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Theory of Ferromagnetic Resonance in Rare Earth Garnets. I. *g* Values*

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Ferromagnetic resonance phenomena in rare earth iron garnets (except Gd iron garnet) are dominated at room temperature and above by the rapid spin relaxation of the rare earth ions. The rare earth ion relaxation controls the *g* values and the line widths, as well as the temperature dependence of these quantities. It is shown that in the appropriate limit the *g* value of the microwave spin resonance line satisfies the relation $g=g_A(M_A+M_B)/M_A$, where g_A and M_A refer to the ferric lattice and M_A+M_B is the net saturation magnetization of the crystal. This relation obtains essentially when the *B* lattice relaxation frequency is high in comparison with the *AB* exchange frequency and relaxation frequency of the *A* lattice. The theory accounts quite well, with no disposable parameters, for the sequence and temperature variation of the *g* values reported for Dy, Ho, Er, Yb, and Sm iron garnets. When at low temperatures the *B* relaxation frequency becomes sufficiently low, the *g* value should approach the usual result for two coupled undamped lattices, in the absence of anisotropy effects on the *B* lattice. The theory predicts further that at ordinary temperatures the exchange frequency resonance will occur at the usual position and its width will be proportional to the damping constant of the *B* lattice.

INTRODUCTION

We present in this paper and in a paper with P.-G. de Gennes and A. M. Portis to appear later a theory of some general aspects of ferromagnetic resonance in the rare earth garnets, in impurity-doped yttrium iron garnet, and more generally in any ferromagnetic or ferrimagnetic crystal with the property that two types of magnetic ions are present, one type relaxing rapidly and one type relaxing slowly. The theory gives natural and simple explanations of the central experimental facts relating to ferromagnetic resonance in rare earth garnets $5Fe_2O_3 \cdot 3M_2O_3$, where M is an appropriate trivalent cation. In the accepted abbreviation, this garnet would be referred to as MIG. The static magnetic properties of these materials have been discussed exhaustively by Pauthenet,[1] who has determined and verified the essential features of the magnetic lattice structure. Our present observations do not apply directly to GdIG, because the Gd ions are in an ^8S state and are not highly damped in the present sense. Our discussion does not apply to ions in $J=0$ states. Wherever we write relaxation, in what follows, the quantity relaxed is the electronic magnetic moment.

The following conclusions may be drawn from the available microwave resonance work on rare earth garnets carried out in several laboratories, notably Harvard:

1. Over a wide temperature range, from below room temperature up to within 10° of the Curie temperature ($\sim 560°K$), the line width decreases monotonically (roughly as T^{-2}) with increasing temperature. This behavior is anomalous because relaxation frequencies usually increase rapidly with increasing temperature. There is some suggestion that the widths may decrease again at very low temperatures: in YIG (presumably slightly contaminated by rare earth ions) the line width goes through a maximum at about 50°K.

2. The widths at room temperature are of the order of hundreds or thousands of oersteds, except in highly pure yttrium iron garnet which does not contain significant amounts of rare earth elements, where the width is less than 1 oersted in a highly polished specimen. Yttrium is diamagnetic in this crystal.

3. The *g*-values range from 2.02 to values less than 1, and in Ho, Er, and Dy garnets have been observed to be markedly temperature dependent, generally increasing with increasing temperature above the compensation temperature.

4. The *g*-values over a wide temperature range are found to satisfy quite well the following relation involving the saturation moments M_s:

$$g(\mathrm{M})=g(\mathrm{Y})M_s(\mathrm{M})/M_s(\mathrm{Y}),$$

where Y refers to YIG and M refers to MIG. This will not apply at low temperatures.

The key to the understanding of the resonance properties of the garnets is the fact that above 100°K or so the relaxation frequency of all paramagnetic rare earth ions[2] in the garnets may be expected to be high in comparison with any other frequency concerned in the problem, including the microwave driving frequency, the relaxation frequency of the ferric lattice, and the exchange frequency associated with the exchange interaction of the M^{+++} lattice with the Fe^{+++} lattice. The relaxation frequency need not necessarily be high in comparison with the exchange frequencies among the ferric ions.

