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Presented in Partial Fulfillment of the Requirements for
the Degree Doctor of Philosophy in the Graduate
School of The Ohio State University

By

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1977

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I. INTRODUCTION

The low lying magnetic excited states of a ferromagnet are called spin waves. These excitations consist of one flipped spin which is distributed coherently throughout all the spins in the crystal lattice and have the effect of reducing the $z$ component of the total spin quantum number by one. For low energy spin waves with wave vector, $k$, much less than the size of the first Brillouin zone, the energy of a spin wave is proportional to $k^2$ where the proportionality constant is the exchange constant.\(^1\)

The exchange constant is related to the exchange integral which couples neighboring spins and it acts as a measure of the overall strength of the magnetic interaction. In a simple ordered magnetic system such as a ferromagnet this relationship is a proportionality. However, in a more complicated system such as a ferrimagnet in which the spins are situated on two or more interpenetrating sublattices, there are different exchange integrals coupling the spins on the various sublattices and the exchange constant is proportional to a linear combination of these exchange integrals.

---

1. If dipolar interactions are considered in the Hamiltonian, the $z$ component of the total, $\hat{S}_z^i$, is not a good quantum number and this statement is true only in an approximate sense.
Because it measures the strength of the magnetic interaction, the exchange constant determines the thermodynamic properties of magnetic systems such as the specific heat and the temperature dependence of the magnetization which follows the famous Bloch $T^{3/2}$ relation. It also determines the properties of domains in magnetic materials such as the domain wall thickness, energy density, and mobility.

In this experiment the exchange constant of calcium and germanium substituted Yttrium Iron Garnet (YIG) thin films has been measured using ferromagnet resonance techniques. The magnetic garnets were chosen for investigation for a number of reasons. First of all they are an excellent example of a local moment system for which the Heisenberg exchange Hamiltonian gives a valid description of the ordering interaction. In addition, the garnets are an example of a two sublattice ferrimagnet with a rather complex structure. Because of this many variations in the composition can be made by substituting nonmagnetic ions for iron on either magnetic sublattice and by replacing the Yttrium ions with magnetic rare earth ions. This allows a good control of the magnetic properties of the system such as the Curie temperature, magnetization, and even the shape of the temperature dependence of the magnetization over a limited range of temperatures. Because of the ability to accurately control their

properties and because they are insulators, the garnets have for many years found extensive application in microwave devices where magnetic materials are needed which have low losses at high frequencies. More recently the garnets have been used in magnetic bubble domain applications. In these applications it is desirable to know the exchange constant and how it varies with the composition of the material because of its strong influence in determining the domain wall properties. Most previous measurements of the exchange constant have relied on techniques which are valid only at very low temperatures or on techniques which measure the dynamic response of the domain walls. In the latter case the results have not always been consistent both because of the statistical nature of the measurements and the approximations made in the theory. The use of ferromagnetic resonance on thin films affords a direct measurement of the exchange constant which can, in principle, be done at all temperatures below the Curie temperature. This technique has been used to measure the exchange constant in substituted garnets over a wide range of compositions.

A number of theories have been proposed to explain the dependence of the exchange constant in the magnetic garnets on the level

of substitution. One of these was proposed by Slonczewski et al.\textsuperscript{10} and uses the results of the molecular field theory and the random phase approximation to predict an approximate relationship between the exchange constant and the Curie temperature. A second theory by Kirkpatrick and Harris\textsuperscript{11} uses a percolation\textsuperscript{12} model to relate the exchange constant directly to the level of substitution. The validity of both of these theories has been tested in this work.

Chapter II of this dissertation presents a brief historical review of the theory of ferromagnetism. The topics discussed include the molecular field theory, the Heisenberg exchange interaction, and the Holstein Primakoff method of diagonalizing the magnetic Hamiltonian which includes exchange and Zeeman terms. Also included are a description of the Néel theory of ferrimagnetism, its application to the magnetic garnets, and a review of some of the work which has been done to explain the effects of nonmagnetic ion substitution in an ordered magnetic material. Finally the dispersion relation for spin waves in a thin film has been derived using a semiclassical approach and the effects of the surface interaction on the spin wave spectra have been discussed.

In Chapter III the sample growth and preparation techniques are discussed. Also a description of the experimental apparatus and techniques is given. Finally in Chapter IV the results of the

\begin{itemize}
  \item \textsuperscript{11} Kirkpatrick, S. and Harris, A. B., AIP Conf. Proc., 24, 99 (1974).
  \item \textsuperscript{12} Kirkpatrick, S., Rev. Mod. Phys., 45, 474 (1973). A review of percolation theory.
\end{itemize}
ferromagnetic resonance and magnetization experiments are discussed and a comparison is made with the models of Slonczewski and Kirkpatrick and Harris. In addition, results are described which can possibly be interpreted as evidence of moment canting in the spin system.
CHAPTER II

A. Molecular Field Theory

The first breakthrough in the attempt to understand magnetically ordered systems was probably the theory proposed by Weiss \(^2,13\) in 1907. At the time the problem in magnetism was twofold. The first was to explain the existence of an ordering temperature below which magnetic materials exhibit a spontaneous magnetization. The second was to explain the fact that for temperatures below the ordering temperature the application of a small field changes the magnetization from a negligibly small value to the saturation value. Weiss made two hypotheses to explain this behavior. The first was that below the ordering temperature the material is divided into domains. In each domain the atomic moments are aligned. However, the random orientation of the individual domains causes a magnetization which is much smaller than it would be in a single domain material. The application of a small magnetic field causes the domains to align resulting in a large magnetization. The second hypothesis was that the moments inside a domain were magnetized or aligned due to the presence of a very strong molecular field of unknown origin.

13. Weiss, P., J. de Physique, 6, 667 (1907).
The size of this molecular field, $H_E$, can be estimated since at the ordering temperature or Curie temperature, $T_C$, the thermal energy of a moment must be of the same order as the Zeeman energy due to the molecular field

$$\mu H_E = K T_C$$

where $K$ is the Boltzmann constant and $\mu$ is on the same order of size as a Bohr magneton, $\mu_B$. For iron which has a Curie temperature of $1043 K$, this expression yields $H_E = 10^7$ Oe. This is much larger than any dipolar field which should be of the order $4\pi M$ which is $\sim 2 \times 10^4$ Oe for iron. Weiss assumed that the molecular field could be written in the form:

$$H_E = \lambda M$$

Using this molecular field, the magnetization of a domain can be calculated. For a paramagnetic system of spin $S$ particles, the magnetization is given by:

$$M = n g \mu_B S B_S(g \mu_B SH/K T)$$

where $n$ is the number density of spins, $g$ is the spectroscopic splitting factor, and $B_S(x)$ is the Brillouin function:

$$B_S(x) = \frac{2S+1}{2S} \coth \left( \frac{2S+1}{2S} \right) x - \frac{1}{2S} \coth \frac{x}{2S}$$

This is the quantum mechanical version of the classical theory of paramagnetism due to Langevin and it takes into account the fact that the $z$ component of each spin is quantized. In the Weiss form the field in Eq. 3 is replaced by an effective field, $H = H_O + \lambda M$, where

\( H_0 \) is the applied field. For high temperatures \( x \ll 1 \) and the Brillouin function may be expanded to give the form

\[
B_S(x) = \frac{S+1}{3S} x \quad x \ll 1
\]  

(5)

Using this expansion the susceptibility may be written in the form of the Curie Weiss law:

\[
x = \frac{M}{H_0} = \frac{C}{T-T_C}
\]

(6)

where \( C \) is the Curie constant:

\[
C = \frac{ng^2 \mu_B^2 S(S+1)}{3K}
\]

(7)

and \( T_C = \lambda C \) is the Curie temperature. For \( T<T_C \) there is a spontaneous magnetization for zero applied field which is given by the simultaneous solution of the equations:

\[
M = ng \mu_B S B_S(x) = M(0) B_S(x)
\]

(8)

\[
x = \frac{g \mu_B S \lambda M}{K T}
\]

Figure 1 shows a plot of the temperature dependence of the magnetization for \( S = 1/2, S = 5/2 \), and the classical limit \( S = \infty \).

In spite of its simplicity the Weiss molecular field theory agrees qualitatively with experimental results. It does break down in the regions near \( T = T_C \) where it predicts that \( M \rightarrow 0 \) as \((T_C-T)^{1/2}\) in contrast to experimental results which measure a critical exponent closer to 1/3. It also fails to predict the low temperature result that \( M(0)-M(T) \sim T^{3/2} \) in a local moment system. This can be explained using spin wave theory.
B. Exchange Hamiltonian

It was discovered by Heisenberg\textsuperscript{15} that the molecular field could have its origin in a purely quantum mechanical effect, the Pauli exclusion principle. Consider a two electron system in which spin orbit coupling is neglected. The exclusion principle says that the two electron wave function must be antisymmetric with respect to exchange of particles. In order to satisfy this condition a symmetric orbital wave function must be coupled with a spin function which is antisymmetric and vice versa. The orbital wave functions will be of the form:

\begin{align*}
\psi_{\text{sym}} &= \phi_{\alpha}(\vec{r}_1)\phi_{\beta}(\vec{r}_2) + \phi_{\beta}(\vec{r}_1)\phi_{\alpha}(\vec{r}_2) \\
\psi_{\text{asym}} &= \phi_{\alpha}(\vec{r}_1)\phi_{\beta}(\vec{r}_2) - \phi_{\beta}(\vec{r}_1)\phi_{\alpha}(\vec{r}_2) \\
\end{align*}

where \(\phi_{\alpha}\) and \(\phi_{\beta}\) are the one electron wave functions and \(\vec{r}_1\) and \(\vec{r}_2\) are the coordinates of the 2 electrons. The energy of these two states is given by

\begin{align*}
E_{\text{sym}} &= E_0 + K - J_{\alpha\beta} \\
E_{\text{asym}} &= E_0 + K + J_{\alpha\beta} \\
\end{align*}

where \(E_0\) is the energy of the two 1 electron states.

\begin{align*}
K &= \iint \frac{e^2}{|\vec{r}_1 - \vec{r}_2|} |\phi_{\alpha}(\vec{r}_1)|^2 |\phi_{\beta}(\vec{r}_2)|^2 \, d\vec{r}_1 \, d\vec{r}_2 \\
\end{align*}

\textsuperscript{15} Heisenberg, W., Z. Physik, \textbf{49}, 619 (1928).
FIGURE 1: Temperature dependence of the spontaneous magnetization of a $S = 1/2$, $S = 5/2$, and a classical $S = \infty$ ferromagnet.
is the Coulomb energy and

\[ J_{\alpha \beta} = \int \int \phi_\alpha^*(\mathbf{r}_1) \phi_\beta^*(\mathbf{r}_2) \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \phi_\alpha(\mathbf{r}_2) \phi_\beta(\mathbf{r}_1) \, d\mathbf{r}_1 \, d\mathbf{r}_2 \]

(12)

is the exchange energy. This energy in Eq.10 can be written in a somewhat different form to include the spin. If \( \hat{S}_1 \) and \( \hat{S}_2 \) are the spin of the two electrons, the total spin will be given by \( \hat{S} = \hat{S}_1 + \hat{S}_2 \). The eigenvalues of the operator

\[ S^2 = S_1^2 + S_2^2 + \frac{1}{2} \hat{S}_1 \cdot \hat{S}_2 \]

(13)

will then be

\[ S(S+1) = 2 S_1(S_1+1) + 2 \frac{1}{2} \hat{S}_1 \cdot \hat{S}_2 \]

(14)

Since \( S = 1 \) for the symmetric state and 0 for the antisymmetric state, the operator \( \frac{1}{2} + 2 \frac{1}{2} \hat{S}_1 \cdot \hat{S}_2 \) has eigenvalue 1 for symmetric state and -1 for the antisymmetric state. The energy can then be written in terms of this spin operator:

\[ E = E_0 + K - \left( \frac{1}{2} + 2 \frac{1}{2} \hat{S}_1 \cdot \hat{S}_2 \right) J_{\alpha \beta} \]

(15)

This shows that the electrons behave as if there were a strong magnetic coupling of the form \( \hat{S}_1 \cdot \hat{S}_2 \). In reality this coupling is due to the Coulomb interaction and the exclusion principle. For \( J_{\alpha \beta} > 0 \) the spins will tend to align parallel to each other or ferromagnetically, while for \( J_{\alpha \beta} < 0 \) they will tend to align antiferromagnetically.

For a many electron system this interaction can be written as an exchange Hamiltonian.
Since the exchange integral depends on orbital wave function overlap between electrons, the interaction is probably of very short range. The sum in Eq. 16 therefore will be restricted to nearest neighbor and in some cases next nearest neighbor terms. An approximate connection can be made with the Weiss molecular field by assuming that the spin \( S_1 \) interacts with the average value \( \bar{S} \) of its neighbor spins. For a simple cubic lattice with \( z \) nearest neighbors the Hamiltonian can be approximated by:

\[
H_E = -\frac{1}{2} \left( 2NzJ \right) \bar{S}^2
\]  

(17)

where \( N \) is the number of spins in the system and the factor \( 1/2 \) is introduced to avoid double counting in the sums. Since the magnetization is related to the average spin by \( M = ng_\mu_B \bar{S} \), this Hamiltonian can be rewritten in the form

\[
H_E = -\frac{1}{2} \lambda M^2
\]  

(18)

where

\[
\lambda = \frac{2zJ}{ng_\mu_B^2}
\]  

(19)

is the molecular field constant and

\[
\frac{K_T}{J} = \frac{2}{3} zS(S+1)
\]  

(20)

gives the relationship between the Curie temperature and the exchange integral. The very approximate method used to arrive at Eq. 20 gives a result which is within a factor of 2 of the results from more precise calculations.\(^2\)
Although it is only an approximate form for handling the magnetic interaction between spins, the Heisenberg Hamiltonian has had great success in describing the magnetic properties of materials including the spin wave excitations which account so well for low temperature magnetic effects.

**C. Néel Theory of Ferrimagnetism**

The Weiss molecular field along with Heisenberg's exchange interaction provide a very good qualitative description of the properties of a ferromagnet. In 1948 Néel predicted another type of ordering which might exist. The conditions for this type of ordering are that the exchange interaction between nearest neighbor magnetic atoms is negative and the crystal structure is such that the magnetic atoms can be divided up into two interpenetrating sublattices with atoms on the a sublattice having only neighbors on the b sublattice and vice versa. Under these conditions it is possible that each sublattice is ordered magnetically but the two are aligned antiparallel to each other. If the sublattices are identical, there is no net magnetic moment and the structure is called antiferromagnetic. If the two sublattices are not identical, containing either different types of magnetic atoms or different numbers of magnetic atoms, or if there are more than two sublattices, the ordered system may have a nonzero magnetic moment. Néel called this ordering ferrimagnetic after the ferrites which had been developed at about this time and which had properties consistent with this type

of ordering. In both the antiferromagnetic and ferrimagnetic cases the ordering spontaneously occurs below a certain temperature called the Néel temperature, $T_N$. Smart and Standley have both reviewed Néel's treatment of ferrimagnetism.

