With mismatches of the sort encountered in practical applications (typically less than two to one), an isolator using the materials discussed in this paper should function satisfactorily at peak power levels of several megawatts, if the necessary precautions are taken to suppress the first order nonlinear effect. Experiments using typical isolator configurations instead of the resonance cavity have shown that the onset of nonlinear effects in isolators can be predicted with fair accuracy from cavity measurement.

A further improvement of the power handling capacity of microwave ferrites appears quite feasible. In particular, it is not difficult at all to increase the linewidth and thus increase the critical field (h_{crit} or h_∞). This is not a promising line of attack, however, since the ratio of reverse to forward attenuation of resonance isolators decreases rapidly with increasing linewidth.[17] To improve the power-handling capacity one must, therefore, according to (5), decrease the saturation magnetization and/or increase the spin-wave linewidth ΔH_k. This can generally be achieved by suitable substitution of the magnetic ions in ferrites of the spinel and garnet type.

ACKNOWLEDGMENT

The authors would like to thank L. G. Rubin for his help in constructing the equipment used in this work. They are also indebted to Dr. S. L. Blum, D. A. Belforti and Miss S. Georgian for the preparation of the materials and for preliminary measurements.

[17] B. Lax, "Frequency and loss characteristics of microwave ferrite devices," Proc. IRE, vol. 44, pp. 1368–1386; October, 1956.

High Power Ferromagnetic Resonance at X-Band in Polycrystalline Garnets and Ferrites*

J. J. GREEN† AND E. SCHLÖMANN‡

Summary—**Resonance experiments have been performed at X-band on spherical samples of polycrystalline yttrium garnet, yttrium-gadolinium garnet, yttrium-holmium garnet and nickel-cobalt ferrite. The RF field strength extended up to 60 Oersted. In the case of yttrium garnet the samples differed considerably in density and hence in linewidth. At fairly low power levels the susceptibility at resonance varies linearly with the square of the RF magnetic field strength. At high power levels the susceptibility is inversely proportional to the amplitude of the microwave magnetic field. The "spin-wave linewidth" ΔH_k is inferred by extrapolation from the behavior at very high powers. It is found that ΔH_k is, to a large extent, independent of the linewidth ΔH observed by the usual low power experiments. In particular ΔH_k was found to be essentially the same (approximately 4 Oe) for all yttrium iron garnets (single crystals and polycrystals with linewidth varying between 1.8 Oe and 450 Oe). On the other hand, ΔH_k increases very rapidly if the yttrium is partially substituted by holmium ($\Delta H_k \sim 11$ Oe for 1 per cent substitution.)**

I. Introduction

It was explained in the preceding paper[1] that at X-band frequencies and for conventional ferrimagnetic materials, the saturation of the resonance line involves the excitation of spin waves which have the same frequency as the signal (second-order nonlinear process). In addition, at these frequencies, one observes a subsidiary absorption peak below the main resonance which is attributed to the excitation of spin waves with frequencies equal to half the signal frequency (first-order nonlinear process). For spherical samples of the materials investigated in this paper the subsidiary peak lies considerably below the main resonance and (for this reason) becomes noticeable only at quite high power levels. This means that one never encounters a situation in which both nonlinear processes (first and second order are simultaneously important. Experiments at X-band are, therefore, better suited for a detailed investigation of the second-order nonlinear process than the L-band experiments reported in the preceding paper.[1]

The materials used in this investigation belong to four different families. One of these families comprises yttrium iron garnets of varying densities and hence varying linewidths (between 47 and 450 Oe). Two other families are derived from yttrium iron garnet by partial substitution of gadolinium or holmium for the yttrium. The fourth family is derived from nickel ferrite by partial substitution of cobalt for nickel.

