Contribution of the Two-Magnon Process to Magnetostatic-Mode Relaxation*

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The linewidths of the magnetostatic modes have been measured at X band with the static magnetic field along the [111] direction in several highly polished spheres of single-crystal pure yttrium iron garnet (YIG). The large variations of the magnetostatic-mode linewidths previously found in larger linewidth spheres were not observed in the highly polished YIG spheres. However, there is a residual variation of mode linewidths at both 300°K and 4.2°K, the variation being somewhat larger at 4.2°K. It was found that the essential features of the variation of mode linewidths in a highly polished YIG sphere can be explained by the surface-pit-scattering theory of Sparks, Loudon, and Kittel when the linewidth predicted by this theory is multiplied by an enhancement factor F_{nmr} to include the effect of the spatial variation of magnetization intensity for the individual magnetostatic modes. The results of calculations based on the Sparks-Loudon-Kittel theory for the linewidths due to scattering by fractional-micron-size pits are presented, as well as computed values of the enhancement factor F_{nmr} for most of the low-order modes. From the measurements it is estimated that at both temperatures the contribution of the two-magnon process to the uniform-precession linewidth is 0.10 Oe or less. This indicates that the major relaxation channel for the uniform-precession mode is not via degenerate spin-wave interactions. However, at 4.2°K this may be due to rare-earth impurities broadening the linewidth. The conclusions reached regarding the contribution of the two-magnon process to magnetic resonance relaxation of the uniform-precession mode in highly polished YIG spheres are in substantial agreement with the modulation experiments of Fletcher, LeCraw, and Spencer.

I. INTRODUCTION

THE measurement of the linewidths of the multiple magnetic resonances known as magnetostatic modes provides a means by which one may assess the relative contributions of certain processes to magnetic resonance relaxation of ferrimagnetic dielectrics.[1] Degenerate with each magnetostatic mode in a spherical sample are a number of shorter wavelength spin waves. Since the density of these spin waves will in general be different for each magnetostatic mode, by comparing magnetostatic-mode linewidths one can in principle determine the extent to which the two-magnon process, that is, the scattering of the long-wavelength magnetostatic modes into shorter wavelength degenerate spin waves, is contributing to magnetic resonance relaxation. The primary purpose of this work is to determine the extent to which the two-magnon process is contributing to the magnetostatic-mode relaxation in highly polished spheres of the ferrimagnetic dielectric yttrium iron garnet (YIG). In an effort to make such a determination, magnetostatic-mode linewidth measurements were made at about 9.5 Gc/sec on highly polished spheres of pure YIG at 300°K and 4.2°K with the static field along the [111] direction.

It is generally accepted that the major relaxation channel for the uniform precession mode in rough spheres of YIG at room temperature involves the two-magnon process.[2,3] However, for highly polished spheres the results of the modulation experiment of Fletcher, LeCraw, and Spencer[4] showed that the two-magnon process is not very important for relaxation of the uniform precession mode at room temperature. Several years ago measurements by White[1] revealed that for a spherical sample with a uniform precession linewidth of about 1 Oe there was considerable variation in the magnetostatic-mode linewidths. These variations appeared to be attributable to a two-magnon-type process although no theory had been formulated at that time to adequately explain the results. Since the highly polished samples of Fletcher, LeCraw, and Spencer had linewidths of 0.5 Oe or less, it was felt that it would be of considerable interest to see whether the large variations of magnetostatic-mode linewidths observed by White persisted in highly polished samples. For the highly polished samples, the results of Fletcher, LeCraw, and Spencer would lead one to expect only a residual variation of the magnetostatic-mode linewidth at room temperature due to the two-magnon process.

Further experiments on a highly polished pure YIG sphere using the modulation technique were performed at 4.2°K by Spencer and LeCraw.[5] At this temperature the contribution of the two-magnon process to relaxation of the uniform precession mode was greater than had been found in the earlier experiment of Fletcher, LeCraw, and Spencer at 300°K, although the total uniform precession linewidth had decreased. In fact, at 4.2°K the two-magnon process was the dominant relaxation process for the uniform precession mode. It therefore seemed that it would be of further interest to measure the magnetostatic-mode linewidths in the same sample at 4.2°K and 300°K. In view of the in-

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[1] R. L. White, Fourth Symposium on Magnetism and Magnetic Materials, Philadelphia, 1958 (unpublished); J. Appl. Phys. Suppl. **30**, 182S (1959); **31**, 86S (1960).