Néel's work began by extending the molecular field theory to a multisublattice system. The molecular field acting on the $i^{th}$ sublattice is then:

$$\mathbf{H}_i = \mathbf{H}_0 + \sum_{j} \gamma_{ij} \mathbf{M}_j$$

where $\mathbf{H}_0$ is the external field and $\gamma_{ij}$ represents the interaction coupling the $i^{th}$ and $j^{th}$ sublattice. By symmetry $\gamma_{ij} = \gamma_{ji}$. Consider a two sublattice system consisting of sublattices $a$ and $b$. If $\gamma_{ab}$ is negative the two sublattices will tend to align antiparallel. In the following description $\gamma_{ab}$ will be considered a positive quantity and a minus sign in front of it will represent the negative interaction. The molecular fields can then be written:

$$\mathbf{H}_a = \mathbf{H}_0 + \gamma_{ab} [a \mathbf{M}_a - b \mathbf{M}_b]$$
$$\mathbf{H}_b = \mathbf{H}_0 + \gamma_{ab} [b \mathbf{M}_b - a \mathbf{M}_a]$$

The quantities $\alpha$ and $\beta$ represent $\gamma_{aa}/|\gamma_{ab}|$ and $\gamma_{bb}/|\gamma_{ab}|$ respectively. Let $\lambda$ be the fraction of the magnetic atoms which are on $a$ sites and $\mu = 1 - \lambda$ be the fraction on $b$ sites. The magnetization of each sublattice can then be written:

17. In the literature the ordering temperature of a ferrimagnet is referred to both as $T_N$ and $T_C$.
\[ \hat{M}_a = \lambda g \mu_B S_B (g^B \mu_B \hat{H}_a / KT) \]
\[ \hat{M}_b = \mu g \mu_B S_B (g^B \mu_B \hat{H}_b / KT) \]  

(23)

just as in the Weiss model. If \( \lambda = \mu \) and \( \alpha = \beta \), both sublattices will be identical. Each one will have a temperature dependence which is identical to that of a ferromagnet but the net magnetization will be zero.

The Néel temperature can be evaluated by calculating the susceptibility \( \chi = (M_b - M_a) / H_0 \) in the high temperature range where the argument of the Brillouin function is small. Using the expansion of Eq. 5 the susceptibility is:

\[
\chi = \frac{C(T + \theta)}{(T - T_N)(T - T_N')} 
\]

where \( C \) is the Curie constant defined by Eq. 7:

\[
\theta = C \gamma_{ab} \lambda \mu [2 - \alpha - \beta] 
\]

\[
T_N = \frac{1}{2} C \gamma_{ab} [\alpha \lambda + \mu \beta + \{ (\alpha \lambda - \mu \beta)^2 + 4 \lambda \mu \}^{1/2}] 
\]

\[
T_N' = \frac{1}{2} C \gamma_{ab} [\alpha \lambda + \mu \beta - \{ (\alpha \lambda - \mu \beta)^2 + 4 \lambda \mu \}^{1/2}] 
\]

\( T_N' \), the higher of the two temperatures \( T_N \) and \( T_N' \) defines the ordering temperature. Unlike a ferromagnet where the inverse susceptibility is linear in temperature and goes to zero at \( T_C \), the inverse susceptibility for a ferrimagnet has the shape of a hyperbola and asymptotically approaches a linear form for \( T >> T_N \).

The zero field temperature dependence of the magnetization can be determined by solving Eq. 23 along with Eq. 24 and identifying \( M = |M_b - M_a| \). This process must in general be done numerically.
although many features of the shape of the magnetization curve can be determined without doing the calculation. It is instructive to look at a plane in which $\alpha$ and $\beta$ are plotted with $\lambda$ as an adjustable parameter. This is done in Figure 2 with $\lambda = 0.4$ and $\mu = 0.6$ which are the appropriate conditions for Yttrium Iron Garnet. In the various regions of this plane the shape of the magnetization curve will have various forms which are sketched in Figure 3. The boundaries of the first region can be investigated by looking at Eq. 25 which determines the ordering temperature. If both $\alpha$ and $\beta$ are negative $T^*_N$ will be negative. The hyperbola $\alpha\beta = 1$ with $\alpha$ and $\beta$ both negative, which defines the condition $T^*_N = 0$, is plotted in Figure 2. To the left of this curve, which corresponds to the situation $\alpha\beta < 1$ and is labeled $G$ in the figure, $T^*_N$ is negative. Néel assumed that in this case the system would remain paramagnetic at all temperatures. What is more likely the case however, is that with the large value of $\alpha$ or $\beta$ in this situation, the intrasublattice interaction will be greater than the intersublattice interaction for one of the sublattices and the moments within this sublattice will align antiparallel to each other. In the remainder of the $\alpha-\beta$ plane, $T^*_N$ is positive and the system will order spontaneously below $T^*_N$.

Further subdivision of the $\alpha-\beta$ plane can be made by using the requirement from the third law of thermodynamics that $\partial M/\partial T = 0$ as $T \to 0$. This requires that both sublattices be saturated at $T = 0$. Néel found that for $\alpha < -\mu/\lambda$ the $a-a$ interaction is strong enough to prevent saturation of the $a$ sublattice and that for $\beta < -\lambda/\mu$ the $b-b$ interaction is strong enough to prevent saturation of the $b$ sublattice.
FIGURE 2: Division of the $\alpha - \beta$ plane of a ferrimagnet. The letters P, Q, and V refer to regions for which the $\sigma/\tau$ curves are shown in Figure 3 (after Smart).
FIGURE 3: Shape of the magnetization curve of a ferrimagnet in the regions of the $\alpha - \beta$ plane illustrated in Figure 2 (after Smart).
These conditions, which are represented by the lines ab and be in Figure 2, define two more regions, $M_1$ and $M_2$. Yaffet and Kittel$^{19}$ predicted that in region $M_1$ the strong negative a-a interaction would cause the a sublattice to break up into two equivalent sublattices which would align almost antiparallel but at some angle other than $180^\circ$. The net magnetization of these two sublattices would then line up antiparallel to the b sublattice. The analogous situation should occur in region $M_2$. A phenomenon at least similar to this has occurred in some of the garnets and will be discussed later. In the remaining region of the $\alpha$-$\beta$ plane, the magnetization curve will be similar to that of a ferromagnet. There is however some further subdivision which can be done.

Along the line bc in Figure 2 which is given by $\lambda(1 - \alpha) = \mu(1 - \beta)$ it can be shown from Eq. 22 that the magnitudes of the molecular fields acting on the two sublattices are equal. Under this condition the magnetization of both sublattices will have exactly the same temperature dependence and the overall temperature dependence of the magnetization will be exactly of the Brillouin form. For points above the line bc in region P the magnetization will be an increasing function of temperature near $T = 0$ although it will still have zero slope at $T = 0$. The maximum value of the magnetization will then occur at a nonzero temperature. This is due to the fact that the magnetization of the a sublattice decreases much faster with temperature than the magnetization of the b sublattice. This type of

temperature dependence has been seen by Maxwell and Prichart\textsuperscript{20} in Aluminum substituted Nickel-Ferrites.

The region below line bc can be further subdivided by the line fd which specifies the condition \( \lambda(l+a) = \mu(l+b) \). Under this condition \( \partial M/\partial T = 0 \) at \( T = T_N \). For points in region Q between lines bc and fd the magnetization has negative slope just below the Néel temperature and the direction of the magnetization will be the direction of the b sublattice for all temperatures. The magnetization curves in this region will have roughly the Brillouin form. Below the line fd in region V the slope of the magnetization is positive for temperatures just below the Néel temperature. This means that near \( T_N \) the direction of the magnetization will be that of sublattice a while at \( T = 0 \) the direction is that of sublattice b. Somewhere between \( T = 0 \) and \( T_N \) there is a temperature at which \( M = 0 \) known as a compensation temperature. This phenomenon has been seen in many of the garnets.

Figure 3 shows a sketch of the reduced magnetization plotted against the reduced temperature for typical points in the regions P, Q, and V where the molecular field theory is valid. It should be noted that in region V the measured form of the magnetization will be given by the solid curve since experiments are sensitive to the magnitude of \( M \) only.

The molecular field theory of ferrimagnetism breaks down, just as the Weiss model did, in the low temperature region where it cannot predict the \( T^{3/2} \) temperature dependence of \( M \) and in the high

temperature region where it cannot predict the critical exponents as $T \to T_N$. Nevertheless, it has been used quite successfully in quantitatively describing the temperature dependence of the magnetization outside these regions and in estimating the sizes of the various exchange constants.

D. Spin Wave Theory

A more correct description of ordered magnetic systems is found in the quantum mechanical treatment. Consider a Hamiltonian which includes both Zeeman and Exchange interactions.

$$H = -g \mu_B H \sum_i S_i^z - \frac{1}{2} \sum_{i \neq j} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

(26)

The magnetic field $H$ defines the $z$ axis of quantization and the factor of $1/2$ has been included in the second term to avoid double counting of the spin pairs. The case of a ferromagnet with $J_{ij} > 0$ is considered first. In this case the ground state is that state with all spins aligned parallel to $H$ so that the total $z$ component of the spin system is $NS$ where $N$ is the number of spins. It can easily be shown that this state is an eigenstate of (26). It might be expected that the first excited state is one in which the $z$ component of one of the spins is reduced by one. This state however is not an eigenstate of (26). The true eigenstate is a wavelike disturbance of the spin system with amplitude such that the $z$ component of the total spin of the system is reduced by one.
All of this can be demonstrated by using a series of transformations first used by Holstein and Primakoff\textsuperscript{21} to diagonalize the Hamiltonian. In their original treatment a dipole-dipole interaction term was included. The effect of this term is to introduce an anisotropy in the spin wave dispersion relation which gives rise to the spin wave band. This term has been dropped from the following discussion in the interest of simplicity and will be included later by the introduction of the demagnetization field. The first step in diagonalizing the Hamiltonian is to expand the \( \mathbf{\hat{S}}_i \cdot \mathbf{\hat{S}}_j \) term into its component parts and to replace \( S^x \) and \( S^y \) by the spin raising and lowering operators \( S^+ \) and \( S^- \) which are defined by

\[
S^\pm = S^x \pm i S^y \tag{27}
\]

This replacement yields the result:

\[
H = -g_\mu_B H \sum_i S_i^z - \frac{1}{2} \sum_{i \neq j} 2 J_{ij} \left( \frac{1}{2} S_i^+ S_j^- + \frac{1}{2} S_i^- S_j^+ + S_i^z S_j^z \right) \tag{28}
\]

The first transformation is to introduce harmonic oscillator type creation and annihilation operators\textsuperscript{22} \( a_i^+ \) and \( a_i^- \). The operator \( a_i^+ \) creates a spin deviation at site \( i \) while \( a_i^- \) destroys a spin deviation at site \( i \). Then \( n_i \), the number of spin deviations at \( i \), is given by

\[
n_i = S - S_i^z. \]

Let the state \( |n_i\rangle \) represent the state with spin deviations \( |n_1, n_2, n_3, \ldots, n_i, \ldots, n_N\rangle \). The action of the creation and annihilation operators on \( |n_i\rangle \) is given by:

\[\text{References:}\]
where the operators $a_1$ and $a_1^+$ obey the commutation relations:

$$[a_1, a_1^+] = \delta_{ij}, \quad [a_1, a_j] = [a_1^+, a_j^+] = 0$$

(30)

The operator $S^\pm$ has the raising and lowering properties:

$$S^\pm |S^Z\rangle = (S(S + 1) - S^Z(S^Z \pm 1))^{1/2} |S^Z \pm 1\rangle$$

(31)

It can be seen that the operator $S_1^+$ is very similar in action to the operator $a_1$ while $S_1^-$ is similar to $a_1^+$. Using this fact and the relationship $n_1 = S - S^Z$, the effect of the operators $S^\pm$ on the state $|n_1\rangle$ can be seen to be:

$$S^+ |n_1\rangle = (2S)^{1/2}(1 - \frac{n_1 - 1}{2S})^{1/2} (n_1)^{1/2} |n_1 - 1\rangle$$

(32a)

$$S^- |n_1\rangle = (2S)^{1/2} (n_1 + 1)^{1/2} (1 - \frac{n_1 + 1}{2S})^{1/2} |n_1 + 1\rangle$$

(32b)

A comparison between Eq. 29 and Eq. 32 gives the relationships between $S_1^\pm$ and $a_1$ and $a_1^+$:

$$S_1^+ = (2S)^{1/2}(1 - \frac{a_1 a_1^+}{2S})^{1/2} a_1$$

(33a)

$$S_1^- = (2S)^{1/2} a_1^+(1 - \frac{a_1 a_1^+}{2S})^{1/2}$$

(33b)

$$S_1^Z = S - a_1^+ a_1$$

(33c)
The quantity in the parentheses has the effect of reducing the space of the operators $a$ and $a^+$ from an infinite one to a finite one. This must necessarily be done since the maximum possible value of $<a^+a>$ is $2S$.

At this point Holstein and Primakoff made what is known as the quasisaturation approximation. At low temperatures the magnetization differs little from its zero temperature value. This difference can be equated to the average value of $n_i$, $\langle n_i \rangle$. Therefore the condition:

$$1 \gg \frac{1}{2} \frac{M(0) - M(T)}{M(0)} = \frac{\langle n_i \rangle}{2S} = \frac{\langle a^+_i a_i \rangle}{2S}$$

should hold true. Under this approximation the term $(1 - \frac{a^+_i a_i}{2S})^{1/2}$ in Eq. 33a and Eq. 33b can be set equal to unity. After this is done and Eq. 33 is substituted into Eq. 28, the Hamiltonian has the form:

$$H = -\sum_{ij} J_{ij} S_i^2 - g\mu_B N_S + g\mu_B H \sum_i a^+_i a_i - \sum_{i \neq j} 2J_{ij} S_i^2 (a^+_i a_j - a^+_j a_i)$$

The first two terms in Eq. 35 represent constant terms in the energy of the ground state and can be neglected. The terms containing three and four magnon operators of the form $a^+_i a_j a^+_j a_i$ have also been dropped since they should be small by the quasisaturation approximation. The Hamiltonian in Eq. 35 is still not in diagonal form since the last term has operators which connect different sites of the type $a^+_i a_j$. This type of operator has the effect of shifting the spin deviation from site $j$ to site $i$ which suggests that a wavelike spin deviation must be used.
The second transformation is then a Fourier transformation from
the \( a_i \) and \( a_i^\dagger \) to wavelike operators \( b_k^\dagger \) and \( b_k^\dagger \).

\[
\begin{align*}
    a_i &= \frac{1}{N^{1/2}} \sum_k e^{-i\mathbf{k} \cdot \mathbf{R}_i} b_k^+ \\
    a_i^\dagger &= \frac{1}{N^{1/2}} \sum_k e^{i\mathbf{k} \cdot \mathbf{R}_i} b_k^+
\end{align*}
\]

(36)

The \( \mathbf{R}_i \) is the lattice vector from an arbitrary origin to the site \( i \)
and \( \mathbf{k} \) is summed over its allowed values in the first Brillouin zone.

