Nuclear-Magnetic-Resonance Studies of Fe^{57} in Rare-Earth Iron Garnets

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Nuclear-magnetic-resonance studies of Fe^{57} have been carried out in several rare-earth iron-garnet single crystals (EuIG, TmIG, ErIG, GdIG) and in polycrystalline samples of SmIG and DyIG. The studies were made using the spin-echo technique at 4.2 °K and in some cases at 77 °K. External dc fields of several kG were used in the single-crystal studies to drive out domain walls and were of sufficient magnitude in most cases to align the magnetization along hard as well as easy directions. From studies with the magnetization along ⟨111⟩ and ⟨100⟩ directions, anisotropic effective fields at octahedral and tetrahedral Fe sites could be deduced. The anisotropic effective fields could be largely accounted for by considering the dipolar fields of neighboring magnetic ions. Effects of covalency and transferred hyperfine fields on the isotropic effective fields are also considered.

I. INTRODUCTION

Nuclear-magnetic-resonance (NMR) studies in rare-earth iron garnets are of interest because they can give very detailed information concerning the magnetic properties and hyperfine interactions of the magnetic ions. Recently we have been studying the NMR of rare-earth ions in several of the iron garnets,[1-6] and in this communication we report on Fe^{57} NMR studies in these same garnets (preliminary accounts of portions of this work have been reported previously[4,7]).

The effective fields associated with the Fe^{+3} ions at the octahedral (a) and tetrahedral (d) sites of the iron garnets have previously been discussed in detail. The effective fields are composed of isotropic parts, arising mainly from the core polarization, and anisotropic parts, arising mainly from the classical dipolar fields of neighboring magnetic ions. The dipolar fields in YIG have been discussed by Robert and Hartmann-Boutron.[8] The Fe^{+3} ions on a sites possess axes of threefold symmetry along the ⟨111⟩ directions, while the iron ions on d sites have symmetry axes along the ⟨100⟩ directions. If we consider the case of the iron magnetization $\vec{M}$, along a ⟨111⟩ direction, we expect the Fe^{57} NMR from a sites to exhibit two resonances at $\nu_{a'}$ and $\nu_{a''}$, the a' resonance being due to sites whose local symmetry axis is along $\vec{M}$, and the a'' resonance being due to sites whose symmetry axes are along the other three ⟨111⟩ directions. The d-site NMR will exhibit only one line at ν_d for $\vec{M}$ along ⟨111⟩, but for $\vec{M}$ along ⟨100⟩ it splits into two lines at $\nu_{d'}$ and $\nu_{d''}$ with relative intensities of 1:2, corresponding to the symmetry axes being parallel and perpendicular to $\vec{M}$, respectively.

With the exception of YIG where studies have been carried out with single crystals and externally applied dc magnetic fields,[8,9] all the previous studies of rare-earth iron garnets have been carried out with polycrystalline samples in zero field. Gonano and co-workers[9,10] and Myers and co-workers[11,12] have observed the zero-field NMR of Fe^{57} in several rare-earth iron garnets from nuclei in both domains and domain walls. Since the easy direction of magnetization in most of the garnets is ⟨111⟩, they could obtain the a-site resonances. Since, however, the d-site domain resonance for $\vec{M}$ along ⟨111⟩ is unsplit, and since the domain-wall spectrum from d sites was not understood, they could not determine the dipolar splittings at (d) sites. In the present study we have used single crystals and dc fields of sufficient magnitude to drive out domain walls and align $\vec{M}$ along various crystallographic directions and have been able to obtain dipolar splittings at d as well as a sites. A brief discussion of the theory of the dipolar fields at a and d sites is given in Sec. II. Experimental techniques are discussed in Sec. III. Results are presented and discussed in Secs. IV and V.

II. THEORY OF DIPOLAR FIELDS

As already noted, Robert and Hartmann-Boutron[8] have discussed the dipolar fields at the a and d sites of the garnets. Their discussion applies specifically to YIG. They find that the dipolar field H_{dip} at these sites can be written as

$$H_{dip} = (3\cos^2\theta - 1)\sum_{i\neq 0}\frac{1}{2}\left(\frac{3\cos^2\theta_i - 1}{r_i^3}\right)\mu_i \,. \qquad (1)$$

Here θ is the angle between the Fe magnetization vector $\vec{M}$ and the symmetry axis (between $\vec{M}$ and ⟨111⟩ for a sites or between $\vec{M}$ and ⟨100⟩ for d

sites). In the lattice sum, r_i is the distance to the ith neighboring magnetic atom, θ_i is the angle between the symmetry axis and r_i, and μ_i is the moment of the atom at r_i. Equation (1) was derived assuming all moments to be colinear. If one neglects the canting of the rare-earth moments and assumes that they are all magnetically equivalent, then Eq. (1) remains valid if one considers the presence of rare-earth ions.

