 dq \int_{-1}^1 \frac{\delta[f(q, \epsilon_q)]}{\Omega_q^4} d\epsilon_q \quad (3.14)$$

The limits of integration, k_1 and k_2 , are discussed later, but are chosen such that the argument of the delta function is always zero in the range of the integration (see Fig. 3-17). The factor of one half appears because of normalization. Introduction of ϵ_q means the following must hold

$$\frac{V}{2\pi^2} \int q^2 dq \int_{-1}^1 d\epsilon_q = N$$

The above is the mathematical requirement that the number of spinwave states must be constant regardless of the introduction of ϵ_q . The integral over ϵ_q is performed first using the fact that

$$\int_{\alpha_1}^{\alpha_2} g(x) \delta[f(x)] dx = \frac{g(x_0)}{\left| \frac{df(x)}{dx} \right|_{x=x_0}}$$

where the bars indicate absolute value and x_0 is the root of $f(x)$. This holds as long as $\alpha_1 \leq x_0 < \alpha_2$. Such is the case for the integration at hand because of the judicious choice of the limits (see Fig. 3-17). With this in mind, the integral over ϵ_q yields

Fig. 3-17 Broadening of nuclear spinwave spectrum by inhomogeneities in the electronic system.

$$\int_{k_1}^{k_2} q^2 dq \int_{-1}^1 \frac{\delta[f(q, \epsilon_q)]}{\Omega_q^4} d\epsilon_q = \frac{1}{B\Omega_{10}\Delta\Omega_{10}\Omega_K^2} \times \int_{k_1}^{k_2} q^2 dq$$

Next the integral over q can be performed with ease

$$\frac{1}{B\Omega_{10}\Gamma\Delta H\Omega_K^2} \int_{k_1}^{k_2} q^2 dq = \frac{k_2^3 - k_1^3}{3B\Omega_{10}\Gamma\Delta H\Omega_K^2} \quad (3.15)$$

The values of k_1 and k_2 are those values of k corresponding to the largest and smallest values of ϵ_k compatible with a constant ω_k as shown in Fig. 3-10. The relationship relating ω_k and k is

$$\propto k^2 = \frac{\omega_n H_E H_N}{\omega_n - \omega_K} - \Omega_{10}^2 - \Omega_{10} \Gamma \Delta H \epsilon_K \quad (3.16)$$

where use is made of Eq. (3.11).

The values of k_1 and k_2 are

$$k_1 = \left(\frac{\Omega_K^2 - \Omega_{10}^2 - \Omega_{10} \Gamma \Delta H}{\alpha} \right)^{1/2} \text{ for } \Omega_K^2 - \Omega_{10}^2 - \Omega_{10} \Gamma \Delta H > 0$$

$$k_1 = 0 \quad \text{otherwise} \quad (3.17)$$

$$k_2 = \left(\frac{\Omega_K^2 - \Omega_{10}^2 + \Omega_{10} \Gamma \Delta H}{\alpha} \right)^{1/2}$$

Combining the above with Eqs. (3.14) and (3.15), one finds the relaxation rate for nuclear spinwaves in the context of Richards' theory to be

$$\eta_k = \frac{1}{2} \left(\frac{\Omega_k^2 B}{\langle I_z \rangle} \right) \frac{I(I+1)}{12\pi} \left(\frac{V}{N} \right) \frac{K_2^3 - K_1^3}{3B\Omega_{10}\Gamma\Delta H\Omega_k^2} \quad (3.18)$$

$\frac{V}{N}$ is taken to be $2a^3$ for RbMnF_3 and $a^2 c/\sqrt{3}$ for CsMnF_3 . The following definition is useful in reducing Eq. (3.16) to a more usable form.

$$\Gamma H_N = A \langle I_z \rangle = \frac{1}{3} A I(I+1) \hbar \omega_n / k_B T \quad (3.19)$$

The high temperature limit has been used. Combining Eqs. (3.18) and (3.19) and using the definition of B yields

$$\eta_k = \frac{1}{2} \left(\frac{A \Gamma H_E}{8\pi} \right) \left(\frac{V}{N} \right) \frac{k_B T}{\hbar} \frac{K_2^3 - K_1^3}{3\Omega_{10}\Gamma\Delta H} \quad (3.20)$$

The factor of one half comes from summing over one half the total number of modes since relaxation via the field independent branch is not included.

