Effect of Grain Size on the Microwave Properties of Polycrystalline Yttrium Iron Garnet

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Data on the parallel pump and subsidiary absorption threshold at 9.1 GHz and effective linewidth ΔH_{eff} at 9.9 GHz have been obtained for polycrystalline YIG materials with an average grain diameter a_0 from 1 to 30 μ. The fine-grain samples (1–10 μ) were made by hot pressing. The parallel pump spin-wave linewidths extrapolated to zero wavenumber, $\Delta H_{k\to 0}$, vary as a_0^{-1}, with $\Delta H_{k\to 0}=25$ Oe at $a_0=1$ μ. The ΔH_k for the fine-grain samples ($a_0\approx 1$ μ) show an inverse k-dependence. A simple model in which the spin-wave transit time across individual grains limits ΔH_k appears to explain the a_0^{-1} dependence but not the observed k-dependences for ΔH_k. The high-field ΔH_{eff} is 5.5 Oe for $a_0=1$ μ and varies as $a_0^{-1/2}$. The low-field values are slightly larger, due to a small two-magnon contribution arising from coupling to degenerate short-wavelength spin waves. Even though the high-field ΔH_{eff} contains no two-magnon contribution, it is significantly larger than linewidths in single crystals. A recently proposed magnetostriction mechanism is consistent with the $a_0^{-1/2}$ dependence, but the predicted magnitude of ΔH_{eff} is much smaller than observed.

I. INTRODUCTION

It is generally well known that fine graining can improve the power-handling capability of polycrystalline microwave ferrites.[1] However, the numerous studies of grain-size influences on microwave properties[1–7] have not provided any clear understanding of the physical mechanisms which are involved. These investigations were carried out (a) at various stages of the evolution of present-day ferrite technology, (b) on a large variety of different materials, (c) under widely varied experimental conditions, and (d) with emphasis on different properties. In the present investigation a more unified picture of grain-size influences is obtained by concentrating on a single material, polycrystalline yttrium iron garnet, with good intrinsic microwave properties and a well-understood microstructure,[8] and by obtaining extensive microwave data on samples prepared in a similar manner but with a wide range of grain size.

Parallel pump and subsidiary absorption butterfly curves, resonance linewidth, off-resonance effective linewidth, and magnetization were measured for samples with an average grain diameter a_0 from 1–30 μ. The thresholds show an a_0^{-1} dependence and the butterfly curves for a_0 in the 1-μ range have no minimum. The off-resonance effective linewidth varies as $a_0^{-1/2}$ and is substantially larger than single-crystal linewidths. After a brief description of the experimental procedures in the next section, the high-power data are presented and discussed in Sec. III, and the effective linewidth results are considered in Sec. IV.

II. EXPERIMENT

The microwave measurements were performed on spherical samples of polycrystalline YIG material made by hot pressing conventionally prepared powders. In the powder preparation yttrium oxide with less than one part in 10^7 residual rare-earth impurities was used. The basic material was single-phase polycrystalline YIG with good microwave properties, as indicated by a resonance linewidth ΔH of 20 Oe and a spin-wave (parallel pump) linewidth ΔH_k of 0.6 Oe at 9.1 GHz on a large-grain sample. One series of samples was obtained by hot pressing the powder to obtain ceramic pieces with a 1-μ grain size and then annealing small cubes cut from the piece to obtain the final desired grain size. After determining the average grain diameter a_0 from photographs of the etch pattern on polished surfaces of the cubes, spheres were ground for microwave measurements. A second series of samples was obtained by eliminating the annealing operation and hot pressing directly to the final grain size.

TABLE I. Average grain diameter $a_0(\mu)$, average saturation induction $4\pi M$(G), and resonance linewidth ΔH(Oe) at 9.1 GHz for the YIG materials used in the present investigation.