Consider now the a-site splitting between lines at $\nu_{a'}$ and $\nu_{a''}$ that occurs when $\vec{M}$ is along the $\langle 111\rangle$ direction. Following Myers and co-workers[11] one can show that

$$\nu_{a''}(\text{obs}) - \nu_{a'}(\text{obs}) = (\gamma/2\pi)\tfrac{8}{3} A[\sigma_d + (M_{0c}/M_{0d})\sigma_c] + \Delta_a . \quad (2)$$

Here $\nu_{a''}$ (obs) and $\nu_{a'}$ (obs) are the observed (uncorrected) a-site frequencies. The gyromagnetic ratio $\gamma/2\pi$ is 137.4 Hz/G for Fe^{57}.[11,13] The factor A is a constant depending on the dipolar lattice sum for d sites with respect to a sites and on the magnetic moment of the Fe ion. The quantities σ_d and σ_c are the reduced magnetizations for the d and c sublattices, respectively, while M_{0c} and M_{0d} are the magnetizations of these sublattices at absolute zero. The first term in parentheses is due to the dipolar fields of d-site iron ions. The second term is the dipolar contribution from the c sites and takes this form because the lattice sum for c sites with respect to a sites is just the negative of the sum for d sites. The parameter Δ_a takes into account effects not due to dipolar fields. For YIG from the calculations of Robert and Hartmann-Boutron[8] or Burns[14] one obtains $A = 2044$ G at 0 °K.

Similarly by taking into account the relative magnitudes of the appropriate lattice sums[14] one can write for the d-site splitting for $\vec{M}$ along $\langle 100\rangle$:

$$\nu_{d''}(\text{obs}) - \nu_{d'}(\text{obs}) = (\gamma/2\pi)3A \times [0.29\,\sigma_a - 0.21\,\sigma_d - 1.03(M_{0c}/M_{0d})\sigma_c] + \Delta_d . \quad (3)$$

Note that the d sites the dipolar fields from the two iron sublattices nearly cancel.

Although the preceding discussion is valid for uncanted rare-earth ions, for cases like Dy and Er where the canting is found to be rather large[15] and where the rare-earth moments are not magnetically equivalent, the situation becomes considerably more complex. Some simplication can be made, however, by observing that for the case of $\vec{M}$ along a symmetry axis the components of the canted rare-earth moments perpendicular to $\vec{M}$ will cancel and only the component along $\vec{M}$ need be considered. For the case of $\vec{M}$ along $\langle 111\rangle$ two types of rare-earth sites c and c' occur in equal abundance, each with a different moment and different angle of canting out of the $\langle 111\rangle$ direction. If one considers a given neighbor shell of an a site one finds that, on the average, a' or a'' atoms see equal numbers of c and c' neighbors. Consequently, the average dipolar fields at a' and a'' sites should still satisfy Eq. (1), and the average a-site splitting should still be given by Eq. (2) if we use for M_{0c} an average of the c- and c'-moment components parallel to the $\langle 111\rangle$ direction. For most of the rare-earth iron garnets the easy direction of magnetization is $\langle 111\rangle$. Consequently, we can use for M_{0c} in Eq. (2) the values deduced from bulk-magnetization studies.[16,17] Similar considerations apply to the d-site splittings.

III. EXPERIMENTAL METHODS

A. Samples

The garnets were obtained from Airtron,[18] and consisted of four single crystals (Eu, Tm, Er, Gd) and five finely divided polycrystalline powder samples (Eu, Sm, Tm, Er, Dy). About 15 g of each powder was sealed in a glass tube. A typical single crystal weighed about 6 g and was used as grown without further shaping. Each crystal was mounted on a Bakelite rod so that it could be rotated about a [110] axis so that each of the principal axes were contained in the plane of rotation.

B. NMR Spin-Echo Equipment

The samples were maintained at 4.2 °K in exposed-tip helium Dewars which fitted into the tunable tank coil. The coil consisted of several turns of wire with a 2.5-cm diam for the single crystals, and a 3.8-cm coil was used for the larger powder samples. The axis of the coil was colinear with the crystal [110] axis and perpendicular to the external magnetic field (0–8 kG). The input and output coupling loops consisted of a single turn of wire.

