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Ferrimagnetic Resonance Line Width in Rare Earth Doped Yttrium Iron Garnet

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Measurements of ferrimagnetic resonance line width have been made for YIG single crystals doped with many of the rare earths. Starting from the lowest temperatures, the line width increases to a maximum in the range 30 to 150°K, then decreases with further increase of temperature. Some structure is discernible in these curves; they are anisotropic; and they are frequency dependent (at least in the case of YIG(Tb)). The losses are discussed briefly in terms of transitions within the ground multiplet of the rare earth ions in the magnetic crystal.

Introduction

In the course of studies of the ferrimagnetic resonance line width of rare earth doped YIG, it has become apparent that these experiments should be considered in terms of the energy levels of the rare earth ions. In this paper we will very briefly report our results and their interpretation with particular emphasis on the case of YIG doped with Tb. Experimentally, we have been concerned with single crystal spheres of YIG in which say 0.01 to 0.2% of the Y ions have been replaced by one of the $4f$ rare earth ions. We may consider our system to consist of three parts: the ferric spin lattice, the rare earth ion lattice, and the crystal lattice.

In a resonance experiment energy is put into the almost lossless ferric spin lattice. In these crystals, the energy is passed onto the lossy rare earth lattice. From there it finds its way to the crystal lattice, presumed to be in thermal contact with the outside world.

Rare earth energy levels

Clear effects of the rare earth energy level scheme were seen in the low temperature field for resonance measurements on crystals such as those used here[1]. Along various directions, often along arcs, the low temperature field for resonance shows very high peaks, some of which are exceedingly sharp. The field required for ferrimagnetic resonance is determined by the curvature of the energy surface for the magnetization. The energy of the rare earth ion, being coupled to the magnetization, appears as an additive term in that energy. The peaks in H_{res} are associated with directions in which this energy surface shows a large curvature[2]. L. R. Walker has considered the problem analytically for the case of Tb^{+++} in YIG[3,4]. He took a wave function for the ion, subjected it to a plausible Hamiltonian containing both crystal and exchange terms and solved for the eigenstates as well as the field for re-

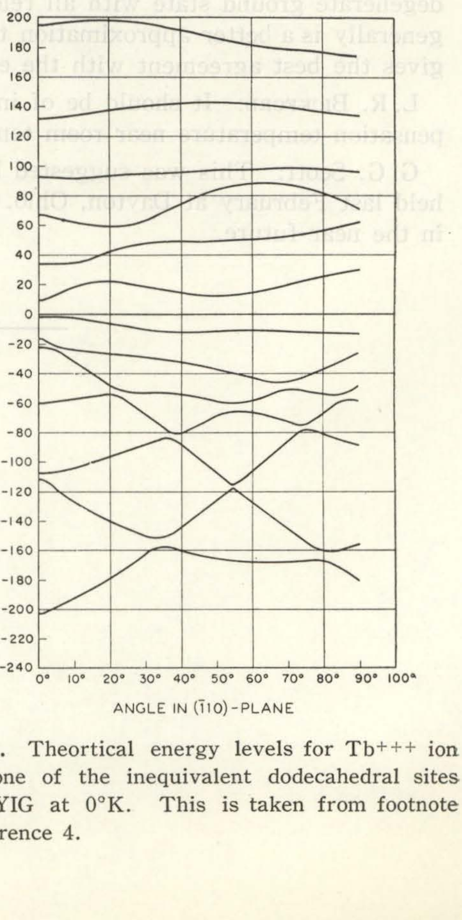

Fig. 1. Theortical energy levels for Tb^{+++} ion on one of the inequivalent dodecahedral sites of YIG at 0°K. This is taken from footnote reference 4.

sonance. The problem contains nine parameters of which only four were optimized. It was done specifically for the garnet lattice with all its inequivalent sites. A typical set of energy levels is shown in Fig. 1[5]. These are a theoretical approximation to the splitting of the Tb^{+++} ground multiplet for one of the dodecahedral sites in YIG as the direction of the magnetization is rotated through the (110) plane. The field for resonance data calculated for this set of parameters agrees fairly well with the experimental observations. Note that the total splitting here is about 400°K. At the lowest temperature accessible