[2] C. Kittel, J. Phys. Soc. Japan Suppl. B1, **17**, 396 (1962).

[3] C. W. Haas and H. B. Callen in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. 1, p. 450.

[4] R. C. Fletcher, R. C. LeCraw, and E. G. Spencer, Phys. Rev. **117**, 955 (1960).

[5] E. G. Spencer and R. C. LeCraw, Phys. Rev. Letters **4**, 130 (1960).

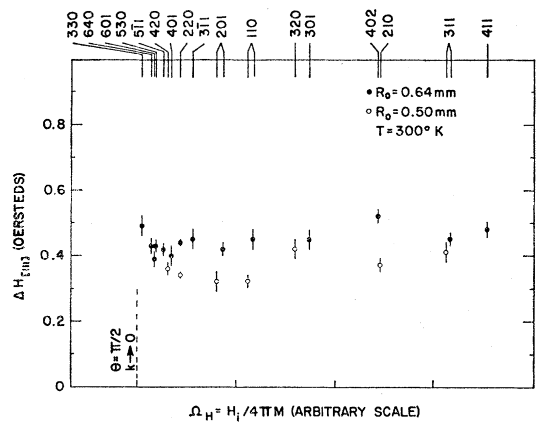

FIG. 1. Magnetostatic-mode linewidths at 300°K for two highly polished YIG spheres with the static magnetic field along the [111] direction. The mode indices according to Walker are listed at the top of the figure. The sample with $R_0=0.64$ mm was measured with $\Omega=\omega/\gamma 4\pi M=1.961$ and for the $R_0=0.50$-mm sample, $\Omega=1.887$.

crease of the contribution of the two-magnon process at 4.2°K, one would expect that dependence of the magnetostatic-mode linewidths on the number of degenerate spin-wave states would be accentuated at the lower temperature.

In Sec. II a brief description is given of the technique used for the measurement of the magnetostatic-mode linewidths at 300 and at 4.2°K. The results of the measurements on several samples are given in Sec. III. A residual variation is found in the magnetostatic-mode linewidths and it is shown that the essential features of the observed variation can be accounted for by the pit-scattering theory of Sparks, Loudon, and Kittel.[6]

II. EXPERIMENTAL TECHNIQUES

To be able to draw any meaningful conclusions about the relative linewidths of the various magnetostatic modes, measurements must be made of a sufficient number of mode linewidths with fields for resonance extending over most of the range that is theoretically possible for a sphere. This requires the measurement of modes of various orders and, in general, for a given exciting microwave field, these modes will have widely varying intensities. On the other hand, one would like the various modes to be measured to have roughly the same strength so that the relative accuracy of the linewidth measurements be as high as possible. Cavity perturbation techniques were used to make the measurements of the magnetostatic-mode linewidth reported here. The intensity of absorption of most of the modes was adjusted to approximately the same level by positioning the sample within a TE_{102} rectangular cavity and by varying the angle that the static magnetic field made with the broad face of the cavity. In general the linewidths of the modes were measured by means of a point-by-point technique. A few of the modes were too weakly absorbing to measure by this method however, and the measurement was made by modulating the static magnetic field and using superheterodyne detection. The field modulation amplitude was kept low enough to avoid line-broadening and the two methods of linewidth measurement were checked against one another by measuring several modes by both methods. The external magnetic field was measured with a nuclear resonance magnetometer.

The highly polished YIG spheres[7] were positioned in the microwave cavity by placing them in the hollow tip of either a polystyrene rod or a high-density styrofoam rod. The sphere was allowed to rotate freely within the tip of the sample holder in order that the easy magnetic axis (the [111] direction for YIG) was always aligned with the static magnetic field.[8]