The inverse transformation is:

\[
\begin{align*}
    b_k^+ &= \frac{1}{N^{1/2}} \sum_i e^{i\mathbf{k} \cdot \mathbf{R}_i} a_i \\
    b_k^\dagger &= \frac{1}{N^{1/2}} \sum_i e^{-i\mathbf{k} \cdot \mathbf{R}_i} a_i^\dagger
\end{align*}
\]

(37)

It can be shown that \( b_k^+ \) and \( b_k^\dagger \) satisfy the same commutation relations
as \( a_i \) and \( a_i^\dagger \). Substituting this transformation into Eq. 35 leads
to the Hamiltonian:

\[
H = g\mu_B H \sum_k b_k^+ b_k^- + \sum_k \left[ \sum_{ij} \frac{2J_{ij} S}{N} \right] b_k^+ b_k^- - \sum_{ij} \sum_{kk'} \frac{2J_{ij} S}{N} e^{i(\mathbf{k} \cdot \mathbf{R}_i - \mathbf{k'} \cdot \mathbf{R}_j)} b_k^+ b_k^- \]

(38)

Since the exchange integral \( J_{ij} \) depends only on the separation between
sites \( i \) and \( j \), i.e. \( J_{ij} = J(|\mathbf{R}_i - \mathbf{R}_j|) \), the replacement \( \delta \simeq \mathbf{R}_i - \mathbf{R}_j \) can
be made. This allows the last term in Eq. 38 to be simplified by use
of the relationship
The Hamiltonian is then in the diagonal form:

$$H = \sum_{\mathbf{k}, \mathbf{k}'} A_{\mathbf{k}} b^+_{\mathbf{k}} b_{\mathbf{k}'}$$  \hspace{1cm} (40)$$

where the dispersion relation $A_{\mathbf{k}}$ is given by:

$$\hbar \omega_{\mathbf{k}} = A_{\mathbf{k}} = g_{\mathbf{u}} B + \sum_{\mathbf{k}} 2J(\delta) S(1 - e^{i\mathbf{k} \cdot \delta})$$  \hspace{1cm} (41)$$

and the sum runs over all lattice vectors $\mathbf{\delta}$. The exchange interaction is due to the Pauli exclusion principle and is therefore expected to act over only a short range. The sum can then usually be restricted to nearest neighbor or at most next nearest neighbor terms. For long wavelength low energy spin waves with $\lambda$ much greater than the lattice constant, the exponential in Eq. 41 can be expanded. The term linear in $\mathbf{k}$ cancels for most crystal symmetries and the first term to contribute is order $k^2$. For a simple cubic lattice of lattice constant $a$, the dispersion relation in zero field becomes:

$$\hbar \omega_{\mathbf{k}} = D k^2$$  \hspace{1cm} (42)$$

where the dispersion constant $D$ is given by:

$$D = 2JSa^2$$  \hspace{1cm} (43)$$

In a two sublattice system the results are similar but the analysis is much more difficult. An excellent review of this case has been written by Keffer. The first problem that must be dealt with in treating the two sublattice systems is the fact that the
state with all spins on one sublattice aligned antiparallel to all spins on the other sublattice is not an eigenstate of the Hamiltonian Eq. 26. Nevertheless this state is assumed to be an approximate ground state and the Holstein Primakoff transformations are used to diagonalize the Hamiltonian. Due to the antiparallel ordering of the assumed ground state, spin deviation operators are defined separately for the spins on each sublattice. Each of these operators is separately Fourier transformed into spin wave creation and annihilation operators and a third transformation which makes linear combinations of these spin wave operators diagonalizes the Hamiltonian.

Since the two sublattice system has more than one magnetic atom per unit cell, the spin wave dispersion relation, in analogy to phonons, has an accoustical branch and optical branches. The optical branches have a large exchange energy and can be neglected at low temperatures. For antiferromagnets Hulthén\textsuperscript{23} and Anderson\textsuperscript{24} found that for small $k$ the spin wave dispersion relation is linear in $k$ unlike the ferromagnetic case. Kaplan\textsuperscript{25} has found that the acoustic branch for ferrimagnets behaves like a ferromagnet and is quadratic in $k$. However, for a ferrimagnet, the dispersion constant contains one intersublattice exchange integral and two intrasublattice exchange integrals. Harris\textsuperscript{26} has calculated the dispersion constant for the ferrimagnetic garnet YIG ($\text{Y}_3\text{Fe}_5\text{O}_{12}$) and found the result:

\begin{equation}
\end{equation}

\begin{itemize}
\item \textsuperscript{25} Kaplan, H., Phys. Rev., \textit{86}, 121 (1952).
\item \textsuperscript{26} Harris, A. B., Phys. Rev., \textit{132}, 2398 (1963).
\end{itemize}
\[ D = \frac{5}{16} (8J_{aa} - 5J_{ad} + 3J_{dd})a^2 \]  

\( J_{aa} \) and \( J_{dd} \) are the intrasublattice exchange integrals and \( J_{ad} \) is the intersublattice exchange integral and \( a \) the lattice constant.

One of the early successes of the spin wave theory was its ability to predict the low temperature thermodynamic properties of ferromagnetic systems. Since spin waves are bosons, the number of spin waves of wave vector \( k \) excited at a temperature \( T \) is:

\[ n_k = \frac{1}{\exp(h\omega_k/KT)-1} \]  

and the average number of spin waves at a temperature \( T \) is:

\[ \langle n_k \rangle = \sum_k n_k \]  

This sum may be converted into an integral over the first Brillouin zone using the appropriate density of states.

\[ \sum_k \rightarrow \frac{V}{(2\pi)^3} \int_{B.Z.} 4\pi k^2 dk \]  

For low temperatures \( n_k \) is so small for large \( k \) that the integral may be extended to infinity. This yields the result:

\[ \langle n \rangle = V \left( \frac{KT}{4\pi D} \right)^{3/2} \zeta(3/2) \]  

where \( \zeta(3/2) = 2.612 \) is the Riemann zeta function. Since the excitation of one spin wave is equivalent to changing the \( z \) component of the total spin by one and reducing the magnetic moment by \( g\mu_B S \), the temperature dependence of \( M \) is given by:
This result is known as the Bloch $T^{3/2}$ law after Bloch who first introduced the spin wave approach and predicted the $T^{3/2}$ dependence of $M$. It has been verified in both ferromagnetic and ferrimagnetic systems. The specific heat per unit volume also shows the same $T^{3/2}$ dependence:

$$C_V = \frac{1}{V} \frac{\partial E}{\partial T} = \frac{15}{4} \frac{K}{4\pi D} T^{3/2} \zeta(5/2)$$

where $E$ is given by $\sum_{k} n_k \hbar \omega_k$.

Both specific heat and magnetization measurements have been used to measure the dispersion constant for various magnetic materials. Anderson has calculated $D$ in YIG by measuring the shape of the magnetization curve between $T = 0$ and $T_N$, fitting the shape to the molecular field theory to find the exchange integrals, and using Eq. 44 to find $D$. This allows a comparison to be made between the spin wave theory and the molecular field theory. It is found that the molecular field data yields a value of $D$ which is roughly a factor of two smaller than the specific heat measurements.

E. Magnetic Garnets

The iron garnet system, first identified by Bertaut and Forrat provides an excellent example of an insulating ferrimagnetic material. The chemical formula is $R_3Fe_5O_{12}$ with $R$ either Yttrium

or one of the rare earths. The system is cubic belonging to the space group $^{10}_h$-Ia3d and has 8 formula units per unit cell. The lattice constant of $Y_3Fe_5O_{12}$ (YIG) is 12.376Å while that of the rare earth iron garnets range from about 12.53Å in $Sm_3Fe_5O_{12}$ to about 12.28Å in $Lu_3Fe_5O_{12}$. Within the unit cell the 96 oxygen ions surround three different types of cation sites. The smallest of these is the tetrahedral or "d" site which has 4 nearest neighbor oxygen ions forming a tetrahedron. The next larger site is the octahedral or "a" site which has 6 oxygen ions around it on the corners of an octahedron. The largest site is the dodechedral or "c" site which has 8 oxygen ions surrounding it and forming an 8-cornered, 12-sided polyhedron. The shapes of these sites are shown in Figure 4.

In the unit cell the 40 Fe$^{3+}$ ions fill the a and d sites with 24 on the d sites and 16 on the a sites. The 24 rare earth ions which are larger than Fe$^{3+}$ fill the c sites. The magnetic properties of high temperatures are determined almost exclusively by the Fe$^{3+}$ ions while at low temperatures the rare earth ions may play a significant role. There exists a strong negative exchange interaction between the Fe$^{3+}$ ions on the a sites and those on the d sites. This causes the a site ions and d site ions to align on sublattices which are aligned antiparallel. Since there are 3 d sites and 2 a sites per formula unit, there results a net magnetic moment of 1 ferric ion ($S = 5/2$) or $5\mu_B$ per formula unit. There is also a negative interaction between ions within each sublattice. Harris$^{26}$ has reported the approximate sizes of these exchange interactions to be
FIGURE 4: Shape of the 3 cation sites of the garnet structure.
\[ \alpha = \frac{J_{aa}}{|J_{ad}|} \approx -0.2 \text{ and } \beta = \frac{J_{dd}}{|J_{ad}|} \approx -0.3 \text{ with } J_{ad} = -32 \text{ cm}^{-1}. \]

This corresponds to region Q in Figure 2 and a magnetization curve of the form Q in Figure 3. If the c sublattice ion is the nonmagnetic \( Y^{3+} \), \( La^{3+} \), or \( Lu^{3+} \), the only magnetic interactions will be those from the ferric ions on the a and d sublattices. However if the c sites contain a magnetic rare earth, there exists a negative exchange interaction which causes the rare earth moments to align antiparallel to the net d-a sublattice moment. At zero temperature the net magnetic moment per formula unit is then \[ \pm (3\mu_M - 5\mu_B) \] where \( \mu_M \) is the magnetic moment of the rare earth ion. It is found experimentally that the rare earth ion moment lies somewhere between the value \( g_J \mu_B \), where \( g_J \) is the Landé g factor, and the spin only value of \( 2S\mu_B \). This can probably be attributed to partial quenching of the orbital angular momentum by the crystal field. In the S-state ion \( Gd^{3+} \), the moment is given by the spin only value. The c sublattice magnetization is very weakly aligned at room temperature but the alignment increases very rapidly with decreasing temperature. At low temperatures it can dominate the magnetization. There often exists a compensation temperature at which the rare earth magnetization equals the net ferric ion magnetization and the net magnetization is zero. This is illustrated in Figure 5 where the magnetization curves of various rare earth iron garnets are shown. It is also seen that the small rare earth moment at higher temperatures has a negligible effect on the Curie temperature.

FIGURE 5: Spontaneous magnetization of the rare earth iron garnets (after Pauthenet 29).
The magnetic properties in the iron garnet system can be easily and controllably altered by changing the composition. There are basically two ways in which this is done. When more than one type of rare earth ion is put in the structure the garnet is known as a mixed garnet and its properties are a weighted average of the properties due to the different rare earths. In this way the low temperature moment and compensation temperature can be controlled with little effect on the Curie temperature. The magnetic properties can also be altered by replacing some of the Fe$^{3+}$ ions with nonmagnetic ions. The effect of this is to lower the Curie temperature as well as to change the moment at a given temperature. This type of material is known as a substituted garnet. Due to the different volumes of the a and d sites, ions which are smaller than Fe$^{3+}$ substitute preferentially on the d sites while ions which are larger than Fe$^{3+}$ substitute preferentially on the a sites. Geller et al.\textsuperscript{30} have shown that Ge$^{4+}$, Si$^{4+}$, and V$^{5+}$ substitute almost exclusively into the tetrahedral sublattice while Zr$^{4+}$ and Sc$^{3+}$ substitute almost exclusively into the octahedral sublattice. Some ions like Ga$^{3+}$ substitute primarily into the tetrahedral sublattice although a significant number enter the octahedral sublattice. Substitution into the tetrahedral or d sublattice tends to decrease the net moment per formula unit by making the fraction of magnetic ions on each sublattice more nearly equal while octahedral substitution tends to increase the moment. Both types of substitution lower the Curie

temperature since the strength of the effective exchange interaction is decreased. If the substitution level becomes high enough, the intrasublattice exchange interaction begins to play an important role and the alignment of the sublattices becomes distorted. There has been a large amount of work done in an attempt to explain the effects of substitution on the magnetic properties of the garnet system. A brief review of this work is presented below.

Gilleo\textsuperscript{31} proposed a theory which attempted to account for the observed $T_C$ and 0 K moments of substituted garnets from a statistical point of view. He assumed that in order for an Fe$^{3+}$ ion on one sublattice to participate in the magnetic ordering it must have at least two Fe$^{3+}$ neighbors on the other sublattice with which to interact. This is analogous to a magnetic chain where an atom with less than two neighbors cannot participate in the magnetic ordering of the chain. He considered a system of the type:

$$(Y_{3-x-y}Me_{x+y})(Fe_{3-x}M_x)(Fe_{2-y}M')_{12}$$

(51)

If $M$ or $M'$ is a tetravalent ion such as Si$^{4+}$ or Ge$^{4+}$, then $Me$ must be a divalent ion such as Ca$^{2+}$ to maintain charge balance. If $M$ and $M'$ are trivalent then $Me$ can be Y$^{3+}$, La$^{3+}$, or Lu$^{3+}$. It is assumed that there are no magnetic rare earth ions present. The fractional substitution is defined to be:

$$K_d = x/3 \quad K_a = y/2$$

(52)

The low temperature moment of each sublattice should then be:

\[ n_d(0) = 15\mu_B(1 - k_d)[1 - E_d(k_a)] \]
\[ n_a(0) = 10\mu_B(1 - k_a)[1 - E_a(k_d)] \] (53)

where \(1 - k_i\) is the fraction of the sites on sublattice \(i\) which are magnetic and \(E_j(k_j)\) is the fraction of these which have only 0 or 1 magnetic neighbor on sublattice \(j\). The net moment is then given by:

\[ n = n_d - n_a \] (54)

The Curie temperature is proportional to the number of magnetic interactions per magnetic ion per formula unit. There are 12 \(O^{2-}\) ions per formula unit and each of these interacts with 2 \(c\) sites, 1 \(d\) site, and 1 \(a\) site. Neglecting the \(c\) sites which are assumed to be occupied by nonmagnetic ions, there are then 24 interactions per formula unit in YIG. With nonmagnetic ion substitution this number is given by:

\[ n(k_a, k_d) = 24(1 - k_d)[1 - E_d(k_a)](1 - k_a)[1 - E_a(k_d)] \] (55)

The number of ions per formula unit which participate in the interactions is:

\[ N(k_a, k_d) = 3(1 - k_d)[1 - E_d(k_a)] + 2(1 - k_a)[1 - E_a(k_d)] \] (56)

The Curie temperature should then be given by:

\[ T_C(k_a, k_d) = \frac{n(k_a, k_d)/24}{N(k_a, k_d)/5 \cdot T_C(YIG)} \] (57)
This theory accounts reasonably well for the low temperature moments and the Curie temperatures for low levels of substitution, but it breaks down rapidly as the level of substitution is increased. This is probably due to the fact that it neglects completely the intrasublattice exchange interaction which has a large effect on ions which have few magnetic neighbors on the opposite sublattice. The intrasublattice interaction is negative and causes the moments within the sublattice to cant. The theory also does not account for the temperature dependence of the magnetization.

