Exchange resonances in gadolinium iron garnet at 24.000 MHz
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Résumé. — Une étude de la résonance ferrimagnétique est faite sur des monocrustaux de garnet de gadolinium (5Fe₂O₃.3Gd₂O₃) à 24,000 Mc au voisinage de la température de compensation T₀ = +13°C. Les deux modes de résonance correspondant aux deux sens de précession des sous-réseaux de Fe et Gd couplés sont observés et les résultats comparés avec une théorie qui tient compte de la susceptibilité du Gd et de l'anisotropie cristalline. On déduit de ces expériences que le champ d'échange à T₀ est 231,000 Oe, en bon accord avec la valeur déduite de la courbe d'aimantation du matériau. De plus, on obtient les valeurs numériques de (gFe — gGd) et de l'anisotropie cristalline. Pour des températures voisines de T₀ à un degré près, on observe d'autres modes de résonance. Chacun de ces modes correspond au fait que les sous-réseaux de Fe et Gd sont alignés le long d'un axe (111) du cristal qui n’est pas parallèle au champ magnétique appliqué.

Abstract. — Ferrimagnetic resonance has been experimentally studied in single crystals of gadolinium iron garnet at 24,000 Mc in the vicinity of the magnetic compensation point at T₀ = +13°C. The two modes of resonance corresponding to the normal and abnormal directions of precession of the coupled Fe and Gd sublattices were observed and the results compared with a theory which includes the effect of the susceptibility of the Gd. From these measurements, the exchange field at T₀ was found to be 231,000 Oe, in excellent agreement with magnetization data. In addition, information on (gFe — gGd) and the crystalline anisotropy were obtained. In a one degree region around the compensation temperature additional modes of resonance were observed. Each of these resonances correspond to the Fe and Gd sublattices aligned along a (111) direction which is not parallel to the external magnetic field.

Introduction. — The rare earth iron garnets are, strictly, systems with three sublattices: [1], [2] the antiferromagnetic coupling between the rare earth ions and the iron sublattices is, however, much weaker than that between the latter. For resonance experiments in the applied fields normally encountered in the laboratory, it is sufficient, then, to treat the two iron sublattices as one. Several authors [3], [4] have given the theory of ferrimagnetic resonance in systems with two sublattices. So long as λM is large compared to the applied magnetic field and the anisotropy fields, λ being the molecular field constant and M the net magnetization, it is shown that there are two well separated resonant modes. One involves a flexing of the bond between the lattices and is found in normal fields at a high frequency of order γλM, γ being a typical gyromagnetic ratio. The other shows no flexing, the system behaves as a single lattice with an effective γ value given by the ratio of net magnetization to net angular momentum.

Close to magnetic compensation the earlier analyses predict that the two modes will approach one another in field (or frequency). Specifically, the fields for resonance predicted are

\[ H = -\frac{\omega}{\gamma} \pm \sqrt{\lambda M_0 \left[ \left( \frac{\omega}{\gamma_1} - \frac{\omega}{\gamma_2} \right) + H_a \right]} \]  

(1)

where \( M_0 \) is the magnetization of either sublattice, \( H_a \) is an anisotropy field and \( \gamma_1, \gamma_2 \) are the \( \gamma \)'s of the two sublattices. An important effect in compensated systems which is not taken into account in the above formula is the susceptibility of the sublattices. This causes the applied field to enter the expressions for the sublattice magnetization and considerably alters the form of the field for resonance near compensation as will be shown.

A study has been made of ferrimagnetic resonance in the Gd-Fe garnet system in the immediate neighborhood of the magnetic compensation at 13.1°C, where both resonant modes are accessible at microwave frequencies, and the results compared with a theory which includes the effect of susceptibility. Accurate information has been obtained in this way about the exchange field, the difference of \( \gamma \)'s and the anisotropy fields. Previous work on this system [5], [6] did not cover this region, where such information is apparently best obtained, presumably because of limitations upon the sensitivity of the apparatus used.