III. RESULTS AND DISCUSSION

Measurements were made of the magnetostatic-mode linewidths at room temperature on the three different highly polished YIG spheres. The three samples had radii of 0.50, 0.64, and 0.74 mm, respectively. The sphericity of the samples was checked and the diameters of each sample were found to vary 0.2% or less. The behavior of the mode linewidths in all three samples was essentially the same although more mode linewidths were measured in the 0.64- and 0.74-mm radii samples than in the 0.50-mm sample. The results obtained with the two smaller spheres are shown in Fig. 1. The measurements in the sample with radius $R_0=0.64$ mm were made at a frequency of 9490 Mc/sec, and the $R_0=0.50$-mm sample linewidths were measured at a frequency of 9245 Mc/sec. The numbers at the top of the figure are the mode indices according to the notation of Walker.[9] The modes were identified by comparing the observed spectrum for each sample with the calculated spectrum[10] suitably corrected for propagation effects.[11] For each case the magnetization M of the sample was computed by using the observed difference in the fields for resonance of the 220 and 210 modes and the theoretical difference obtained from the results of Walker[9] and Plumier.[11] Published values of the anisotropy constants[12] were used to correct the

[6] M. Sparks, R. Loudon and C. Kittel, Phys. Rev. **122**, 791 (1961); M. Sparks, Microwave Laboratory Report 932, Stanford University, 1962 (unpublished).

[7] All of the samples used in this experiment were obtained from Microwave Chemical Laboratories, Inc. in the form of highly polished spheres.

[8] For further details see J. Nemarich, Doctoral thesis, University of Maryland, 1964 (available from University Microfilms, Inc., Ann Arbor, Michigan).

[9] L. R. Walker, Phys. Rev. **105**, 390 (1957).

[10] Walker's characteristic equation was programmed for modes of arbitrary order by O. R. Cruzan.

[11] R. Plumier, Physica **28**, 423 (1961).

[12] G. P. Rodrique, H. Meyer, and R. V. Jones, J. Appl. Phys. Suppl. **31**, 376S (1960); R. F. Pearson and R. W. Cooper, J. Phys. Soc. Japan Suppl. B1, **17**, 369 (1962).

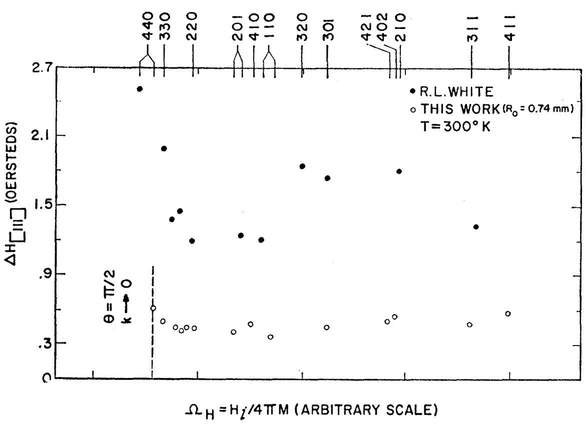

FIG. 2. Behavior of magnetostatic-mode linewidths in two YIG spheres with differing linewidths. The measurements were made at 300°K with the static field along the [111] direction. The data of R. L. White (Ref. 1) were taken at 9.7 Gc/sec. The narrower linewidths were measured at 9.5 Gc/sec with a highly polished sphere with radius $R_0=0.74$ mm. The mode indices according to Walker are shown at the top of the figure.