The rf pulses were supplied by a high-power pulsed oscillator (Arenberg Ultrasonic Laboratory Model No. PG-650-C). The signals were detected by a superheterodyne receiver and displayed on an oscilloscope. Both the pulsed oscillator and receiver were matched to the coupling loops with double-stub tuners.

C. Measurement Techniques

The measurement procedure and parameters depend upon whether one is observing domain-wall signals or domain signals. In the powder samples in zero field one can often detect both kinds of signals.[11] For the wider and more enhanced domain-wall signals one can use a 1-μsec pulse at relatively low power and trace out the line shape which may

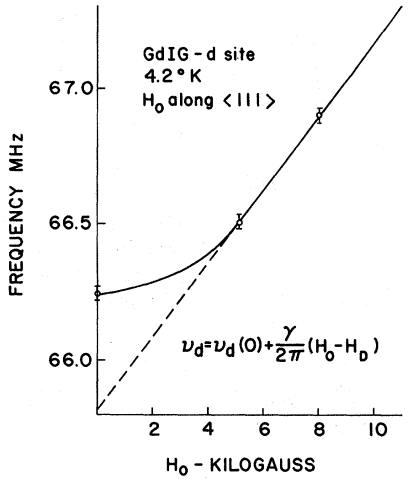

FIG. 1. External magnetic field dependence of d-site frequency at 4.2 °K in GdIG crystal with $\vec{H}_0$ along the ⟨111⟩ direction.

be about a MHz in width. As the pulse frequency is varied, the intensity of the echo is measured by comparison with an auxiliary signal generator, and its frequency is measured by zero beating the echo against the signal generator and measuring the signal-generator frequency with a counter.

The much narrower domain signals require a pulse length of about 10 μsec in order to identify the position at peak accurately. This permits an accuracy of the frequency measurement of about 10-20 kHz. Also, the rf power must be raised to compensate for the smaller enhancement factor inside the domain.

In the single-crystal measurements, the application of a magnetic field causes the domain-wall resonance to vanish and the domain resonance is improved. It was found that this improvement more than made up for the smaller size of the crystal sample. The resonances are measured as the crystal is rotated with respect to the magnetic field. As the crystal is rotated the enhancement factor, as well as the frequency, varies. However, even in the easy direction, which is the position of poorest enhancement, the echo amplitude could be optimized by using 10-μsec pulses and high-rf power.

IV. RESULTS

A. Gadolinium Iron Garnet

Since GdIG has an easy direction of magnetization along ⟨111⟩, the d site therefore exhibits only a single line in zero fields. The d-site frequency in an external dc field $\vec{H}_0$ can be written

$$\nu_d = \nu_d(0) + \gamma/2\pi(H_0 - H_D). \quad (4)$$

Here $\nu_d(0)$ is the *zero-field* d-site frequency corresponding to the isotropic hyperfine field (including Lorentz field), and the second term reflects the corrections arising from external and demagnetizing fields. We have plotted the d-site frequency at 4.2 °K obtained with $\vec{H}_0$ along the ⟨111⟩ direction of the crystal in Fig. 1. The slope of the linear part of the curve yields a value of $\gamma/2\pi$ of 135 Hz/G in approximate agreement with the known value. Since the crystals are approximately spherical in shape we can take H_D (in the fully magnetized specimen) to be $4\pi M_s/3$ or about 2559 G. Extrapolating the linear portion of the curve (Fig. 1) back to $H_0 = 0$ yields a value of ν_d of 65.82 MHz. Correcting this value for H_D gives a value of ν_d of 66.17 MHz. This value is in approximate agreement with the observed value of $\nu_d(0)$ (for nuclei in domains) of about 66.246 MHz. This shows that the nuclei in the domains of the unmagnetized specimen see essentially no demagnetizing field which is an assumption implied in Ref. 11.

Values of $\nu_{a''}(0)$, $\nu_{a'}(0)$, and $\nu_d(0)$ at 4.2 °K are listed in Table I and are in approximate agreement with the values reported in Ref. 11. With $H_0 =$ = 5.1 kG along ⟨111⟩, the a'' and a' frequencies were 75.71 and 73.69 MHz, respectively, the splitting being in approximate agreement with that in zero field.

For GdIG the anisotropy is not too large[19] even at 4.2 °K so that a relatively low value of H_0 is sufficient to orient $\vec{M}$ along the ⟨100⟩ direction. Values of $\nu_{a'}$ and $\nu_{a''}$ observed at 4.2 and 77 °K for various values of applied field are given in Table II. Within experimental error the splitting is independent of applied field.

