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Magnetic relaxation in rare earth garnets

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Abstract. An attempt has been made to review the ferromagnetic resonance and relaxation data on rare earth iron garnets and in the light of it, to analyze the data on Sm and Eu thin films and Gd and Dy bulk polycrystalline samples obtained in our laboratory. A phenomenological approach adopted to explain the temperature and composition dependence of the line width and g_{eff} appears to explain the complex spectra in most cases.

Keywords. Magnetic relaxation; rare-earth garnets; ferromagnetic resonance.

1. Introduction

The ferromagnetic resonance line width in rare earth iron garnets has been extensively studied over almost three decades (De Gennes *et al* 1959; LeCraw *et al* 1963; Sinha and LeCraw 1971). Yet even today some aspects of the magnetic relaxation phenomena in these garnets have not been fully understood. For example, in SmIG the magnetic moment of Sm^{3+} ion is much smaller than what is expected on the basis of the effect of the presence of low lying excited states on the ground state and yet the ferromagnetic line width is large and shows a complex temperature dependence. Likewise, in EuIG three possible microscopic processes have been proposed to explain the presence of a low temperature peak in the ΔH vs T curve. The first is based on the resonant magnon excitation of the ground state ion (LeCraw *et al* 1963), second, on the crystal field and/or spin-orbit splitting of the excited state $J = 1$, (Huber 1964) and the third on the magnon-phonon (Sinha and LeCraw 1971) interaction. None of these theories is found to adequately explain the relaxation phenomena either in bulk EuIG or in thin EuYFeGaG films (Rao 1985). It is only in a system like YbIG with a relatively simple ground state of the rare earth ion that it is possible to explain the structure of the low temperature peak satisfactorily. Relaxation behaviour of rare earth (RE) garnets with ions of S-character like Y^{3+}, Gd^{3+}, La^{3+} and Lu^{3+} is, however, fairly well understood.

A survey of the room temperature line width for $4f^n$ ($0 \leqslant n \leqslant 14$) rare earth garnets ($RE_3Fe_5O_{12}$) shows that it can vary from a few Oersted for LuIG to 3000 Oe for HoIG. Also there are two peaks, one at f^5 (Sm^{3+}) and the other at f^{10} (Ho). The presence of these peaks is yet not fully understood. The $RE_3Fe_5O_{12}$ garnet phase is not formed when the rare earth sublattice is fully occupied by the $4f^n$ ion for $0 \leqslant n \leqslant 4$, so it is not possible to discuss the behaviour of ΔH as a function of the f electron concentration in this region. Some indication of the relaxation behaviour of these ions

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can be obtained from the manner the line width changes with small additions of rare earth ions in YIG. The observed line broadening due to the presence of small amounts of impurities in YIG has not been fully explained due mainly to lack of knowledge about the low lying excited state.

It is possible to explore the dominant mechanism for relaxation only if the ground and low lying excited states of the ion are well understood. The easiest way to obtain these is to study the magnetization and the gyromagnetic ratio. In the next sections, we briefly discuss these for some of the $RE_3Fe_5O_{12}$ garnets.

Recently a programme to study the relaxation phenomena in Sm- and Eu- thin films started in our laboratory when liquid phase epitaxy (LPE) facilities were established for the growth of these films (Mukhopadhyay and Rao 1984). Some results obtained in this study are presented in § 3.

A new technique for the measurement of ΔH_{eff} developed in our laboratory (Srivastava *et al* 1980) has been used to study the relaxation phenomena in Gd- and Dy-iron garnets. It is shown in § 5 that the FMR line width, ΔH, and ΔH_{eff} both depend on the concentration of rare earth ions in a similar manner.

Finally in § 6, we briefly analyse the existing data on the magnetic relaxation in rare earth iron garnets and conclude with some comments on the work remaining to be done in this field.

2. Magnetic structure and gyromagnetic ratio

In the garnet with composition $RE_3Fe_5O_{12}$ each unit cell contains eight formula units and the trivalent metal ions occupy three types of cation sites, 16 octahedral or *a* sites, 24 tetrahedral or *d* sites and 24 dodecahedral or *c* sites. The *a* and *d* sites are occupied by Fe^{3+} ions and *c* is occupied by RE^{3+} ions. According to Neel's theory of ferrimagnetism the magnetic moment on *a* and *d* sites are antiparallel and that of the *c* site is antiparallel to the resultant of the *a* and *d* sites. In the case of the non-spherical configuration of the rare earth ion on the *c*-sublattice, the exchange field occurs only on the spin. As shown in table 1, all experimental values of 0 K magneton number ($n_B = g_J J$) for the rare earth ions with $L \neq 0$ are lower than the calculated values implying that L is partially quenched by the crystal field or their moments are canted with respect to the magnetic axis or both. As La^{3+} through Pm^{3+} do not form the garnet structure with $RE_3Fe_5O_{12}$ composition, mixed compositions like $RE_xY_{3-x}Fe_5O_{12}$ have been attempted and it is found that Pr^{3+} and Nd^{3+} ions have n_B values of 1·6 and 1·25 respectively. The Sm^{3+} and Eu^{3+} ions have nearby excited states which influence their values for n_B through the second order Zeeman effect produced by the exchange field of the iron sublattice (Wolf and Van Vleck 1960; White 1964).

A three sublattice analysis of the magnetization and paramagnetic susceptibility of the heavier rare earth iron $RE_3Fe_5O_{12}$ garnets ($4f^n$, $7 \leqslant n \leqslant 13$) has been carried out by our group (Srivastava *et al* 1982) and the set of six exchange constants obtained from the analysis are given in table 2. It can be seen that $|J_{dc}|$ increases continuously as n increases from 7 to 13. A similar trend is observed also for $|J_{ac}|$. It is surprising that the values of $|J_{ac}|$ and $|J_{dc}|$ are one order of magnitude higher for Yb compared to all the rest i.e. $7 \leqslant n \leqslant 12$.

