Parallel pumping in rare earth doped garnets

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Abstract. The parallel pump instabilities of Yb^{3+} and Tb^{3+} doped YIG crystals have been measured in the helium temperature range. These experiments form a useful adjunct to ferromagnetic resonance in the study of relaxation processes since the instability thresholds measured are independent of static or two-magnon scattering mechanisms which have a marked influence on the resonance linewidth. In general, the rare earth impurities in the garnets act both as static scattering centres and as dynamic relaxers. The parallel pump technique has been used here to distinguish between these two roles of the impurity ions. The results for Yb^{3+} suggest that the temperature independent transverse relaxation process dominates over the longitudinal mechanism below approximately 6°K. In Tb^{3+} doped crystals a comparison of resonance and instability measurements show rather good agreement even near giant anisotropy peaks. Thus it appears that, contrary to theoretical expectation, the static scattering mechanism is probably unimportant for Tb^{3+} dopings.

1. Introduction

In recent years, the ferromagnetic resonance behaviour of rare earth doped yttrium iron garnet has been investigated extensively. For most of the rare earth impurities the low temperature experimental results may be characterized by two distinct features—viz. giant anisotropies in the magnetic field required for resonance and peaks in the temperature dependence of the resonance linewidth. The giant anisotropies have been attributed to 'near crossings' in the energy level structure of the rare earth ion (Kittel 1960). This model has been particularly successful in the interpretation of the anisotropies of Tb^{3+} doped YIG (Dillon and Walker 1961, Huber, private communication). The temperature dependence of the resonance linewidth has been discussed in terms of the relaxation behaviour of the induced magnetization of the rare earth impurity in the oscillatory exchange field associated with the precessional motion of the dominant ferric sub-lattice (deGennes et al. 1960, Van Vleck and Orbach 1963, Van Vleck 1964, Hartmann-Boutron 1963, 1964, Dillon 1962 a, b). The linewidth of Yb^{3+} doped YIG has been the most carefully studied and may be said to be fairly well understood in terms of a relaxation of the longitudinal susceptibility of the Yb^{3+} ion (Teale and Tweedale 1962).

In this paper we present some experimental results on the low temperature longitudinal or parallel pump instability of YIG crystals doped with Tb^{3+} and Yb^{3+} ions. It has been noted that ferromagnetic resonance and parallel pump instability experiments do not necessarily measure the same decay rates (Sparks 1962, 1964, Haas and Callen 1964). The ferromagnetic resonance ΔH_0 results from scattering processes which may be either static (i.e. two-magnon) or dynamic (i.e. magnon–phonon, three-magnon, etc.) in character. However, the threshold microwave field for parallel pump instability h_{crit} is independent of static scattering mechanisms. The parallel pump instability occurs when the longitudinal coupling between the microwave field and a pair of normal modes of the spin system exceeds the intrinsic damping of these modes (Schlömann et al. 1960, Morgenthaler 1960). The correct normal modes arise from a diagonalization of the spin wave Hamiltonian which already accounts for any static coupling terms between spin modes. Thus the only contributions to the intrinsic damping come from time-dependent coupling terms. In general, the randomly distributed rare earth impurity ions in YIG act as both static and dynamic spin wave scattering centres (Hartmann-Boutron, private communication). The bulk of the theoretical treatments of the rare earth problem have been concerned with the dynamic effects of the impurities, although deGennes et al. (1962) have examined the static contributions of the terbium ion and find them to be sizable. The experimental work presented here was undertaken to determine how important a role two-magnon processes play and to test further the dynamic models.