Theory. — If the d.c. magnetic field is applied along an easy (111) or hard (100) direction of magnetization, and the sublattices are lined up along the applied field, the r.f. magnetization is circularly polarized and the relation between the field, \( H_o \), for resonance and the angular frequency, \( \omega \), becomes relatively simple. It may be written in the form:

\[ \frac{\lambda M_{Fe}}{H_o + \omega \gamma_{Fe} + H_{Fe}} + \frac{\lambda M_{Gd}}{H_o + \omega \gamma_{Gd} + H_{Gd}} = 1 \]  

(2)

where \( H_o \) is always taken positive; \( M_{Fe}, M_{Gd} \),...
$H_{Gd}$, $H_{Fe}$ are, respectively, the sublattice magnetizations and the anisotropy fields with signs appropriate to the assumption of a positive $H_c$. $\omega/\gamma$ is taken to be negative for the normal polarization. $\gamma_{Fe}$ and $\gamma_{Gd}$ are the $\gamma$'s of the two sublattices. The magnetizations, $M_{Fe}$ and $M_{Gd}$, will be functions of the temperature and of the applied magnetic field, which, in principle, could be found by using the molecular field theory and the observed magnetization data. $H_0$ could be found numerically in this way as a function of $\omega$ and $T$. Since we are concerned only with the region, $T \sim T_c$, we make a number of valid approximations in order to obtain the simplest expression for the resonant field which is quantitatively accurate. Defining $M_0$ as the common magnitude of $M_{Fe}$ and $M_{Gd}$ at $T = T_c$, in zero applied field, we write, ignoring the susceptibility of the iron sublattice,

$$M_{Fe} = \pm M_0 \pm \alpha (T - T_c)$$

where, here and elsewhere, the $+$ sign is taken if the iron sublattice points along $H_0$, the $-$ sign if the gadolinium sublattice does so. If the gadolinium magnetization is a function of $1/T \times$ (the total field on the sublattice) we have, to the first order in $(T - T_c)$ and in $H_0$,

$$M_{Gd} = \pm M_0 \pm \chi_{Gd} \alpha [\pm (M_0/T_c) (T - T_c) + \chi_{Gd} H_0]$$

where $\chi_{Gd}$ is the differential susceptibility of the gadolinium at compensation. Since, at compensation, in zero applied field,$

$$M_{Gd} = \lambda M_{Fe} \chi = \lambda M_{Gd} \chi$$

we have

$$\lambda \chi = 1$$

Experimentally $\chi$ and $\chi_{Gd}$ differ by only a few per cent at compensation, we, therefore, set $\chi_{Gd} = 1/\lambda$. It should be noted that, although the susceptibility, $\chi_{Gd}$, has been taken into account, it will not appear explicitly in the final result.

$H_0$ is now given by:

$$H_0 = \left[ 1 + \frac{\lambda M_0 (T - T_c)}{T_c (\omega/\gamma_{Fe} + H_{Fe})} \right]^{-1}$$

$$- \frac{\omega}{\gamma_{Gd}} + H_{Gd} \pm \lambda M_0 \left[ \frac{\omega}{\gamma_{Fe}} - \frac{1}{\gamma_{Gd}} + H_{Fe} + H_{Gd} \right]$$

$$\pm \lambda (T - T_c) \left[ \alpha - \left( \alpha - \frac{M_0}{T_c} \frac{\omega}{\gamma_{Fe}} + H_{Fe} \right) \right]$$

It is to be remarked that $H_{Fe} \pm H_{Gd}$, $H_{Gd} = \mp H_{Fe}$, where $H_{Fe}$ and $H_{Gd}$ are the "anisotropy fields", in the usual sense of the term, measured in the direction of the relevant sublattice. Without serious error we may write

$$\frac{1}{\omega/\gamma_{Fe} + H_{Fe}} \sim \frac{\gamma_{Fe}}{\omega}$$

and, in the present case, where we anticipate that

$$\gamma_{Gd} \approx \gamma_{Fe},$$

we put

$$\frac{\omega/\gamma_{Gd} H_{Fe}}{\omega/\gamma_{Fe} + H_{Fe}} = 1.$$ (7)

Finally, then,

$$H_0^+ = \frac{H^+(\omega)}{1 - \frac{\lambda M_{Fe}}{T_c \omega} (T - T_c)}$$

for the Fe lattice in the direction of $H_0$ and

$$H_0^- = \frac{H^-(\omega)}{1 + \frac{\lambda M_{Fe}}{T_c \omega} (T - T_c)}$$

for the Gd lattice in the direction of $H_0$ where

$$H^+(\omega) = -H^-(\omega) = -\omega/\gamma_{Gd} + \lambda M_0 \left[ 1 - \frac{\gamma_{Fe}}{\gamma_{Gd}} \right]$$

$$+ \lambda M_0 \frac{\gamma_{Fe}}{\omega} (H_{Fe} + H_{Gd})$$

$$H^-(\omega) = -H^+(\omega) = -\omega/\gamma_{Fe} - \lambda M_0 \left[ 1 - \frac{\gamma_{Fe}}{\gamma_{Gd}} \right]$$

$$+ \lambda M_0 \frac{\gamma_{Fe}}{\omega} (H_{Fe} + H_{Gd}).$$

In the particular case of GdIG treated here the above expressions are valid to within a few per cent in a ten degree range on either side of compensation.