The Wangsness (1953) relation for the g_{eff} for the three sublattice is

$$g_{\text{eff}} = (M_d - M_a - M_c) \Big/ \left(\frac{M_d}{g_d} - \frac{M_a}{g_a} - \frac{M_c}{g_c} \right) = \frac{1}{e/2mc} \frac{M}{S}. \tag{1}$$

Table 1. Magneton number n_B for trivalent lanthanide group ion (room temperature)

Ion	Configuration ($4f^n$)	Ground level (free ion)	g_J	$n_B = g_J J$ (calc)	n_B (exp)
Ce^{3+}	1	$^2F_{5/2}$	6/7	2·143	—
Pr^{3+}	2	$^3H_4$	4/5	3·2	1·6
Nd^{3+}	3	$^4I_{9/2}$	8/11	3·27	1·25
Pm^{3+}	4	$^5I_4$	3/5	2·40	—
Sm^{3+}	5	$^6H_{5/2}$	2/7	0·71	0·14
Eu^{3+}	6	$^7F_0$	1	0	0·74
Gd^{3+}	7	$^8S_{7/2}$	2	7	7·00
Tb^{3+}	8	$^7F_6$	3/2	9	7·73
Dy^{3+}	9	$^6H_{15/2}$	4/3	10	7·30
Ho^{3+}	10	$^5I_8$	5/4	10	6·73
Er^{3+}	11	$^4I_{15/2}$	6/5	9·0	5·07
Tm^{3+}	12	$^3H_6$	7/6	7·0	2·07
Yb^{3+}	13	$^2F_{7/2}$	10/7	5.0	1·67
Lu^{3+}	14	$^1S_0$			

Table 2. Set of exchange constants (°K) in garnets $RE_3Fe_5O_{12}$

R	$-J_{aa}$	$-J_{ad}$	$-J_{dd}$	$-J_{ac}$	$-J_{dc}$	$-J_{cc}$
Y	6·45	30·40	12·05	0	0	0
Gd	6·45	30·40	12·05	0·60	1·8	0
Tb	6·45	30·40	12·05	1·12	2·0	0
Dy	6·45	30·40	12·05	2·97	2·97	−0·9
Ho	6·45	30·40	12·05	2·64	2·80	0
Er	6·45	30·40	12·05	2·70	3·40	0
Tm	6·45	30·40	12·05	2·64	3·96	0
Yb	6·45	30·40	12·05	21·00	16·1	−0·2

Since the exchange field acts on the spin, S_{RE}, while the equation of motion of the c-sublattice involves the total angular momentum J_{RE}, of the rare earth ion, we have

$$\frac{dJ_{RE}}{dt} = 2\mu_B S_{RE} \times H_{ex}$$

But $(g_J - 1)J_{RE} = S_{RE}$, so

$$\frac{dS_{RE}}{dt} = 2(g_J - 1)\mu_B (S_{RE} \times H_{ex}).$$

Hence, $$g_c = 2(g_J - 1) \tag{2}$$

This indicates that for the lighter rare earths ($1 \leqslant n < 7$) the effective value of the Landé g-factor for the c-sublattice is negative. We obtain from (1) and (2) for $|M_c| \ll |M_d - M_a|$ and $g_a = g_d = g_{Fe}$,

$$g_{eff} = g_{Fe}\left\{1 - \left[1 - \frac{g_{Fe}}{2(g_J - 1)}\right]\frac{M_c}{M_d - M_a}\right\} \tag{3}$$

In case we know the dependence of the sublattice magnetization on temperature we shall be able to obtain the variation of g_{eff} with temperature using (3). From (1) it shows that there can be two types of compensation points, (i) the M-compensation point, t_m, in which $M \to 0$ with S also vanishing and (ii) the S-compensation point, t_s, in which $M \neq 0$ but $S \to 0$. In (i) both g_{eff} and ΔH diverge at t_m while in (ii) g_{eff} diverges but ΔH does not increase at t_s (LeCraw *et al* 1965). It has been possible to prepare high g materials with g exceeding 30 (LeCraw *et al* 1975) using a YEuFeGa garnet which has an s-compensation point close to room temperature. Recently Borghese *et al* (1980) have extensively studied several compositions of $Eu_x Y_{3-x} Ga_y Fe_{5-y} O_{12}$ garnets and have found that t_s and t_m can be separated for y close to 1·2. Since the mobility of the cylindrical domains (Hagedorn 1974) in magnetic bubble materials is directly proportional to g_{eff}, the high g-materials have been an attractive choice to the bubble magnetic memory designers (Eschenfelder 1981; Winkler 1981). The high temperature coefficient of magnetization near the compensation point necessitates, however, use of special biasing circuits in bubble devices (Nielson 1976).

Since the early days of magnetic garnet research, single crystals of YIG have been used for microwave filter devices (Whicker and Young 1978) attempts have been made to reduce the material cost of the device through replacement of YIG single crystals with low line width polycrystalline calcium vanadium garnets but success has been limited.

3. FMR studies

FMR in thin garnet films and single crystals of rare earth garnets has been studied extensively due to their being excellent materials for testing the theoretical models of ferromagnetic relaxation (Sparks 1964). We discuss some of the results obtained in our laboratory on the resonance studies of thin films of Sm- and Eu-films and polycrystalline samples of Eu- and Dy-iron garnets.

3.1 *Eu substituted garnets*

The value of g_{eff} shows a significant departure from 2 for the Eu-film. This departure increases with the concentration of the Eu^{3+} ion. The low temperature value of g_{eff} for $Eu_3 Fe_5 O_{12}$ bulk crystal is 1·10 (LeCraw *et al* 1963). Also, for x varying from 0 to 1·2 in $Eu_x Y_{3-x} Ga_{1\cdot0} Fe_4 O_{12}$, g_{eff} varies from 2·0 to 1·0 at 300 K as shown in figure 1.