This relaxation rate agrees with Richards in the limit of large k when $\Gamma\Delta H$ approaches zero, which it must. There is a question of the functional form of the inhomogeneity, i.e. the distribution of ϵ_q . Using for example a triangular rather than a square distribution of ϵ_q in Eq. (3.13) would give a slightly different rate, but the general features remain the same. The results of Eq. (3.20) are shown in Figs. 3-13 and 3-14.

In the analysis just presented the relaxation rates at high k values are unaffected by the addition of the broadening of the high frequency electronic mode. This is reasonable because the density of states due to the broadening of the electronic mode is considerably reduced for high k values over the density of states at $k = 0$ as can be

Fig. 3-18 (a) High density of states for low wavevector. (b) Reduction in the density of states at high wavevector.

seen in Fig. 3-18 (see also Fig. 3-17). Furthermore, at high k values the density of states due to inhomogeneity broadening becomes constant while the intrinsic density of states increases linearly with k .

It is clear (see Fig. 3-14) that the result of including broadening of the electronic mode did not account for the large discrepancy between the theoretical and experimental values for the relaxation rates in RbMnF_3 . One instructive thing learned is that for low k values the relaxation rate is considerably higher than that calculated by Richards. He assumed that at k values near $k = 0$ the relaxation rate is governed by broadening due to the small effects of the dipolar originated demagnetizing fields. In light of the foregoing, this broadening is completely negligible in comparison to the inhomogeneous broadening caused by the electronic mode. For CsMnF_3 the data, as shown in Fig. 3-13, is reasonably well explained for low k values by this approach. In general the addition of the broadening of the electronic system does not account for the observed k dependence of the nuclear relaxation rates in CsMnF_3 .

In light of the results of Chapter II in which it is seen that three magnon terms are important in the determination of the parallel pump thresholds it might be asked whether three magnon type scatterings can be effective in relaxation. Woolsey and White (WW)²⁷ consider three magnon relaxation processes for electronic spinwaves in a flopped antiferromagnet where the requirement of conservation of momentum is relaxed because of inhomogeneities. Also, three magnon terms cancel to order $\left(\frac{\Gamma^H_E}{\Omega_k}\right)^{\frac{1}{2}}$ as noted in Chapter II, but the effect of inhomogeneities removes this cancellation. It is obvious that a reasonable model for the strains must be formulated before their exact influence on relaxation can be accurately assessed. The model chosen by WW is arrived at because

it is physically reasonable and gives experimentally observed results for electronic relaxation rates.

It is worthwhile to examine which three magnon processes are possible for nuclear spinwave relaxation due to the exchange Hamiltonian discussed in Chapter II. These are three magnon splitting and confluence processes and are shown in Figs. 3-19a and 3-19b respectively.

Conservation of energy for the processes which are of interest requires

$$\omega_1 = \omega_2 + \omega_3 \quad (\text{splitting})$$

$$\omega_1 + \omega_2 = \omega_3 \quad (\text{confluence})$$

where ω_1 is the frequency of a nuclear spinwave and ω_2 and ω_3 are either nuclear or electronic frequencies.

These three magnon relaxation processes due to the exchange are small because such terms vanish to order $\left(\frac{\Gamma_H E}{\Omega_k}\right)^{\frac{1}{2}}$. Most of the possible processes are ruled out by their inability to satisfy conservation of energy while others are excluded because the coefficients of such three magnon terms are rigorously zero if the sample is homogeneous. With the inclusion of the effects of inhomogeneity in the sample three magnon relaxation effects might become appreciable.

E. Conclusion

It does not seem possible to understand in any simple manner the large differences between the nuclear relaxation rates in RbMnF_3 and CsMnF_3 . Also, the k dependence of the nuclear relaxation rates is not precisely understood. Richards' theory provides order of magnitude agreement for the relaxation rates in CsMnF_3 , but the exact agreement must be considered poor. It appears that the temperature dependence of

Fig. 3-19 (a) Three magnon splitting process. (b) Three magnon confluence process.

the nuclear relaxation rates in CsMnF_3 is approximately linear with temperature as predicted by Richards but, because of the narrow range of temperature an exponential dependence is not excluded.