observed spectra for anisotropy. It was assumed that since the measurements were made with the static field along the [111] direction, the only effect of the crystalline anisotropy was to shift the entire spectrum by a constant field.[13] Except for one unidentified mode observed at 4.2°K, the observed spectra at both room temperature and 4.2°K were found to deviate from the computed spectrum by 0.6% or less. With regard to the measurements at 300°K shown in Fig. 1, the sample with 0.64-mm radius was found to have $4\pi M=1728$ G, and the 0.50-mm-radius sample was found to have $4\pi M=1750$ G. Since the values of the reduced frequency $\Omega=\omega/\gamma 4\pi M$ (where ω is the angular frequency and γ is the absolute value of the gyromagnetic ratio) were different for two samples, the mode linewidths were plotted on an arbitrary scale for the reduced field for resonance $\Omega_H=H_i/\gamma 4\pi M$ (H_i being the internal static magnetic field). In this way the corresponding modes and the extrapolated top of the spin-wave manifold have roughly the same abscissa for the two samples. The dashed line marked $\theta=\pi/2$, $k\rightarrow 0$ indicates the extrapolated top of the spin-wave manifold, θ being the angle the spin-wave vector $\mathbf{k}$ makes with the direction of H_i, and modes lying to the right of the dashed line are within the spin-wave manifold. The length of the line through each datum point indicates the rms deviation of each measurement. All of the room-temperature measurements were made with the sample placed in a polystyrene sample holder, while the measurements at 4.2°K were made in a styrofoam holder. Although there is some variation in the mode linewidths the magnitude of the variation is much less than has been observed in a larger linewidth sample. This can be seen in Fig. 2 where the data obtained with a 0.74-mm radius sphere are compared with those reported by White[1] for a sample with a uniform precession linewidth (110 mode) almost four times greater. The size of White's sample was not reported but the measurements were made at 9.7 Gc/sec. Since the measurements on the 0.74-mm-radius sample were made at 9490 Mc/sec, the scale for Ω_H was adjusted for each sample so that corresponding modes as well as the extrapolated top of the spin wave manifold roughly coincided. The depressed variation in magnetostatic-mode linewidth exhibited by the narrow linewidth sample in Fig. 2 tends to bear out the conjecture that the absence of appreciable contribution by the two-magnon process to magnetic resonance relaxation should result in little or no variation of magnetostatic linewidth as the position of the mode with respect to the spin-wave manifold changes. The question now arises as to whether the residual variation of mode linewidths exhibited in the room-temperature data can be shown to be quantitatively consistent with the results obtained for the uniform precession by Fletcher, LeCraw, and Spencer.[4] Also of interest is the behavior of the mode linewidths at 4.2°K.

Prior to presenting these results, the details of possible causes for the variation of magnetostatic-mode linewidths will be discussed. The primary source of the two-magnon process in YIG is believed to originate from scattering by surface and volume imperfections.[2,3] The surface of the samples used in this experiment were examined and no imperfections greater than about 5 μ could be observed. Highly polished samples are usually final-polished with a fine-grit material such as alumina. The individual particles of this substance are irregularly shaped, but for the finest grit generally used the particles have a maximum size of about 0.3 μ. Needless to say, a quantitative description of the character of a highly polished YIG sphere in the 1–2-mm size range would be difficult to obtain and was not known for our samples. The pit-scattering theory of Sparks, Loudon, and Kittel[6] was therefore investigated as a means of interpreting the experimental results. This theory computes the contribution to the magnetic-resonance linewidth from scattering by a spherical diamagnetic inclusion. The effect of a rough surface is approximated by assuming the surface to be equivalent to one uniformly covered by hemispherical pits that scatter independently. For a spherical sample of radius R_0 covered with pits of radius R, the linewidth due to this scattering process is

$$\Delta H_{SLK}=\frac{3}{4}\left[\frac{R}{R_0}\right]4\pi M\int_{k_{\min}R}^{k_{\max}R} d(kR)\frac{\Omega}{\Omega_r(k)} \times[j_1(kR)]^2\frac{(3\cos^2\theta_k-1)^2}{\cos\theta_k}, \quad (1)$$

[13] V. D. Krivchenkov and A. I. Pil'shchikov, Zh. Eksperim. i Teor. Fiz. 43, 573 (1962) [English transl.: Soviet Phys.—JETP **16**, 410 (1963)].

where

M = saturation magnetization,

$\Omega = \omega/\gamma 4\pi M$ = constant for this integration,

ω = angular frequency,

γ = absolute value of the gyromagnetic ratio,

$\Omega_r = \Omega_H + \Omega_{ex} l^2 k^2$,

$\Omega_H = H_i/4\pi M$,

H_i = internal static magnetic field,

$\Omega_{ex} = H_{ex}/4\pi M$,

H_{ex} = effective exchange field,

l = lattice spacing,

$j_1(x) = (\sin x/x^2) - (\cos x/x)$ = spherical Bessel function of order 1,

θ_k = angle the spin-wave propagation vector $\mathbf{k}$ makes with the static magnetic field H_i,

$\cos^2\theta_k = (\Omega_r^2 + \Omega_r - \Omega^2)/\Omega_r$,

$k_{\min} = 0$ if $\Omega^2 \leq \Omega_H(\Omega_H+1)$,

$k_{\min} = \{[(4\Omega^2+1)^{1/2} - (2\Omega_H+1)]/2\Omega_{ex} l^2\}^{1/2}$ if $\Omega^2 > \Omega_H(\Omega_H+1)$,

$k_{\max} = [(\Omega - \Omega_H)/\Omega_{ex} l^2]^{1/2}$.