The argument begins with the remark that the relaxation frequencies of rare earth ions (excepting, always, Gd^{+++}) in resonance experiments on dilute paramagnetic salts at 10 kMc/sec and 20°K are commonly observed to be of the order of 10^{10} sec^{-1}. For Raman relaxation processes the theoretical T^7 temperature dependence would then give 10^{13} sec^{-1} at

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[1] R. Pauthenet, Ann. phys. 3, 424 (1958).

[2] Except Gd^{+++}, which has an ^8S ground state and is known to relax slowly.

60°K, at the same frequency; however, there is not much experimental evidence that the temperature dependence is this marked. The relaxation processes in the garnets, moreover, are carried out in an exchange field of the order of 10^5 oersteds acting from the ferric lattice on a rare earth ion. At 20°K the direct relaxation frequency is proportional to H^2, theoretically, so that even at 20°K the relaxation frequency in the garnets may be higher than 10^{10} sec^{-1}. Evidence on this argument is lacking. The H^2 dependence actually holds only if $kT > \hbar\omega_{\text{ex}}$, while for Raman processes the relaxation rate is not necessarily field-dependent. Here ω_{ex} is the exchange frequency of the rare earth ion in the exchange field ($\sim 10^5$ oersteds) of the ferric ion lattice.

It is convenient to discuss first (Part I) the analysis of the g-values observed in the resonance experiments, and we discuss in a later paper (Part II) the line width observations. A list of the principal symbols employed is given below.

LIST OF PRINCIPAL SYMBOLS

$\mathbf{H}_A = \lambda\mathbf{M}_A =$ molecular field of lattice A at ions of lattice B.

$\mathbf{H}_B = \lambda\mathbf{M}_B =$ molecular field of lattice B at ions of lattice A.

$H = H_z =$ static external magnetic field, directed along the z axis.

$M_A = M_A{}^z =$ magnetization of lattice A; in the garnets of composition $5\text{Fe}_2\text{O}_3 \cdot 3\text{M}_2\text{O}_3$ there are two sublattices of Fe^{+++} ions coupled together strongly to form the A lattice.

$M_A{}^+ = M_A{}^x + iM_A{}^y$; $M_B{}^+ = M_B{}^x + iM_B{}^y$.

$\Delta M = M_A + M_B =$ resultant magnetization. Note that M_A and M_B in the garnets will have opposite signs.

$\alpha_A =$ relaxation constant of lattice A. We assume in Part I that the direct relaxation of lattice A may be neglected.

$\alpha_B =$ relaxation constant of lattice B.

$\alpha_1 = \alpha_B M_B H_B$; $\alpha_2 = \alpha_B M_B(H + H_A)$: associated relaxation frequencies.

$\gamma_A = g_A e/2mc$.

$\gamma_B = g_B e/2mc$.

$\Delta\gamma = \gamma_A - \gamma_B$.

$\lambda =$ molecular field constant describing the exchange interaction of lattices A and B. The associated exchange energy density is $-\lambda\mathbf{M}_A\cdot\mathbf{M}_B$. In the garnets λ is negative.

$\rho = 1/\tau =$ frequency associated with spin correlation time τ of ions on lattice B.

$\omega =$ resonance frequency (complex).

$\omega_A = \gamma_A H$.

$\omega_B = \gamma_B H$.

THEORY OF g-VALUES

We let $\mathbf{M}_A$ be the magnetization vector of the resultant Fe^{+++} lattice, treating the two ferromagnetic Fe^{+++} sublattices as coupled together infinitely tightly. We neglect entirely in the g-value analysis the relaxation of $\mathbf{M}_A$ except as it may be damped by contact with $\mathbf{M}_B$, the magnetization vector of the lattice occupied by the rare earth or the yttrium ions. The interlattice exchange energy density is $-\lambda\mathbf{M}_A\cdot\mathbf{M}_B$, where a negative λ gives antiparallel coupling. The equation of motion for the A lattice is

$$d\mathbf{M}_A/dt = \gamma_A\{\mathbf{M}_A\times\mathbf{H} + \lambda\mathbf{M}_A\times\mathbf{M}_B\}. \tag{1}$$