B. Europium Iron Garnet

Studies were carried out with the single-crystal sample at 4.2 °K. Values of $\nu_{a''}(0)$, $\nu_{a'}(0)$, and $\nu_d(0)$ are given in Table I and are in approximate agreement with the values of Ref. 12. The a-site spectrum was studied at 4.2 °K with an external field of 7.8 kG as a function of angle φ_0 in the (110) plane between $\vec{M}$ and the ⟨100⟩ direction. As dis-

TABLE I. Observed zero-field a''-, a'-, and d-site frequencies in various rare-earth garnets.

Garnet	$\nu_{a''}(0)$ (MHz)	$\nu_{a'}(0)$ (MHz)	$\nu_d(0)$ (MHz)
GdIG[a]	76.012	73.969	66.246
EuIG[a]	76.317	75.287	65.45
TmIG[a]	75.840	74.458	64.858
DyIG[a]	75, 95	74.35	65.80
YIG[b]	76.058	75.057	64.980
LuIG[b]	75.802	74.900	65.560

[a]Values at 4.2 °K.
[b]Values at $T = 0$ °K from Ref. 10.

TABLE II. Values of $\nu_{a'}$ and $\nu_{a''}$ observed at 77 and 4.2 °K for different values of H_0 applied along the ⟨100⟩ direction of a GdIG crystal.

Temp (°K)	H_0 (kG)	$\nu_{a'}$ (MHz)	$\nu_{a''}$ (MHz)	$\nu_{a'} - \nu_{a''}$ (MHz)
77	2.8	65.97	65.28	0.69 ± 0.05
4.2	8.0	67.78	66.60	1.18 ± 0.05
4.2	5.1	67.41	66.28	1.13 ± 0.05

cussed elsewhere[5] this field is sufficient to align the magnetization exactly along the principal cubic directions and approximately along other directions in the (110) plane. From Eq. (1) one can show that

$$\nu_a(\theta_0) = \nu_a(\perp) + \cos^2\theta_0[\nu_a(\parallel) - \nu_a(\perp)]. \quad (5)$$

Here $\nu_a(\parallel)$ and $\nu_a(\perp)$ are the frequencies corresponding to $\vec{M}$ being parallel and perpendicular to the symmetry axis, and θ_0 is the angle between $\vec{M}$ and the symmetry axis. The a-site frequencies $\nu_a(\varphi_0)$ have been calculated for each of the four inequivalent types of a sites (corresponding to symmetry axes along different ⟨111⟩ directions). Experimental and calculated values of $\nu_a(\varphi_0)$ are shown in Fig. 2 where we have taken $\nu_a(\parallel) = 76.26$ MHz and $\nu_a(\perp) = 77.42$ MHz in obtaining the calculated curves. For $\vec{M}$ in the (110) plane the symmetry does not allow for more than three resonances at any angle. In plotting the experimental points we have corrected, as was done in Ref. 5, for the fact that the angles φ_0 between ⟨100⟩ and $\vec{M}$ are not, in general, equal to the angles φ between ⟨100⟩ and $\vec{H}_0$. The agreement between experimental and calculated values is within the limits of experimental error, and the ⟨111⟩ splitting is approximately equal to that in zero field.

The angular dependence of the d-site line was also studied in the ⟨110⟩ plane at 4.2 °K with H_0 equal to 7.8 kG. No shifting or splitting of the d-site line was observed to an accuracy of about 0.01 MHz.

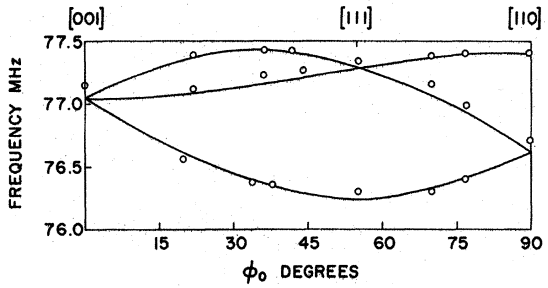

FIG. 2. Fe^{57} a-site frequencies in EuIG at 4.2 °K calculated from Eq. (5) as a function of angle φ_0 in the (110) plane between $\vec{M}$ and the [001] axis together with experimental points.