Since the integration is to be performed at constant frequency, the relationship between θ_k and k has been obtained from the spin-wave dispersion relation, that is, $\Omega^2 = \Omega_r(\Omega_r + \sin^2\theta_k)$. Equation (1) also applied to the linewidth due to a single volume pit of radius R and volume V if the factor $\frac{3}{4}(R/R_0)$ is replaced by a factor $3V/V_0$, where V_0 is the sample volume.

For the uniform precession mode, if $\Omega_H \gg 1$ and $k_{\max} R \gg 10$, Eq. (1) is very well approximated by[6]

$$\Delta H_{SLK} = \frac{\pi}{8}\left[\frac{R}{R_0}\right] 4\pi M \frac{(3\cos^2\theta_0 - 1)^2}{\cos\theta_0}, \qquad (2)$$

where

$$\cos^2\theta_0 = (\Omega_H^2 + \Omega_H - \Omega^2)/\Omega_H.$$

For YIG at 4.2°K the exchange parameter is[14] $\Omega_{ex} l^2 = 2.09 \times 10^{-12}$ cm^2, from which it follows that $k_{\max} < 5 \times 10^5$ cm^{-1}. In this case the approximate formula (2) also describes very well the behavior of the exact integral for any resonance if $\theta_0 < 70°$ and $R > 12.5 \times 10^{-4}$ cm. However, we are interested in cases where R is smaller than the above value and also in cases where the resonance is either at the extrapolated top of the spin-wave manifold ($k \to 0$, $\theta_0 = \pi/2$) or above. For these cases the integral of Eq. (1) must be evaluated numerically. Owing to the nature of the integrand it is useful to use one form of the integral for integrations below and up to the extrapolated top of the spin-wave manifold (region I) and another form in the region above the extrapolated top of the spin-wave manifold (region II). For region I the explicit dependence $\cos\theta_k$ on k may be expressed in the integral as

$$\cos^2\theta_k = \frac{[\Omega_H(\Omega_H+1) - \Omega^2] + \Omega_{ex} l^2 k^2 [\Omega_r + \Omega_H + 1]}{\Omega_r}. \qquad (3)$$

When $\cos^2\theta_k$ is put into this form it is seen that as $k \to 0$, $\theta = \pi/2$, the first bracket in Eq. (3) vanishes and the integral of Eq. (1) is very nearly proportional to an integral of the form

$$\int_0^{k_{\max} R} dx \frac{[j_1(x)]^2}{x},$$

which is easy to evaluate in the region of $x = 0$.

Above the top of the manifold, in region II, $k_{\min} R$ is not zero, and near $\theta = \pi/2$ the integral may prove difficult to evaluate in the form used for region I. An expression equivalent to Eq. (1) and more suitable for numerical integration in region II may be obtained by noting that in deriving Eq. (1) the integration over $d\theta$ may be retained instead of that over dk. The resultant expression is then

$$\Delta H_{SLK} = \frac{3}{4}\left[\frac{R}{R_0}\right] 4\pi M \times \int_{u_{\min}}^{1} du \frac{\Omega R (3u^2-1)^2}{\Omega_{ex} l^2 k [4\Omega^2 + (1-u^2)^2]^{1/2}} [j_1(kR)]^2, \qquad (4)$$

where

$u = \cos\theta$,

$u_{\min} = 0$ if $\Omega^2 \geq \Omega_H(\Omega_H+1)$,

$$u_{\min} = \left[\frac{\Omega_H(\Omega_H+1) - \Omega^2}{\Omega_H}\right]^{1/2} \text{ if } \Omega^2 < \Omega_H(\Omega_H+1),$$

$$k = \left[\frac{[4\Omega^2 + (1-u^2)^2]^{1/2} - [2\Omega_H + (1-u^2)]}{2\Omega_{ex} l^2}\right]^{1/2}.$$

The linewidth due to surface pit-scattering by pits with radii R less 12.5×10^{-4} cm was calculated[15] on a digital computer for a series of pit radii R and reduced field for resonance Ω_H. The results obtained for several values of smaller pit radii are shown in Fig. 3. It is seen that there is a marked change in the character of the linewidth versus Ω_H curve for variations of pit size in this range. For the smallest pit radius the linewidth varies relatively smoothly with Ω_H. As the pit radius increases, the maxima in the curve become more pronounced. The peak in ΔH at the smaller value of Ω_H not only increases in magnitude with pit radius but also moves toward the extrapolated top of the spin wave

[14] R. C. LeCraw and L. R. Walker, J. Appl. Phys. Suppl. 32, 167S (1961).