A different approach was taken by Dionne\textsuperscript{32} who used the molecular field theory to explain the effects of substitution. He defined the molecular field acting on sublattice 1 and molecular field coefficients $N_{ij}$ by the equation:

$$H_i = g\mu_B S \sum_j N_{ij} M_j$$  \hspace{1cm} (58)

The magnetization curves for various substituted garnets were measured and a least squares fit done to the molecular field theory. It was found that the magnetic moment per mole of the two sublattices at $0$ K could be given by:

$$M_d(0) = 3g\mu_B SN(1-k_d)(1-\cdot1k_a)$$

$$M_a(0) = 2g\mu_B SN(1-k_a)(1-k_d^{5.4})$$  \hspace{1cm} (59)

and molecular field coefficients by:

\[
N_{dd} = -30.4(1 - 0.87 k_d) \text{ mole/cm}^3,
\]

\[
N_{aa} = -65.0(1 - 1.26 k_a) \text{ mole/cm}^3,
\]

\[
N_{ad} = 97.0(1 - 0.25 k_a - 0.38 k_d) \text{ mole/cm}^3. \tag{60}
\]

These values give quite a reasonable fit to the magnetization curves and Curie temperature for \( k_d < 0.65 \) and \( k_a < 0.35 \). This work was later extended to garnets with magnetic rare earths and molecular field coefficients were calculated for each of the rare earths.\(^{33,34}\)

Beyond the range \( k_d \lesssim 0.65 \) and \( k_a \lesssim 0.35 \) the effects of canting become quite significant. This causes a reduction in the net magnetization from the aligned state since it always occurs in the sublattice opposite to the one in which substitution occurs. If, for example, a nonmagnetic ion is substituted into one of the \( d \) sublattice sites, the four nearest-neighbor \( a \) sublattice ions feel a smaller effective exchange interaction. This increases the ratio of the effective \( a-a \) exchange to the \( a-d \) exchange and the negative \( a-a \) exchange causes the ion to cant with respect to its \( a \) sublattice neighbors. This phenomenon is illustrated in Figure 6 for a 2-dimensional system.

Yaffet and Kittel\(^19\) first suggested that for certain values of the exchange constants a type of ordering could occur in a two sublattice system which was different from the simple ordering which

Néel had predicted. These conditions correspond to regions G and M in Figure 2. In region G each sublattice should split into two sublattices which should align antiparallel to each other. In region M one of the two sublattices should split into two sublattices which should align at some angle between 0 and 180°. The net moment of these two sublattices should then align antiparallel to the unsplit sublattice moment. In addition it was predicted that a system could go through transitions from one type of ordering to another as the temperature was decreased. While this theory fits the experimental results better than the Néel theory the agreement is still not that good and the multiple phase transitions have not been detected. One of the problems with the theory was that it assumed that even after substitution the lattice retained its translational symmetry, i.e. that the canting angle was the same for all spins on a given sublattice.

Nowik\textsuperscript{35} introduced a model in which the idea of local canting was considered. He assumed that if a spin had no magnetic nearest neighbors, it would be flipped by the second neighbor interaction while a spin with one magnetic nearest neighbor would be canted at some angle $\alpha$ which was treated as an adjustable parameter. In this way he was able to fit the low temperature magnetic moment data for cases with low substitution. For high levels of substitution the quality of the fit decreased quite rapidly. In fact for complete substitution on one sublattice the theory predicted a ferromagnetic ordering of the other sublattice.

FIGURE 6: The effect of substitution on the garnet system showing the canting on the a sublattice induced by nonmagnetic ion substitution on the d sublattice (after Dionne),
A better treatment of this problem was developed by Rosencwaig who used a method analogous to the Bethe-Peiris-Weiss model for ferromagnetism in which a nearest-neighbor cluster of spins is treated in terms of Heisenberg exchange interactions while the atoms outside the cluster are represented by an effective field. In this spirit he solved exactly for the canting angle $\phi$ of a spin in terms of the average canting angle $\bar{\theta}$ of its neighbors on the same sublattice and in terms of its number of magnetic nearest-neighbors on the opposite sublattice. The average canting angle $\bar{\theta}$ was just the canting angle predicted by Yafet and Kittel. Using this theory and treating the ratio of $J_{dd}/J_{ad}$ as an adjustable parameter he was able to fit the low temperature magnetic moment and the Curie temperatures over a large range of substitution for systems in which the substitution occurs on the a sublattice and canting on the d sublattice. For substitution on the d sublattice, $J_{aa}/J_{ad}$ was varied. In all cases the two ratios were determined to be within the range of values predicted by other authors. It was found that the Curie temperatures were not very sensitive to the exchange parameter ratios although the moments were quite sensitive. It was also found that for those systems in which the lattice constant increased with increasing substitution the fit could be improved by allowing the exchange parameter ratio to increase slightly with substitution. Similarly if the lattice constant decreased the fit improved by decreasing the ratio. In all cases this change was less than 10%.

In addition to the magnetization and Curie temperature, another quantity of interest in the substituted iron garnet systems is the dispersion constant $D$ in Eq. 42. It is often convenient to replace $D$ by the exchange stiffness constant or Landau-Lifshitz exchange constant $A$ where:

$$A = \frac{MD}{2g_B}$$

(61)

For a simple cubic ferromagnet, $M = g_B S/a^3$ and $D = 2JSa^2$. This gives $A = JS^2/a$. For YIG there is one net spin moment per formula unit and 8 formula units per unit cell, so $M = 8g_B S/a^3$ and from Eq. 44

$$A = \frac{4SD}{a^3} = \frac{5S}{4a} (8J_{aa} - 5J_{ad} + 3J_{dd})$$

(62)

This simplest model for predicting the variation of $A$ or $D$ with substitution assumes that $|J_{ad}| \gg |J_{dd}|, |J_{aa}|$. This is in effect the assumption that the intrasublattice exchange interaction can be completely neglected. Then from Eq. 25 comes the result:

$$T_C \propto J_{ad}$$

(63)

Combining this expression with Eq. 12 gives the expression:

$$\frac{A}{A(YIG)} = \frac{T_C}{T_C(YIG)}$$

(64)

Henry and Heinz\(^{37}\) have measured the exchange constant of Ga\(^{3+}\) substituted YIG at room temperature using ferromagnetic resonance and found that the exchange constant fell much more rapidly than $T_C$.

---

Slonczewski assumed that Eq. 64 is valid at zero temperature and developed an approximate temperature correction for it. He used the random phase approximation which says that near the Curie temperature, the exchange constant of a ferromagnet is related to its magnetization by:

$$A(T, T_C) = A(0, T_C) [M(T, T_C)/M(0, T_C)]^2$$  \hspace{1cm} (65)

In addition near the Curie temperature the Landau theory of phase transitions predicts that:

$$[M(T, T_C)/M(0, T_C)]^2 = k (T_C - T)/T_C, \quad T \leq T_C$$  \hspace{1cm} (66)

where $k$ is a dimensionless constant which depends on the material. Combining these two equations gives the result:

$$A(T, T_C) = A(0, T_C) k (T_C - T)/T_C$$  \hspace{1cm} (67)

Assuming that $k$ is independent of composition or at most depends very weakly on composition, the same expression can be written for YIG. Then dividing the two Eqs. 67 and making use of Eq. 64 yields the expression:

$$A(T) = A_{YIG}(T) [(T_C - T)/(T_C, YIG - T)]$$  \hspace{1cm} (68)

While this expression was derived for a ferromagnet and is only an approximation, Slonczewski assumed that it should be approximately true for a two sublattice system also. It does have the obvious advantage over Eq. 64 that as the Curie temperature of a material approaches room temperature, the room temperature exchange constant approaches zero. This theory also gives a much better fit to the Henry and Heinz data than the simple expression in Eq. 64.
Kirkpatrick and Harris\textsuperscript{11} have developed a model which uses percolation theory\textsuperscript{12} to predict the dependence of the exchange constant on substitution. Their model begins with the standard Heisenberg Hamiltonian:

\[ H = - \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \]  \hspace{1cm} (69)

where the usual factor of 2 and double counting restriction on the sum have been dropped. In the limit of low temperatures and large spins where the spin wave occupation is low, the spins can be treated classically and \( \vec{S}_i \cdot \vec{S}_j \) becomes \( S^2 \cos (\theta_i - \theta_j) \). The exchange constant, \( A \), is found by calculating the energy necessary to rotate the magnetization through an angle \( \theta \) over the distance \( L \) with the restrictions \( \theta << l \) and \( L >> a \). The Hamiltonian can then be treated as the classical energy of the system and the equilibrium orientation of the spin \( S_i \) can be found by minimizing the energy with respect to \( \theta_i \). This leads to the condition:

\[ \sum_j J_{ij} S^2 (\theta_i - \theta_j) = 0 \]  \hspace{1cm} (70)

The exchange constant is then determined by the relations:

\[ E = \frac{1}{2} \sum_{ij} J_{ij} S^2 (\theta_i - \theta_j)^2 = A\theta^2 V L^{-2} \]  \hspace{1cm} (71)

where \( V \) is the volume of the system.

Eqs. 70 and 71 can be treated by noticing that Eq. 70 is just Kirchhoff's law for a network of conductances \( J_{ij} S^2 \) with voltages \( \theta_i \) and \( \theta_j \) at the nodes of the network. Eq. 71 then sums the power dissipated in each conductance and the exchange constant \( A \) is proportional to the bulk conductivity. Eq. 70 can be inverted to give:
Using first and second nearest neighbor interactions which were estimated by van der Ziel et al. to be $J_{ad} = -20 \text{ cm}^{-1}$, $J_{dd} = -2 \text{ cm}^{-1}$, and $J_{aa} = 0$, a computer simulation of a resistor network was done. The values of the resistances were scaled to give the proper ratios for the conductances and Eq. 72 was solved by an iterative procedure over a system of over 5000 spins with periodic boundary conditions in 2 directions. Sites were then removed at random from the network and all the conductances connecting these sites were set equal to zero. Eq. 72 was then solved and the variation of the exchange constant with composition numerically determined. Kirkpatrick and Harris predict that for the system defined in Eq. 51 the exchange constant should be

$$A(x,y)/A(YIG) = 1 - 0.43x - 0.73y - 0.25xy$$

for levels of substitution well below the percolation threshold.

In this work the relationship in Eq. 73 has been tested on CaGe substituted YIG where it is assumed that the Ge substitution is confined to the tetrahedral sublattice and a good agreement has been found.

\[ F. \text{ Dispersion Relation for Spin Waves in a Thin Film} \]

The dispersion relation for spin waves in a thin film can be derived using a semiclassical approach similar to the one used by

Soohoo.\footnote{Soohoo, R. F., Magnetic Thin Films, New York: Harper and Row, 1965.} This approach starts with the Hamiltonian for an assembly of exchange coupled spins in the presence of an applied field and transforms into a classical continuum theory from the quantum mechanical equations of motion. This treatment is valid for spin waves with wavelengths much greater than the lattice spacing and the result is an equation of motion for the magnetization which can be solved using standard techniques.

The Hamiltonian for an assembly of exchange coupled localized spins is the presence of a magnetic field is from Eq. 26:

\[
H = -g\mu_B \sum_i H_{\text{eff}} \cdot \hat{s}_i - 2J \sum_{ij} \hat{s}_i \cdot \hat{s}_j
\]  

(74)

\(H_{\text{eff}}\) represents the effective field acting on the spin \(\hat{s}_i\) and contains contributions from the applied field, demagnetizing fields, and anisotropy fields. In the second term \(J\) represents the isotropic exchange interaction connecting spins \(\hat{s}_i\) and \(\hat{s}_j\) and the sum is restricted to nearest neighbor interactions. For small deviations from the aligned state, the exchange term in the Hamiltonian can be simplified somewhat by treating the spins as classical vectors and expanding the spin \(\hat{s}_j\) in a Taylor series about its neighbor position \(\hat{s}_1\).

\[
\hat{s}_j = \sum_i \left( \hat{s}_1 + [(\hat{r}_j - \hat{r}_1) \cdot \hat{v}] \hat{s}_1 \right) + \frac{1}{2} \left[(\hat{r}_j - \hat{r}_1) \cdot \hat{v} \right)^2 \hat{s}_1 + \ldots
\]  

(75)
Assuming that the site \( j \) is a center of inversion symmetry, the second term in the expansion will not contribute. The exchange term in the Hamiltonian is then:

\[
H_{\text{EX}} = -2zJ \sum_i S_i^2 - J \sum_{i,j} \cdot (\vec{r}_i - \vec{r}_j) \cdot \vec{v} \cdot ^2\vec{S}_i \tag{76}
\]

where \( z \) is the number of nearest neighbors of the site \( i \). The first term just shifts the energy of the ground state by a constant amount and can be dropped. The second term can be further simplified according to the lattice type. For a simple cubic lattice of lattice constant \( a \), the Hamiltonian becomes:

\[
H = -g\mu_B \sum_i \vec{H}_{\text{eff}} \cdot \vec{S}_i - 2Ja^2 \sum_i \vec{S}_i \cdot \nabla^2 \vec{S}_i \tag{77}
\]

A similar expression results for other lattice types.

This Hamiltonian can be used to obtain a quantum mechanical equation of motion for an individual spin, \( \vec{S}_m \), in the usual manner:\[22\]

\[
i\hbar \frac{d}{dt} \vec{S}_m = [\vec{S}_m, H] \tag{78}
\]

Using the relationship \( \vec{S} \times \vec{S} = i\hbar \), the commutator can be evaluated yielding the expression:

\[
\hbar \frac{d}{dt} \vec{S}_m = -g\mu_B \vec{H}_{\text{eff}} \times \vec{S}_m - 2Ja^2 (\nabla^2 \vec{S}_m) \times \vec{S}_m \tag{79}
\]

Since \( \vec{S}_m \) represents any of the spins in the bulk of the sample, this expression can be rewritten in terms of the magnetization, by the replacement, \( \vec{S} = \vec{M} \frac{\hbar}{N} \). The equation of motion of \( \vec{M} \) then becomes:

\[
\frac{1}{\gamma} \frac{d}{dt} \vec{M} = \vec{M} \times \vec{H}_{\text{eff}} + \frac{2A}{\hbar^2} \vec{M} \times \nabla^2 \vec{M} \tag{80}
\]
where γ is the gyromagnetic ratio $\gamma = g\mu_B/\hbar$ and $A = JS^2/a$ is the exchange constant. The two terms on the right of Eq. 80 represent torques acting on the magnetization. The first term represents the torque on a unit volume of material with magnetization $\mathbf{\vec{M}}$ placed in a magnetic field $\mathbf{H}^\text{eff}$ while the second term can be thought of as the torque due to an exchange field $\mathbf{H}^\text{ex} = \frac{2A}{M^2} \mathbf{\vec{v}}^2 \mathbf{\vec{M}}$.

The torque due to $\mathbf{H}^\text{eff}$ can be most easily treated by expressing it in terms of the free energy density of the system:

$$\mathbf{\vec{r}} = \mathbf{\vec{r}} \times (-\nabla E) \quad (81)$$

The effects of the applied field, demagnetization field, and anisotropy fields can then be handled as scalar contributions to the energy density rather than as vector fields. The equation of motion can then be solved in a relatively straightforward method similar to the one used by Soohoo. \textsuperscript{39} A coordinate system is chosen such that the equilibrium direction of the magnetization is the $\mathbf{\hat{r}}$ direction and small deviations from equilibrium are in the $\mathbf{\hat{\theta}}$ and $\mathbf{\hat{\phi}}$ directions.