The distinction between ferromagnetic resonance and parallel pump instability results may be developed further in a schematic way by writing the equation of motion for the spin wave amplitudes a_k appropriate to a doped garnet system

$$\dot{a}_k = -i(A_k - C_k)a_k - i(B_k - D_k)a^*_{-k} + i\sum_{k' \neq k} (E_{k'}a_{k'} + F_{k'}a^*_{-k'}). \qquad (1)$$

A_k and B_k are the usual magnetostatic-exchange coefficients (Suhl 1957) for the composite ferric ion sublattice. The coefficients C_k, D_k, E_k and F_k arise from the back reaction produced on the ferric ion spins by their exchange interaction with the rare earth impurities and are complex quantities proportional to the product of the square of the Fourier transform of the rare earth ferric exchange interaction times the susceptibility of the rare earth ion. The k' sum is taken over the reciprocal lattice vectors of the chemical cell. Since the rare earth ferric exchange interaction is anisotropic the longitudinal as well as transverse susceptibilities contribute to the back reaction (Van Vleck and Orbach 1963, Van Vleck 1964, Hartmann-Boutron 1963, 1964). In ferromagnetic resonance the linewidth will be determined not only by the imaginary part of C_k but also by the scattering out of the uniform precessional mode represented by the k' sum in equation (1). However, in principle, equation (1) can be transformed into normal mode form to account for

the time-independent parts of E_k and F_k. The normal modes under this transformation become spatially modulated plane waves. In most cases this transformation would have a negligible effect on the complex resonance frequency of the normal modes which may be approximated as

$$\Omega_k = \{(A_k - C_k')^2 - |B_k - D_k|^2\}^{1/2} - iC_k'' \qquad (2)$$

if we ignore the frequency dependence of C_k and D_k. C_k' and C_k'' are respectively the real and imaginary parts of C_k. Since the phase of B_k depends on the ratio k_y/k_x, we see that one important effect of impurities is to remove, through the term $|B_k - D_k|$, the degeneracy of spin waves having the same value of θ_k. A similar lifting of the transverse degeneracy has been discussed previously in connection with the inclusion of magnetocrystalline anisotropy in the spin-wave problem (Haubenreisser and Linzen 1962, Sethares et al. 1964).

Again ignoring the effect of the coupling terms in equation (1) and any frequency dependence of C_k and D_k the condition for the parallel pump instability becomes (Hartmann-Boutron, private communication)

$$h_{crit} = \frac{\omega}{\gamma} \frac{C_k''}{|B_k - D_k|}. \qquad (3)$$

The factor $|B_k - D_k|$ not only determines the splitting of the transverse spin waves, but is also a measure of the 'ellipticity' of the modes. Thus the lowest transverse spin wave branch has the smallest threshold. In most cases (except at giant anisotropy peaks or in concentrated rare earth garnets) $D_k \ll B_k$ and equation (3) reduces to the famous expression (Schlömann et al. 1960, Morgenthaler 1960)

$$h_{crit} = \frac{\omega}{\omega_M} \frac{\Delta H_k}{\sin^2\theta_k} \qquad (4)$$

where we have taken $C_k'' = {}^1/_2\gamma\Delta H_k$. Because of the short range of the rare earth ferric exchange, C_k and D_k are essentially independent of k value so that we should expect h_{crit} to be independent of the applied d.c. magnetic field for values below the 'corner' field (i.e. the field value at which the frequency of the $k = 0$ transverse spin waves is equal to one-half the drive frequency). This expected behaviour is in marked contrast to the strong field dependence of h_{crit} associated with long range relaxation interactions such as the three-magnon process (Haas and Callen 1964).

The parallel pump instabilities discussed below have been observed in an X band reflection type spectrometer using a pulsed magnetron source. The h_{crit} were measured at variable pulse lengths and extrapolated to infinite pulse length to avoid possible errors in the measurement of small ΔH_k. In attempting to carry through these experiments in a systematic way one is greatly hampered by the tremendous range of critical powers which must be covered for only small variations in temperature or orientation. Even with very small doping concentrations the threshold power may quickly vary from above the gaseous breakdown level down to small levels of the order of the pulse length correction.