Sample	Average grain diameter a_0 (μ)	Saturation induction $4\pi M$ (G)	Resonance linewidth ΔH (Oe)
HP1.0	1.0	1709	40
HP3.1	3.1	1752	83
HP3.4	3.4	1755	28
HP9.0	9.0	1810	29
HP10.1	10.1	1815	27
HA2.4	2.4	1700	105
HA3.1	3.1	1707	191
HA3.2	3.2	1775	225
HA3.4	3.4	1732	179
HA5.2	5.2	1700	93
HA9.7	9.7	1670	206
CF30.1	10.1	1756	20

The following magnetic data were obtained: average saturation induction $4\pi M$, resonance linewidth ΔH at 9.1 GHz, parallel pump and subsidiary absorption threshold h_{crit} for static fields between 600 and 2200 Oe at 9.1 GHz, and effective linewidth ΔH_{eff} as a function of field from 600 to 7000 Oe at 9.9 GHz. The magnetic induction was measured with a vibrating sample magnetometer. The spin-wave thresholds were determined from susceptibility data obtained with a standard reflection cavity spectrometer and a pulsed

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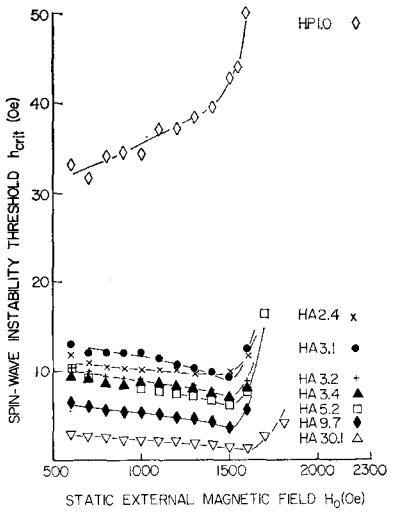

FIG. 1. Spin-wave instability threshold h_{crit} as a function of the static external magnetic field H_0 for parallel pumping at 9.1 GHz for the HA series samples.

magnetron source at 9.1 GHz.[9] The pulse duration and repetition rate were 4 μsec and 60 Hz, respectively. Effective linewidth values were calculated from susceptibility data as discussed in Ref. 10. Increased sensitivity for the susceptibility measurements was achieved by stabilizing the reference microwave source with a crystal oscillator, stable to one part in 10^8, and using a high-Q TE_{011} cylindrical cavity ($Q \approx 20\,000$). Changes in the 3-dB frequency width of the cavity as small as 100 Hz could be measured. In addition, an extrapolation procedure was used to determine the cavity parameters at infinite field for use in the perturbation calculation for susceptibility. With these improvements in the instrumentation and procedures, the errors in ΔH_{eff} were less than 0.5 Oe for a 2-mm-diam sample.

The average grain size, saturation induction ($4\pi M$ extrapolated to infinite field), and 9.1-GHz resonance linewidth for the materials are listed in Table I. The labels HP1.0, HP3.1, etc. denote samples which were hot pressed directly to the final grain size with no subsequent annealing treatment. The labels HA2.4, HA3.1, etc. denote samples which were hot pressed to obtain an average grain diameter a_0 of about 1 μ and then annealed at 1400°C to achieve the desired grain size. CF30.1 denotes a conventionally fired large-grain size (30.1 μ), narrow-linewidth YIG. The numbers following the HP, HA, and CF labels denote the average grain diameters.

Several differences were noted between the HP and HA series materials. Annealing after hot pressing appears to induce porosity in the material, as indicated by the relatively large 100–200-Oe linewidths for the HA series samples. This porosity was also evident from density measurements on the individual spherical samples. The HP series materials, which were hot pressed directly to the final grain size, have much smaller linewidths, on the order of 30 Oe. The instability threshold h_{crit}, spin-wave linewidth ΔH_k, and off-resonance effective linewidth ΔH_{eff} were also slightly smaller for the HP series samples. The important point to be emphasized here is that other factors besides the average grain size are important. The manner in which a particular microstructure is obtained has an important influence on the microwave properties of polycrystals.