M.H. Seavey¹⁹ recently observes in parallel pumping experiments magnon phonon interactions in CsMnF_3 at temperatures below 2°K. In these experiments two electronic spinwaves are pumped. Such an interaction is difficult to observe in the parallel pumping of two nuclear spinwaves. Such an effect is detected by an increase in the observed relaxation rate at a particular wavevector. For nuclear spinwaves the wavevector where such an effect could be observed is

$$k = \frac{\omega}{V}$$

where ω is the frequency of the phonon and V its velocity. Using typical numbers for CsMnF_3 yields¹⁹ a k value of 10^4 cm^{-1} . This is such a low value of k that it would be reasonable that the increase in relaxation rate due to the magnetoelastic crossover is masked by the large relaxation rate due to the inhomogeneities.

This work represents the first excitation of two nuclear spinwaves by parallel pumping and hence, the first direct measurement of nuclear relaxation rates. In performing the parallel pumping experiments one obtains the characteristic "butterfly" curve when the critical rf fields are plotted against external field. These "butterfly" curves show a minimum at the predicted values of external field and this gives confidence in the experiments. In general, the parallel pumping of two nuclear spinwaves can be a useful tool for the study of nuclear relaxation rates.

IV. PARALLEL PUMPING OF ONE ELECTRONIC AND ONE NUCLEAR SPINWAVE

A. Introduction

The theory of Chapter II makes two very apparent predictions. First, that it is impossible to excite simultaneously spinwaves by parallel pumping if the magnons, either nuclear, electronic, or both, are of the field independent type. Secondly, the threshold rf field for parallel pumping one electronic and one nuclear spinwave of the field dependent branch becomes very small as the pump frequency approaches the frequency of the field independent electronic mode. These two predictions are a direct consequence of the inclusion of a Suhl first order type process and hence, the reasonableness of this assumption can be tested. The sample used is RbMnF_3 because its field independent mode lies conveniently at 10.7 GHz, a frequency easily accessible to experiment.

B. Experimental Method

The sample of RbMnF_3 examined is approximately a cube whose sides have a length of 3 mm. This sample is placed in the bottom of a TE_{10n} cavity made from modified standard X band copper waveguide. The sample is attached to the bottom by double sided "scotch tape" and removed by dissolving the tape with ethyl acetate. The sample is mounted such that the $\langle 100 \rangle$ direction is parallel to the rf as well as the external magnetic fields. The cavity is attached to a reflection type bridge spectrometer. The spectrometer is shown in Fig. 4.1. Rf power is provided by a varian X-3B klystron which is amplified by a Litton Model 539 pulsed traveling wave tube (TWT) amplifier. The TWT is operated slightly saturated to produce flat pulses. Frequency modulation of

Fig. 4-1 Pulsed X band spectrometer

different parts of the pulse is undetectable. The pulses are twenty microseconds in length and occur at a repetition rate of less than fifty pulses per second. The cavity is immersed in the liquid helium bath to minimize sample heating. Two X band cavities are used, each resonant at two frequencies in a TE_{101} and a TE_{102} mode. The wide dimension of the waveguide as well as the cavity length is adjusted to make the cavity resonate at the desired frequency and to maintain a reasonable Q . Q 's are measured by observation of the ring time of the cavity. The cavity field is computed using the relation

$$h^2 = \frac{.16 P_{abs} Q_0}{f V [1 - (\lambda_g / \lambda_c)^2]}$$

where P_{abs} is in watts, f in GHz, V in cm^3 and Q_0 is the unloaded cavity Q . λ_g and λ_c are the guide and cut off wave lengths respectively.

The measurement at 13 GHz is made by using K_u waveguide components in a configuration shown in Fig. 4-2. The cavity is a TE_{102} type. Microwave power is provided by a Varian X-12 klystron and amplified by a Varian 617-G TWT. AEL model SWB141A diode switches are used to switch the rf. These diode switches are driven by a Hewlett Packard model 212A pulser. Again, the cavity is immersed in the liquid helium.

In both systems it is found that spectrometer sensitivity is significantly enhanced when the cavity is not critically coupled. This allows reflected power to bias the detecting crystal into a more sensitive region. The threshold power is taken to be that power level at which the reflected pulse obtains its characteristic nonlinear shape.

