FERRIMAGNETIC RESONANCE OF DILUTE RARE-EARTH DOPED IRON GARNETS*

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The present note is concerned with the resonance behavior of yttrium iron garnet (YIG) in which a small percentage of the yttrium ions have been replaced by rare-earth (RE) ions. The latter will be assumed (and hence restricted) to possess a low-lying isolated pair of levels split by the exchange field of the iron sublattice. An example is the ytterbium ion, where this splitting is 12 to 31 cm^{-1}, being highly direction dependent,[1] and where the excited Kramers doublets are 550 cm^{-1} or more above the ground one.[2] We exclude from consideration cases such as terbium where there is a crossing or near crossing of energy levels.

To explain the pronounced effect of small amounts of RE ions on the linewidth and shift of YIG, two distinct mechanisms have been proposed, utilizing RE relaxation frequencies fast and slow, respectively, compared with the splitting of the ground doublet. The fast relaxing theory has been presented in some detail by Kittel, de Gennes, and Portis.[3] The possibility of a slow relaxing mechanism was suggested by Dr. Y. Obata to one of the writers, who mentioned it briefly at the Kyoto conference on magnetism.[4] The distinction between the fast and slow theories has been stressed for the ferrites in a paper by Galt and Spencer,[5] and the slow one has implicitly been employed by Teale and Tweedale[6] in adapting to their data on YbIG a formula for linebreadth given by Clogston[7] in a different context. Various applications to the garnets of essentially the slow relaxing mechanism are also treated by LeCraw and Kasuya.[8]

Quite apart from considerations of the expected magnitude of the RE ion's relaxation frequency, experimental evidence appears to favor the slow relaxing theory, as the observed linewidths and frequency shifts depend on the ferrimagnetic resonance frequency,[9] even though the latter is small compared with the exchange frequency.

For the slow relaxing theory to be effective, it is necessary that the Fe-RE exchange field be anisotropic. The essential feature is that, because of the anisotropy, the oscillating part of the exchange field exerted on the RE ion by the iron has a component parallel to the large constant, or average, exchange field which is responsible for the spatial quantization of the RE moment. The relaxation process by which the RE ion absorbs low-frequency quanta is thus reminiscent of the Gorter aperiodic type of paramagnetic relaxation, or more still, the Bloembergen process by which small nuclear quanta (here represented by the Zeeman energy of the $k = 0$ ferrimagnetic spin-wave mode) are absorbed by paramagnetic impurities even though the electronic Zeeman quanta (here represented by the exchange splitting of the RE ion's ground doublet) are enormously large compared to the nuclear Zeeman quanta. That this "longitudinal" relaxation process was the mechanism underlying the Clogston formulas used by Teale and Tweedale is demonstrated in a forthcoming paper by Mme. Hartmann-Boutron, to whom we are indebted for seeing her manuscript in advance of publication. Our results, obtained independently, in some ways do not go as far as hers, as she treats the nuclear resonance of Fe^{57}, but we feel our specialization to a low-lying doublet displays illuminating explicit formulas for the dependence of the linewidth and frequency shift on angle and temperature.

The model we have used assumes an iron-rare-earth exchange interaction of the form

$$\mathcal{H} = \sum_n (K_1 S_{x'} J_{nx'} + K_2 S_{y'} J_{ny'} + K_3 S_{z'} J_{nz'}), \qquad (1)$$

where S is the spin of the iron sublattice, and J_n the fictitious spin of the nth RE ion. The local principal axes for the RE ions are denoted by x', y', z', and are different for the six types of RE sites. We make the customary assumptions regarding the iron atoms, namely, that the Fe-Fe exchange coupling is much greater than the Fe-RE exchange interaction, and we neglect any "back-action" of the RE ions that mixes the spin modes of the iron, an assumption certainly valid for very small RE concentrations. To transform (1) back to the direction of the net iron spin, we use the direction-cosine relations

$$S_{\alpha'} = \sum_\beta \lambda^{\alpha\beta} S_\beta, \qquad (2)$$

where the x, y, z system is so chosen that the z axis is parallel to the magnetization of the iron, along which a static field H is directed. If second-order terms in S_x, S_y are neglected, the equa-