The magnetization can then be written:

$$\mathbf{\vec{M}} = M \mathbf{\hat{r}} + m_\theta \mathbf{\hat{\theta}} + m_\phi \mathbf{\hat{\phi}}$$

where

$$m_\theta = M \delta \theta \quad m_\phi = M \sin \theta \delta \phi \quad (82)$$

and $\mathbf{\hat{r}}$, $\mathbf{\hat{\theta}}$, and $\mathbf{\hat{\phi}}$ are the standard unit vectors of a spherical coordinate system and $\delta \theta$ and $\delta \phi$ are small deviations from equilibrium. Figure 7 illustrates the coordinate system and film geometry used in this derivation where the applied field is chosen to be in the x-y plane. Using Eq. 81, Eq. 80 can be rewritten in the form:
\[
\frac{1}{\gamma} \frac{d}{dt} \mathbf{M} = -\frac{\mathbf{M}}{M^2} \times \mathbf{\nabla} \mathbf{E} + \frac{2A}{M^2} \mathbf{M} \times \mathbf{\nabla}^2 \mathbf{M} \tag{83}
\]

Many authors introduce a phenomenological damping term on the right side of Eq. 83. This term usually has one of three forms:

**Landau-Lifshitz form:**

\[-\frac{1}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}})\]

**Gilbert form:**

\[-\frac{a}{M} \mathbf{M} \times \mathbf{M} \tag{84}\]

**Bloch-Bloembergen form:**

\[-\frac{M_x \mathbf{Y}}{\tau_2} - \frac{M_z - M}{\tau_2}\]

These damping terms are related to the relaxation processes and will be neglected in the rest of this work.

The equilibrium orientation of the magnetization can be obtained by solving the static case with Eq. 83 set equal to zero. This results in the condition:

\[-\phi \mathbf{E}_{\theta} + \phi \frac{1}{\sin \theta} \mathbf{E}_{\phi} = 0 \tag{85}\]

where the notation \(\mathbf{E}_{\theta} = \frac{\partial}{\partial \theta} \mathbf{E}\) and \(\mathbf{E}_{\phi} = \frac{\partial}{\partial \phi} \mathbf{E}\) has been used. Eq. 85 is just a statement of the fact that zero net torque acts on the magnetization in equilibrium. The equation of motion is then linearized by expanding Eq. 83 about equilibrium and retaining only terms first order in the deviation from equilibrium.

\[\frac{1}{\gamma} \delta \left( \frac{d}{dt} \mathbf{M} \right) = \delta (-\frac{\mathbf{M}}{M} \times \nabla \mathbf{E}) + \frac{2A}{M^2} \delta (\mathbf{M} \times \nabla^2 \mathbf{M}) \tag{86}\]
FIGURE 7: Coordinate system used in deriving the spin wave dispersion relation for a thin film.
Evaluating Eq. 86 term by term

\[
\delta\left(-\frac{\dot{\hat{M}}}{M} \times \vec{v}E\right) = \frac{-1}{M} \delta\hat{M} \times \vec{v}E - \frac{\dot{\hat{M}}}{M} \times \delta(\vec{v}E) \tag{87}
\]

The first term on the right hand side is zero from Eq. 85 and

\[
-\frac{\dot{\hat{M}}}{M} \times \delta(\vec{v}E) = \frac{1}{M \sin \theta} \left( E_{\phi} m_\theta + \frac{E_{\phi\phi} m_\phi}{\sin \theta} \right) \delta - \frac{\dot{\hat{M}}}{M} \left( \dot{\hat{E}} \dot{\hat{m}}_\theta + \frac{E_{\theta\phi}}{\sin \theta} \right) \tag{88}
\]

The fact that \( E_{\phi}/\sin \theta = 0 \) from Eq. 85 and the identification \( M_\theta = M \delta \theta \) and \( M_\phi = M \sin \theta \delta \phi \) from Eq. 82 have been used to derive Eq. 88. Using Eq. 82, the second term in Eq. 86 becomes:

\[
\frac{2A}{M} \delta(\dot{\hat{M}} \times \vec{v}^2 \hat{M}) = \frac{2A}{M} (\vec{v}^2 m_\phi - \vec{v}^2 m_\theta) \tag{89}
\]

The equation of motion linearized form then reduces to:

\[
\frac{1}{\gamma} \frac{d}{dt} m_\theta = \frac{1}{M \sin \theta} \left( E_{\phi} m_\theta + \frac{E_{\phi\phi} m_\phi}{\sin \theta} \right) - \frac{2A}{M} \vec{v}^2 m_\phi \tag{90}
\]

\[
\frac{1}{\gamma} \frac{d}{dt} m_\phi = -\frac{1}{M} \left( E_{\theta\phi} m_\theta + \frac{E_{\theta\phi} m_\phi}{\sin \theta} \right) + \frac{2A}{M} \vec{v}^2 m_\theta
\]

Rather than solving these equations for \( m_\theta \) and \( m_\phi \) directly, plane wave solutions of the form

\[
m_\theta = \alpha e^{i(ky - \omega t)} \quad m_\phi = \beta e^{i(ky - \omega t)} \tag{91}
\]

are assumed and used to find the dispersion relation. The characteristic equation is obtained from the expression

---

40. A complete solution also contains terms of the form \( e^{\pm k'y} \). However, since these terms have a very short decay length and have no measurable effect on the spectrum, they have been neglected.
\[
\begin{pmatrix}
\frac{E_{\theta\phi}}{M \sin \theta} + \frac{i \omega}{\gamma} & \frac{E_{\phi\phi}}{M \sin^2 \theta} + Dk^2 \\
\frac{E_{\theta\phi}}{M} + Dk^2 & \frac{E_{\phi\phi}}{M \sin \theta} - \frac{i \omega}{\gamma}
\end{pmatrix}
\begin{pmatrix}
m_{\theta} \\
m_{\phi}
\end{pmatrix} = 0 \tag{92}
\]

where \(D = 2A/M\) is the spin wave dispersion constant. Evaluating the characteristic equation by setting the determinant of the coefficients of Eq. 92 equal to zero gives the dispersion relation:

\[
(\omega)^2 = \left(\frac{E_{\theta\phi}}{M} + Dk^2\right)\left(\frac{E_{\phi\phi}}{M \sin^2 \theta} + Dk^2\right) - \left(\frac{E_{\theta\phi}}{M \sin \theta}\right)^2 \tag{93}
\]

Further evaluation of this dispersion relation requires an explicit form for the energy density in the system.

The energy density of a magnetic material consists of Zeeman, demagnetization, uniaxial anisotropy, and crystalline anisotropy terms. The Zeeman energy density is given by:

\[
E_Z = -\hat{M} \cdot \mathbf{H} = -MH \sin \theta \cos(\phi - \phi_H) \tag{94}
\]

where \(\mathbf{H}\) is the applied field and the coordinate system is illustrated in Figure 7. The demagnetizing energy is given by:

\[
E_D = \frac{1}{2} \hat{M} \cdot \mathbf{N} \cdot \hat{M} \tag{95}
\]

where \(\mathbf{N}\) is the demagnetization tensor. For the thin film shown in Figure 7 which is the limiting case of an ellipsoid in which the ratio of the y axis to the x and z axes approaches zero, the demagnetization energy can be expressed as:
The uniaxial anisotropy energy density can be written as:

$$E_U = -K_U \sin^2 \theta \sin^2 \phi$$

(97)

Since the demagnetizing energy and the uniaxial anisotropy energy have the same angular form, the uniaxial term will be dropped and wherever $2\pi M^2$ is written it will be understood to mean $2\pi M^2 - K_U$.

The final energy term, the crystalline anisotropy energy, can be written for a cubic structure as:

$$E_C = \frac{K_1}{M^4} (M_i^2 M_j^2 + M_i^2 M_k^2 + M_j^2 M_k^2)$$

(98)

where $K_1$ is the first order anisotropy constant and $i$, $j$, and $k$ refer to the principal crystal axes $[100]$, $[010]$, and $[001]$ respectively. Expressing this energy in terms of the coordinate system in Figure 7 the expression becomes:

$$E_C = \frac{K_1}{M^4} \left( \frac{1}{4} M_x^4 + \frac{1}{3} M_y^4 + \frac{1}{4} M_z^4 + \frac{1}{2} M_x^2 M_z^2 + \frac{\sqrt{2}}{3} M_x^3 M_y - \sqrt{2} M_x M_y M_z \right)$$

$$= K_1 \left( \frac{\sin^4 \theta \cos^4 \phi}{4} + \frac{\sin^4 \theta \sin^4 \phi}{3} + \frac{\cos^4 \theta}{4} + \frac{\sin^2 \theta \cos^2 \phi}{8} \right)$$

(99)

$$+ \frac{\sqrt{2}}{3} \sin^4 \theta \sin \phi \cos^3 \phi - \frac{\sqrt{2}}{8} \sin^2 \phi \sin 2 \phi$$

The total energy density is then the sum of the four terms contained in Eqs. 94, 96, 97, and 99.
The equilibrium orientation of \( \vec{M} \) is found by setting both \( E_\theta \) and \( \frac{1}{\sin \theta} E_\phi \) equal to zero. It is easily shown that the equation \( E_\theta = 0 \) implies that the equilibrium value of \( \theta \) is \( \pi/2 \). Setting \( E_\phi = 0 \) with \( \theta = \pi/2 \) yields the condition:

\[
M H \sin (\phi - \phi_H) + 2\pi M^2 \sin 2\phi + K_1 \left[ \frac{1}{12} \sin 2\phi (1 - 7 \cos 2\phi) \right] \\
+ \frac{\sqrt{2}}{3} \cos^2 \phi (2 \cos 2\phi - 1) = 0
\]  

(100)

There is in general no analytic solution to this equation and it will be carried along with the dispersion relation in this form. Evaluating the second derivatives of the energy allows the dispersion relation to be solved explicitly. It is easily shown that \( E_{\theta \phi} = 0 \). In addition the transformations \( \alpha - \frac{\pi}{2} - \phi \) and \( \beta - \frac{\pi}{2} - \phi_H \) are made. \( \alpha \) and \( \beta \) then become the angles between \( \vec{M} \) and \( \vec{H} \) and the film normal, a [111] crystallographic direction. \( \vec{H} \) and \( \vec{H} \) are chosen to lie in a (110) plane. Evaluating the derivatives yields the dispersion relation from Eq. 93:

\[
\left( \frac{\omega}{\gamma} \right)^2 = [H \cos (\beta - \alpha) - 4\pi M \cos^2 \alpha + g_1(\alpha) + Dk_1^2] \chi \\
+ [H \cos (\beta - \alpha) - 4\pi M \cos 2\alpha + g_2(\alpha) + Dk_2^2]
\]  

(101)

In addition the equilibrium orientation of \( \vec{M} \) is given by:

\[
H \sin (\beta - \alpha) + 2\pi M \sin 2\alpha + g_3(\alpha) = 0
\]  

(102)
where

\[ g_1(\alpha) = H_K \left[ \frac{1}{8} \sin^2 2\alpha - \frac{2}{3} \cos^4 \alpha - \frac{\sqrt{2}}{2} \sin 2\alpha (1 + \frac{2}{3} \sin^2 \alpha) \right] \]

\[ g_2(\alpha) = -H_K \left[ \frac{1}{12} \cos 2\alpha + \frac{7}{12} \cos 4\alpha + \frac{\sqrt{2}}{6} (\sin 2\alpha - 2 \sin 4\alpha) \right] \]

\[ g_3(\alpha) = H_K \left[ \frac{1}{24} \sin 2\alpha (1 + 7 \cos 2\alpha) - \frac{\sqrt{2}}{6} \sin^2 \alpha (1 + 2 \cos 2\alpha) \right] \]

(103)

and \( H_K = 2K_1 / M \) is the effective cubic anisotropy field.

Setting \( k = 0 \) in Eq. 101 allows the position of the uniform mode resonance to be found.

\[ H_U \cos(\beta - \alpha) = 2\pi M (\cos^2 \alpha + \cos 2\alpha) - \frac{1}{2} (g_1(\alpha) + g_2(\alpha)) \]

\[ + \frac{1}{2} \left[ [4\pi M (\cos^2 \alpha - \cos 2\alpha) + g_2(\alpha) - g_1(\alpha)]^2 \right. \]

\[ + 4 \left( \frac{\omega}{\gamma} \right)^2 \left]^{1/2} \right. \]

(104)

The position of the \( n^{th} \) spin wave mode with \( k = k_n \) is then found from Eq. 101 by:

\[ H_n = \frac{H_U - Dk^2}{\cos(\beta - \alpha)} \]

(105)

The equilibrium condition in Eq. 102 shows that if the field is applied along the film normal, \( \alpha = \beta = 0 \) and the expression simplifies to

\[ H_n = \frac{\omega}{\gamma} + 4\pi M + \frac{2}{3} H_K - Dk_n^2 \]

\[ = H_U - Dk_n^2 \]

(106)

In the above derivation of the spin wave dispersion relation, it was assumed that the film was ferromagnetic. For a ferrimagnet,
the effect of the two antiparallel sublattices must be considered.
Wangsness\textsuperscript{41} has done this by solving the equation of motion of a
system of two exchange coupled sublattices in an applied field and
found that the dispersion relation is essentially the same. However,
if the two sublattices have different gyromagnetic ratios, $\gamma$ must
be replaced by a $\gamma^\text{eff}$ where:

$$\gamma^\text{eff} = \frac{M_1 - M_2}{M_1 \gamma_1 - M_2 \gamma_2}$$  \hspace{1cm} (107)

and $M_{1,2}$ and $\gamma_{1,2}$ are the magnetization and gyromagnetic ratios of
the individual sublattices. If the material has a compensation
point where $M_1 = M_2$ and if $\gamma_1 \neq \gamma_2$, there will be a point at which
$\gamma^\text{eff}$ diverges. This effect has been seen in a number of ferrites
and substituted garnets.\textsuperscript{42} The gyromagnetic ratio is determined by
measuring the field position of the first spin wave mode at two or
more frequencies with the applied field along the film normal. Then
from Eq. 106

$$\gamma^\text{eff} = \frac{\mu_B}{h} g^\text{eff} = \frac{\omega_2 - \omega_1}{H_2 - H_1}$$  \hspace{1cm} (108)

The other film parameters can also, in principle, be determined
from the spin wave spectrum. The cubic anisotropy field, $H_K$, and
the effective demagnetizing field, $4\pi M - 2K_U/M$, can be measured by
fitting the angular variation of the resonance fields to the

\textsuperscript{42} von Auloch, W. H., ed. Handbook of Microwave Ferrite Materials.
dispersion relation, Eq. 101. In practice this measurement is made quite difficult and inaccurate because of the influence of the $Dk^2$ term in the dispersion relation. The allowed values of the wave vectors, $k$, of the spin waves are determined by boundary conditions at the film surfaces. These boundary conditions are often anisotropic and suitable care must be taken to account for this anisotropy since it can have an effect on the resonance field of a given spin wave mode which is just as large as the effect due to the bulk anisotropy fields, $H_K$ and $4\pi M - 2K_u/M$.

Finally the dispersion constant can be determined from the spectrum if the boundary conditions which determine the allowed values of $k$ are known. These boundary conditions are discussed in the next section.