Fig. 4-2 Pulsed Ku band spectrometer

C. Discussion of Results

The value of the critical field at $k = 0$ at different frequencies is plotted in Fig. 4-3. Obviously, there exists a minimum in the vicinity of the field independent mode. This alone gives a strong support of the validity of the theory of Chapter II. To check details of the results, the following procedure is used. The expression in Table 2.1 for the critical fields versus relaxation rates is used together with known electronic and nuclear relaxation rates to provide a prediction for the critical fields for the excitation of one electronic and one nuclear field dependent spinwave. This predicted critical field is then compared to the measured values.

The first question one must ask is, do the relaxation rates themselves depend upon frequency? Richards' theory predicts that nuclear relaxation rates are independent of frequency.^{21,30} Thus, one can use with some degree of confidence the values for nuclear relaxation rate measured for RbMnF_3 in Chapter III for all the pumping frequencies used here even though the value of the nuclear spinwave frequency changes. The nuclear frequencies are always above 500 MHz in the experiments performed in this chapter.

The three following equations must hold in the electronic-nuclear experiment for RbMnF_3

$$\omega_p = \omega_k + \Omega_k \quad (4.1)$$

$$\omega_k^2 = \omega_n^2 \left(1 - \frac{2\Gamma^2 H_E H_N}{\Omega_k^2} \right) \quad (4.2)$$

$$\Omega_k^2 = \Omega_{10}^2 + \alpha k^2 \quad (4.3)$$

Ω_k and ω_k are the frequencies of the electronic and nuclear spinwave

Fig. 4-3 Critical field data taken near the field independent mode resonance frequency of 10.7 GHz. Solid line is predicted critical field (see text).

respectively, while ω_p is the pumping frequency. Eqs. (4.2) and (4.3) are discussed in Chapter III. Eq. (4.1) is simple conservation of energy. These three equations determine uniquely the electronic and nuclear frequencies for a given pumping frequency. The nuclear relaxation rates of RbMnF_3 reported in Chapter III are measured at 1.15°K. Since the present work is done at 4.2°K the relaxation must be corrected accordingly. They are assumed to follow the empirically determined $T^{1.3}$ behavior noted in CsMnF_3 rather than the linear dependence predicted by Richards.

One must decide the exact form for the electronic spinwave relaxation rates. For the frequency range concerned here the electronic spinwave relaxation rates for RbMnF_3 are taken to follow an exponential frequency dependence of the form

$$\eta_k^e = 5.5 \times 10^9 \exp(2\pi f / 1.05 \times 10^{10}) \quad (4.4)$$

This follows from the work of Woolsey and White in which they considered three magnon confluence processes involving only electronic spinwaves when³¹ $ka = .001$. The electronic relaxation rates are shown in Fig. 4-4. This theoretical expression for the electronic relaxation rates can be checked at one frequency. Seavey measures the relaxation rate of $1.1 \times 10^7 \text{ sec}^{-1}$ at a pump frequency of 16.9 GHz. To infer the relaxation rate from the critical rf field he uses the relationship of Hinderks and Richards. When one makes corrections, as pointed out in Chapter II, he obtains $1.5 \times 10^7 \text{ sec}^{-1}$ which is in fairly reasonable agreement with Woolsey and White's theory. Their theory is thus used for the electronic relaxation rate.

Combining their results for electronic relaxation rates with measured nuclear relaxation rates yields the solid curve shown in Fig. 4-3. In this graph the shape of the critical fields versus frequency is affected little by the value of η_{20} , the phenomenological damping factor of the field independent mode except for the region extremely near the frequency of the field independent mode. The value of η_{20} used in Fig. 4-3 corresponds to a full width of 200 Oe. The fit of the experimental points is remarkable considering the assumptions and the fact that there are no adjustable parameters. This shows the necessity for the inclusion of the Suhl first order type process and gives confidence in the frequency dependence of the electronic relaxation rates predicted by Woolsey and White.

Further support for the theory of Chapter II is provided by the inability to excite one nuclear and one electronic field independent spinwave. To make an estimate of the critical fields required for the excitation of one electronic and one nuclear spinwave from the field independent branch, the following values are inserted in the appropriate formula in Table II-1: $\eta_{2k}^e = 1.0 \times 10^7 \text{ sec}^{-1}$, $\eta_{2k}^n = 5.0 \times 10^6 \text{ sec}^{-1}$, $T = 4.2^\circ\text{K}$, $f_p = 11.5 \text{ GHz}$, $H_E = 8.1 \times 10^5 \text{ Oe}$, $H_N = 9.43/T \text{ Oe}$, $H = 4.5 \times 10^3 \text{ Oe}$ and $f_{2k} = 540 \text{ MHz}$. The result is a threshold field of the order of 30 Oe. Only slightly discernable nonlinear processes are observed for parallel pumping at a frequency of 11.5 GHz and input powers on the order of 200 watts. At this pumping frequency the cavity is highly perturbed by the large linear susceptibility of the field independent mode and analysis is unclear. At any rate, there is no easily detectable

nonlinear effect, as is expected from the preceeding estimate.