tion of motion for S_x is

$$\hbar\dot{S}_x = -2H_\beta S_y - \sum_n \sum_{q=1}^{3} F(\omega_n, T) \times \{(S_z^2 \lambda_n^{q2} \lambda_n^{q3} + S_z S_x \lambda_n^{q2} \lambda_n^{q1})(K_q^2 - K_1^2) + S_y S_z [(\lambda_n^{q2})^2 - (\lambda_n^{q3})^2] K_q^2\},$$

where

$$F(\omega_n, T) = [\tanh(\hbar\omega_n/2kT)]/2\hbar\omega_n,$$

and $\hbar\omega_n$ is the exchange splitting of the lowest RE ion doublet on the nth site, viz.,

$$\hbar\omega_n = (K_1^2 S_{x'}^2 + K_2^2 S_{y'}^2 + K_3^2 S_{z'}^2)^{1/2}.$$

The equation for $\dot{S}_y$ can be found in a similar manner. We now linearize the equations in the usual way, so that, for example, ω_n has the structure

$$\omega_n = \omega_{n0} + aS_x + bS_y,$$

with ω_{n0}, a, and b functions of S_z, and the direction cosines. In addition, in order to allow for the fact that the redistribution in population may not be able to keep up with the oscillations in the effective longitudinal field, we insert a factor $1/(1+i\tau_n\omega)$ in the a,b terms of ω_n when ω_n enters in the argument of the tanh terms.

This procedure gives two simultaneous homogeneous equations in S_x, S_y from which one determines the ferrimagnetic resonance frequency ω. For N iron atoms and a rare-earth concentration c, we find that its frequency shift $\Delta\omega_S$ and linewidth $\Delta\omega_W$ are given by

$$\Delta\omega_S = \frac{Nc}{12}|S_z| \sum_{n=1}^{6} \left\{ -\frac{1}{kT} \frac{Q_n}{1+\tau_n^2\omega^2} \operatorname{sech}^2\left(\frac{\hbar\omega_0}{2kT}\right) + F(\omega_{n0}, T)[4Q_n + 2K_1^2 - K_2^2 - K_3^2 + 3(\lambda_n^{33})^2 (K_3^2 - K_1^2) + 3(\lambda_n^{23})^2 (K_2^2 - K_1^2)] \right\},$$

$$\Delta\omega_W = \frac{Nc}{12kT}|S_z| \sum_{n=1}^{6} \frac{Q_n \tau_n \omega}{1+\tau_n^2\omega^2} \operatorname{sech}^2\left(\frac{\hbar\omega_{n0}}{2kT}\right),$$

where the sum now goes over the six inequivalent RE ion sites instead of over all the RE ions in the body. The symbol Q_n stands for

$$Q_n = -S_z^2 (1/2\hbar\omega_{n0})^2 \times (\lambda_n^{33})^2]^2 \{[(K_2^2 - K_1^2)(\lambda_n^{23})^2 + (K_3^2 - K_1^2) - (K_3^2 - K_1^2)^2 (\lambda_n^{33})^2 - (K_2^2 - K_1^2)^2 (\lambda_n^{23})^2\}.$$

The angular dependence is quite interesting. With complete isotropy Q_n vanishes. If the exchange field has axial symmetry the RE ion's principal axis ($z' \sim x, y, z$ in the garnets), so that $K_1 = K_2 = 0$, then Q_n is a maximum along a $\langle 111 \rangle$ direction, and vanishes along a $\langle 100 \rangle$ direction. Hence, measurements of $\Delta\omega_W$ with the iron magnetization along the $\langle 100 \rangle$ direction will give direct information regarding the deviations of the local exchange interaction from axial symmetry, and a detailed analysis of the entire angular dependence at constant temperature would yield the three principal components of the exchange field tensor, *if* one assumes a weak angular dependence for τ_n. The predicted dependence is, in general, in reasonable agreement with experiment, especially if the constants are given somewhat different values than those obtained by Wickersheim.[1] For comparison with the experimental curves, the reader is referred to the paper of Teale and Tweedale.[6] The predictions of the theory upon the temperature dependence of the line shift in the adiabatic ($\tau_n\omega \sim \infty$) and isothermal ($\tau_n\omega \sim 0$) limits should also be noted. If the exchange splitting is small compared to kT, one finds after summing over the RE ion sites that $\Delta\omega_S \sim 1/T$ if $\omega\tau_n = \infty$ and $\sim 1/T^3$ if $\omega\tau_n = 0$. It is not surprising that in the isothermal case the deviations occur as $1/T^3$ rather than $1/T$, for this is a close analog of the fact that in a powder a crystalline potential small compared to kT first affects the susceptibility χ in a term in $1/T^3$ when χ is developed as a series in $1/kT$.