II. Experimental Procedure

Ferromagnetic resonance measurements were made at 9250 mc using a TE_{202} transmission cavity. The samples were mounted on a quartz rod and placed in the cavity at a point of maximum RF magnetic field. Sample absorption as a function of dc magnetic field was determined from the change in incident power necessary to maintain a constant transmitted power. The magnetic field strength at the sample was obtained from

* Manuscript received by the PGMTT, August 3, 1959. The portion of the work performed by J. J. Green was supported by the AF Cambridge Research Center, Contracts AF 19(604)-1084 and AF 19(604)-5487.

† Gordon McKay Lab., Harvard University, Cambridge, Mass.
‡ Research Div., Raytheon Co., Waltham, Mass.

[1] E. Schlömann, J. Saunders, and M. Sirvetz, "L-band ferromagnetic resonance experiments at high peak power levels," this issue, pp. 96–100.

a knowledge of incident power, cavity coupling, loaded Q and the cavity mode. This was done at one power level and all other power levels were compared to this reference level by means of a calibrated attenuator. The duty cycle was always less than 5×10^{-5}. At power levels where heating effects might occur, single pulses of one microsecond duration were used. A 2J51 tunable magnetron provided the microwave power which was sufficient to obtain cavity fields up to 60 Oe. Calibrated attenuators preceding each crystal made it possible to take readings with the crystals always at the same power level. All measurements were performed at room temperature on spherical samples. With increasing power level a slight shift of the resonance was observed in many cases. If this happened, the dc magnetic field was adjusted for resonance at each power level.

III. Results

Fig. 1 shows a set of typical saturation curves (susceptibility at resonance as a function of the amplitude of the microwave magnetic field on a logarithmic scale). The two curves which saturate at the lowest power levels (curves a and b) are obtained from measurements on single crystals of yttrium iron garnet. The two crystals differed in their surface treatment, since one (a) was polished (and had a linewidth of 3.9 Oe); and the other (b) had a rough surface (and a linewidth of 9.1 Oe). Curves (c) and (d) are typical saturation curves obtained with polycrystalline yttrium iron garnet and holmium-substituted yttrium iron garnet, respectively. In these cases the saturation occurs at higher power levels as might be expected in view of the larger linewidth. Curve (e) is a simplified theoretical curve. It was obtained from the results described in the previous paper by matching the two theoretical expressions (expected to be valid at moderate and high powers, respectively) in such a way that the resulting curve had a continuous slope.[2] It is seen that the theoretical curve can be fitted moderately well to any of the experimental curves by sliding it along the horizontal axis. A good fit, however, is not obtainable in this way. This is not surprising since the behavior of χ'' at moderate and at very high power levels is determined by different physical consideration. In order to obtain a good fit one needs at least two adjustable parameters, like C and h_∞ of the preceding paper.[1]

In order to demonstrate the validity of the theoretical formulas derived for moderate and very high power levels [(3) and (4) of the preceding paper[1]] the susceptibility should be plotted on a linear scale vs h^2 and vs $1/h$. The phenomenological parameters C and h_∞ can be obtained from such plots by simple extrapolation. In the present paper, we shall concentrate on the behavior at very high powers and hence use primarily the $1/h$ representation. Fig. 2 shows two typical sets of measurements obtained on polycrystalline yttrium

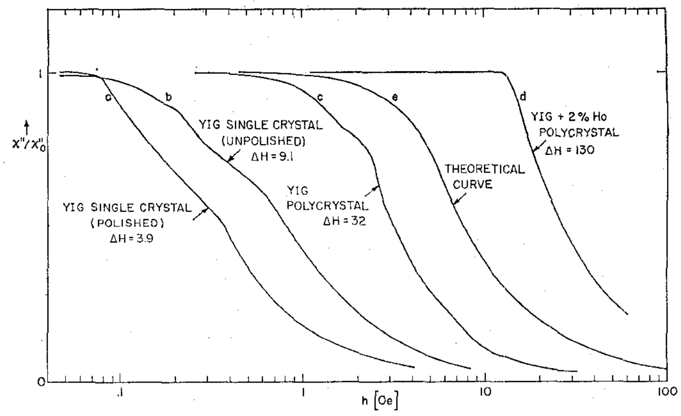

Fig. 1—Saturation curves for various yttrium garnets.