[15] The programming for this calculation was performed by A. Hausner.

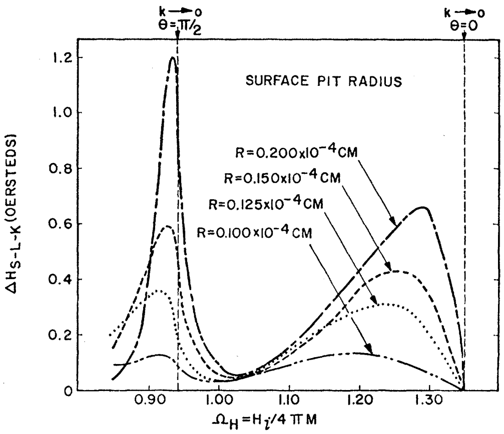

FIG. 3. Magnetic resonance linewidths for a sphere of radius $R_0=0.74$ mm covered with hemispherical surface pits of radius R according to the theory of Sparks, Loudon, and Kittel. Constants assumed for the computation were $4\pi M=2481$ G, $\Omega=\omega/\gamma 4\pi M=1.352$, and $\Omega_{ex}l^2=2.086\times10^{-12}$ cm².

manifold and becomes narrower. For larger values of pit radii than shown in the figure the peak is right at the top of the spin wave manifold and drops abruptly above the top of the manifold (that is, for smaller Ω_H). For R greater than about 5×10^{-4} cm and up to $R=12.5\times10^{-4}$ cm the height of the peak is approximated very well by $\Delta H_{SLK}(\text{peak})=37.75\times10^8R^2$. For values of $R<5\times10^{-4}$ cm the peak rises slightly faster than R^2. The maximum in the curve at the larger values of Ω_H also rises with increasing R and the dropoff at the extrapolated bottom of the spin-wave manifold becomes increasingly steeper. For values of R greater than 1.25×10^{-4} cm the dropoff is essentially coincident with the bottom of the spin-wave manifold. Since the expression for the linewidth due to scattering by a volume pit is the same as for surface pits excepting for a scale factor, the above results describe equally well the qualitative behavior of the linewidth as the volume pit radius is increased. The distinctive shape of these curves for various pit radii can be used to delineate clearly the region of volume or surface pit size that may be assumed to be present in the sample and will still give agreement with experimental observations.

When considering the coupling of the magnetostatic modes to degenerate spin waves by surface imperfections, it has been pointed out by Jones[16] that one must consider the fact that the amplitude of the precession angle for a magnetostatic mode is generally larger at the surface of a sphere than it is inside the sphere, and the amount by which it is larger will vary from mode to mode. It would then be expected that the contribution of degenerate spin-wave coupling from surface interactions would also in general vary from mode to mode. The enhancement of surface scattering is considered here in conjunction with the simple pit-scattering theory of Sparks, Loudon, and Kittel. In this theory the contribution of the surface interaction is assumed to be independent of the angle the static magnetic field makes with the surface. The effect of the spatial variation of the mode precession angle (or transverse component of magnetization) on the linewidth due to surface interactions therefore manifests itself as a factor multiplying the Sparks-Loudon-Kittel result. This factor, which we have designated F_{nmr}, may be set equal to the ratio of the energy loss due to surface scattering for a mode with indices (n,m,r) to that for a mode with a uniform distribution of magnetization, providing the modes have the same field and frequency for resonance and the energy stored in the resonant modes are equal. Since the energy loss at the surface is proportional to the integral of the magnitude squared of the transverse component of magnetization over the surface,[17] it is easily shown that

$$F_{nmr}=\Delta H_{SLK}(n,m,r)/\Delta H_{SLK}=\langle|m_t|^2\rangle_{\text{surface}}/\langle|m_t^3|^2\rangle_{\text{volume}}, \quad (5)$$