A number of attempts have been made to describe the temperature dependence of g_{eff} in $Eu_3 Fe_5 O_{12}$ bulk single crystals. According to Sekerka (1965)

$$g_{\text{eff}}(T) = 2[1 - \tfrac{3}{5} \times 0{\cdot}75 \times m_c(T)] \tag{4}$$

$$m_c(T) = M_{\text{Eu}}(T)/M_{\text{Eu}}(0) \tag{5}$$

where $M_{\text{Eu}}(T)$ and $M_{\text{Eu}}(0)$ are the magnetization of the c-sublattice at T and 0 K respectively. This expression has been modified by Sinha and LeCraw (1971) and they find

$$g_{\text{eff}}(T) = 2[1 - 0{\cdot}45\, Z(T)^{-1}] \tag{6}$$

where

$$Z(T) = [1 + 3\exp(-500/T)] \tag{7}$$

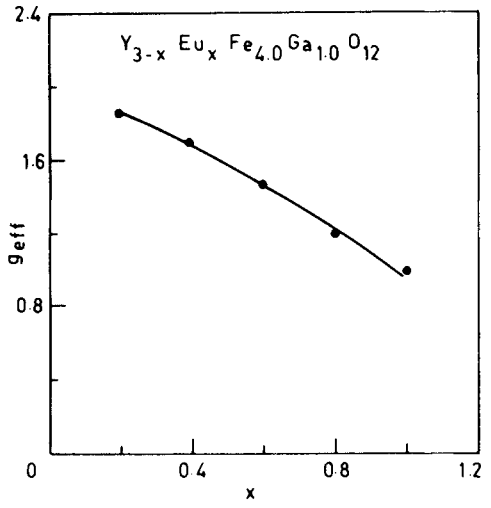

Figure 1. Variation of g_{eff} with Eu^{3+} ion concentration at 300 K.

Table 3. The values g_c and g_J obtained as a function of x in $Y_{3-x}Eu_xFe_{4.0}Ga_{1.0}O_{12}$

x	$-g_c$	g_J
0·2	0·46	0·77
0·4	0·46	0·77
0·6	0·46	0·77
0·8	0·31	0·85
1·0	0·25	0·88

Here, $Z(T)$ is the partition function truncated at the first excited level of Eu^{3+} ($E_{J=1} - E_{J=0} = 500$ K). Neither (4) nor (6) gives a microscopic description of the g-factor in EuIG.

From (3), it follows that for $Eu_3Fe_5O_{12}$ since $M_c(0) = 3 \times 0{\cdot}75\,\mu_B$ and $M_d(0) - M_a(0) = 5\,\mu_B$, we have

$$g_{\text{eff}} = 2\{1 - [(g_J - 2)/(g_J - 1)]\tfrac{3}{5} \times 0{\cdot}75\} \tag{8}$$

On comparing (4) with (8), we find that $(g_J - 2)/(g_J - 1)$ varies as $m_c(T)$. Since g_J is a complex function of the ground state and the first excited state of Eu^{3+} this cannot be easily explained.

The values of g_J for the composition $Eu_xY_{3-x}Fe_{4.0}Ga_{1.0}O_{12}$ ($0{\cdot}2 < x < 1{\cdot}2$) are given in table 3. The dependence of g_{eff} on temperature for $x = 0{\cdot}6$ has been compared with theory using (3) and is shown in figure 2. Here, $g_J = 0{\cdot}77$ and is taken as constant. The agreement is good.

3.2 *Sm substituted garnets*

The g_{eff} in $Sm_xY_{3-x}Fe_{5-y}Ga_yO_{12}$ ($x = 0{\cdot}4$ and $0{\cdot}46$, $y = 1$) has been measured as a function of concentration of Sm and temperature. The g_{eff} in this system shows a very

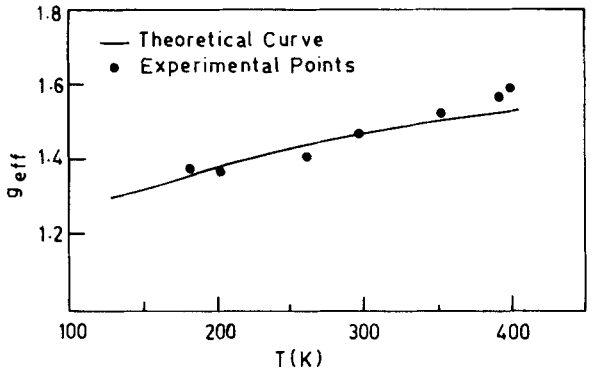

Figure 2. Variation of g_{eff} with temperature for $Eu_{0\cdot6}Y_{2\cdot4}Fe_{4\cdot0}Ga_{1\cdot0}O_{12}$.

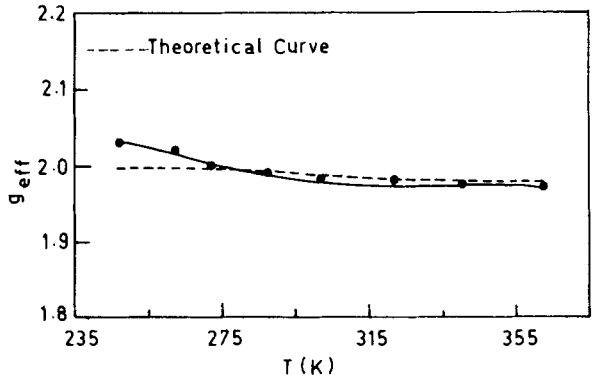

Figure 3. Variation of g_{eff} with temperature for $Sm_{0\cdot4}Y_{2\cdot6}Fe_{4\cdot0}Ga_{1\cdot0}O_{12}$

weak but interesting temperature dependence in the range 230 to 370 K. Using (3) and $g_J = 2/7$ we obtain

$$g_{eff} = g_{Fe}[1 \pm (12/5)M_c/(M_d - M_a)] \tag{9}$$

where the $\pm$ sign arises on account of the crossover temperature, T_{CO}, which lies close to room temperature. In estimating M_c, we take the Sm^{3+} moment to be 0·1 μ_B near T_{CO}. In that case (Rao 1985) we obtain

$$g_{eff} = 1{\cdot}985[1 + 2{\cdot}57 \times 10^{-2} \times (312{\cdot}5 - T)/T] \tag{10}$$

A comparison of the experiment with (10) is shown in figure 3. The agreement is good.