For the B lattice

$$d\mathbf{M}_B/dt = \gamma_B\{\mathbf{M}_B\times\mathbf{H} + \lambda\mathbf{M}_B\times\mathbf{M}_A\} - \alpha_B\mathbf{M}_B\times\{\mathbf{M}_B\times(\mathbf{H}+\lambda\mathbf{M}_A)\}, \tag{2}$$

where the relaxation term is essentially in the Landau-Lifshitz form. The magnetization $\mathbf{M}_B$ relaxes toward the direction of the effective field $\mathbf{H}+\lambda\mathbf{M}_A$ acting on B. Whatever phenomenological form is used to describe the relaxation, Wangsness[3] has pointed out that the form should describe relaxation toward the instantaneous field, as otherwise the second law of thermodynamics might be violated. We want the eigenfrequencies of (1) and (2) in the usual linear approximation. Our problem is a special case of a very general problem considered by Wangsness,[3] but it is well worth solving the special case for the unusual insight afforded, an insight concealed in the general solution.

Forming $M^+ = M^x + iM^y$, the equations of motion of the free system reduce, for time dependence $e^{i\omega t}$, to

$$\omega M_A{}^+ = -\omega_A M_A{}^+ - \gamma_A H_B M_A{}^+ + \gamma_A H_A M_B{}^+; \tag{3}$$

$$\omega M_B{}^+ = -\omega_B M_B{}^+ - \gamma_B H_A M_B{}^+ + \gamma_B H_B M_A{}^+ - i\alpha_1 M_A{}^+ + i\alpha_2 M_B{}^+, \tag{4}$$

where $H_A = \lambda M_A$; $H_B = \lambda M_B$; $\alpha_1 = \alpha_B M_B H_B$; $\alpha_2 = \alpha_B M_B \times (H + H_A)$. The secular equation is

$$\begin{vmatrix} \omega+\omega_A+\gamma_A H_B & -\gamma_A H_A \\ -\gamma_B H_B + i\alpha_1 & \omega+\omega_B+\gamma_B H_A - i\alpha_2 \end{vmatrix} = 0. \tag{5}$$

Low-Frequency Resonance

The usual microwave ferromagnetic resonance solution satisfies $\omega \ll H_A, H_B$. If we drop terms of order ω^2, $\omega_A\omega$, etc., the secular equation for the lower eigenfrequency of (5) reduces to

$$\omega(\gamma_B\lambda M_A + \gamma_A\lambda M_B - i\alpha_B\lambda M_A M_B) + \omega_A\Delta M(\gamma_B\lambda - i\alpha_B\lambda M_B) = 0, \tag{6}$$

and the factor λ may be cancelled out; here $\Delta M = M_A + M_B$.

If the damping of lattice B is sufficiently high so that $\alpha_B\lambda M_A M_B \gg \gamma_B\lambda M_A + \gamma_A\lambda M_B$, then the eigenfrequency of (6) may be expanded as

$$-\omega = (\gamma_A\Delta M/M_A)H[1 - i\lambda\gamma_A/\alpha_B\lambda M_A]. \tag{7}$$

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[3] R. K. Wangsness, Phys. Rev. **111**, 813 (1958).

As $\alpha_B \rightarrow \infty$,

$$-\omega \rightarrow \gamma_A(\Delta M/M_A)H, \tag{8}$$

which is reduced from the usual ferromagnetic resonance frequency $\gamma_A H$ of lattice A by the factor $\Delta M/M_A \equiv (M_A+M_B)/M_A$. The result (8) has a simple physical interpretation. Our general theory[4] of g values applied to two coupled *undamped* lattices gives, as is well known,