2. Ytterbium doped YIG

Certainly YbIG is the most carefully studied of the rare earth garnets and by now it has become the 'textbook' case. The particular magnetic characteristics of the Yb dopings are associated with the properties of the lowest Kramers doublet of the Yb^{3+} ion which is split by the rare earth ferric exchange interaction. If we assume that the longitudinal relaxation mechanism is dominant and that the rare earth relaxation is determined by the 'direct process' it has been shown that the dynamic linewidth should vary as

$$\Delta H = \frac{C\hbar}{3kT} \frac{1}{\gamma} \sum_{n=1}^{6} \frac{\omega_n^2}{\tau_0 \omega} \operatorname{cosech}\left(\frac{\hbar\omega_n}{kT}\right) f_n(\theta, \varphi) \qquad (5)$$

below the peak in the temperature dependence of the linewidth (Van Vleck 1964). In this equation C is the rare earth concentration, f_n a temperature independent angular function, τ_0 the $T = 0°K$ rare earth relaxation time, and $\hbar\omega_n$ the doublet splittings of the six inequivalent rare earth sites. Although the ferromagnetic resonance data are in good agreement with the longitudinal model near the peak (Teale and Tweedale 1962, Dillon 1962 a, b), Seiden (1964) has found that ΔH_0 approaches a residual constant value at low temperature in striking disagreement with equation (5).

To determine whether or not this residual effect is associated with any static inhomogeneities we have studied the parallel pump instability in some detail in the helium temperature range. Figure 1 shows the temperature variation of ΔH_k in the [111] direction (i.e. easy direction) for a 81 mil spherical sample of YIG doped with 0.5% Yb^{3+}. We consider here, mainly, the [111] data since the possibility of non-alignment of the magnetization along the applied field complicates the experimental interpretation in other directions. The large size of the sample was required to give adequate sensitivity to the parallel pump instability. The ΔH_0 measured on this same large sample is also shown in figure 1. It is interesting to note the

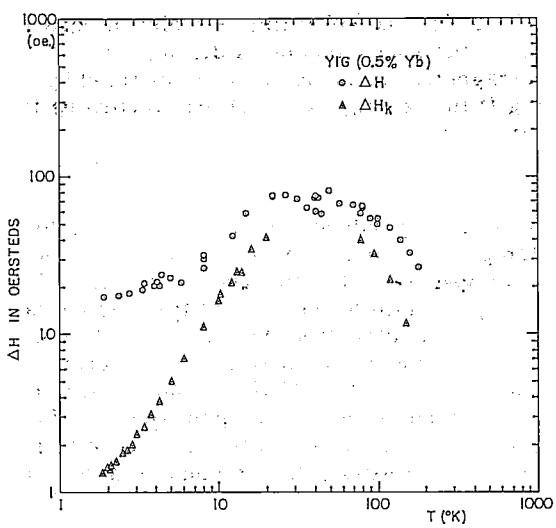

Figure 1. Temperature variation of ΔH_k and ΔH_0 in the [111] direction measured at 8.77 Gc/s on a 81 mil sphere of YIG (0.5% Yb^{3+}).

poor correlation between the values of ΔH_k and ΔH_0. The sample has several surface pits and probably some internal pores which seem to make ΔH_0 much broader than values measured on smaller, more perfect crystals. As discussed earlier, however, these

inhomogeneities do not affect ΔH_k which is in good agreement—above 10°K at least—with more reliable ΔH_0 measurements when we use equation (5) to account for frequency differences.

ΔH_k shows evidence of a peak near 50°K even though the peak h_{crit} is above cavity breakdown. The ΔH_k data are replotted in figure 2 and compared with