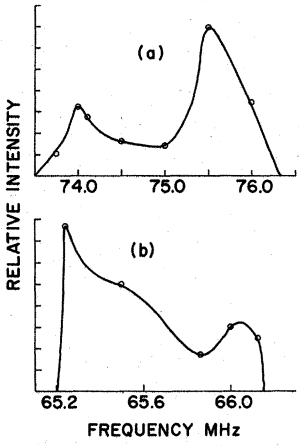

FIG. 3. ErIG zero-field (domain wall) spectrum at 4.2 °K from (a) a sites and (b) d sites.

C. Thulium Iron Garnet

Studies were carried out with the single-crystal sample at 4.2 °K. Values of $\nu_{a''}(0)$, $\nu_{a'}(0)$, and $\nu_d(0)$ are listed in Table I and are in approximate agreement with the values of Ref. 11. The anisotropy of TmIG is sufficiently high at 4.2 °K that we could not align $\vec{M}$ along the hard ⟨100⟩ direction with the available field. With $H_0 = 8.0$ kG along ⟨111⟩, the a'' and a' frequencies were 76.84 and 75.48 MHz, the splitting being approximately equal to that measured in zero field.

D. Erbium Iron Garnet

For ErIG the ⟨100⟩ and ⟨111⟩ directions are both relatively easy directions of magnetization[20,21] at 4.2 °K. The ⟨100⟩ direction is easiest but an external field of only a few kilogauss applied to a single crystal is sufficient to turn the magnetization into the ⟨111⟩ direction. The NMR was studied using both polycrystalline powder and a single crystal. The domain-wall a-site spectrum at 4.2 °K obtained with $H_0 = 0$ with low rf power levels is shown in Fig. 3(a) and the d-site spectrum in Fig. 3(b). On increasing the peak rf power level and the width of the exciting pulses, a single narrow domain resonance emerged at 75.45 MHz from the a-site spectrum, consistent with the easy direction of magnetization being ⟨100⟩. For d sites, domain resonances appeared at 66.15 and 65.22 MHz with relative intensities of 1:2 corresponding to $\nu_{a'}$ and $\nu_{a''}$, respectively.

A plot of the a-site frequency ν_a as a function of H_0 oriented along the ⟨100⟩ axis of the single crystal is shown in Fig. 4. Correcting the linear portion of the curve for H_D gave a value of $\nu_a(0)$ in good agreement with the zero-field value. With $H_0 = 5.6$ kG (along ⟨100⟩) the d-site frequencies were found to be 66.719 and 65.779 MHz, the splitting being approximately equal to that in zero field.

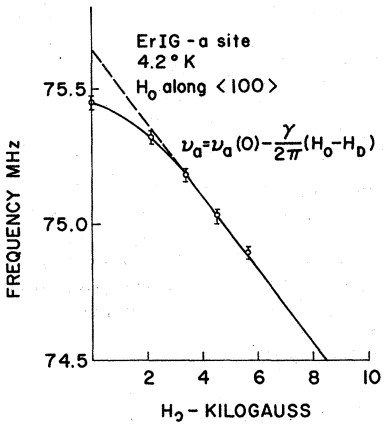

FIG. 4. External magnetic field dependence of a-site frequency at 4.2 °K in ErIG crystal with $\vec{H}_0$ along the ⟨100⟩ direction.

With $H_0 = 5.6$ kG along ⟨111⟩, the a'' and a' frequencies were found to be 75.44 and 73.41 MHz, respectively, while ν_d was found to be 66.166 MHz.

E. Dysprosium Iron Garnet

For DyIG no single crystals were available. The a-site zero-field powder spectrum is shown in Fig. 5(a) and the d-site spectrum in Fig. 5(b). While the signals evidently arise largely from nuclei in domain walls, the peaks may reflect at least approximate values of $\nu_{a'}(0)$, $\nu_{a''}(0)$, and $\nu_d(0)$, and are entered in Table I.

F. Samarium Iron Garnet

For SmIG we could not observe an a-site resonance at 4.2 °K but a weak d-site resonance was observed at 65.66 MHz. For SmIG the easy direction of magnetization is believed[22] to be along ⟨110⟩. For this direction the d-site resonance should exhibit two lines with relative intensity 2:1, but since the Sm moment is small we might expect the splitting to be small as in YIG. Mössbauer studies,[22] however, indicate two d-site hyperfine fields corresponding to frequencies of 66.5 and 64.1 MHz with relative intensities of 2:1. The weighted average yields a frequency 65.7 MHz which is in good agreement with our value.

G. Other Garnets

For convenience, values of $\nu_{a''}(0)$, $\nu_{a'}(0)$, and $\nu_d(0)$ for LuIG and YIG (from Ref. 9) are listed in Table I.