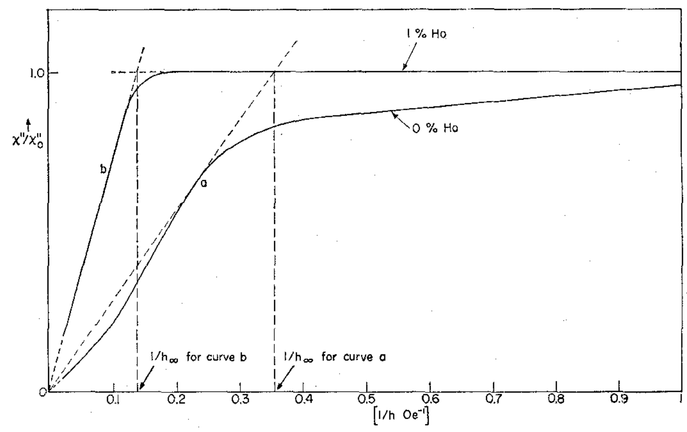

Fig. 2—Saturation curves for polycrystalline yttrium garnet and holmium-substituted yttrium garnet.

garnet (a) and yttrium-holmium garnet (b). It is seen that the latter obeys the $1/h$-law quite well almost over the complete nonlinear range. The former, however, shows appreciable deviations from the theoretical $1/h$-law. In cases such as these the phenomenological parameter h_∞ was obtained by drawing a tangent in the way indicated in the figure. It should be noticed that for any point on the curve (χ'' vs $1/h$) the slope of a line connecting it with the origin is proportional to the amplitude of the precessing magnetic moment. The angle between the precessing magnetic moment and the direction of the dc field is (in radians and using the notation of the previous paper[1])

$$u_0 = \frac{\chi''}{\chi_0''} \frac{h}{\Delta H} \cdot \tag{1}$$

Here h is the amplitude of the RF magnetic field in the absence of the sample and the RF field is assumed linearly polarized. χ'' is the imaginary part of the effective susceptibility (which relates the magnetization of the sample to the magnetic field outside the sample). ΔH is the linewidth (total width). In case (a) of Fig. 2, the amplitude u_0 reaches a maximum with increasing power level and then decreases again. The maximum of u_0 is obviously given by

$$u_{0_{\max}} = \frac{h_\infty}{\Delta H} \cdot \tag{2}$$

[2] This means that the two curves are joined at the point at which χ''/χ_0'' equals $\frac{2}{3}$, and that $C(h_\infty/\Delta H)^2 = 4/27$ in the notation of the preceding paper.

According to (5) of the preceding paper,[1] $u_{0_{max}}$ is related to the "linewidth" of the unstable spin waves by

$$u_{0_{max}}{}^2 = \frac{\Delta H_k}{4\pi M_s}. \tag{3}$$

The experimental results obtained with polycrystalline yttrium garnet are summarized in Fig. 3. The linewidth of these materials varied between 47 Oe and 450 Oe. It increases with decreasing density in an approximately linear fashion. This effect has been observed previously and is attributed to the presence of stray fields produced by nonmagnetic inclusions (such as pores).[3–6] Fig. 3 shows that for these materials the critical field h_∞ is very nearly proportional to the linewidth. Measurements of h_∞ on single crystals (with linewidths ranging between 1.8 Oe and 9.1 Oe) gave results which also agree quite well with the straight line drawn in the figure. We thus conclude that the maximum amplitude of the precessing magnetic moment is independent of the porosity. This implies that the spin wave linewidth ΔH_k is also independent of the porosity.[7]