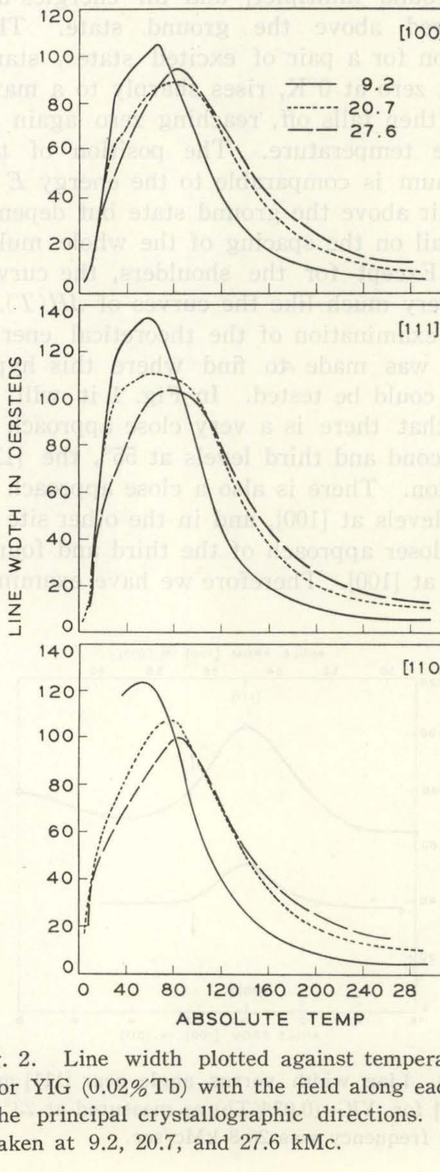

Fig. 2. Line width plotted against temperature for YIG (0.02% Tb) with the field along each of the principal crystallographic directions. Data taken at 9.2, 20.7, and 27.6 kMc.

to us, 1.5°K, very nearly all the ions will be in the lowest state. About 35° from [100] the lowest and next to lowest levels approach closely, and then repel each other. This is the region of a high peak in the field for resonance. Finally note that the whole energy level scheme varies with angle all across the plot.

Line width data

Let us now examine line width data. Fig. 2 shows $\Delta H(T)$ for YIG (Tb) at three frequencies. We have measured $\Delta H(T)$ curves for each of the rare earths in YIG (except Ce, Pm, Gd, and Lu). In general they are similar to that shown here. There is a peak somewhere in the range 30 to 150°K; above the peak, the line width decreases on warming till well above room temperature. Usually there is some structure in the form of shoulders in the curve below the peak. In the case of Tb, the peaks move upward in temperature with increasing frequency, but stay at about the same height. Corresponding data at several frequencies for other ions are not available. For the sake of brevity the few cases in which the behavior of $\Delta H(T)$ is not similar to Fig. 2 must be discussed elsewhere.

Loss mechanisms

It seems appropriate to consider two separate mechanisms for the losses in rare earth substituted YIG. These we will designate "Relaxation Process" and "Direct Transition."

Relaxation Process—We have seen that the energy level scheme for the rare earth ions in the magnetic garnet lattice is strongly dependent on the orientation of the magnetization. For a given temperature and a given direction of the magnetization, there will be an equilibrium distribution of the population of the states for a particular site. If the magnetization direction were to change suddenly the equilibrium populations would change, but the ions would approach this new distribution by some relaxation process. However, we would expect a number of relaxation times to be important for a single site. Ferrimagnetic resonance entails a precession of the magnetization, and the distribution among levels will be relaxing toward the equilibrium distribution for the instantaneous orientation of the magnetization. If any of the relaxation times are at all comparable to the reciprocal of the microwave frequency, we expect that this relaxation process will contribute to the losses in an important way. This is very similar to the problem considered by Galt[6] and by Clogston[7] for the losses in ferrites containing divalent iron. In fact, Clogston's whole formalism seems directly applicable to the present problem. His assumption of a single relaxation time, however, does not seem appropriate. We expect here several relaxation times within the levels of a single site as mentioned above, and different relaxation times for the different sites. Thus there might be something of the order of a dozen relaxation times in the problem. It is believed that this relaxation process accounts for the general character of the $\Delta H(T)$ curves shown in Fig. 2. By general character here is meant the peak itself, the shoulders corresponding to a number of relaxation times, the upward shift in temperature with increasing frequency, and the near constancy of the maximum height with change in frequency.