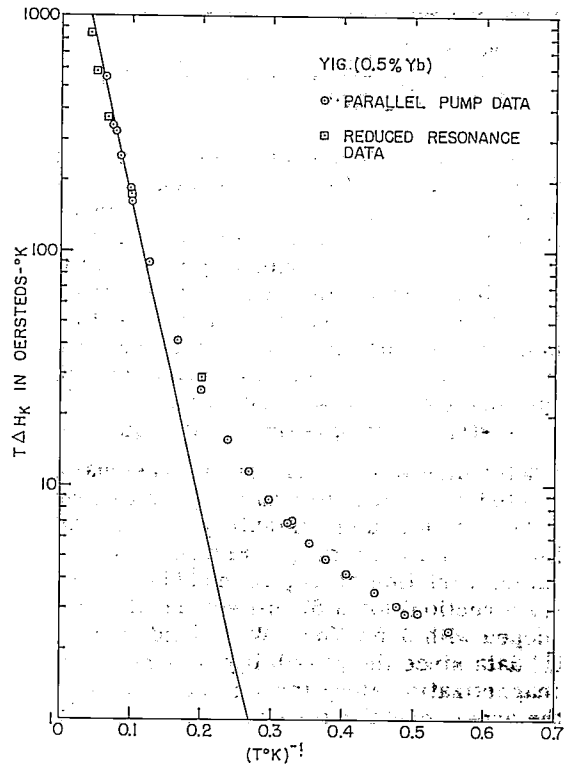

Figure 2. A comparison of the temperature variation of the longitudinal relaxation mechanism and the parallel pump instability threshold data at 8.77 Gc/s. The solid line is a plot of equation (5) using the known doublet splittings of Yb^{3+}. The ferromagnetic resonance data is obtained by reducing Dillon's ΔH_0 values (1962) according to equation (5).

the temperature dependence of equation (5). In deducing this temperature dependence we have used the optically derived doublet splittings of 32°K and 37°K which are appropriate to the case of the magnetization aligned in the [111] direction (Wickersheim 1961). Also shown in figure 2 are some ferromagnetic resonance data points (Dillon 1962 a, b) adjusted in concentration and frequency according to equation (5). Above about 0°K the ferromagnetic resonance and parallel pump instability data are consistent with each other and with the predictions of the longitudinal mechanism. Below this temperature the experimental ΔH_k values decrease more slowly with temperature and seem to be approaching a constant value. To explain this residual width we might postulate all kinds of impurity effects, but it is not unreasonable to invoke the transverse relaxation mechanism which leads to a temperature-independent width at low temperatures (Van Vleck 1964). In fact, it may be estimated that the cross-over temperature between these two mechanisms is near 5°K. The residual value of ΔH_k seems to be about a factor of five smaller than Seiden's value. A factor of two would be more reasonable on the basis of the transverse process.

Figure 3 shows the field dependence of h_{crit} at several temperatures. The data show that ΔH_k is essentially independent of k as expected. However, near the 'corner' field there seems to be some structure in the curve which grows with increasing tem-

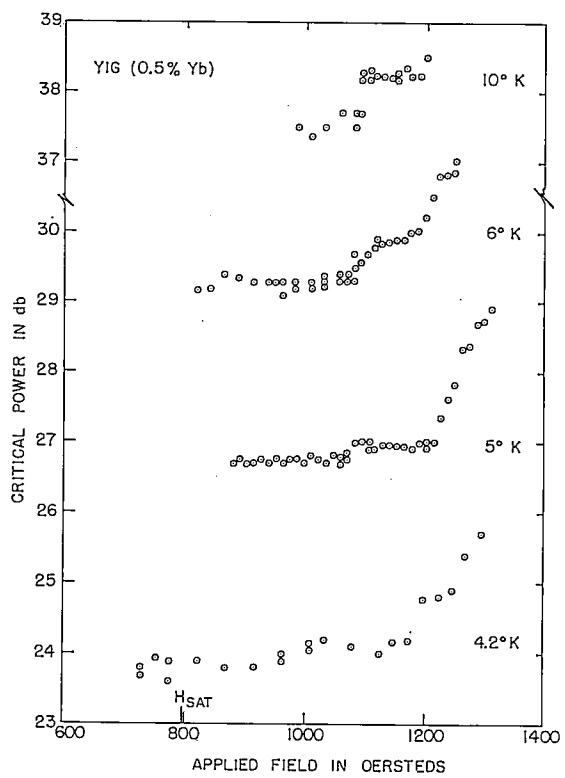

Figure 3. The field dependence of the parallel pump instability threshold at 8.77 Gc/s in the [111] direction at various temperatures. H_{sat} is the field required for saturation in the easy direction (i.e. $4\pi M/3$).

perature. This structure is probably associated with the splitting of the transverse spin waves caused by the term $|B_k - D_k|$ in equation (2).