III. PARALLEL PUMP THRESHOLD AND SPIN-WAVE LINEWIDTH

The parallel pump threshold h_{crit} is shown as a function of static external field for the HA series samples in Fig. 1 and for the HP materials in Fig. 2. Data on the smallest grain-size and the largest grain-size samples, HP1.0 and CF30.1 are shown in both figures for comparison. Similar data were obtained for subsidiary absorption. These data clearly show that the threshold increases substantially as the grain size is reduced. The h_{crit} for the 1-μ-grain-size sample (HP1.0) is over thirty times greater than the threshold for the 30-μ-grain-size material (CF30.1). Furthermore, the shape of the butterfly curve (h_{crit} versus static field H_0) is quite different for HP1.0. The curve does not exhibit the usual minimum near $H_0 = 1500$ Oe. Instead, h_{crit} increases monotonically with H_0. The data for HP3.1 and HP3.4 in Fig. 2 show a similar but less pronounced behavior. None of the HA series samples show this effect. It appears that the effect is related to the small grain size but is eliminated by the grain growth which occurs during annealing.

The different shapes of the butterfly curves for the small-grain samples (HP1.0, HP3.1, HP3.4) lead to a spin-wave linewidth k (wavenumber) dependence that is much different than observed for large-grain samples. The ΔH_k for the data in Fig. 2 are shown as a function

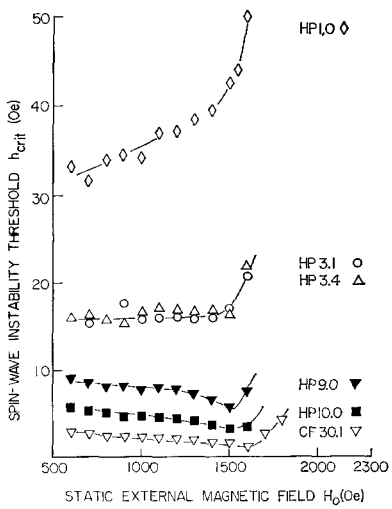

FIG. 2. Spin-wave instability threshold h_{crit} as a function of the static external magnetic field H_0 for parallel pumping at 9.1 GHz for the HP series samples.

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of k in Fig. 3. The curves were obtained by applying parallel pump instability theory[11] and making the usual assumption that the instability occurs for spin-waves propagating at right angles to the static field.[12] The ΔH_k for the small-grain sample (HP1.0) has a large inverse k-dependence, ΔH_k for the intermediate-grain-size samples (HP3.1, HP3.4) has a weak inverse k-dependence, and the ΔH_k for the large-grain samples (HP9.0, HP10.1, CF30.1) has the usual weak linear k-dependence.

These results lead to a number of interesting observations about the physical origin of ΔH_k in fine-grain materials. In Fig. 4, a log-log plot of ΔH_k extrapolated to $k=0$ ($\Delta H_{k\to 0}$) as a function of the average grain diameter a_0 shows that

$$\Delta H_{k\to 0} \propto a_0^{-1}. \tag{1}$$

This result can be qualitatively explained by a model, recently proposed by Vrehen *et al.*[13] in which the lifetime of the excited spin wave is limited by the transit time for the mode across an individual grain, $\tau_k \approx a_0/|v_g|$, where v_g is the spin-wave group velocity. The transit-time-limited linewidth $\Delta H_k^{(t)}$ is given by

$$\Delta H_k^{(t)} = 1/\gamma\tau_k = |v_g|/\gamma a_0. \tag{2}$$

The a_0^{-1} dependence of $\Delta H_{k\to 0}$ indicates that the model is qualitatively correct and that $|v_g|$ for the minimum k mode which can be excited does not change appreciably with grain size. Assuming that the $\Delta H_{k\to 0}$ in Fig. 4 arise from a transit-time mechanism, Eq. (2) gives values of $|v_g|$ on the order of 10^4 cm/sec.

Since $v_g = \nabla_k \omega_k$, where ω_k is the spin-wave frequency, the transit-time model can also be used to calculate the k-dependence of $\Delta H_k^{(t)}$ through the usual dispersion relation[14] for ω_k. The group velocity is given by

$$v_g = 2\gamma D k\mathbf{k} + \gamma 4\pi M \sin\theta_k \cos\theta_k/k\boldsymbol{\theta}, \tag{3}$$

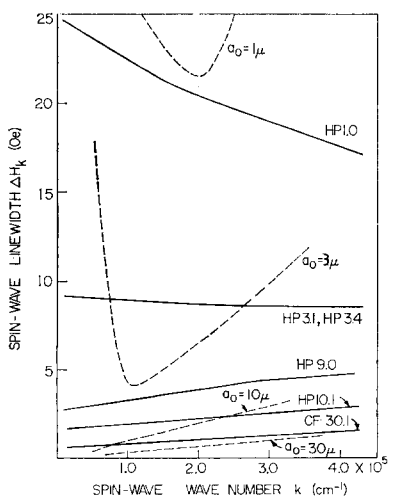

FIG. 3. Spin-wave linewidth ΔH_k as a function of wavenumber k for parallel pumping at 9.1 GHz, calculated from the data in Fig. 2.