$$\gamma_{\text{eff}} = \frac{M_A+M_B}{(M_A/\gamma_A)+(M_B/\gamma_B)}. \tag{9}$$

But the new result (8) for heavy damping of lattice B is that

$$\gamma_{\text{eff}} = \frac{M_A+M_B}{(M_A/\gamma_A)}, \tag{10}$$

so that the damped lattice contributes fully to the magnetization but not to the angular momentum density. This result is what one would expect, and is in good agreement with the experiments reviewed in the following section. The B system, being highly damped, does not respond to an applied torque as a normal gyroscopic system. We may think of the angular momentum of the B system as largely imaginary. We note that *no angular momentum compensation point* should be observed in spin resonance *in this limit*; this prediction probably accounts for the observations on ErIG. However, angular momentum compensation points may be observed if they occur at sufficiently low temperatures so that the B lattice relaxation is not fast in the present sense. Equation (39) of the paper by Wangsness reduces to (10) in the appropriate limit. We note that for our g value considerations the condition of heavy damping of lattice B is that the relaxation frequency $\alpha_B\lambda M_A M_B$ be large in comparison with the frequency ω and with the exchange frequency associated with magnetic fields of value λM_A or λM_B. At very low temperatures the relaxation frequency may become low enough so that (9), rather than (10), will be applicable.

This belief in the largeness of $\alpha_B\lambda M_A M_B$ may account for the width of the resonance; the widths are discussed in detail in Part II. The width of the resonance described by (7) is

$$\frac{\Delta H}{H} \cong \frac{\gamma_A}{\alpha_B M_A} \equiv \frac{\gamma_A \lambda M_B}{\alpha_B \lambda M_A M_B} \equiv \gamma_A \lambda M_B \tau, \tag{11}$$

so the width will be *narrower* as the relaxation frequency of lattice B increases; this accounts for the observation that ΔH decreases with increasing temperature over a well-defined temperature range.

Examination of the general expression for γ_{eff} given by Wangsness shows that our limit (10) for γ_{eff} is very accurately satisfied for ratios α_A/α_B of the order of 10^{-3} as envisaged by us at sufficiently elevated temperature.

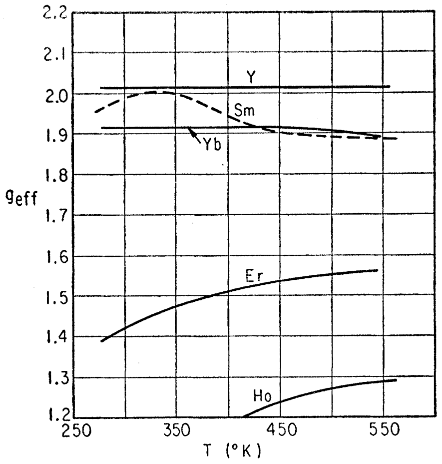

FIG. 1. Experimental g-factors as a function of temperature in deg K for polycrystalline garnets at 9200 Mc/sec, according to Rodrigue, Pippin, Wolf, and Hogan, reference 6. Single-crystal measurements (not shown) on ErIG are in general agreement with the measurements on polycrystals [R. V. Jones (private communication)].

Exchange Frequency Resonance

In the limit $H=0$ the high-frequency root of (5) is

$$\omega = -\lambda(\gamma_A M_B + \gamma_B M_A) + i\alpha_B \lambda M_B H_A, \tag{12}$$

where α_A is still neglected. This equation describes the exchange frequency resonance at the approximate frequency

$$-\omega_{\text{ex}} = \lambda(\gamma_A M_B + \gamma_B M_A),$$

as given originally by Kaplan and Kittel.[5] The damping of lattice B produces a line width

$$\Delta\omega = \alpha_B \lambda M_A M_B, \tag{13}$$

where we neglect α_A. The damping of the exchange frequency resonance is directly proportional to α_B. The result (17) suggests that low temperatures may be most favorable for the detection of the exchange frequency resonance in ferrimagnetic rare earth compounds.

EXPERIMENTAL SITUATION ON g-VALUES

We now confront the result (8),

$$\gamma_{\text{eff}} = \gamma_A(\Delta M/M_A) = \gamma_A(M_A+M_B)/M_A,$$

with the experimental facts known to us at present. In Fig. 1 we have reproduced the experimental results of Rodrigue, Pippin, Wolf, and Hogan,[6] showing g as a function of temperature for polycrystalline garnets at 9200 Mc/sec. The g factors have not been corrected for porosity; the correction should be small. The actual line shapes are somewhat asymmetric, but anisotropy energy measurements by Jones, Rodrigue, and Wolf[7]

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[4] C. Kittel, Phys. Rev. **76**, 743 (1949).

[5] J. Kaplan and C. Kittel, J. Chem. Phys. **21**, 760 (1953).