Fig. 4 summarizes the experimental results obtained with gadolinium-substituted and holmium-substituted yttrium garnet. Here the square of the maximum amplitude is plotted as a function of the gadolinium and holmium content (characterized by the composition parameter t). The cluster of points at $t=0$ represents the yttrium garnet data, which was also shown in Fig. 3. It is seen from Fig. 4 that $u_{0_{max}}{}^2$ increases with increasing gadolinium and holmium content. In the case of the gadolinium-substituted yttrium garnet most of the increase is attributable to the decrease in the magnetic moment. Pure gadolinium garnet has a very small saturation magnetization at room temperature, and, it is known that in the yttrium-gadolinium garnets the saturation magnetization decreases approximately linearly with the gadolinium content. At $t=1$ the saturation magnetization is, therefore, roughly half as large as it is at $t=0$. Fig. 4 shows that between $t=0$ and $t=1$ the square of the maximum amplitude increases by a factor somewhat larger than two. Thus ΔH_k has also increased but only by a rather small factor.

If holmium rather than gadolinium is substituted for the yttrium, a much more rapid increase of $u_{0_{max}}{}^2$ is observed. In this case the saturation magnetization is practically independent of the composition in the range of interest, so that the increase in $u_{0_{max}}{}^2$ must be attributed to an increase in the linewidth ΔH_k of the unstable spin waves. For yttrium garnet ΔH_k is approximately 4 Oe and it increases with holmium content at the rate of approximately 7 Oe per cent.

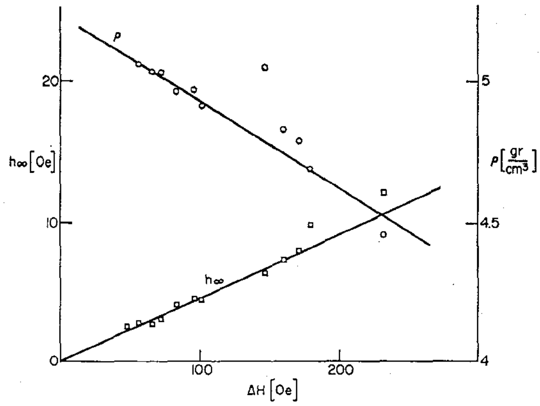

Fig. 3—High power effects in polycrystalline yttrium garnet. The critical field strength h_∞ is proportional to the linewidth ΔH, which increases with decreasing density ρ. The same dependence of h_∞ on ΔH was also observed in single crystals.

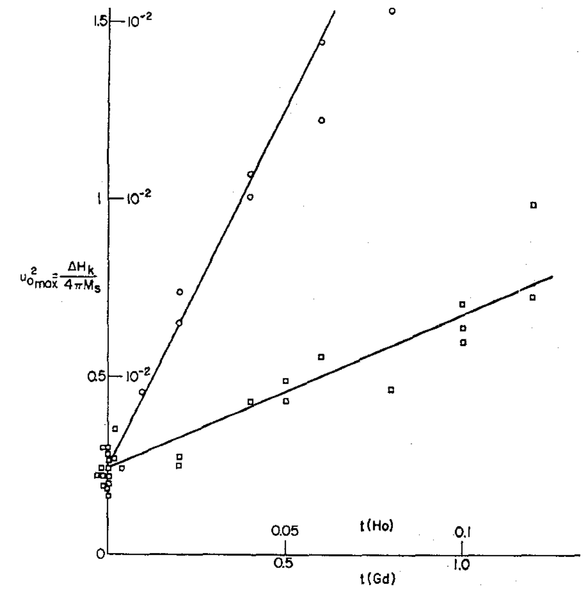

Fig. 4—Maximum amplitude of the uniform mode for polycrystalline samples of $5Fe_2O_3 \cdot 3Y_{2-t}Me_tO_3$ where Me=Gd (□) or Ho (○) as a function of the composition. Note the difference in the t-scales.

The last family of materials to be described in this paper consists of cobalt-substituted nickel ferrites. It is known that in these materials the linewidth first decreases with cobalt content, then reaches a minimum at approximately 3 per cent substitution, and finally increases again. This has been attributed to the fact that small cobalt substitutions tend to decrease the effects of crystalline anisotropy.[3,8,9] Since nickel ferrite has a

[3] E. Schlömann, "The microwave susceptibility of polycrystalline ferrites in strong dc fields and the influence of nonmagnetic inclusions on the microwave susceptibility," *Proc. Conf. on Magnetism and Magnetic Materials*, Boston, Mass., October, 1956.