A process in which two nuclear spinwaves from the field independent branch of RbMnF_3 are excited by parallel pumping was tried unsuccessfully at $T = 1.2^\circ\text{K}$ and with rf fields estimated to be up to 50 Oe. This also is an expected result of Chapter II since the critical field required for the excitation of two field independent nuclear spinwaves is a factor of 60 greater than the critical fields for the excitation of two field dependent spinwaves of the same frequency.

It does not seem possible to excite two electronic field independent spinwaves with $k > 0$. When $\Omega_{2k}/\Omega_{20} < 2$, the threshold field is actually 30 times that of the corresponding field dependent process if the frequencies of the spinwaves excited are the same, and the relaxation rates are assumed approximately equal.

Fig. 4-4 Woolsey and White's predicted electronic relaxation rates as a function of frequency for RbMnF_3 at 4.2°K . The point is Seavey's inferred value from parallel pumping.

D. Other Effects Near Resonance Of Field Independent Mode

The data of Fig. 4-3 taken near the frequency of 10.7 GHz is of questionable accuracy. Because of slight sample misalignment, the field dependent and independent electronic modes are coupled slightly. Thus new modes are formed from combinations of the old modes. This can be seen in Fig. 4-5 where the measured field for resonance is plotted against frequency. It can be seen that whenever the frequency of the field independent and field dependent mode are approximately equal, care must be taken in analyzing critical field data. Such is the case for the excitation of nuclear and electronic spinwaves when the pump frequency is in the neighborhood of the field independent mode.

An interesting effect, previously unnoted, is observed in the foregoing experiment. There exist two distinct thresholds approximately 3 dB in power apart at high k values, and the two thresholds merge into one at $k = 0$. This double threshold occurs at the two frequencies near 10.7 GHz. No data is presented because the effect changed from one experiment to the next, suggesting orientation dependence. Excitation of two electronic spinwaves is ruled out because the field for $k = 0$ is approximately the spin flopping field H_{SF} , which is 2400 Oe. Above this field such excitation is impossible. For the present case the fields are at least 1000 Oe above H_{SF} . It is known that excitation of two electronic spinwaves at fields above the field for $k = 0$ is possible, but such smearing out does not extend more than 300 Oe above $k = 0$.

Fig. 4-5 Resonance field as a function of frequency near the frequency of the field independent mode (dashed line). Deviation from predicted value (long dash, short dash) is caused by sample misalignment which can couple the two modes.

E. Conclusion

It appears that there is good justification for the theory of Chapter II. The ability to predict the critical field for the electronic-nuclear process in RbMnF_3 from the nuclear and electronic relaxation rates gives one confidence in the experiments and theory which are used for the prediction.

V. CONCLUSION

This work predicts that it is possible to excite by parallel pumping electronic, electronic-nuclear and nuclear spinwaves in canted low anisotropy antiferromagnets with a large hyperfine interaction. To check this possibility electronic-nuclear and nuclear spinwaves have been excited by parallel pumping by this author and electronic spinwaves by M.H. Seavey in CsMnF_3 and RbMnF_3 . Experimental results give confidence in the interpretation that nuclear spinwaves are being excited. In particular the position of the minimum in the butterfly curves is where it should be, and large rf fields are required to excite the instability when the external field is greater than the field corresponding to $k = 0$.

It is shown that the Holstein-Primakoff formalism applied to a canted antiferromagnet predicts generally the same results as Platzker and Morgenthaler (MP). The difference between this work and MP manifests itself only when $(\omega_p^2 - \omega_n^2)/(\omega_p^2 - \omega_{20}^2)$ deviates significantly from unity for acceptable pumping frequencies. This situation only arises in RbMnF_3 when parallel pumping the field dependent mode at frequencies near ω_{20} . MP predict that pumping at this frequency would require infinitesimally small rf fields, a result which should be possible to check experimentally in future experiments.