3.3 *Gd substituted garnets*

Polycrystalline samples of $Gd_xY_{3-x}Fe_5O_{12}$ ($0 \leqslant x \leqslant 1{\cdot}5$) have been prepared and their g_{eff} have been measured at 300 K. The g_{eff} is both independent of x as well as T and has a constant value of 2 since Gd^{3+} is an S-state ion.

3.4 *Dy substituted garnets*

In $Dy_xY_{3-x}Fe_5O_{12}$ the magnetic moment of the Dy^{3+} ion is expected to be 10 μ_B at 0 K. The observed value is 7·3 μ_B. This indicates reasonable amount of orbital angular momentum quenching due to crystal field and exchange interactions. Due to the large value of the magnetic moment arising from the orbital angular momentum, Dy^{3+} is a high relaxing ion. In this case g_{eff} has to be calculated with emphasis more on damping. Using the three sublattice model (Rao 1983) as an extension of Kittel's treatment (1959) based on the two sublattice we obtain

$$\gamma_{\text{eff}} = \gamma_d[1 - M_c/(M_d - M_a)]\{1 + (n\tau)^2\gamma_c^2(M_d - M_a)^2 \times [1 - (\gamma_d/\gamma_c)M_c/(M_d - M_a)]\}\{1 + (n\tau)^2\gamma_c^2(M_d - M_a)^2 \times [1 - (\gamma_d/\gamma_c)M_c/(M_d - M_a)]^2\}^{-1} \tag{11}$$

$$\text{where } n = (n_{ac}M_a + n_{dc}M_d)/(M_a + M_d) \tag{12a}$$

$$\tau = 1/[\lambda_c(M_a - M_d)M_c n] \tag{12b}$$

Here M_i and γ_i $(i = a, c, d)$ are the sublattice magnetizations and gyromagnetic ratios of the sublattice i, τ is the relaxation time on the c sublattice and n_{ij} $(i, j = a, c, d)$ are the molecular field constants and λ_c is the Landau Lifshitz damping parameter for the c sublattice. n_{ij} are related to J_{ij} through the equation

$$n_{ij} = (2z_{ij}(g_j - 1)J_{ij})/(N_j g_i g_j \mu_B^2) \tag{13}$$

where z_{ij} is the number of nearest neighbour j sublattice ions for an ion on the ith sublattice, N_j the number of ions per mole on the jth site and g_i the g factor for the ion on the ith site.

The values of g_{eff} for $Dy_xY_{3-x}Fe_5O_{12}$ $(0{\cdot}3 < x < 2{\cdot}1)$ have been taken on polycrystalline bulk samples at 300 K and are shown as a function of x in figure 4. The data is compared with theory using (11) and the values of M_a, M_c and M_d using the exchange constants are given in table 2. The value of τ has been taken as 10^{-12} sec. The agreement of experiment with theory is good.

In figure 5 is plotted the variation of g_{eff} with temperature for $Dy_{0\cdot3}Y_{2\cdot7}Fe_5O_{12}$ using (11) and the experimental data obtained by Rao (1983). The agreement is satisfactory.

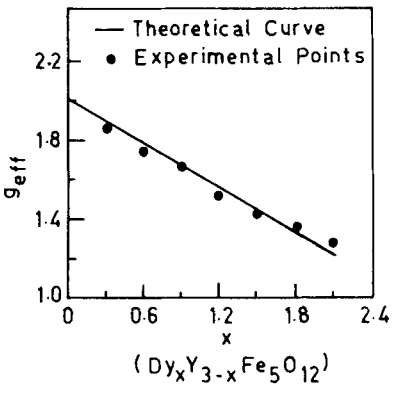

Figure 4. g_{eff} as a function of concentration of Dy^{3+} ions in $Dy_xY_{3-x}Fe_5O_{12}$.

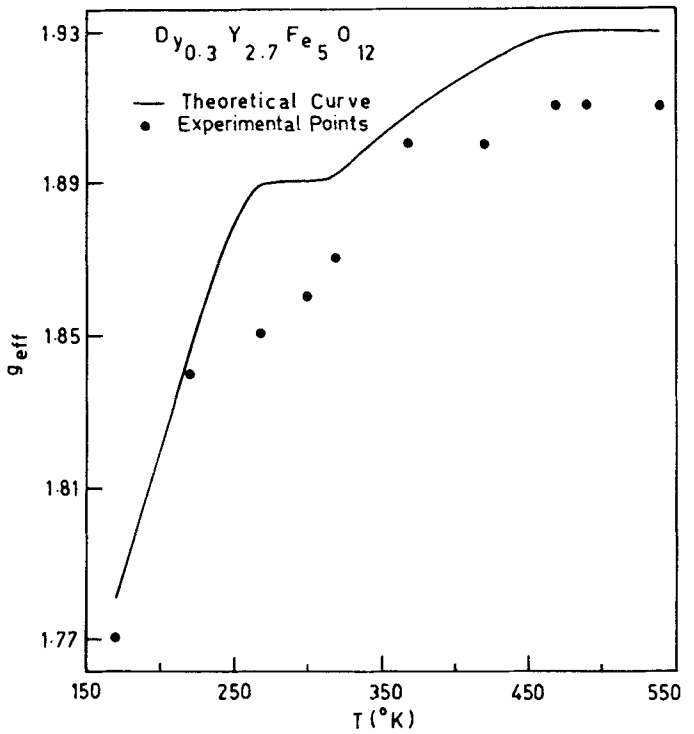

Figure 5. Variation of g_{eff} with temperature in $Dy_{0.3}Y_{2.7}Fe_5O_{12}$.

4. Relaxation in rare earth iron garnets

The line width in most rare earth iron garnets $RE_3Fe_5O_{12}$ often shows a low temperature peak lying in the range 30 to 250 K. Beyond this the FMR line width, ΔH, decreases with temperature till the Curie temperature is reached when ΔH begins to increase with increase in temperature.

The existence of the low temperature peak is attributed to the presence of low lying excited states in the energy levels of the inequivalent rare earth ions in iron garnets. The energy levels of the two inequivalent sites obtained using specific heat data for Sm-, Tb-, Dy- and Ho-iron garnets have been obtained by Harris and Meyer (1962).