G. Spin Wave Boundary Conditions

The allowed values of $k$ for spin waves in a thin film excited by a uniform rf field parallel to the film surface, depend on the boundary conditions existing at the film surfaces. Over the past twenty years there has been considerable discussion in the literature about these boundary conditions and their physical origins. Kittel first predicted that exchange dominated spin waves could be excited in a thin film by a uniform rf field. The equation of motion of the spin system was solved with the applied field along the film normal. A surface field, parallel to the applied field, was assumed

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to act only on the surface spins and an equation of motion for the surface spins was written. In addition to the inclusion of the surface field, the effect of the reduced symmetry of the surface spins was accounted for by retaining the gradient term in the Taylor's expansion of the spin system Eq. 2. Using plane wave solutions for the transverse magnetization of the form:

\[ m = a \sin \beta y + \beta \cos \beta y \quad (109) \]

and demanding that these solutions satisfy the equations of motion of the surface spins, it was concluded that the surface spins were completely pinned \((m = 0\) at both surfaces). The allowed values of \(k\) under this condition are:

\[ k = \frac{n\pi}{L}, \quad n = 1, 3, 5, \ldots \quad (110) \]

where \(L\) is the film thickness. Solutions with even \(n\) are allowed, but having an even number of half wavelengths across the film thickness they have zero net dipole moment and cannot be excited by a uniform rf field. The solutions with odd \(n\) have an intensity which is proportional \(1/n^2\).

Pincus extended the work of Kittel and showed that a mixed boundary condition of the form:

\[ \frac{\nabla \cdot \mathbf{m}}{m} = C \quad (111) \]

at the film surfaces was more appropriate. $\nabla_n$ refers to a gradient along the outward film normal at each surface. The constant $C$ is a ratio of the energy due to the surface field to the exchange energy. Satisfying this condition with plane wave solutions of the form (109) leads to symmetric and antisymmetric solutions. The antisymmetric solutions have no dipole moment and cannot be excited while the symmetric solutions satisfy the condition:

$$\frac{C}{k} = \tan \left(\frac{kL}{2}\right)$$

(112)

which gives solutions of the form:

$$k = (n - \Delta(n))\frac{\pi}{L} \quad n = 1, 3, 5, -\ldots$$

(113)

In the limit of zero surface field $\Delta(n) = 1$ for all $n$ and only the $k = 0$ mode has a nonzero dipole moment. In the limit where the surface field is much greater than the exchange field, $\Delta(n) = 0$ for all $n$ and the Kittel perfect pinning condition is reproduced.

In order to explain the angle dependence of the spin wave spectrum, Soohoo introduced the concept of an uniaxial surface anisotropy. Also the case in which the transverse magnetization is not circularly polarized was considered. This led to boundary conditions of the form:

$$\nabla_n \frac{m_\phi}{m_\phi} = -\frac{K_S}{Ak^2} \cos 2\alpha$$

$$\nabla_n \frac{m_\theta}{m_\theta} = -\frac{K_S}{Ak^2} \cos 2\alpha$$

(114)

where \( m_\theta \) and \( m_\phi \) are the two components of the transverse magnetization, \( \alpha \) is the angle between the saturation magnetization and the film normal, and \( K_S \) is an uniaxial anisotropy with its easy axis along the film normal. In addition to treating the angle dependence of the spectrum this model also explained the occurrence of a critical angle at which the spins become unpinned at the surface and only the \( k = 0 \) mode can be excited.

At about this same time, a volume inhomogeneity in the internal fields was proposed\(^{46-48}\) to account for the non-quadratic behavior of the lower order spin wave modes and the position of the critical angle.

The surface inhomogeneity model\(^{49}\) introduced the concept of an unidirectional surface anisotropy and predicted a nonpropagating surface mode with imaginary \( k \) localized at the film surface. For long wavelength spin waves this model predicts a surface boundary condition of the form:

\[
\frac{V_n m}{m} = \frac{K}{A} \cos (\alpha_0 - \alpha) \tag{115}
\]

where $\alpha$ is the angle between the film normal and the anisotropy direction. The nonpropagating surface modes predicted by this model have been observed in YIG films and an analysis of these modes indicates that the anisotropy must be generalized to a tensorial form $K_\perp \cos^2 \alpha + K_\parallel \sin^2 \alpha$. $K_\perp$ is the magnitude of the anisotropy when \( M \) is perpendicular to the film surface and $K_\parallel$ is the magnitude when $M$ lies in the film plane. The surface inhomogeneity model also considered nonsymmetric boundary conditions by the introduction of a different anisotropy constant for each film surface. The boundary conditions can then be written in the form:

$$\frac{V_m}{m_1,2} = \frac{K_{1,2}(\alpha)}{A}$$  \hspace{1cm} (116)

where the subscripts refer to the two film surfaces. In analyzing the spin wave spectrum it is convenient to define surface pinning parameters $a_1$ and $a_2$ where:

$$a_{1,2} = 1 - \frac{K_{1,2}(\alpha)}{A}$$  \hspace{1cm} (117)

A number of general features of the spectrum can be determined from the size of the pinning parameters in a film with symmetric boundary conditions. If $a$ is less than unity, the surface anisotropy assists the applied field and the surface spins are pinned. For this case a family of sinusoidal body spin wave modes similar to those in Eq. 38 can be excited. If $a$ is unity the surface anisotropy

has no effect on the spectrum and only the $k = 0$ mode can be excited. This corresponds to the condition $\varphi_{n,m} = 0$. If $a$ is greater than unity the anisotropy opposes the applied field and in addition to the sinusoidal body modes a symmetric and an antisymmetric surface mode can be excited. These surface modes have imaginary wave vectors and their amplitudes decay exponentially into the film.

For arbitrary boundary conditions Puszkarski\textsuperscript{51} has shown that the allowed values of $k$ for body modes are given by:

$$\tan(kL) = \frac{r \sin k}{\cos k - p}$$ (118)

where $r = (a_1a_2 - 1)/(a_1a_2 + 1)$ and $p = (a_1 + a_2)/(a_1a_2 + 1)$ and $L$ is the thickness of the film measured in lattice constants. For surface modes with $k = it$, the allowed values of $t$ are given by:

$$\tanh(Lt) = \frac{r \sinh t}{\cosh t - p}$$ (119)

A computer subroutine solution of these equations is given in Appendix I.

CHAPTER III

Experimental Procedure

The experimental work done in this study involved the use of a microwave spectrometer and a vibrating sample magnetometer. Both of these systems are described below. A block diagram of the microwave spectrometer is shown in Figure 8 and a diagram of the magnetometer system is in Figure 10.

A. Microwave Spectrometer

The microwave spectrometer used in the spin wave resonance experiments is a standard reflection cavity spectrometer of the balanced bridge type. The microwaves, generated by a Varian reflex klystron, pass through a ferrite isolator and an attenuator into one port of a circulator. The isolator prevents any reflected power from altering the voltage standing wave ratio (VWSR) at the output port of the klystron and the attenuator is used to control the power level. The circulator is a three port device which transmits power according to the scheme 1 to 2, 2 to 3, and 3 to 1. The microwave power entering port 1 of the circulator then exits at port 2 and passes through a slide-screw tuner, essentially an adjustable complex impedance, into the reflection cavity which contains the sample. The cavity is a TE_{101} or TE_{102} cavity with the sample placed on the narrow wall where the microwave field is most intense, Figure 9.
FIGURE 8: Block diagram of the microwave resonance spectrometer.
FIGURE 9: Microwave magnetic fields in the resonant cavity.
This insures maximum sensitivity and that the microwave field is parallel to the plane of the film. The cavity is normally critically coupled (impedance matched) to the waveguide so that no power is reflected back. Upon sweeping the magnetic field through resonance, the sample absorbs power loading the cavity and changing its Q. This causes an impedance mismatch between the cavity and waveguide and power is reflected back from the cavity, through the circulator, into the third arm of the spectrometer which contains a frequency meter, diode detector, and an adjustable short. The purpose of the adjustable short is to alter the VWSR in the detector arm of the bridge and in so doing to place the detector in the region of maximum microwave field. In an actual experiment the slide-screw tuner is used to slightly decouple the cavity from the waveguide. This causes a small amount of power to be reflected back to the detector diode, biasing it into its most sensitive range.

The signal to noise ratio of the detector signal is improved by modulating the magnetic field with a small audio frequency field and using standard lock-in amplifier techniques. The source of the modulation signal is the reference oscillator from a Princeton Applied Research (PAR) model 124 lock-in amplifier. This signal is amplified by an audio amplifier and fed into a pair of Helmholtz coils mounted on the magnet pole faces. The choice of modulation frequency is a compromise between the noise spectrum of the diode detector, which is proportional to the inverse of the frequency, which would suggest using a high frequency, and the skin depth of the dewar and cavity
walls which decreases with increasing frequency. In this work a modulation frequency of approximately 450 Hz was used.

The modulated absorption signal from the detector is amplified and demodulated by the PAR lock-in amplifier whose output is then fed into the y-axis of an x-y recorder. The PAR demodulator is a full wave rectifier whose phase is adjustable with respect to the reference signal. This method of detection rejects all noise outside of a narrow bandwidth centered about the modulation frequency and also suppresses all noise which is 90° out of phase with the signal at the modulation frequency. Hence the name "phase sensitive detector."

Because of the modulation techniques used, the output of the PAR is the derivative of the absorbed power, dP/dH.  

Resonance experiments were done at two frequencies, X band (9.5 GHz) and K band (23.5 GHz). The microwave power at these two frequencies was generated by Varian V-58 and EM-1188 reflex klystrons respectively. The klystrons were powered by a Hewlett Packard 716B Klystron Power Supply and the frequency was stabilized by an automatic frequency control designed in this laboratory. The AFC supplies a small 10 KHz frequency modulation to the reflector of the klystron. This induces a small 10 KHz signal at the detector which the AFC detects and amplifies using the same phase sensitive detection techniques as the lock-in amplifier. The rectified output of

53. This modulation must be less than the equivalent frequency linewidth γΔH of the resonance signal.
the AFC is then applied to the klystron reflector with the correct polarity to always reduce the frequency difference between the klystron and cavity resonance.

Two different microwave spectrometers were used in this work. One was mounted on a 15 inch Varian V-3400 electromagnet controlled by a Mark II Fieldial which used a temperature stabilized Hall probe to control and measure field. The other spectrometer was mounted on a 12 inch Harvey Wells magnet controlled by a Walker Magnion FFC-4D field controller which used a temperature stabilized rotating coil gaussmeter as its sensing element. Both magnets were calibrated with a Ventron nuclear magnetic resonance gaussmeter. This allowed fields to be measured to within an accuracy of about 1 part in 5000.

The microwave frequency was measured by a Hewlett-Packard X532B wavemeter at X band and a K532A wavemeter at K band. Both wavemeters were calibrated to within 1 part in 1000 by the use of DPPH as a resonance standard \( (g = 2.0036) \)\(^{54} \) and the NMR gaussmeter.

The sample temperature in the spectrometer system could be controlled between 300 K and 4 K by the use of a variable temperature dewar. The dewar is a three-chambered device. The outermost chamber contains liquid nitrogen which acts as a precooling agent and radiation shield for the inner chambers. The middle chamber is the liquid helium reservoir and is connected by a throttle valve and capillary tube to the inner chamber which contains the cavity and sample. The sample cooling is regulated by the throttle valve which controls the

---

amount of helium flowing from the reservoir into the bottom of the sample chamber where it vaporizes. The temperature is controlled by a PAR model 152 Cryogenic Temperature Controller which uses a GaAs diode mounted on the cavity to sense temperature. The temperature controller compares the voltage drop across the GaAs diode with an internally generated reference voltage. Any deviation between these two voltages indicates a deviation in temperature from the set temperature and causes the temperature controller to increase or decrease the current passing through a heater which is mounted on the cavity.

The sample temperature could also be raised above room temperature to about 250°C. This technique involves the use of a system designed in this laboratory. The sample is mounted next to a copper-constantan thermocouple inside a quartz tube which passes through a cylindrical microwave cavity. An exchange gas, heated by passing it over a nichrome heater, flows through the tube and heats the sample. The temperature is controlled by varying either the current in the heater or the flow rate of the exchange gas. This system was not stabilized by the use of a feedback loop temperature controller although its stability could probably have been improved by doing so.

B. Magnetometer

The magnetization measurements reported in this work were made on a PAR model 155 Vibrating Sample Magnetometer. A block diagram of this system is shown in Figure 10. The sample, whose moment is to be measured, is mounted on a rod and suspended between the pole
pieces of the magnet and between a pair of pickup coils mounted on the magnet pole pieces. The other end of the sample rod is connected to a vibrating head mounted above the magnet. When the sample and rod are vibrated, any moment in the sample induces a signal in the pickup coils which is proportional to the sample moment, vibration amplitude, and vibration frequency. In order to reduce any errors due to variations in the vibration frequency and amplitude, a feedback system with a capacitor mounted in the vibrating head is used. One set of the capacitor plates is mounted in the vibrating head and vibrates with it while the other set of plates is mounted in a fixed position. A dc voltage proportional to the magnetic moment of the sample is applied to the vibrating plates of the capacitor. A signal at the vibration frequency and proportional to the moment of the sample, vibration amplitude, and vibration frequency is then induced on the fixed plates of the capacitor. The signals from the pickup coils and fixed capacitor plates are input to the two inputs of a differential amplifier. Since both of these signals depend on the vibration frequency and amplitude in the same way and the differential amplifier amplifies only the difference between these signals, its output is independent of these variations. This output signal is detected and amplified by a lock-in amplifier whose reference is the same oscillator which drives the vibrating head. The dc output of the lock-in amplifier is fed to the output display circuitry and back to the vibrating capacitor plates. The effect of this feedback is to automatically adjust the dc signal to
FIGURE 10: Block diagram of the vibrating sample magnetometer.
the level required to maintain the capacitor output signal at the same level as the pickup coil signal.

The magnetometer was calibrated with a high purity nickel sample (saturation moment = 55.01 emu/gm at 20°C) to within an accuracy of about 1%. The useable sensitivity is approximately $10^{-4}$ emu. For measurements below room temperature the sample is mounted inside a variable temperature dewar similar to the one used with the microwave resonance spectrometer.

C. Sample Preparation

The films used in this work were grown by the technique known as Liquid Phase Epitaxy (LPE) at Bell Laboratories. In the LPE process, the oxides of the garnet film constituents are dissolved in a flux of PbO and $B_2O_3$ at approximately 1000°C. The solution is cooled to about 10°C below its saturation temperature and a single crystal substrate is lowered into it while simultaneously being rotated about a vertical axis. Epitaxial growth of the garnet onto the substrate occurs at a rate of about $\mu$m/min. In order for growth to occur it is necessary that the substrate have the same crystal structure as the film and that the film-substrate lattice constant mismatch be no greater than a few hundredths of an Å. The substrates used in the sample growth were 2 inch diameter single crystal plates of $[111]$ oriented $Gd_3Ga_5O_{12}$.