Nuclear relaxation rates measured experimentally generally are in disagreement with theory. For CsMnF_3 the predicted k dependence of the nuclear relaxation rates is not in exact agreement with that which is observed. The disagreement is more serious in RbMnF_3 . Predicted nuclear relaxation rates are more than an order of magnitude lower than the

observed ones. It appears, therefore, that the effects of strains must be included in a theory which attempts to explain nuclear relaxation rates. The observed temperature dependence of $T^{1.3}$ is not too far removed from the predicted linear dependence, however.

It would be interesting to measure electronic relaxation rates as a function of frequency as well as critical fields over a wide range of frequencies for the electronic-nuclear process in RbMnF_3 . The general problem of relaxation in canted antiferromagnets using parallel pumping as the mechanism for measuring the relaxation rates appears to be a field which lends itself to much more investigation.

REFERENCES

1. P.G. deGennes, P.A. Pincus, F. Hartman-Boutron, and J.M. Winter, Phys. Rev. 139, 1105 (1963).
2. S. Ogawa, J. Phys. Soc. Japan 15, 1475 (1960).
3. A.J. Heeger, A.M. Portis, D.T. Teaney and G. Witt, Phys. Rev. Letters 7, 307 (1961).
4. H. Suhl, Phys. Rev. 109, 606 (1958); J. Phys. Radium 20, 333 (1960).
5. T. Nakamura, Prog. Theoret. Phys. (Kyoto) 20, 547 (1958).
6. For example, M. Sparks, Ferromagnetic Relaxation Theory, (McGraw-Hill Book Co., New York, 1964).
7. F. Ninio and F. Keffer, Phys. Rev. 165, 735 (1968).
8. L.W. Hinderks and P.M. Richards, J. Appl. Phys. 39, 824 (1968).
9. L.W. Hinderks and P.M. Richards, Phys. Rev. 183, 575 (1969).
10. M.H. Seavey, J. Appl. Phys. 40, 1597 (1969).
11. B.T. Adams, L.W. Hinderks and P.M. Richards, J. Appl. Phys. 41, 931 (1970).
12. A. Platzker and F. Morgenthaler, J. Appl. Phys. 41, 927 (1970).
13. T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).
14. F. Keffer and C. Kittel, Phys. Rev. 85, 329 (1952).
15. W.J. Ince and P.H. Cole, Phys. Rev. 150, 1122 (1966).

16. K. Lee, A.M. Portis, and G.L. Witt, Phys. Rev. 132, 144 (1963).
17. M.H. Seavey, Thesis, Harvard (1970).
18. M.J. Freiser, P.E. Seiden, and D.T. Teaney, Phys. Rev. Letters 10, 293 (1963).
19. M.H. Seavey, Phys. Rev. Letters 23, 132 (1969).
20. H.J. Suhl, J. Phys. Chem. Solids, 1, 209 (1957).
21. A. Platzker reports in a private conversation that there is a frequency dependence for the nuclear relaxation rates in RbMnF_3 as measured by phonon created instabilities (see Ref. 12). These results will appear in his Ph.D. thesis.
22. W.J. Ince, Thesis, MIT, 1969.
23. L.B. Welsh, Phys. Rev. 156, 370 (1967).
24. R.M. White, M. Sparks and I. Ortenburger, Phys. Rev. 139, A450 (1965).
25. F.R. Morgenthaler, J. Appl. Phys. 31, 95S (1960).
26. E. Schlomann, J.J. Green and J. Milano, J. Appl. Phys. 31, 386S (1960).
27. R.B. Woolsey and R.M. White, Phys. Rev. 188, 813 (1969).
28. R.M. White and M. Sparks, Phys. Rev. 130, 632 (1963).
29. D.T. Teaney, M.J. Freiser and R.W.H. Stevenson, Phys. Rev. Letters 9, 212 (1962).
30. P.M. Richards, Phys. Rev. 173, 581 (1968).

31. Private communication with R.B. Woolsey and R.M. White in conjunction with their work in Ref. 27.
32. R.E. Watson and A.J. Freeman, Phys. Rev. 123, 2027 (1961).
33. J.R. Pierce, Traveling Wave Tubes, (Van Nostand Book Co., New York, 1950).
34. For proper evaluation of C_{24} , one cannot make the approximation that $j_2 = m_2$.