**Description of apparatus.** — As a result of the small value of the net magnetization in the region

- Electron spin resonance spectrometer using a balanced microwave bridge and superheterodyne detection.
of the compensation point, the absorptions are several orders of magnitude smaller than those usually encountered in ferromagnetic resonance. This made necessary the use of a fairly sensitive electron spin resonance apparatus characteristically used in paramagnetic resonance (fig. 1). The experiments were done at 24,000 Mc using a \( \text{TE}_{2.4.3} \) microwave cavity as well as a \( \text{TE}_{1.1} \) cavity, the latter excited in circular polarization to help identify the sense of polarization of the resonances. In addition, because of the rapid variation of the resonance fields with temperature in the vicinity of the compensation point, an automatic temperature controller was built which kept the cavity temperature constant to within 0.01 °C. The samples were single crystal spheres of GdIG approximately 0.040" in diameter, taken from different preparations.

Discussion of results. — The experimental results are shown in fig. 2. We initially confine ourselvess to the case of \( H_0 \) along a (111) direction. From \( H^- \) and \( H^+ \),

\[
\lambda M \left( 1 - \frac{\gamma_{Fe}}{\gamma_{Gd}} \right) \quad \text{and} \quad \lambda M(H^{0}_H + H^{0}_G)
\]

can be determined from Eqs. (9) and (10) assuming \( \gamma_{Fe} \approx \gamma_{Gd} \sim 2 \). From the temperature variation of \( H^{0}_H \), \( \lambda M/T_c \) can be determined using Eq. (7). That the locus for \( H^{0}_H(-\omega) \) is almost a straight line at this frequency due to the near equality of

\[
(H^{0}_H + H^{0}_G)/(\omega/\gamma_H) \quad \text{and} \quad \left( 1 - \frac{\gamma_{Fe}}{\gamma_{Gd}} \right).
\]

Note that \( H^- \) (111) was extrapolated from this locus, instead of taking the value actually observed at compensation. The reason for this is that we believe that the break in the curve appearing at 11.5 °C arises from the spin system tending towards other (111) directions not along \( H_0 \). With \( \lambda M \) thus determined,

\[
(H^{0}_H + H^{0}_G) \quad \text{and} \quad \left( 1 - \frac{\gamma_{Fe}}{\gamma_{Gd}} \right)
\]

can be found. The locus of the abnormal polarization \( H^{0}_H(\pm \omega) \) should be the negative of \( H^{0}_H(-\omega) \) as it is seen to be. The abnormal polarization mode was found from 5.8 °C to 7.0 °C, the points falling very nicely on the theoretical curve. The intensity of absorption of this mode is among other things proportional to \( (\gamma_{Fe} - \gamma_{Gd})^2 \) and is considerably weaker than the normal polarization (fig. 3). The locus of \( H^+(\pm \omega) \) will be at

\[
\begin{align*}
6.0 \text{°C} & \quad \text{A} \\
6.3 \text{°C} & \quad \text{B}
\end{align*}
\]

Fig. 2. — Field for resonance vs. temperature in GdIG in the region of compensation for the cases \( H^{0}_H \) (111) and \( H^{0}_H \) (100).

\[
\begin{align*}
6.0 \text{°C} & \quad \text{A} \\
6.3 \text{°C} & \quad \text{B}
\end{align*}
\]

Fig. 3. — Resonance with abnormal polarization displaced relative to normal polarization which is at least 20 db greater.
to assume positions along the (111) directions. The results are listed below, and are assigned an error of a few per cent. The values given here differ from those reported at the Conference; the latter involved a computational error. We are indebted to Dr. R. Pauthenet for comments which led us to find this mistake.