The linewidth is predicted to be proportional to $(1/T)\exp(-\hbar\omega_{n0}/kT)$ at low temperatures where τ_n is constant due to its proportionality[10] to $\tanh(\hbar\omega_{n0}/2kT)$. All of these features appear to be exhibited by the experimental data, though a detailed comparison has yet to be carried out.

The analysis which we have described applies regardless of whether the relaxation parameter τ_n is caused by interaction of the RE ions with phonons or with magnons resonant to the RE doublet splitting. We hope to discuss later the relative efficacy of these two processes.

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MAGNETIC BREAKDOWN IN RHENIUM*

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In the course of a detailed study of the de Haas–van Alphen (dHvA) effect in rhenium,[1] we have observed a number of oscillatory terms which can be interpreted as due to electron orbits in reciprocal space resulting from magnetic breakdown similar to that envisaged by Pippard.[2] The results also provide possible experimental evidence for the existence of a degeneracy in the energy spectrum along a symmetry line A-L of the reduced Brillouin zone. The existence of this degeneracy line in hcp metals and its consequences on the electronic structure of nontransition metals have been pointed out by Falicov and Cohen[3] on the basis of spin-orbit coupling calculations.[4]

Pippard's two-dimensional model consists basically of coupled electron orbits of the form shown in Fig. 1(a). His analysis of such a model indicates that when partial magnetic breakdown occurs from one orbit to another [across the gap b-f and d-h in Fig. 1(a)], there exists a possibility of a large number of closed orbits. Two of these possible orbits are shown in Figs. 1(c) and 1(d), and the case of complete breakdown is shown in 1(b). These orbits can, in principle, be observed as dHvA oscillations whose frequencies are proportional to the areas of the orbits. The relative amplitudes of the oscillations would be dependent on the probability of occurrence of magnetic breakdown.

Detailed orientation studies of the dHvA effect by means of null-deflection torsion-balance techniques in fields up to 40 kG have indicated the existence of at least two closed sections of the Fermi surface in rhenium. From the symmetry of the period versus orientation curves in the $(10\bar{1}0)$, $(11\bar{2}0)$, and (0001) planes and two nonsymmetry planes, we deduce that both segments of the Fermi surface lie on a line parallel to the [1010] direction and are located near or at the lateral face of the reduced Brillouin zone. If further we assume that both segments have a common center which is located on the A-L line, the interpretations discussed above follow im-

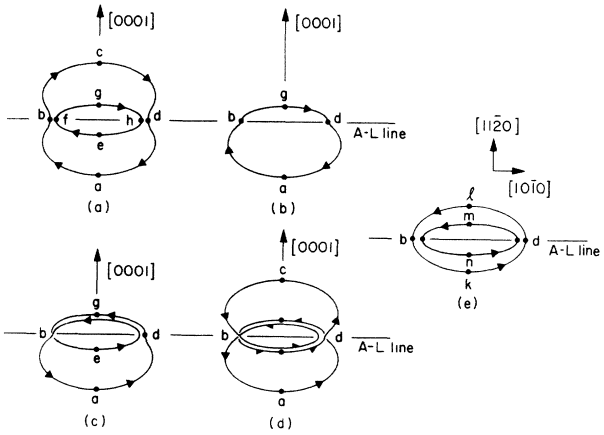

FIG. 1. Extremal cross sections of Fermi surface in rhenium shown schematically, indicating various possible electron orbits. Cross sections in the $(10\bar{1}0)$ plane are shown in (a), (b), (c), and (d), and a cross section in the (0001) plane is sketched in (e).