[4] G. P. Rodrique, J. E. Pippin, W. P. Wolf, and C. L. Hogan, "Ferrimagnetic resonance in some polycrystalline rare earth garnets," IRE Trans. on Microwave Theory and Techniques, vol. MTT-6, pp. 83–90; January, 1958.

[5] W. P. Wolf and G. P. Rodrique, "Preparation of polycrystaline ferrimagnetic garnet materials for microwave applications," *J. Appl. Phys.*, vol. 29, pp. 105–108; January, 1958.

[6] J. Snieder, "Ferromagnetic resonance in polycrystalline ferrites," *Appl. Sci. Res.*, vol. B7, pp. 185–232; 1958.

[7] The saturation magnetization varies only by a small amount in this series.

[8] M. H. Sirvetz and J. H. Saunders, "Resonance widths in polycrystalline nickel-cobalt ferrites," *Phys. Rev.*, vol. 102, pp. 366–337; April 15, 1956.

[9] J. E. Pippin and C. L. Hogan, "Resonance measurements on nickel-cobalt ferrites as a function of temperature and on nickel ferrite-aluminates," IRE Trans. on Microwave Theory and Techniques, vol. MTT-6, pp. 77–82; January, 1958.

negative anisotropy constant K_1 and cobalt ferrite has a very large positive one, a compensation of the crystalline anisotropy is expected from a substitution in the range of a few per cent. Fig. 5 shows ΔH and $u_{0_{max}}{}^2$ as a function of the composition. The linewidth ΔH shows the extraordinary dependence on the cobalt content, which was described above. $u_{0_{max}}{}^2$ however, (and, therefore, ΔH_k since M_s is nearly constant) increases approximately linearly with the cobalt content.

IV. Discussion

The nonlinear effects discussed in this paper have important practical applications. The power-handling capacity of isolators and circulators is in large measure determined by the onset of nonlinearity. The optimum performance of ferrite parametric amplifiers depends on the high power properties of the materials used in such devices.[10] In most cases the nonlinear effects must be avoided but in some cases they can be used advantageously, as in parametric amplifiers and limiters. It is, therefore, very important to understand the factors that influence the onset of nonlinearities and to be able to control these factors.

The critical field h_∞, which characterizes the onset of nonliner effects can obviously be increased by increasing the linewidths ΔH and ΔH_k and by decreasing the saturation magnetization. In practically all applications a small linewidth ΔH is desirable. A useful improvement in the power-handling capacity can, therefore, be obtained only by increasing the spin-wave linewidth ΔH_k or by decreasing the saturation magnetization. The present experiments have shown that ΔH and ΔH_k very often differ by an order of magnitude and that, to a certain extent, they can be controlled independently.

The fact that ΔH_k is approximately independent of the porosity (and independent of the surface roughness of single crystal samples), suggests that this linewidth is characteristic of the intrinsic losses of the material. It has previously been observed by Le Craw and Spencer[11] that in single crystals of yttrium garnet ΔH_k is essentially independent of the surface roughness, whereas ΔH increases rapidly with the surface roughness.[12]

The fact that in the yttrium-holmium garnet ΔH_k increases rapidly with the holmium content can be understood in terms of a theory recently developed by