V. DISCUSSION

A. Dipolar Splittings

Values of $\nu_{a''} - \nu_{a'}$ calculated (for $T = 0$ °K) using the first term in Eq. (2) (the dipolar term) are listed in Table III for several rare-earth iron garnets together with experimental values at 4.2 °K (at 0 °K for YIG and LuIG). For ErIG the splitting has been obtained from values of $\nu_{a''}$ and $\nu_{a'}$ at 5.6 kG. The values of A have been obtained by taking A = 2044 G for YIG and by taking into account the difference in lattice constants[23] for the other garnets. The quantities M_{0c}/M_{0d} have been obtained from bulk magnetization studies[16,17] except for ErIG where we have taken $M_{0c}/M_{0d} = 1$, consistent with $(5.0 \pm 0.2)\ \mu_B$ for the average Er moment[21] for $\vec{M}$ along ⟨111⟩. We see that in all cases except for DyIG the values of Δ_a are positive. The values are approximately consistent with those reported in Ref. 11. The apparent anomaly in the case of Dy may be due to the fact that the splittings were deduced from the powder spectrum or may be associated with the large canting of the Dy moments out of the ⟨111⟩ direction.[15]

A similar comparison between calculated and observed values of $\nu_{d''} - \nu_{d'}$ is summarized in Table IV. For ErIG we have used the average c-site moments[21] obtained with $\vec{M}$ along ⟨001⟩ to calculate M_{0c}/M_{0d}. Note that the value of Δ for d sites appears to be less than that for a sites and within the experimental error is zero for GdIG and ErIG.

B. Isotropic Hyperfine Fields

The observed resonance frequencies (in zero field) must be corrected for the effects of anisotropic and Lorentz fields in order to obtain ν_a(corr) and ν_d(corr), the frequencies associated with the isotropic hyperfine fields at a and d sites. The anisotropy correction for a sites has been made by taking a weighted average over the a' and a'' frequencies (Table I). (For ErIG we have taken an appropriate weighted average over the ⟨100⟩ frequencies to obtain ν_a while ν_d can be obtained directly.)

Values of ν_a(corr) and ν_d(corr) at 4.2 °K are plotted in Fig. 6 vs lattice parameters (at room temperature) obtained from the data of Bertaut and

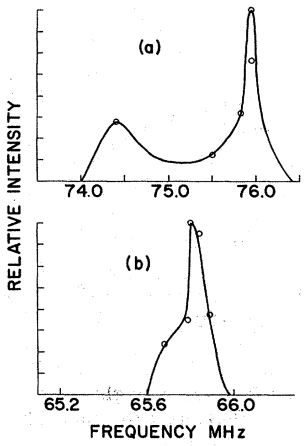

FIG. 5. DyIG zero-field spectrum at 4.2 °K from (a) a sites and (b) d sites.

Forrat.[23] For a sites the hyperfine field varies linearly with lattice parameter with a slope of about 2.7 ± 0.9 MHz/Å. For d sites the slope appears to be larger (about 4.1 ± 1.3 MHz/Å). The a-site slope would be approximately equal to the value obtained in Ref. 11 and corresponds to $K = d\ (\ln A)/d\ (\ln V) = 0.15 \pm 0.05$, where A is the hyperfine coupling constant and V is the sample volume. The d-site slope corresponds to $K = 0.23 \pm 0.07$. The variation of hyperfine field with lattice constant must be due largely to effects of covalency. The larger value of K for d sites may be related to the greater covalency of these sites, the effect having been observed previously at higher temperatures.[24,25]

Note that in the case of d sites the points for GdIG, ErIG, and DyIG appear to fall significantly above the straight line as we have drawn it. One effect that could cause the variation of hyperfine fields with lattice parameter to depart from a linear relation would be the transferred hyperfine fields at iron sites arising from the rare-earth ions. Furthermore, the effect would be expected to be greater for d sites than for a sites due to the relatively larger c-d interaction and would be greatest for those ions like Gd with large magnetic moments and exchange interactions.