[6] Rodrigue, Pippin, Wolf, and Hogan, IRE Trans. on Microwave Theory Tech. **MTT-6**, 83 (1958).

[7] Jones, Rodrigue, and Wolf, J. Appl. Phys. **29**, 434 (1958).

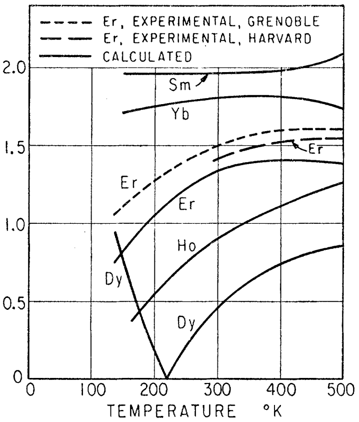

FIG. 2. Theoretical g factors as a function of temperature, as calculated from Eq. (10); also shown are experimental results for ErIG after Rodrigue *et al.*, reference 6, and Paulevé *et al.*, reference 8.

show that the crystalline anisotropy energy is much too small to contribute significantly to the line widths and shapes in the temperature range of Fig. 1. Further, the single-crystal g-values at room temperature obtained by these workers agree with the polycrystalline values within 2%.

To calculate γ_{eff} using the relation (10) we take $\gamma_A = 2.02$, as this is representative of YIG. We take $\Delta M/M_A$ as the ratio of the observed saturation moment for the appropriate rare earth garnet to the observed saturation moment for YIG, using the data of Pauthenet. These data may be significantly in error because of porosity, nonstoichiometry, and foreign magnetic inclusions. Such errors, if present, would have somewhat unpleasant effects on the ratio. We have calculated moments per unit volume from Pauthenet's tables as if the lattice constants were equal to that of YIG; errors of ±3% are introduced in this way. Further, the temperature dependence of the saturation moment for the ferric lattice in a rare earth garnet will not be precisely the same as in YIG, and errors in our input data from this cause are surely important near 500°K and may account for the bending down at high temperature of the calculated g-factor curves for YbIG and ErIG. The curves calculated on this basis are shown in Fig. 2. On Fig. 2 we also give the experimental curves for ErIG from Harvard[6] and from Grenoble.[8]

The general features of the experimental results are reproduced fairly well in the calculated curves. One further fact may be added: In the experiments of Rodrigue *et al.*[6] the lower wing of the resonance line of DyIG was observable at their highest obtainable fields (6100 oersteds) above 510°K which puts 1.2 as an *upper* limit on g for DyIG in this temperature range. This is compatible with the calculated curve and thus the downward sequence of g-values (Sm, Yb, Er, Ho, Dy) is accounted for by the theory.

Note added in proof.—With a relaxation term for the B lattice of the Gilbert form, $-\alpha_B \mathbf{M}_B \times (d\mathbf{M}_B/dt)$, it is not possible to account for the observed g-values of the garnets. With this relaxation term one finds $g_{\text{eff}} \rightarrow 0$ as $\alpha_B \rightarrow \infty$. Indeed, the Gilbert form applied to a single uncoupled spin system gives the resonance condition $\omega = -\gamma H/(1+i\alpha M)$, so that both $g \rightarrow 0$ and $1/\tau \rightarrow 0$ as $\alpha \rightarrow \infty$. Thus such a relaxation term is intrinsically unable to describe a rapidly relaxing spin system.

ACKNOWLEDGMENTS

I am very much indebted to Dr. R. L. White for persistently drawing my attention to this problem in helpful conversations and for summarizing and evaluating the experimental situation in a clear and revealing way. White[9] has suggested that the ferrous ion in ferrites is a rapidly relaxing ion for present purposes and he has pointed out that many properties of ferrites can be understood in this way. I am grateful also to Dr. R. V. Jones and his colleagues at Harvard University for communicating the results of unpublished measurements on single crystals. Professor A. M. Portis and Dr. P.-G. de Gennes made helpful suggestions on the problem.

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[8] Paulevé, Dreyfus, and Soutif, J. phys. radium **20**, 355 (1959); J. Paulevé, Compt. rend. **245**, 408 (1957).

[9] R. L. White, Phys. Rev. Letters **2**, 465 (1959).