The effect of the low lying excited state on magnetization has been discussed in great detail for EuIG by Wolf and Van Vleck (1960) and for YbIG by Teale and Tweedle (1968) and Clarke *et al* (1965). For YbIG the theory gives a fairly good description of the microscopic processes which are responsible for the low temperature peak.

In figure 6 is shown the room temperature FMR line width as a function of n, the number of $4f$ electrons in RE for $RE_3Fe_5O_{12}$ with n varying from 5 to 12. Since in $Gd_3Fe_5O_{12}$ the compensation point occurs at room temperature the data for $n = 7$ has been taken at $T = 200$ K. For $n \leqslant 4$ the garnets are not formed on account of the large size of the rare earth ions and hence the data is not available. We discuss the origin of the low temperature peak in $Eu_3Fe_5O_{12}$ in the next section. In other garnets the mechanism for the occurrence of this peak is similar and will not be discussed due to limitation of space.

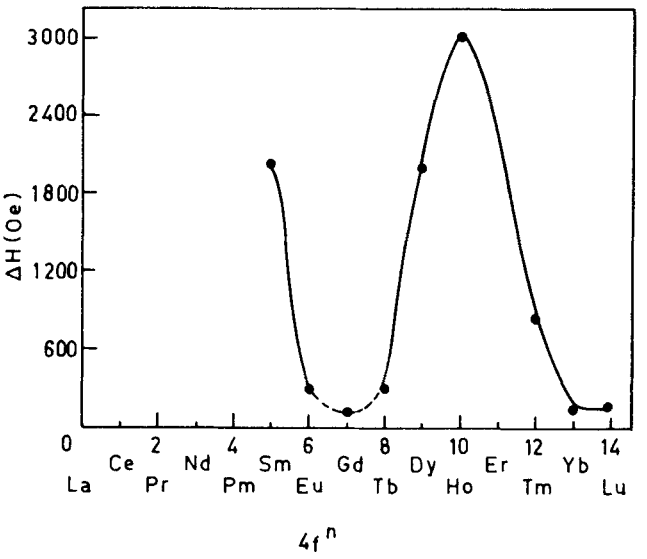

Figure 6. Variation of room temperature FMR line-width with the number of $4f$ electrons in $RE_3Fe_5O_{12}$. Since for $n = 7$, the compensation point lies at room temperature, the data for GdIG is taken at 200 K.

4.1 *Low temperature peak in ΔH vs T curve for EuIG*

The complex behaviour of the dependence of the resonance line width, ΔH, on temperature for $Eu_3Fe_5O_{12}$ has been generally explained using two different basic assumptions, the fast relaxation process ($\tau_{RE} \ll \omega^{-1}$) and the slow relaxation process ($\tau_{RE} \sim \omega^{-1}$). The former has been discussed by De Gennes *et al* (1959) and the latter by Galt (1954), Clogston (1955), Teale and Tweedle (1962), Van Vleck and Orbach (1963) and Hartmann-Boutron (1963). It has been concluded by LeCraw *et al* (1963) that the relaxation in EuIG can be described by the slow relaxation process. According to them the excited state $J = 1$ close to ground state $J = 0$, is assumed to be split into two states by $\hbar\omega_{\parallel}$ on account of exchange and crystal field effects. The energy separation E_1 between $J = 1$ and $J = 0$ states is large and equal to 480 K while $\hbar\omega_{\parallel}$ is small and is of the order of 50 K. For this case assuming excitation of ions from $J = 0$ to $J = 1$ states by iron lattice magnons resonant to E_1 the line width given by LeCraw *et al* (1963) is

$$\Delta H = \frac{C\hbar\omega_{\parallel}^2}{\gamma k_B T}\phi\,\omega\tau\,\frac{\exp(-E_1/k_BT)}{1+3\exp(-E_1/k_BT)} \tag{14}$$

Here C is the atomic ratio of Eu to Fe i.e. 3/5 for EuIG, ϕ is the dimensionless constant arising from the angular dependence and vanishes for all angles only if the state $J = 1$ is unsplit by the crystal field and if, in addition, the exchange is isotropic.

In (14) the relaxation time τ is temperature dependent and is given by

$$\tau = \tau_0 \tanh(E_i/2k_BT) \tag{15}$$

If the relaxation is by the resonant magnon mode $E_i = E_1$ while for the spin-orbit relaxation between the $J = 1$ manifold, $E_i = \hbar\omega_{\parallel}$ (Huber 1964). The possibility of both

processes coexisting is large. In the two cases it has been shown that the peak in ΔH can shift by 20 to 30 K (Rao 1985).

A new phonon-magnon interaction mechanism which arises from a microscopic formulation of the exchange modulation by crystal field oscillations has been discussed by Sinha and LeCraw (1971). The processes considered involve two magnons, one of the acoustical mode and the other of the exchange (optical) mode and one phonon of the longitudinal optical mode. The expression for the line width has the right temperature and frequency dependence.

None of the three microscopic processes explain the data on thin films in which Fe is partially replaced by Ga. We have observed the variation of $\Delta H_\perp$ and $\Delta H_\parallel$ in $Eu_xY_{3-x}Fe_{5-y}Ga_yO_{12}$ thin films for ($0{\cdot}2 \leqslant x \leqslant 0{\cdot}8$, $y = 1{\cdot}0$) with temperature in the range 100 to 400 K. The films $x = 0{\cdot}2$ to $0{\cdot}6$ show generally two peaks whose position depends on the composition. For $x = 0{\cdot}8$, no peak exists. The data for $x = 0{\cdot}4$ are shown in figure 7 for $\Delta H_\perp$. It has been proposed that since the exchange field is considerably weakened with Ga substitution, the exchange modulation of the excited state manifold ($J_z = 0, \pm 1$) may be the dominant mechanism for relaxation. In that case

$$\Delta H = \frac{C\hbar\omega_{ex}^2}{\gamma k_B T}\phi\omega\tau_0 \tanh\frac{\hbar\omega_{ex}}{k_B T}\frac{\exp(-E_1/k_B T)}{1+3\exp(-E_1/k_B T)} \tag{16}$$

with $$\omega_{ex} = \omega_{ex}(0)m_{Fe}(T) \tag{17}$$

Here $\omega_{ex}(0)$ is the exchange frequency at 0 K and $m_{Fe}(T)$ is the reduced magnetization of the iron sublattice i.e., $m_{Fe}(T) = m_d(T) - m_a(T)$. In the present case $\omega_{ex}(0) = 2{\cdot}4k_B/\hbar$. In that case (16) yields

$$\Delta H = \frac{A}{T}m_{Fe}^2(T)\tanh\frac{1{\cdot}2m(T)}{T}\frac{\exp(-480/T)}{1+3\exp(-480/T)} \tag{18}$$