The magnitude of the effect can be estimated by considering the transferred hyperfine field at Ga atoms[26] on d sites in Ga-substituted YIG. The field is about 20 kG and arises primarily from Fe atoms on neighboring a sites. The sign of the field was found to be along the direction of the a-site moments. Since the Ga and Fe ions have similar structures we would expect a similar field at Fe d sites arising from neighboring iron and rare-earth ions. The existence of this field has been shown by Gonano and co-workers,[9] who find the total transferred field in YIG at a sites from the neighboring d sites to be about 25 ± 25 kG and the field at d sites from neighboring a sites to be about 10 ± 10 kG. The sign in both cases is such that the field adds to that of the parent ion. Assuming a crude proportionality between the transferred hyperfine field at a given site and molecular-field coefficients,[27] we can estimate the transferred hyperfine fields at Fe d sites from Gd ions on c sites to be about 2 kG corresponding to a frequency of about 0.3 MHz. The sign of the field would be such as to increase the d-site frequency as is observed.

It is interesting that the apparently smaller transferred hyperfine field at d sites relative to a sites is in agreement with what is found in the spinel and hexagonal ferrites.[28]

The anisotropic fields represented by the quantity Δ may also arise from transferred-hyperfine-field effects. The value of Δ_a which is typically 0.3 MHz corresponds to a field of about 2 kG. This is larger than the value of 1 kG calculated in Ref. 8 where admixtures of $3d^4 4s$ states with the $3d^5$ states were considered. Contributions to Δ could also arise

TABLE III. Comparison of calculated and observed values of $\nu_{a''} - \nu_{a'}$.

Garnet	A (G)	M_{0c}/M_{0d}	Calc[a] $\nu_{a''} - \nu_{a'}$ (MHz)	Observed $\nu_{a''} - \nu_{a'}$ (MHz)	Δ_a (MHz)
YIG	2044	0.00	0.75	1.00[b]	0.25
LuIG	2094	0.00	0.77	1.10[b]	0.33
EuIG	1975	0.16	0.84	1.03[b]	0.19
TmIG	2069	0.25	0.96	1.38[b]	0.42
ErIG	2054	1.00	1.51	2.03[c]	0.52
GdIG	1994	1.40	1.75	2.04[b]	0.29
DyIG	2034	1.46	1.78	1.6[b]	−0.18

[a]See text.
[b]Obtained from values in Table I.
[c]Obtained for $H_0 = 5.6$ kG (see text).

TABLE IV. Comparison of calculated values of $\nu_{d''} - \nu_{d'}$ with the observed values.

Garnet	Temp (°K)	M_{0c}/M_{0d}	Calc[a] $\nu_{d''} - \nu_{d'}$ (MHz)	Observed $\nu_{d''} - \nu_{d'}$ (MHz)	Δ_d (MHz)
YIG	4.2	0.00	0.07	0.13[b]	0.06
EuIG	4.2	0.16	−0.07	0.00 ± .01	0.07
GdIG	77	0.85	−0.66	−0.69 ± .05	−0.03
ErIG	4.2	1.08	−0.88	−0.93 ± .05	−0.05
GdIG	4.2	1.40	−1.12	−1.15 ± .05	−0.03

[a]See text.
[b]Obtained from Ref. 8.

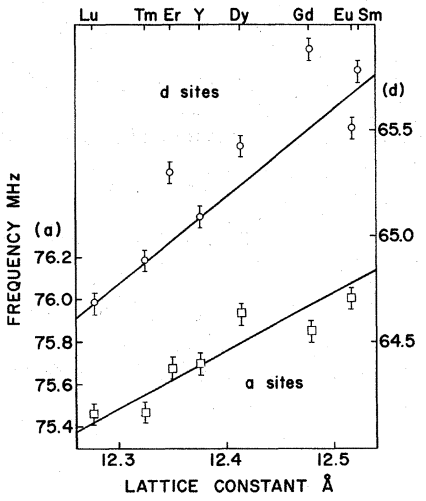

FIG. 6. Plots of a- and d-site frequencies (corrected for anisotropic effects and Lorentz fields) at 4.2 °K vs lattice parameter for several rare-earth iron garnets.

from admixtures of p wave functions from the ligands into the Fe orbitals which would represent the anisotropic part of the transferred hyperfine field. Note that the anisotropic transferred hyperfine field would only need to be about 10% of the isotropic field to account for the observed effect. Furthermore, the lower value of Δ observed for d sites may be related to the lower transferred hyperfine field at these sites.