\[
\left( \frac{\gamma_{Fe}}{\gamma_{Gd}} - 1 \right)_{111} = + .0103 \quad \left( \frac{\gamma_{Fe}}{\gamma_{Gd}} - 1 \right)_{100} = + .0122
\]

\[
H^0_{Fe} + H_{Gd} = + 103 \text{ Oe} \quad (T = T_c)
\]

\[
H^0_{Fe} + H_{Gd} = - 103 \text{ Oe} \quad (T = T_c)
\]

\[
\lambda M(T_c) = 230,000 \text{ Oe.}
\]

The difference in g-values is in reasonable agreement with the values of Fe\(^{3+}\) and Gd\(^{3+}\) from para...

magnetic resonance data [7]. The slight anisotropy of the g-value difference, although surprising, may arise from the fact that the Gd is in a site of relatively low local symmetry. This could also explain the fact that a first order anisotropy constant can not fit the data, i.e.

\[
H_{anis}(111) \neq - \frac{2}{3} H_{anis}(100).
\]

From our data we can predict \(H_0\) to be expected at 9300 Mc.

At \(-3\) °C we predict \(H_0(111) = 2150\) and \(H_0(100) = 5700\) in fairly good agreement with the experimental results of Jones and Rodrigues [8] who get \(H_0(111) = 2444\) and \(H_0(100) = 5390\). A few degrees difference in \(T_c\) between samples could bring these values into better agreement.

In a neighborhood of approximately one degree around compensation, further resonances are observed. We are led to assume that the sample breaks up into domains with spins lying along the different (111) directions. Because of the small magnetization, there will be no demagnetizing effects for these domains so that they will resonate independently of each other. The number of resonances to be expected will depend upon the number of inequivalent (111) directions with respect to \(H_0\), allowing for two opposite orientations of the sublattices. In general, one expects...

**Fig. 4.** — Resonances observed at \(T = T_c\) believed to arise from spin systems pointing along the different allowable (111) directions including those cocked relative to the applied field.

**Fig. 5.** — Resonances observed in the immediate vicinity of \(T_c\), with \(H_0\) along a (111) direction showing growth of domains with spins along a particular (111) orientation relative to those with spins in other (111) directions, until finally we approach the conditions of a single domain with Gd pointing down above \(T_c\) (the line at low field).
a maximum of six resonances for an arbitrary direction of $H_0$ in the (110) plane. As many as five can be identified in some of the traces shown in fig. 4. With $H_0$ along (100), $\theta = 0^\circ$, all the (111) directions are equivalent and one finds only two resonances corresponding to the cases of Fe making acute or obtuse angles respectively with respect to $H_0$.

If one postulates a very slight variation in $T_c$ in different parts of the sample, then domains with different orientations will exist simultaneously over a slight temperature range around $T_c$. As the temperature decreases, those with Fe down will grow at the expense of those with iron up as shown in fig. 5. Calculation of the frequencies of the resonant modes with the spin system cocked with respect to $H_0$, as well as a discussion of line widths will appear in a forthcoming publication.

REFERENCES

[8] JONES (R. V.) and RODRIGUES (R. P.) (Private communication).

DISCUSSION

Mr. Kittel. — Are the g-value ratios in reasonable agreement with those found in paramagnetic salts containing these ions?

Mr. Clogston (for Geschwind). — The g-value deviations from the spin-only value are similar to those obtained by paramagnetic resonance experiments but seem to be different by experimentally significant amounts.

Mr. Schlömann. — In the theory of ferrimagnetic resonance it is usually assumed that the two sublattice magnetizations are aligned parallel and anti-parallel to the field if no anisotropy forces are present and if no driving field is applied. One can show, however, that this assumption is not correct, if the applied field exceeds a critical value equal to the net magnetization times the exchange parameter (paper presented at the Solid State Conference in Brussels, 1958). In the compensation region this critical field strength becomes very small. I should like to ask whether the applied field has always been smaller than this critical field in the present experiments.

Mr. Geschwind. — It must be borne in mind that just as the anisotropy of the sublattices and the susceptibility of the Gd strongly influence the resonance frequency in the region of the compensation temperature, $T_c$, so do they affect the so called critical field. The net result, is to make the critical field larger than one might expect by neglecting the anisotropy and susceptibility. This will be reported on in greater detail in a subsequent publication. Nonetheless, the critical field was indeed exceeded in our experiments, as shown by the breaks in the curves in figure 2. However, this occurred so close to $T_c$, that we were able to extrapolate to $T_c$, to obtain the fields for resonance at $T_c$, assuming the sublattices had remained aligned along the external magnetic field.