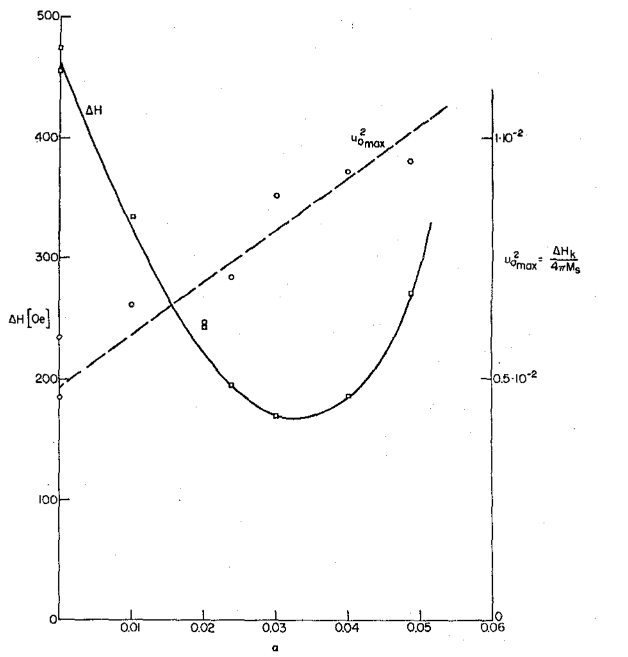

Fig. 5—Linewidth and maximum amplitude of the uniform mode for spherical samples of $Ni_{1-\alpha}Co_\alpha Mn_{0.02}Fe_{1.9}O_4$ as a function of the composition.

de Gennes, et al.[13] According to this theory the magnetic moment of the rare earth ions (with the exception of gadolinium) relaxes extremely rapidly because of strong spin orbit interaction. For this reason small amounts of any rare earth metal (with the exception of gadolinium) in yttrium garnet should increase ΔH_k very rapidly. ΔH should be effected in essentially the same way, but for small substitution the increase in ΔH is not as readily and unambiguously observable because the linewidth is still predominantly caused by inhomogeneity broadening. Experiments on samarium substituted yttrium garnet have qualitatively confirmed the theoretical expectations.

Similar arguments apply to the nickel-cobalt ferrite series. The magnetic moment of the cobalt ion is believed to relax appreciably faster than that of the nickel ion.[14] This can be inferred from the large linewidth observed in single crystal cobalt ferrite[15,16] and is also consistent with the large crystalline anisotropy of this material.

Acknowledgment

One of the authors (J. J. Green) is particularly indebted to Dr. L. C. Hogan for his suggestion to investigate the high power phenomena in ferrites and garnets. Both authors would like to acknowledge many stimulating discussions with Prof. R. V. Jones and Dr. M. H. Sirvetz. The technical assistance of Mrs. L. Lin, F. Molea, and C. Quadros is also gratefully acknowledged.

[10] R. W. Damon and J. R. Eshbach, "Theoretical limitations of ferrite parametric amplifier performance," presented at PGMTT meeting, Cambridge, Mass., June 1–3, 1959; G. E. Tech. Rept. 59-RL-2230E.

[11] R. C. Le Craw and E. G. Spencer, "Surface-independent spin-wave relaxation in ferromagnetic resonance of yttrium iron garnet," *J. Appl. Phys.*, vol. 30, pp. 185S–186S; April, 1959.

[12] In Le Craw and Spencer, *Ibid.*, ΔH_k is determined from the first onset of nonlinear effects, rather than by extrapolation from the high power behavior as in the present paper. The procedure followed in Le Craw and Spencer leads to much smaller values of ΔH_k, and it is believed to be in error, since it does not take into account the influence of inhomogeneities on the high power behavior. In the present context the difference between the two methods of obtaining ΔH_k is not very important, because the two values generally differ by a constant factor.

[13] P. G. deGennes, C. Kittel and A. M. Portis, to be published.

[14] R. L. White, "Ferromagnetic resonance line widths and g-factors in ferrites," *Phys. Rev. Letters*, vol. 2, pp. 465–466; June 1, 1959.

[15] J. O. Artman, "Microwave resonance relations in anisotropic single crystal ferrites," Proc. IRE, vol. 44, pp. 1284–1293; October, 1956.

[16] P. E. Tannenwald, "Multiple resonances in cobalt ferrites," *Phys. Rev.*, vol. 99, pp. 463–464; July 15, 1955.