A comparison of (18) with the experimental data on the Eu film with $x = 0{\cdot}4$ is given in figure 7. The agreement is satisfactory.

In conclusion it can be said that in EuIG and Eu based thin films there may exist more than one mechanism for relaxation which can account for the observed dependence of ΔH on ω and T.

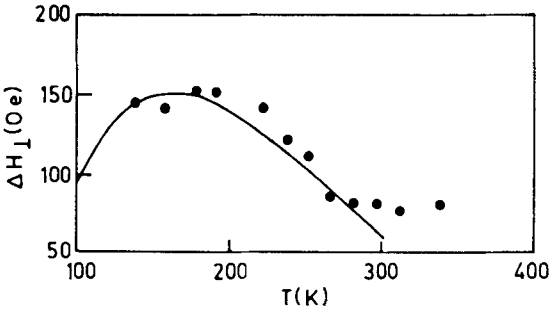

Figure 7. Variation of ΔH with temperature for $Eu_{0{\cdot}4}Y_{2{\cdot}6}Fe_{4{\cdot}0}Ga_{1{\cdot}0}O_{12}$ film.

4.2 *Three sublattice model for relaxation*

It is often convenient to describe the temperature dependence of ΔH in garnets on a phenomenological model first proposed by Kittel (1959) using a two sublattice structure and later extended by Rao (1983) to a three sublattice model. The expression for the three sublattice model is

$$\Delta H = \frac{\omega_0 \tau n M_c [M_d - M_a]}{M_d - M_a - M_c} \frac{1 + (n\tau)^2 [\gamma_c (M_d - M_a) - \gamma_d M_c]^2}{1 + n^2 \tau^2 \gamma_c [(M_d - M_a)][\gamma_c (M_d - M_a) - \gamma_d M_c]} \tag{19}$$

Here $\omega_0 = \gamma_{\text{eff}} H_0$ and other symbols are as described in (11).

We have used the sublattices magnetization obtained for $Dy_{0.3}Y_{2.7}Fe_5O_{12}$ by Aiyar (1982) to calculate the ΔH as a function of temperature using $\tau = 10^{-12}$ sec in (19) and values of J_{ij} given in table 2. The experimental data has been obtained from Rao (1983) and compared with theory in figure 8. The agreement is good.

Rao (1983) has shown that this approach gives good fit with theory for $Dy_xY_{3-x}Fe_5O_{12}$ with $x = 0{\cdot}6$, $0{\cdot}9$, $1{\cdot}2$, $1{\cdot}5$ and $1{\cdot}8$ also.

4.3 *Dependence of ΔH on composition and temperature*

The explicit dependence of ΔH on the concentration of the rare earth ion in the c-sublattice in $RE_xY_{3-x}Fe_5O_{12}$ or $RE_xY_{3-x}Fe_{5-y}Ga_yO_{12}$ thin films cannot be

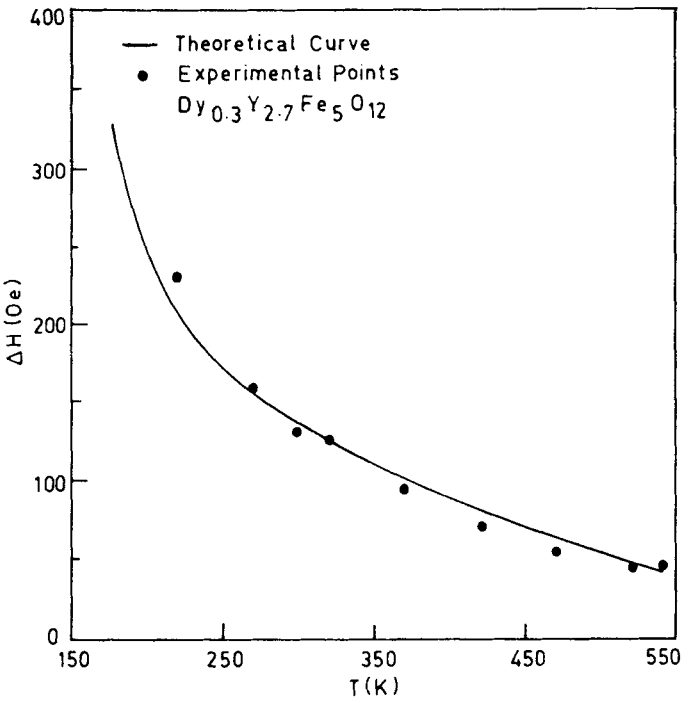

Figure 8. Variation of ΔH with temperature in $Dy_{0.3}Y_{2.7}Fe_5O_{12}$.

obtained from (19). We have developed a relation between ΔH and x (Srivastava 1984)

$$\Delta H = A + Bx + Cx^2 \tag{20}$$

where A, B, C are constants depending on temperature and are given by

$$\Delta H = \xi_\Lambda (T/T_c)^p + \eta_\Lambda [1 - (T/T_c)^p] \tanh(\hbar\omega/k_B T) \times \frac{N_r}{N_0} \frac{\exp(-\hbar\omega/k_B T)}{1 + \exp(-\hbar\omega/k_B T)} + \zeta_\Lambda (T - \theta)^n \tag{21}$$

Here $\Lambda = A$, B, C and ξ_Λ, η_Λ and ζ_Λ are constants which can be determined from experiment.