C. Erbium Iron Garnet

As we pointed out previously in a preliminary account of the ErIG studies,[4] our results as well as those of Refs. 20 and 21 indicate without doubt that the easy direction of magnetization in ErIG at 4.2 °K is the ⟨100⟩ direction. Furthermore, since our results were obtained by cooling the powder from room temperature, it is apparent that the ⟨100⟩ direction becomes the easy direction of magnetization at 4.2 °K, even when cooling from room temperature, in contrast to the statement made in Ref. 15 (note that we observed only ⟨100⟩ lines at 4.2 °K in zero field from ^{57}Fe nuclei in domains of ErIG). Tcheou and co-workers[15] have interpreted their ErIG neutron-diffraction results on the basis of $\vec{M}$ being along ⟨111⟩ at 4.2 °K, an interpretation which must be incorrect. It is interesting that they find c-site moments of 6.7 μ_B and 4.4 μ_B: moments which agree well with those reported by Orlich and Hufner[21] for $\vec{M}$ along ⟨100⟩ [(6.9 ± 0.2) μ_B and (4.8 ± 0.2) μ_B], but not with the ⟨111⟩ moments [(5.3 ± 0.2) μ_B and (4.7 ± 0.2) μ_B].

VI. CONCLUSIONS

The anisotropy of the hyperfine field at a sites appears to be somewhat larger than can be explained by the dipolar effects alone in agreement with the findings of Myers and co-workers. The splitting at d sites appears to be closer to that which one would calculate from the dipolar effects alone. While the general trend of the isotropic hyperfine fields as a function of lattice constant can be explained on the basis of covalency, there is also some evidence for transferred hyperfine fields at Fe ions arising from rare-earth ions.

[1] R. L. Streever, Phys. Letters 29A, 710 (1969).
[2] R. L. Streever and P. J. Caplan, Phys. Letters 31A, 169 (1970).
[3] R. L. Streever and P. J. Caplan, Phys. Rev. Letters 24, 978 (1970).
[4] R. L. Streever and P. J. Caplan, Phys. Letters 33A, 413 (1970).
[5] R. L. Streever and P. J. Caplan, Phys. Rev. B 3, 2910 (1971).
[6] R. L. Streever and P. J. Caplan, Phys. Letters 35A, 8 (1971).
[7] R. L. Streever and P. J. Caplan, Bull. Am. Phys. Soc. 16, 628 (1971).
[8] C. Robert and F. Hartmann-Boutron, J. Phys. Radium 23, 574 (1962).
[9] R. Gonano, E. Hunt, and H. Meyer, Phys. Rev. 156, 521 (1967).
[10] R. Gonano, E. Hunt, H. Meyer, and A. B. Harris, J. Appl. Phys. 37, 1322 (1966).
[11] S. M. Myers, R. Gonano, and H. Meyer, Phys. Rev. 170, 513 (1968).
[12] S. M. Myers, J. P. Remeika, and H. Meyer, Phys. Rev. 170, 520 (1968).
[13] P. R. Locher and S. Geschwind, Phys. Rev. 139, A991 (1965).
[14] G. Burns, Phys. Rev. 124, 524 (1961).
[15] F. Tcheou, E. F. Bertaut, and H. Fuess, Solid State Commun. 8, 175 (1970).
[16] S. Geller, H. J. Williams, R. C. Sherwood, J. P. Remeika, and G. P. Espinosa, Phys. Rev. 131, 1080 (1963).
[17] S. Geller, J. P. Remeika, R. C. Sherwood, H. J. Williams, and G. P. Espinosa, Phys. Rev. 137, A1034 (1965).
[18] Airtron, Division of Litton Precision Products Inc., Morris Plains, N.J.
[19] G. P. Rodrique, H. Meyer, and R. V. Jones, J. Appl. Phys. 31, 3765 (1960).
[20] F. W. Harrison, J. F. A. Thompson, and G. K. Lang, J. Appl. Phys. 36, 1014 (1965).
[21] E. Orlich and S. Hufner, Z. Physik 232, 418 (1970).
[22] J. Atzmony, E. R. Bauminger, A. Mustachi, I. Nowik, S. Ofer, and M. Tassa, Phys. Rev. 179, 514 (1969).
[23] F. Bertaut and F. Forrat, Compt. Rend. 244, 96 (1957).
[24] M. Buyle-Bodin and Le Dang Khoi, J. Phys. Radium 23, 565 (1962).
[25] S. Ogawa and S. Morimoto, J. Phys. Soc. Japan 17, 654 (1962).
[26] R. L. Streever and G. A. Uriano, Phys. Rev. 139, A305 (1965).
[27] E. E. Anderson, in *Proceedings of the International Conference on Magnetism, Nottingham, England*, 1964 (The Institute of Physics and the Physical Society, London, 1965), p. 672.
[28] R. L. Streever, Phys. Rev. 186, 285 (1969).