After growth, the thickness of the films was measured by an optical interference technique. If light is reflected off of a

thin film at an angle $\theta$, the condition for destructive interference of the light reflected from the front and back sides of the film is:

$$2nL \cos \theta' = m\lambda \quad (120)$$

where $L$ is the film thickness, $n$ the index of refraction, $\theta'$ the angle of refraction in the film, $\lambda$ the free space wavelength of the light, and $m$ the integer order of the interference. The thickness of the film is computed by measuring the intensity of the reflected light as a function of wavelength. This can be done by either illuminating the film with monochromatic light from a monochromator and detecting the intensity of reflected light as the wavelength is scanned or by illuminating the sample with white light and detecting the reflected light with a monochromator. The second method is less sensitive to noise from ambient light. The spectrum consists of a series of interference minima and maxima. If a quantity

$$T(\lambda) = \frac{2n\cos \theta'}{\lambda} \quad (121)$$

is defined, the film thickness is given by:

$$L = \frac{N}{T(\lambda_2) - T(\lambda_1)} \quad (122)$$

where $\lambda_1$ and $\lambda_2$ are the wavelengths of two of the interference fringes and $N = m_2 - m_1$ is the difference in the order numbers of the two fringes. For film thicknesses on the order of 10$\mu$m this method is quite accurate. The absolute accuracy is limited by how well $n(\lambda)$ is known. As the film thickness is decreased to about 1$\mu$m the
accuracy decreases since there are fewer fringes in a given wavelength range. For the substituted garnets the problem is further complicated by the fact that the index of refraction varies with composition. The index of refraction for CaGe substituted YIG was estimated by linearly interpolating between the measured value of \( n(\lambda) \) for YIG and \((YLuCaSm)_3(Fe_{4.1}Ge_{0.9})_{12}\). The overall accuracy is estimated to be about 5%.

The samples were prepared for measurement by cutting them into small rectangles and applying a mask of Silicone RTV rubber adhesive mask to the surface. After the mask was cured, the samples were etched in concentrated \( H_3PO_4 \) at 150°C to remove the unwanted film from around the mask and from the back side of the substrate. The mask was then removed by dissolving it in dichloromethane and the samples were subjected to a short etch in \( H_3PO_4 \) at 100°C to clean the surface. The resonance samples were cut to approximately 3 x 3 mm and the mask was 1 mm in diameter. The magnetization samples were 10 x 5 mm and the mask was placed over the entire surface.

CHAPTER IV
Results and Discussion

Microwave resonance and magnetization measurements have been made on CaGe substituted YIG with Ge concentrations ranging from 0.1 to 1.4 moles per formula unit. Calcium and germanium were chosen for substitution for a number of reasons. It was desirable to test the dependence of the exchange constant on substitution on a particular sublattice. Geller et al. have shown that a number of ions substitute either predominantly or exclusively on only one of the magnetic sublattices. For example, Zr$^{4+}$, Sc$^{3+}$, and In$^{3+}$ all have ionic radii which are larger than Fe$^{3+}$ and substitute into the larger octahedral sites while Si$^{4+}$ and Ge$^{4+}$, which have ionic radii smaller than that of Fe$^{3+}$, substitute into the smaller tetrahedral sites. When tetravalent ions are substituted, the divalent Ca$^{2+}$ is added for charge compensation. The Ca$^{2+}$ ion has a radius which is almost twice the size of the Fe$^{3+}$ ion and it enters the dodecahedral site which is the largest one available.

The ions which substitute on the octahedral sublattice cause the lattice constant to change more rapidly than those which substitute on the tetrahedral sublattice. This limits the range of compositions

for which single crystal films can be grown. In addition, the effects of canting begin to show up at lower levels of substitution on the octahedral sublattice than on the tetrahedral sublattice. This complicates the analysis somewhat as will be seen later. For these reasons it was decided to substitute on the tetrahedral sublattice.

Of the two major candidates for substitution, Si\(^{4+}\) substitutes exclusively on the tetrahedral sites while Ge\(^{4+}\) has a small fractional substitution of about 2% on the octahedral site. However the variation of the lattice constant from that of YIG to the end member of the substitution Ca\(_3\)Fe\(_2\)M\(_3\)O\(_{12}\) is much greater when M = Si\(^{4+}\) than when M = Ge\(^{4+}\). In fact the lattice constant ranges from 12.376\(\text{Å}\) in YIG to 12.048\(\text{Å}\) in Ca\(_3\)Fe\(_2\)Si\(_3\)O\(_{12}\) while it only decreases to 12.320\(\text{Å}\) in Ca\(_3\)Fe\(_2\)Ge\(_3\)O\(_{12}\). The CaGe:YIG films can therefore be grown over a much larger range of substitution.

A. Composition Measurements

The samples studied in this work and their measured properties are listed in Table 1. The compositions were estimated by measuring the drop in the Curie temperature from that of pure YIG and comparing with the data of Winkler et al.\(^{59}\) which is plotted in Figure 11. The solid line represents Winkler's data and the points are samples used in this work. For low levels of substitution there is a problem with this method due to the different methods of sample preparation.

Winkler made measurements on sintered polycrystalline samples while

<table>
<thead>
<tr>
<th>Sample</th>
<th>Composition $x$</th>
<th>Thickness (μm)</th>
<th>$T_C$</th>
<th>$4\pi M \pm 5%$ (Gauss)</th>
<th>$D \times 10^8$ (Gauss cm$^2$)</th>
<th>$A \times 10^7$ (erg/cm)</th>
<th>$g \pm .005$</th>
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<tr>
<td>B3158</td>
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<td>1.05</td>
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<td>1517</td>
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<td>533</td>
<td>1258</td>
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<td>3.14±.25</td>
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<td>1.31</td>
<td>530</td>
<td>1143</td>
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<td>922</td>
<td>0.805</td>
<td>2.95±.18</td>
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<td>B2956</td>
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<td>768</td>
<td>0.862</td>
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<td>463</td>
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<td>1.36±.18</td>
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</tr>
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</table>
FIGURE 11: Variation of the Curie temperature with composition in CaGe: YIG (after Winkler59).
the samples used here were grown by the LPE process. In LPE films there is inevitably a small amount of lead impurity which has the effect of lowering the Curie temperature. In fact the Curie temperature of a typical LPE YIG film is 543K rather than the standard 553K value. However, as soon as Ca is substituted in the film the level of Pb impurity drops rapidly. There is at present no good explanation for this effect. For films with Curie temperatures less than 530K it was assumed that the lead level was low enough to be neglected and the composition was determined directly from the graph. To estimate the compositions of the two samples with $T_C > 530$, the graph was extrapolated to give a $T_C$ for pure YIG of 543K which is the value of the Curie temperature of a YIG film which was grown from the melt before the Ca and Ge were added.

B. Magnetization Measurements

The room temperature magnetization of all samples was measured and the results of this measurement are plotted in Figure 12. Since the magnetometer actually measures the magnetic moment of the sample not the magnetization (moment per unit volume), there is an uncertainty in the magnetization due to the uncertainty in the film thickness. The error bars on the graph represent only the uncertainty in the film thickness rather than the total uncertainty which is expected to be similar in all samples except the one at $x = 1.03$. This sample is very near the compensation point and its moment is approaching the limit of sensitivity of the magnetometer. The magnetization of the last two points is represented as being negative since these two samples have compositions which are beyond the compensation
FIGURE 12: Variation of the room temperature magnetization with composition in CaGe: YIG.
point and their magnetizations are dominated by the octahedral sub-
lattice. For complete tetrahedral substitution it would be expected that the compensation point is at $x = 1$. Geller\textsuperscript{30} has shown that at liquid helium temperatures the compensation point is slightly beyond $x = 1$ and the Faraday rotation on the sample at $x = 1.03$ indicates that its moment is dominated by the tetrahedral sublattice. The compensation point at room temperature is estimated to be between 1.03 and 1.1 moles per formula unit.

The solid line in the Figure is the best straight line fit to all the data except the last two points. The deviation of last two points from the line is possibly an indication of canting on the octahedral sublattice. Geller's low temperature data and Winkler's room temperature data both show this same departure from linearity in this range of substitution.

C. Resonance Data

A typical room temperature spin wave spectrum of a CaGe:YIG film is shown in Figure 13. This spectrum was measured at X band with the applied field oriented along the film normal. The large number of spin wave modes detected indicates that the pinning at the film surfaces is quite strong ($m \sim 0$) while the fact that some even numbered modes have been detected indicates that there is some asymmetry in the boundary conditions. Recall that in Eq. 112 it was predicted that for symmetric boundary conditions, a set of $k$'s was allowed corresponding to spin wave excitations which were antisym-
metric about the center of the film and had zero intensity. As the
Typical CaGe: YIG spin wave spectrum measured at 9.4 GHz with the applied field perpendicular to the film plane.
boundary conditions become asymmetric these spin waves are no longer antisymmetric and have a small but nonzero intensity. Under these conditions the allowed values of $k$ are approximately given by $n\pi/L$. The modes with even $n$ have an intensity envelope which increases with increasing $n$ to a peak value then decreases while the modes with odd $n$ have an intensity envelope which falls off roughly as $1/n^2$. For large $n$ both even and odd numbered modes lie under the same intensity envelope. This phenomenon has been seen in pure YIG and in many of the CaGe:YIG films investigated in this work. The unique form of the spectrum under this type of boundary condition simplifies greatly the interpretation of the spectrum.

1. Resonance Linewidth

In addition to the strong pinning, another factor which has enabled many spin wave modes to be detected is the narrow resonance linewidth. At room temperature the linewidth of all samples was between 1 and 3 Gauss except for the sample at $x = 1.03$ which had a 20 Gauss linewidth. Since this sample is quite near the compensation point it is thought that inhomogeneities in the film have caused the large linewidth. As a result of this broadening only four spin wave modes were detected in this sample and the resultant uncertainty in the exchange constant is quite large.

It has previously been reported that the resonance linewidth in YIG increases with decreasing temperature to a maximum at about

40K and then decreases. This is thought to be due to rare earth
impurities which are exchange coupled to the iron spins and aid the
iron spins in relaxing to the lattice. A similar behavior was seen
in the CaGe:YIG films where linewidths as large as 50 Gauss were
measured at 40K. However as the temperature was decreased to 4K, the
linewidths remained between a factor of 5 and 10 larger than the room
temperature linewidth. As a result of this the number of spin wave
modes which could be detected at low temperatures was much less than
at room temperature and the accurate measurement of the exchange
constant at low temperatures became impossible. Therefore only room
temperature data has been presented here.

2. Data Analysis

The customary method of measuring the dispersion constant
D from spin wave spectra has been to assume that the surface spins
are completely pinned \( k_n = n\pi/L \) and to plot the resonance field
positions against \( n^2 \). If the first few modes are neglected, this
plot yields a straight line with a slope proportional to D. Using
the surface inhomogeneity model it has been shown that a plot of this
type is nearly linear over a large range of surface boundary con­
ditions. However the slope of this line varies strongly with the
boundary conditions. This fact can introduce quite sizeable errors
into the measurement of D.

Another method of analysis relies on the positions of the low
order modes. Since these depend strongly on the boundary conditions,

61. Henry, R. D., Brown, S. D., Wigen, P. E., and Besser, P. J.,
they can be used to determine the actual boundary conditions in the film using the surface inhomogeneity model. Once the boundary conditions are known the values of $k$ for all the modes can be generated and the dispersion constant determined from the slope of a graph of $H$ vs $k^2$. There are two principal disadvantages to this method. First of all it is quite time consuming. This by itself would not be a significant disadvantage and this method of analysis has been used with good success in pure YIG films. The second disadvantage is that this method relies on the low order modes which not only are quite sensitive to the surface boundary conditions, but also are quite sensitive to any volume inhomogeneities in the internal fields of the film. It was found that a number of the CaGe:YIG samples had spin wave spectra which could not be adequately fit to the surface inhomogeneity model. It is suspected that this poor fit in the substituted garnets is due to volume inhomogeneities in the film caused by stress and concentration gradients near the substrate.

A third method of analysis has been found which is insensitive to the boundary conditions and uses only spin wave modes with $n>3$ which are not very sensitive to small volume inhomogeneities. If the separation between adjacent spin wave modes is plotted against the difference in the squares of the wave vectors of these modes, the plot is nearly linear over a wide range of boundary conditions and the slope is only weakly sensitive to the boundary conditions. In

addition any volume inhomogeneities which have the effect of shifting the spectrum to higher or lower $k$ will show up by shifting only one or two points on the plot which can then be neglected in the analysis. An example of the utility of this method can be seen by examining a few possible pinning cases for which the allowed values of $k$ can be easily written down. As the boundary conditions go from the case in which the spins precess freely at both surfaces ($\nabla m = 0$), to free precession on one surface and complete pinning on the other, to complete pinning ($m = 0$) on both surfaces, the allowed values of $k$ for the $n^{th}$ mode go through the range $(n-1)\pi/L + (2n-1)^2\pi/L + \frac{n\pi}{L}$. However the difference in the squares of consecutive wave vectors $(k_n^2 - k_{n-1}^2)$ is, in units of $(\pi/L)^2$, $2n-3$, $2n-2$, and $2n-1$ for these three cases. Therefore a plot of the field separation between mode $n$ and mode $n-1$ against $2n$ is linear and has a slope which is at most weakly dependent on boundary conditions. A complete analysis shows that the slope of this plot gives the correct value of $D$ within about 3% over the entire range of physically reasonable boundary conditions. In Figure 14 an example of the first and third methods of analysis is shown for a film with Ge substitution at $x = .39$. It can be seen that the first method gives a slope which is 10% lower than the third method due to the lack of complete pinning in the film.

3. Dispersion Constant

Figure 15 shows the results of the measurement of the dispersion constant plotted against composition. The spectra were all measured at X band and the dispersion constant values are all normalized to
FIGURE 14: Example of the difference in the two methods of determining the dispersion constant, D, from the spin wave spectra.
FIGURE 15: Variation of the dispersion constant with composition in CaGe: YIG.
the bulk value for YIG which is taken to be $5.16 \times 10^{-9}$ Oe cm$^2$.

The error bars represent the uncertainty in the film thickness. Since $D = 2A/M$, the solid line is obtained by assuming a linear variation in the magnetization through a compensation point at $x = 1.08$ and using the model of Kirkpatrick and Harris to estimate the exchange constant $A$. The divergence in the curve is due to the existence of the compensation point. As was stated earlier the data from the sample near the compensation point is of questionable value since the linewidth was large and only a few spin wave modes were detected. The reason for the last two points falling above the line is the fact that the magnetization departs from linearity in this region of composition and the exact location of the compensation point is uncertain. From this graph it is difficult to draw many conclusions about the behavior of the exchange constant since the major influence on $D$ comes from the variation in the magnetization.

4. Exchange Constant

Upon combining the measured value of the magnetization $M$ and the dispersion constant $D$, a value of the exchange constant $A = MD/2$ is obtained. This has been done with the X band resonance data and the magnetometer data and the results are plotted in Figure 16. The solid line is the best straight line fit to the data with the sample at $x = 1.03$ excluded from the analysis. The value of the room temperature exchange constant of pure YIG is taken to be $A = 3.75 \times 10^{-7}$ erg/cm.

FIGURE 16: Variation of the exchange constant with composition in CaGe: YIG.
There is a considerable disagreement in the literature about the size of $A_{\text{YIG}}$ and this number falls well within the range of previously reported values. LeCraw and Walker, for example, have measured $A_{\text{YIG}} = 3.61 \times 10^{-7} \text{erg/cm}$ while Henry et al. report the value $4.15 \times 10^{-7} \text{erg/cm}$.