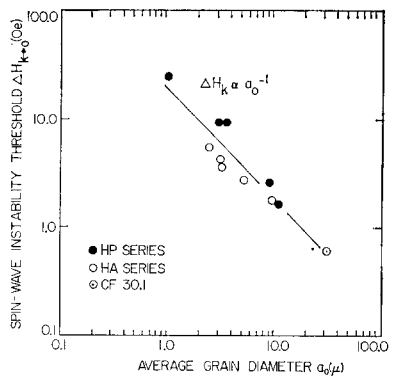

FIG. 4. Parallel pump spin-wave linewidth ΔH_k extrapolated to $k=0$ as a function of the average grain diameter a_0. The data were obtained at 9.1 GHz.

where $\mathbf{k}$ and $\boldsymbol{\theta}$ are unit vectors in the $\mathbf{k}$ and $\mathbf{k}\times(\mathbf{k}\times\mathbf{H}_0)$ directions, respectively, $D=5\times10^{-9}$ Oe-cm^2 is the exchange parameter for YIG, $\gamma=1.76\times10^7$ Oe^{-1} sec^{-1} is the gyromagnetic ratio, and $4\pi M=1750$ G is the saturation induction. Even though $\theta_k \approx \pi/2$ is usually assumed for parallel pumping, the case of interest here, the deviation of θ_k from $\pi/2$ because of the nonzero k-component in the field direction (k_z) leads to a significant v_g contribution from the $\hat{\theta}$ term in Eq. (3). Because of the finite size of the region supporting the mode, k_z can never be zero. In the present independent grain model $k_z \approx 2\pi/a_0 \approx 10^5$ cm^{-1} is a reasonable assumption. When k is on the order of k_z, the second term in Eq. (3) contributes significantly to the transit-time linewidth. Equations (2) and (3) have been used to calculate $\Delta H_k^{(t)}$ versus k for $k_z \approx 2\pi/a_0$ and the results are indicated by the broken lines in Fig. 3. One can see that the agreement is rather poor. The theory for $a_0=1$ μ and $a_0=3$ μ gives distinct minima in the 1–2×10^5 cm^{-1} range and the ΔH_k increase sharply at higher k-values. The theoretical curves for $a_0=10$ μ and $a_0=30$ μ have the right k-dependence, but the increase is more rapid than indicated by the data.

It is not clear, however, that a comparison of the ΔH_k obtained from parallel-pump data with the $\Delta H_k^{(t)}$ from the transit time model is entirely legitimate. If Eqs. (2) and (3) are applied to the $\Delta H_{k\to 0}$ data in Fig. 4 to determine the actual lower limit on k ($k_{\min}$) for the unstable modes, $k_{\min}$ turns out to be about 2×10^5 cm^{-1} independent of grain size. This means that the portions of the experimental curves for $k<2\times10^5$ cm^{-1} are meaningless. These values of $k_{\min}$, moreover, correspond to a wavelength λ_k in the 0.5-μ range. If the transit-time approach is correct, it appears that a submicron-scale inhomogeneity operates to limit the wavelength of the unstable modes. It should also be noted that the instability theory[11] was developed under the assumption that the dimensions of the region supporting the mode were much larger than the wavelengths of the relevant spin waves. If the modes are confined to single grains with a_0 in the micron range