Direct Excitation — An examination of Fig. 1 will show several instances in which there are very close approaches of the energy levels. If these levels approach within a microwave quantum of each other, the direct excitation of transitions across the gap with attendent losses would be expected. Such direct transitions appear to have been seen in the case of YIG (Tb). There, measurements were made at 1.5°K of H_{res} in (110) as a function of frequency in the range from 10 to 75 kMc/sec. The primary data consisted of the height of the peaks for each frequency. For one of the peaks, the height increased somewhat faster than linearly with frequency, then turned steeply upward at about 65 kMc/sec. At this point the line width suddenly became so broad that the line was no longer detectable in our apparatus. This behavior is described in detail in footnote reference 4. Our interpretation is that the microwave photon energy reached the energy interval between the lowest and next to lowest levels. It is as if one were dealing with a "paramagnetic resonance" with a zero field splitting of 65 kMc/sec. This resonance is enhanced by the high resonant susceptibility of the ferric lattice.

The suggestion here is that similar direct transitions would arise for close approaches of levels other than the lowest two if there is a significant population difference, and if the microwave quanta reach across the gap. The Boltzmann distribution shows that the population difference between two states ΔE apart, whose average is E above the ground state is given by

$$\frac{\Delta E}{kT}\,\frac{e^{-E/kT}}{\sum_i e^{-E_i/kT}}$$

where the summation is over the states of the ground multiplet, and all energies are measured above the ground state. This function for a pair of excited states, starts out at zero at 0°K, rises sharply to a maximum, then falls off, reaching zero again at infinite temperature. The position of the maximum is comparable to the energy E of the pair above the ground state but depends in detail on the spacing of the whole multiplet. Except for the shoulders, the curves look very much like the curves of $\Delta H(T)$.

An examination of the theoretical energy levels was made to find where this hypothesis could be tested. In Fig. 1 it will be seen that there is a very close approach of the second and third levels at 55°, the [111] direction. There is also a close approach of these levels at [100], and in the other site an even closer approach of the third and fourth levels at [100]. Therefore we have examined

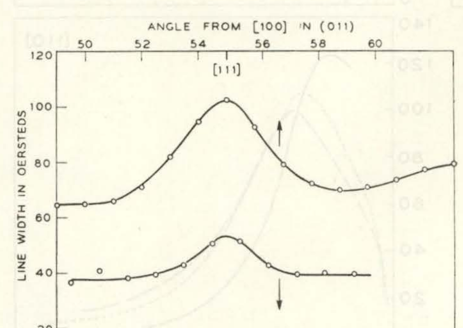

Fig. 3. Line width versus angle near [111] and [100] for YIG (0.02% Tb) as measured at 23°K. The frequency was 20.8 kMc/sec.

the linewidth as a function of angle near these two directions at a temperature such that a reasonable difference in population might be expected. The results are given in Fig. 3. The temperature was chosen out of convenience rather than as being that corresponding to the largest population difference. It will be seen that the line width increases by 50% along [111], and the peak is perhaps 3° wide. Along [100] the line width increases by about 30%, and the width is about 2°. Similar line width versus angle measurements made at 1.5°K or 295°K showed no such narrow peaks. Clearly, a large fraction of the line width at 23°K is associated with the close approach of levels well above the lowest level of the multiplet.

Conclusion

We conclude that at least in the case of Tb doped YIG the general character of the line width variation with temperature and frequency can be accounted for by assuming that the rare earth magnetization relaxes with a number of relaxation times, some of which are comparable with the microwave frequency. In addition, the excitation of direct transitions between closely approaching excited states of the rare earth ground multiplet produces a significant effect at intermediate temperatures. The data do not seem to be at all compatible with the fast relaxation theory given by DeGennes, Kittel, and Portis[8].

References

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DISCUSSION

J. C. Slonczewski: Is it possible to associate the calculated energy levels with a crystal field symmetry higher than that actually present which would have the effect of replacing the near approaches by actual crossings?

L. R. Walker: The only symmetry assumed was that of the site, namely orthorhombic. We have not examined the best field used to see whether it approximates one of higher symmetry or not.