The best fit straight line for the data in Figure 16 has the equation:

$$\frac{A(x)}{A(0)} = 1 - (0.48 \pm 0.03)x$$

It is interesting to compare this result with the prediction of the Kirkpatrick and Harris model. According to the model the variation of the exchange constant with substitution should be:

$$\frac{A(x)}{A(0)} = 1 - 0.43x$$

when the substitution is completely on the tetrahedral sublattice. The agreement is quite reasonable. The small difference between the model and the experimental results cannot be explained on the basis of incomplete substitution on the tetrahedral sublattice since it would require a full 20% substitution on the octahedral sublattice to explain the difference. That large a degree of octahedral substitution would have a significant effect both on the shape of the magnetization curve and on the location of the compensation point. What is more likely the case is that the disagreement is due to the fact that the model is a zero temperature model and the data is room temperature data. Since the room temperature exchange constant must go to zero as the Curie temperature approaches room temperature and
since the Curie temperature decreases with increasing substitution, it would be expected that the room temperature exchange constant decrease with substitution more rapidly than the zero temperature exchange constant. Because of this it is somewhat surprising that the agreement between the model and the data is as good as it is.

It is very difficult to make an estimate of the strength of the temperature dependence of $A$. Slonczewski\textsuperscript{10} attempted to do this in a very approximate theory which was discussed in the previous chapter. In order to compare this theory with the experimental results the exchange constant is plotted against the Curie temperature for all the samples in Figure 17. The straight line is the prediction of the Slonczewski model from Eq. 68.

\[
A(T) = \frac{A_{\text{YIG}}(T)}{(T_C - T)/(T_{C,\text{YIG}} - T)}
\]

$A_{\text{YIG}}(T)$ is taken to be the room temperature value used in this work ($3.75 \times 10^{-7}$ erg/cm) and $T$ is 295K. As can be seen from the graph, the data points seem to fall on a non-linear curve. As $T_C$ decreases from $T_{C,\text{YIG}}$, $A$ first falls off more rapidly than the model predicts and then flattens out with the theory and experiment curves crossing at a Curie temperature just under 400K. This same type of behavior was reported by Henry and Heinz\textsuperscript{37} who measured the exchange constant in gallium substituted YIG. Although their values of $A$ were somewhat higher than those reported here, the shape of the curve was the same. The disagreement between the model and the experiment is probably due to the fact that the model assumes relationships between $A$ and $M$ and between $M$ and $T$ which are valid only near the Curie
FIGURE 17: Variation of the exchange constant with Curie temperature in CaGe: YIG. The straight line is the prediction of Slonczewski et al.\textsuperscript{10}
temperature. The agreement seems to improve significantly as the Curie temperature approaches room temperature.

5. Frequency Dependence of D

In addition to the X band data, the dispersion constant of all samples was measured at K band and for two of the samples at KA band (34GHz). The results of these measurements are listed in Table II. In all samples the measured value of D decreased with increasing frequency. Unfortunately there was no trend in this effect which could be correlated to the composition and the exact cause is as of yet unknown. A number of factors could have influenced this result. However, none of these seem sufficient to cause a decrease in D of up to 10% which was seen in some of the samples.

One cause for a change in the measured value of D with frequency is the increase in the linewidth at higher frequencies. Because of this the uncertainty in the measurement is higher at higher frequencies. This effect would be expected to cause random changes in the measurement at different frequencies, not the decrease which was seen in all samples. Another cause of the frequency variation could be a frequency dependence in the boundary conditions. It is difficult to estimate the size of this effect, if it occurs, but it is not expected that this could cause a change in the measurement of D by more than a percent or two since the overall shape of the spectrum remained the same for each sample as the frequency changed. This indicates that any change in the boundary conditions must have been a small one. A third cause of the measurement of a frequency
<table>
<thead>
<tr>
<th>Sample</th>
<th>Composition ( x )</th>
<th>X Band ( D \times 10^8 ) (Gauss cm(^2))</th>
<th>K Band ( D \times 10^8 ) (Gauss cm(^2))</th>
<th>KA Band ( D \times 10^8 ) (Gauss cm(^2))</th>
</tr>
</thead>
<tbody>
<tr>
<td>B3158</td>
<td>.16</td>
<td>0.578</td>
<td>0.549</td>
<td></td>
</tr>
<tr>
<td>B2843</td>
<td>.32</td>
<td>0.614</td>
<td>0.560</td>
<td></td>
</tr>
<tr>
<td>B3246</td>
<td>.39</td>
<td>0.685</td>
<td>0.638</td>
<td></td>
</tr>
<tr>
<td>B3160</td>
<td>.49</td>
<td>0.805</td>
<td>0.777</td>
<td></td>
</tr>
<tr>
<td>B2956</td>
<td>.62</td>
<td>0.862</td>
<td>0.829</td>
<td></td>
</tr>
<tr>
<td>B2971</td>
<td>.73</td>
<td>1.23</td>
<td>1.09</td>
<td></td>
</tr>
<tr>
<td>B3162</td>
<td>.92</td>
<td>3.07</td>
<td>2.84</td>
<td></td>
</tr>
<tr>
<td>B3272</td>
<td>.96</td>
<td>4.37</td>
<td>4.05</td>
<td>3.88</td>
</tr>
<tr>
<td>B2981</td>
<td>1.03</td>
<td>7.29</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B3177</td>
<td>1.25</td>
<td>2.02</td>
<td>1.98</td>
<td></td>
</tr>
<tr>
<td>B3286</td>
<td>1.37</td>
<td>9.84</td>
<td>9.02</td>
<td>8.23</td>
</tr>
</tbody>
</table>
dependence could be the existence of a measurable static susceptibility in the samples. At higher frequencies the resonance field is larger and any susceptibility will cause $M$ to increase. Since $D = 2A/M$, this would show up as a decrease in $D$. The size of this effect can be estimated for pure YIG. Anderson\textsuperscript{27} has measured the temperature dependence of the static susceptibility of YIG below the Curie temperature. At room temperature he finds $\chi = 0.02$ emu/g/KOe. An increase of the applied field by 5 KOe, which is the approximate difference in the resonance field between X band and K band, will cause an increase in $M$ of 0.3\% and a decrease in $D$ of approximately the same amount. For substituted YIG the susceptibility should be larger since the ratio of the applied field to exchange field is larger and since the measurement temperature is closer to the Curie temperature where the susceptibility diverges. It is difficult to estimate how large the effect of the susceptibility would be.

6. Effective $g$ value

In addition to measuring the dispersion constant, the effective $g$ value of all samples was calculated by measuring the resonance field of the first spin wave mode at two or more different frequencies and using Eq. 108. The results of this measurement are plotted in Figure 18. The uncertainty in the measurement is estimated to be no greater than ±0.005 for all samples. It is seen that $g_{\text{eff}}$ diverges at a point near $x = 1$. This result is in agreement with the Wangsness\textsuperscript{41} model for a two sublattice system in which the two sublattices have different $g$ values. The location of the point at $x = 1.03$ is explained by the fact that the experiment actually measures not $g$ but
FIGURE 18: Variation of the effective $g$ value with composition in CaGe: YIG.
its absolute value. This sample then has a composition which is just above the composition for which \( g \) diverges. It can also be seen that the curve rises much less rapidly after the compensation point than before it. A similar effect has been seen in a series of compounds known as nickel ferrite-aluminates \((\text{Ni}_2\text{Fe}_2-x\text{Al}_x\text{O}_3)\). This effect can also be possibly explained by the existence of a measurable static susceptibility. If the magnetization is given by

\[
M = M_0 + \chi H
\]

where \( M_0 \) is the spontaneous magnetization and \( H \) is the applied field, the resonance equation, neglecting the spin wave energy and anisotropy terms, becomes:

\[
\omega = H\gamma(1 - 4\pi\chi) - 4\pi\gamma M.
\]

The measured value of \( g \) is then reduced by a factor of \( 1 - 4\pi\chi \). The size of this effect can be roughly estimated. The value of the susceptibility necessary to lower the \( g \) value of the last sample from 2.02 to its measured value of 1.99 is \( \chi \approx 0.2 \text{ emu/g/kOe} \). This is a factor of 10 greater than the measured room temperature susceptibility of YIG. It is then reasonable to suspect that at least part of the shape of the curve is due to this effect.

**D. Summary**

The room temperature exchange constant in CaGe substituted YIG has been measured over a broad range of compositions using ferromagnetic resonance techniques. In addition an attempt was made to measure the exchange constant at liquid helium temperatures but the increase in resonance linewidth over its room temperature value made
this measurement quite difficult. The results of the room temperature measurement have been shown to be in good agreement with the predictions of a percolation model by Kirkpatrick and Harris when it is assumed that the substitution is made exclusively on the tetrahedral sublattice. This model is insensitive to variations of a few percent in the fractional substitution on the sublattices. Although it is designed for low temperatures, the model can be used at room temperatures to give a quantitative estimate of the variation of the exchange constant with substitution. It would be interesting to test this model on a system in which the substitution is confined to the octahedral sublattice and plans are presently being made to do so.

The exchange constant data has also been compared with a theory by Slonczewski which predicts the relationship between the room temperature exchange constant and the Curie temperature. In this case it has been found that the agreement is qualitative at best and that the relationship between $T_C$ and $A$ is not a simple linear one. This result is in agreement with previously reported results on the measurement of the exchange constant in Ga substituted YIG.

The dispersion constant was measured at two frequencies and in some cases three and it was found that in all cases $D$ decreased with increasing frequency. The interpretation of this result is not clear. It is suspected that this is due in part to the existence of a susceptibility and that at higher frequencies there is an increase in the magnetization due to the higher resonance fields. However, this interpretation is not unambiguous and more work must be done before anything definite can be said.
Lastly the effective g value has been measured and it has been shown to follow the predicted behavior of the g value in a two sub-lattice system in which each sublattice has a different g value.
BIBLIOGRAPHY


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APPENDIX

This is a computer subroutine written for a Hewlett-Packard model HP-9820 calculator. It solves the characteristic equation of the surface inhomogeneity model to find the allowed values of $k$ for spin waves in a thin film. In this model the allowed values of $k$ for bulk spin waves are given by the solutions to the equation:

$$\tan kL = \frac{r \sin k}{\cos k - p}$$  \hspace{1cm} (A1)

where

$$r = \frac{ab - 1}{ab + 1} \hspace{1cm} p = \frac{a + b}{ab + 1}$$  \hspace{1cm} (A2)

and $a$ and $b$ are the two surface parameters. $L$ is the film thickness measured in lattice constants. For surface spin waves with $k = \pi t$, the allowed values of $t$ are solutions to the equation:

$$\tanh tL = \frac{r \sinh t}{\cosh t - p}$$  \hspace{1cm} (A3)

In this subroutine the surface parameters $a$ and $b$ are replaced by new parameters $x$ and $y$ where:

$$a = 1 - x \hspace{1cm} b = 1 - y$$  \hspace{1cm} (A4)

and the small $k$ approximation, $\sin k \approx k$ and $\cos k = 1 - \frac{1}{2}k^2$, is made. Under this approximation the characteristic equation becomes

$$\tan kL = \frac{\frac{2a}{2 - \alpha} k}{k^2(2 - \alpha) - 2xy}$$  \hspace{1cm} (A5)
where $a = x + y - xy$. A similar expression results for the surface mode characteristic equation.

The solutions of these equations fall into three regions according to the values of $x$ and $y$. If

$$x < 0 \quad y < 0$$

and

$$-a < Lxy$$

the first two solutions are surface modes. If

$$-a > Lxy$$

the first solution is a surface mode and all others are bulk modes. If neither of the conditions determined by Eq. A6 and Eq. A7 hold, all solutions are bulk spin wave modes.

The values of $k$ for surface modes are stored as negative numbers to indicate that they are imaginary.
0: "FIND K"; "5"
1: CFG 1; CFG 2; 1→C; X+Y→XY=R0; SFG 14; TBL 2
2: IF -R0>A; SFG 1
3: IF (0>X)(0>Y)(-R0≤AXY); SFG 2
4: GSB "M3"; IF C≤2; GSB "M2"; IF C=1; GSB "M1"
5: IF (C+1→C)<R1; GTO -1
6: RET
7: "M1"
8: IF (0>XY)(-R0=AXY); 0+B→R(5+C); RET
9: (2-R0)/(2-R0-XY)→R2; LN (R2+√(R2R2-1))→R2
10: IF FLG 2; R2→B+Z; GTO +3
11: IF FLG 1; 1E-10→B; 10/A+Z; GTO +2
12: GTO +3
13: GSB "T1"
14: -B→R(5+C); RET
15: 1E-10→B; π/2A+Z; GSB "B1"
16: B→R(5+C); RET
17: "M2"
18: IF(0>X)(0>Y)(-R0=AXY); 0+B→R(5+C); RET
19: (2-R0)/(2-R0-XY)→R2; LN (R2+√(R2R2-1))→R2
20: IF FLG 2; -.5(R2→B)+Z; GTO +5
21: IF FLG 1; 1E-10→B; GTO +2
22: R(4+C)+π/4A+B
23: π/2A+Z; GSB "B1"
24: B→R(5+C); RET
25: GSB "T1"
26. \(-B+R(5+C)\); RET

27: "M3"

28: IF FLG 2(C=3); 1E-10+B; GTO +2

29: \(R(4+C)+\pi/4A+B\)

30: \(\pi/2A+Z\); GSB "B1"

31: \(B+R(5+C)\); RET

32: "B1"

33: CFG 3; CFG 4

34: \((BB(2-R0)-2XY) \sin (AB)-2BR0 \cos(AB)\rightarrow R3\)

35: IF FLG 3=0; SFG 3; R3+R4; B+Z+B; GTO -1

36: IF 0>R3R4; R3+R4; SFG 4; B+(-.5Z+Z)\rightarrow B; GTO -2

37: IF ABS \((Z/B)\)\(\leq\)1E-4; RET

38: IF B>20\pi/A; GTO +2

39: \((1-.5 \text{FLG 4})Z+Z; B+Z+B; R3\rightarrow R4; GTO -5\)

40: DSP "ERROR IN B1"; STP

41: "T1"

42: CFG 3; CFG 4

43: \(.5(\exp(B+\exp(-B))\rightarrow R14; \ .5(\exp(B-\exp(-B))\rightarrow R5\)

44: \(((2-R0)(R14-1)+XY)(1-\exp(-2AB))/(1+\exp(-2AB))\rightarrow ROR5\rightarrow R3\)

45: IF FLG 3=0; SFG 3; R3\rightarrow R4; B+Z+B; GTO -2

46: IF 0>R3R4; R3\rightarrow R4; SFG 4; B+(-.5Z+Z)\rightarrow B; GTO -3

47: IF ABS \((Z/B)\)\(\leq\)1E-4; RET

48: IF \((O>B)+(B>\text{FLG 1+100 R2 FLG 2})\); GTO +2

49: \((1-.5 \text{FLG 4})Z+Z; B+Z+B; R3\rightarrow R4; GTO -6\)

50: DSP "ERROR IN T1"; STP

51: END
MEMORY MAP

A  Thickness L
B  k
C  counter
X  
Y  Surface Parameters
Z  delta k

R0  X + Y - XY
R1  Maximum Mode Number
R2  
R3  temporary storage
R4  
R5  

R6-R(13)  \( k_1 \)
R14  temporary storage