We note in passing that a sharp peak in the variation of h_{crit} with angle occurs at the point (32° from [100] in the (110) plane) where Dillon (1961) has observed the perplexing anomaly in ΔH_0. Our observation rules out experimentally any kind of static inhomogeneous scattering source for the peak (Van Vleck 1964). The field dependence of h_{crit} is complicated in the vicinity of the anomaly and will be discussed in more detail elsewhere.

3. Terbium doped YIG

It has been suggested that the static scattering mechanism has a strong effect on ΔH_0 near the giant anisotropy peaks in Tb^{3+} doped samples (deGennes et al. 1962). To explore this idea we have looked at the parallel pump instability in a 74 mil sphere of YIG doped with 0.025% Tb^{3+}. The angular variation of the field for resonance is in very good agreement with the results given by Dillon and Walker (1961). Figure 4 shows the field dependence of h_{crit} at several orientations near the [100] direction. Pearson

(1962) has pointed out that Tb^{3+} even in very small concentrations has a profound effect on the magnetocrystalline anisotropy causing the [100] direction to become the easy direction. The minimum in h_{crit}

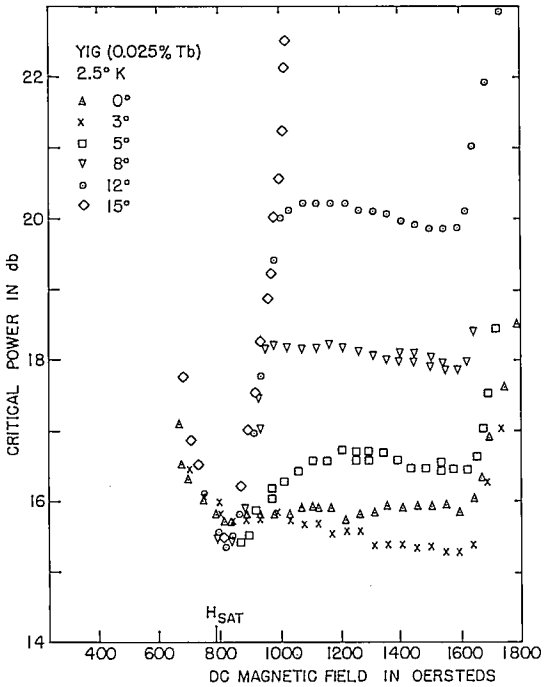

Figure 4. The field dependence of the parallel pump instability threshold at 8.77 Gc/s in the [100] direction at various temperatures. H_{sat} is the field required for saturation in the easy direction (i.e. $4\pi M/3$).

near H_{sat} (i.e. $4\pi M/3$) seems to correspond to the parallel pump instability for the magnetization along the [100] direction. As the field is increased the magnetization becomes polarized along the field and the threshold rises to a new plateau appropriate to the direction of the magnetization. As the magnetization is rotated towards the giant anisotropy peak labelled I by Dillon and Walker (1961) (i.e. 19° from the [100] in the (110) plane) the plateau h_{crit} increases rapidly to power levels above microwave breakdown. Similar behaviour is observed near the other anisotropy peaks. Over the range covered ΔH_k seems to vary as rapidly in angle as ΔH_0. These results suggest that the dominant linewidth mechanism even at the anisotropy peaks is a dynamic process. This suggestion is confirmed by the observation that ΔH_0 at the peaks and in the troughs between the peaks has essentially the same temperature dependence.

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