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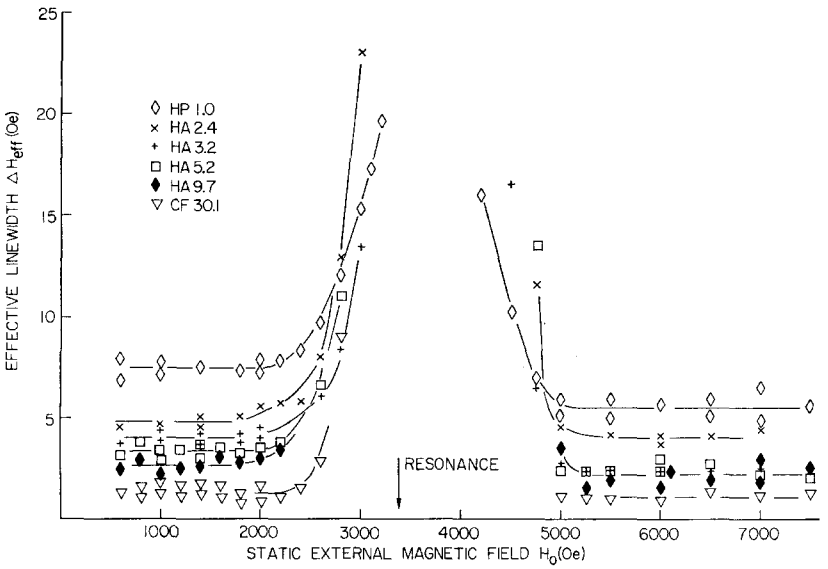

FIG. 5. Effective linewidth ΔH_{eff} as a function of static external magnetic field H_0. The ΔH_{eff} were obtained from susceptibility data at 9.9 GHz.

as assumed in the present model, this assumption is no longer justified. On the positive side, it should be pointed out that k-dependences for ΔH_k which are qualitatively similar to the theoretical curves in Fig. 3 have been observed for porous YIG materials.[15] In fact, a 20% porous sample with $a_0 \approx 1$ μ exhibits a ΔH_k versus k with a minimum value of 15 Oe at $k = 1.7 \times 10^5$ cm^{-1}. It is to be expected that transit-time effects should be more apparent in porous samples, where the grains are separated, than in dense materials.

Vrehen *et al.*[13] reported that a transit-time approach qualitatively explains parallel-pump data for hot-pressed magnesium–manganese ferrites with $a_0 < 1$ μ. However, they considered only the $\hat{\mathbf{k}}$ component of $\mathbf{v}_g$ in Eq. (3). For $k_z \approx 5 \times 10^4$ cm^{-1}, the $\hat{\boldsymbol{\theta}}$ component of v_g is quite large, especially for low k. The $\hat{\boldsymbol{\theta}}$ component is very important for $a_0 < 1$ μ and cannot be neglected.

Several of the early investigations alluded to two-magnon scattering as a likely source of the high thresholds in fine-grain materials.[1,4,5] The basic argument was that the coupling between degenerate modes will lead to a broadening of ΔH_k in much the same way that porosity and anistropy-induced scattering increases the uniform and precession resonance linewidth in polycrystals. It has been recognized for some time, however, that two-magnon interactions should not have a large effect on spin-wave linewidths.[14] A recent theoretical analysis by Sage[16] shows that the maximum increase in the threshold caused by two-magnon interactions is roughly a factor of 2 for YIG. More detailed analyses indicate that for polycrystalline ferrite materials the increase under normal circumstances should be, at most, a few percent,[17] as compared to the increase by a factor of 30 for the present data. Moreover, data for porous YIG [15] strongly suggest that two-magnon interactions do not influence the parallel pump ΔH_k even for materials where the contribution to the resonance losses is extremely large.

IV. EFFECTIVE LINEWIDTH

The effective-linewidth results are summarized in Fig. 5. A detailed discussion of the meaning of ΔH_{eff} is presented in Ref. 10. Essentially, ΔH_{eff} expresses the microwave losses in linewidth units and is useful in the comparison of off-resonance loss data with conventional linewidth results. In the vicinity of resonance (indicated by the arrow in Fig. 5), ΔH_{eff} is on the order of ΔH (see Table I). The present data show that far from resonance the effective linewidth is quite small compared to ΔH. Below 2000 Oe and above 5000 Oe, ΔH_{eff} is field independent. The low-field values are slightly larger than the high-field values. The off-resonance ΔH_{eff} does exhibit a significant dependence on grain size. The low-field ΔH_{eff} decreases from 7.5 Oe for HP1.0 to 1.5 Oe for CF30.1.