In figure 9 is plotted the variation of ΔH for 300 K as a function of the concentration of Eu ions in $Eu_x Y_{3-x} Fe_{4\cdot0} Ga_{1\cdot0} O_{12}$ thin films. The theoretical curve is based on (20). In figure 10 is shown the temperature dependence of ΔH for $Sm_{0\cdot4} Y_{2\cdot6} Fe_{4\cdot0} Ga_{1\cdot0} O_{12}$ thin films. The theoretical curve is plotted using (21).

In figures (11) and (12) we have plotted the variation of ΔH with x in $Dy_x Y_{3-x} Fe_5 O_{12}$ and $Gd_x Y_{3-x} Fe_5 O_{12}$ respectively and compared them with (20). The agreement is satisfactory.

5. Effective line width

The effective line width, ΔH_{eff}, outside the magnon manifold in $RE_x Y_{3-x} Fe_5 O_{12}$ should have a similar dependence on x as in (20) because in this region the dominant

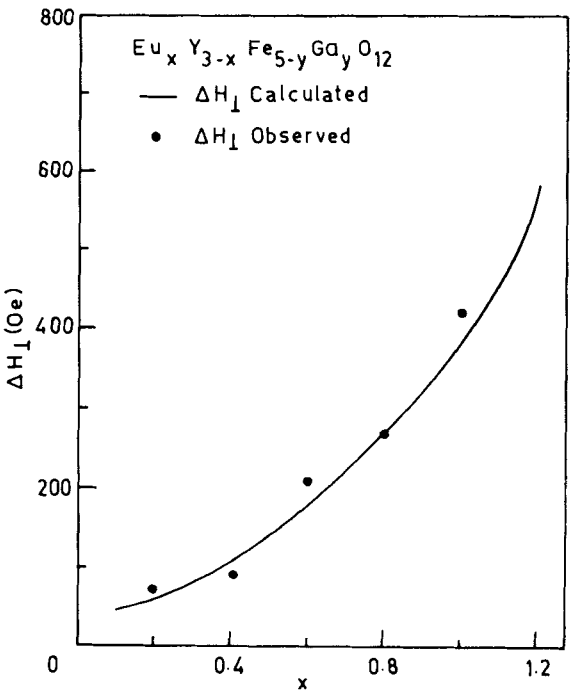

Figure 9. Variation of ΔH with Eu ion concentration.

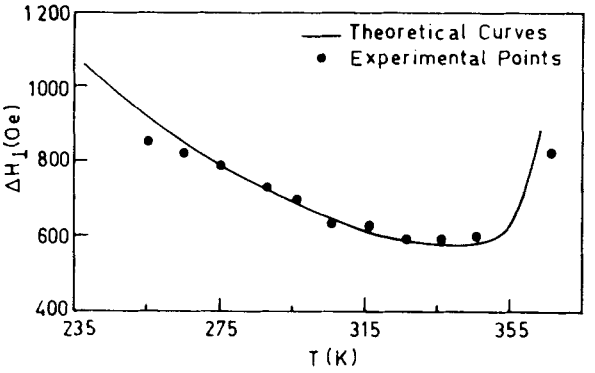

Figure 10. Variation of ΔH with temperature for $Sm_{0.4}Y_{2.6}Fe_{4.0}Ga_{1.0}O_{12}$.

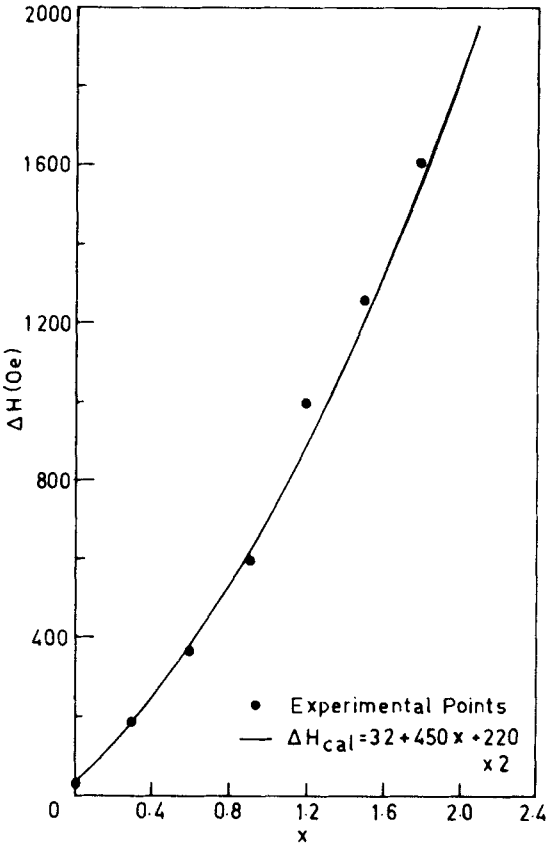

Figure 11. ΔH as a function of x in $Dy_xY_{3-x}Fe_5O_{12}$.

relaxation arises from the scattering of the uniform precession made by the spin-orbit interaction. This is indeed found for Dy- and Gd-substituted iron garnets. In figure 13 the experimental data is obtained using the technique developed by Srivastava *et al* (1980). The fit to (20) is satisfactory.

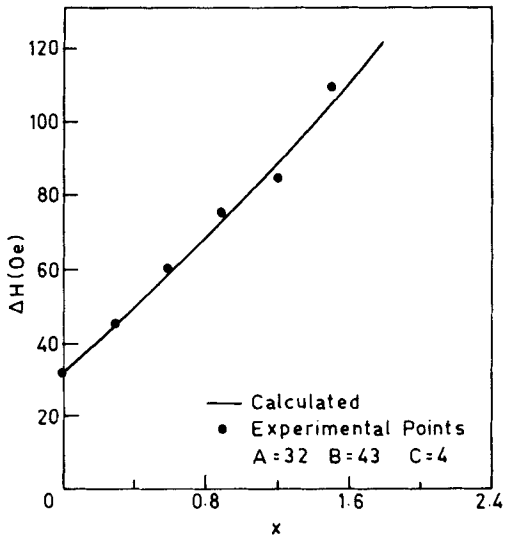

Figure 12. ΔH as a function of x in $Gd_xY_{3-x}Fe_5O_{12}$.