The large increase in ΔH_{eff} for the field interval 2000–4500 Oe is due to two-magnon interactions between the uniform precession mode at the driving frequency and degenerate long-wavelength spin waves. It is presumably due to the residual porosity (which was quite large in some of the HP series samples) and the anisotropy in the randomly oriented crystallites of the polycrystal. The effective linewidth in this field region of spin-wave degeneracy or near degeneracy with the uniform precession has been discussed extensively by several authors[10,18,19] and will not be considered further.

When the sample is biased at high field there are no spin-waves degenerate with the uniform precession mode and two-magnon interactions cannot contribute to the losses. Consequently, the high-field ΔH_{eff} is a measure of the microwave losses in the absence of any

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two-magnon contribution. Even so, the lowest ΔH_{eff} observed, 1 Oe for CF30.1, is still substantially larger than single-crystal linewidths, which are on the order of 0.5 Oe. This high-field ΔH_{eff} also exhibits a strong grain-size dependence. It varies as $a_0^{-1/2}$, compared to the a_0^{-1} dependence for ΔH_k. The small increase in ΔH_{eff} at low field, compared to the high-field values, is probably due to a small two-magnon contribution because of the high-k modes which are degenerate with the uniform precession at low field. For H_0 less than 2000 Oe, the wavelengths of the degenerate modes are less than 0.1 μ, much smaller than the grain size of the materials, so that the coupling and the linewidth contribution should be quite small. The data confirm this expectation since the largest increase in ΔH_{eff} from high field to low field is only 2 Oe for the 1-μ sample and is generally much smaller for the larger-grain materials.

The origins of the relatively large high-field effective linewidths, compared to the 0.5 Oe (or less) typically observed in single crystals, remain unexplained. The $a_0^{-1/2}$ dependence appears to exclude the transit-time explanation considered in the last section and the nature of the measurement excludes two-magnon contributions. Schlömann has recently proposed that magnetoelastic scattering due to the random orientations of the crystallites in the polycrystal may be important.[20] His theory does predict a weak inverse grain-size dependence which is consistent with the observed $a_0^{-1/2}$ relationship, but the calculated values are several orders of magnitude smaller than indicated by the data.

V. SUMMARY AND CONCLUSION

In this investigation, the microwave properties of polycrystalline YIG have been measured for samples with average grain diameters from 1 to 30 μ. The resonance linewidth ΔH at 9.1 GHz is essentially grain-size independent and is on the order of 20–30 Oe for the "best" samples. Residual porosity is responsible for the large linewidths in the other materials. The spin-wave instability threshold h_{crit}, a strong function of the average grain diameter a_0 for both parallel pumping and subsidiary absorption, goes approximately as a_0^{-1}. Values of the spin-wave linewidth ΔH_k for the parallel-pump data show a decrease with increasing wavenumber for the small-grain samples ($a_0 < 5$ μ) and a small linear increase with k for $a_0 > 9$ μ. The near-resonance effective linewidth is independent of grain size and approximately equal to ΔH. At high field ($H_0 \approx 6000$ Oe) ΔH_{eff} varies as $a_0^{-1/2}$, increasing from 1.0 Oe or $a_0 = 30$ μ to 5.5 Oe for $a_0 = 1$ μ. The ΔH_{eff} at low field are slightly higher than the high-field values.

A transit-time model in which the propagation time for the potentially unstable spin wave across individual grains qualitatively explains the a_0^{-1} dependence of the parallel pump ΔH_k. The model does not adequately explain the k-dependences of ΔH_k for the different grain size samples. The difficulty probably lies both in the interpretation of parallel pump thresholds for fine-grain samples and the crude nature of the transit-time model. Two-magnon scattering would raise h_{crit} by a factor of two at most and can definitely be eliminated as a source of the high thresholds. Apart from a small two-magnon contribution to the low-field ΔH_{eff}, the origins of the off-resonance effective linewidth are not understood. Magnetostriction may be important, although predicted values are much smaller than those observed experimentally.

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