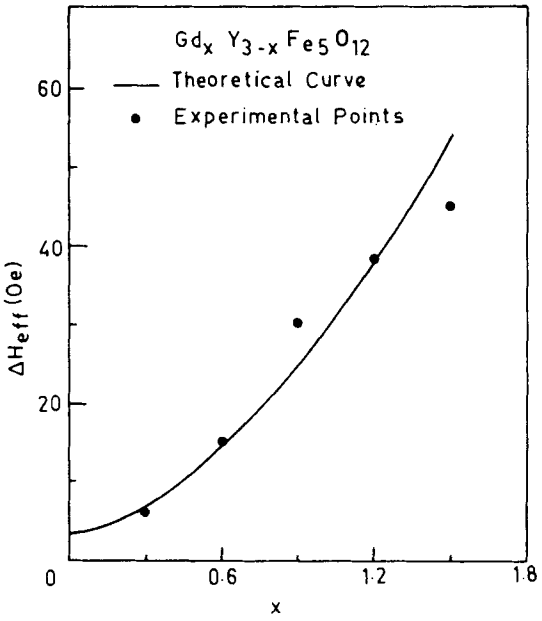

Figure 13. Variation of ΔH_{eff} in $Gd_xY_{3-x}Fe_5O_{12}$ as a function of x.

6. Relaxation and the low lying excited states

It is now fairly clear that relaxation in rare earth iron garnets is intimately connected to the presence of low lying excited states. The maximum in ΔH at room temperature is obtained for HoIG and for this the ground has the highest value of J. Similarly Sm^{3+} has the highest value of J amongst the lighter rare earth ions for which the $RE_3Fe_5O_{12}$ garnet phase is possible. Our group is now attempting to obtain a quantitative

relationship between the relaxation frequency and the structure of the low lying states of the rare earth ions.

7. Conclusion

The complex phenomena of resonance and relaxation in rare earth iron garnets has been reviewed with a view to establish correlation between the temperature frequency dependence of the FMR line width and the ground state configuration of the rare earth ion. A phenomenological approach to describe the dependence of line width using three sublattice model has been shown to give good agreement for Dy substituted YIG garnet. Another attempt to explain the relaxation on the basis of coexistence of two channels of relaxation, one a collective mode and the other an independent spin mode, gives good agreement for thin films as well as bulk polycrystalline samples of mixed garnets.

References

Aiyar R 1982 *Studies on exchange constants and magnetic relaxation in ferrimagnetic garnets*, Ph. D. thesis, Indian Institute of Technology, Bombay

Borghese C, Cosmi R, De Gosperis P and Tappa R 1980 *Phys. Rev.* **B21** 183

Clarke B H, Tweedle K and Teale R W 1965 *Phys. Rev.* **A139** 1933

Clogston A M 1955 *Bell Syst. Tech. J.* **34** 739

De Gennes P G, Kittel C and Portis A H 1959 *Phys. Rev.* **11** 323

Eschenfelder A H 1981 *Magnetic bubble technology* (Berlin, Heidelberg, New York: Springer-Verlag)

Galt G K 1954 *Bell Syst. Tech. J.* **33** 1023

Hagedorn F B 1974 *AIP Conf. Proc.* **18** 222

Harris A B and Meyer H 1962 *Phys. Rev.* **127** 101

Hartmann-Boutron F 1963 *Compt. Rend.* **256** 4412

Huber D L 1964 *Phys. Rev.* **A136** 500

Kittel C 1959 *Phys. Rev.* **115** 1587

LeCraw R C, Nielsen W G, Remeika J P and Van Vleck J H 1963 *Phys. Rev. Lett.* **11** 490

LeCraw R C, Remeika J P and Mathews H W 1965 *J. Appl. Phys.* **36** 901

LeCraw R C, Blank S L and Vella Coleiro G P 1975 *Appl. Phys. Lett.* **26** 722

Mukhopadhyay P and Rao B U M 1984 *Bull. Mater. Sci.* **6** 47

Nielsen J W 1976 *IEEE Trans. Magn.* **MAG-12** 327

Rao N S H 1983 *Studies of microwave magnetic relaxation processes in rare earth substituted garnets using a new technique*, Ph. D. thesis, Indian Institute of Technology, Bombay

Rao B U M 1985 *Ferromagnetic relaxation studies in Sm- and Eu-based bubble garnet films*, Ph. D. thesis, Indian Institute of Technology, Bombay

Sekerka R F 1965 Doctoral Thesis, Harvard

Sinha K P and LeCraw R C 1971 *J. Phys. Chem. Solids* **32** 373

Sparks M 1964 *Ferromagnetic relaxation theory* (New York: McGraw Hill)

Srivastava C M, Patni M J and Hanumantha Rao N S 1980 Proc. Int. Conf. Ferrites, ICF-3, Kyoto (eds) H Watanabe, S Iida and M Sugimoto (Japan: Centre for Academic Publications) p. 824

Srivastava C M, Srinivasan C and Aiyar R 1982 *J. Appl. Phys.* **53** 781

Srivastava C M 1984 Proc. Int. Conf. Ferrites, ICF-4, San Francisco (to be published) *Adv. Ceramics* **15** (Ohio: American Ceramic Society)

Teale R W and Tweedle K 1968 *Phys. Lett.* **1** 298

Van Vleck J H and Orbach R 1963 *Phys. Rev. Lett.* **11** 65

Wangsness R K 1953 *Phys. Rev.* **91** 1085

Whicker L R and Young C W 1978 *Microwave J.* **21** (11) 33

White J A 1964 *Rare earth research II* (ed) K S Vorres (New York and London: Academic Press) p. 407

Winkler G 1981 *Magnetic garnets* (Braunschwaig: Friedr Vieweg and Sohn)

Wolf W P and Van Vleck J H 1960 *Phys. Rev.* **118** 1490