where $|m_t|^2=|m_x|^2+|m_y|^2$ is the magnitude squared of the transverse component of magnetization for the mode with indices (n,m,r), $\langle\ \rangle_{\text{surface}}$ indicates the surface average, and $\langle\ \rangle_{\text{volume}}$ indicates the volume average. The published values[18] of m_x and m_y for the magnetostatic modes in a sphere were used to compute the values of F_{nmr}. In most cases $F_{nmr}=(2n+1)/3$ and is therefore independent of the index r and hence of the field and frequency at which the mode is resonant. Table I gives the values of F_{nmr} computed for the modes on which linewidth measurements were made when $\Omega=1.893$. The results are similar when $\Omega=1.365$, the value appropriate to the measurements at 4.2°K.

The foregoing considerations regarding the variation of magnetostatic-mode linewidths were used in the analysis of the measurements on the 0.74-mm radius sample at 300 and 4.2°K. The assumption was made that the observed magnetostatic mode linewidths can

TABLE I. Computed values of F_{nmr}, the factor expressing the degree of relative concentration of transverse magnetization at the surface of a sphere for the magnetostatic modes with indices (n,m,r). The F_{nmr} are independent of r excepting for the cases footnoted and these were computed for $\Omega=1.893$ and the indicated values of r. N.C. indicates these F_{nmr} were not computed since measurements were not made on the corresponding modes.

n \ m	0	1	2	3	4	5
			F_{nmr}			
1	...	1	...	...	...	...
2	5/3	5/3	5/3	...	...	...
3	7/3	7/3	7/3	7/3	...	...
4	7.3/3[a]	9/3	9/3	9/3	9/3	...
5	N.C.	7/3[b]	N.C.	11/3	11/3	11/3
6	N.C.	N.C.	16/3[c]	N.C.	13/3	13/3

[a] $r=2$. [b] $r=1$. [c] $r=0$.

[16] G. R. Jones, Bull. Am. Phys. Soc. 9, 113 (1964).

[17] G. R. Jones, Bull. Am. Phys. Soc. 8, 360 (1963); G. R. Jones, Doctoral thesis, Catholic University of America, 1963 (available from University Microfilms, Inc., Ann Arbor, Michigan).

[18] R. C. Fletcher and R. O. Bell, J. Appl. Phys. 30, 687 (1959).

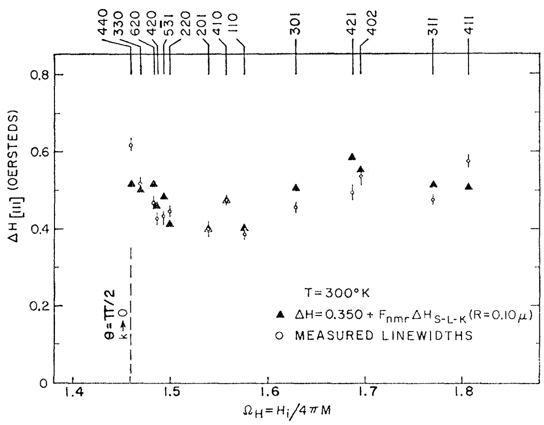

FIG. 4. Magnetostatic-mode linewidths at 300°K in a highly polished YIG sphere of 0.74 mm with the static field along the [111] direction. The mode indices according to Walker are listed at the top of the figure. Parameters used for obtaining Ω_H are $4\pi M=1790.5$ G, $H_{\rm ANIS}[111]=60$ Oe, $\gamma/2\pi=2.80$ (Mc/sec)/Oe, and $\Omega=\omega/\gamma 4\pi M=1.893$. Data for this sample at 4.2°K are shown in Fig. 5.

be expressed as a sum of a constant contribution plus a part that depends on the mode indices and the position of the mode with respect to the spin-wave manifold (in this case, the Sparks-Loudon-Kittel result); that is, for a mode with indices (n,m,r) and reduced field for resonance Ω_H, the linewidth is assumed to have the form

$$\Delta H=\Delta H_0+F_{nmr}\Delta H_{SLK}(\Omega_H,R). \qquad (6)$$

The best fit to the measured linewidths was sought by varying R, the assumed effective pit radius, and ΔH_0, the constant contribution. ΔH_0 was not assumed to be constant with temperature, but the value of R was chosen to be the same at both temperatures. For the data obtained at both temperatures, the best fit resulted when the residual linewidth variations were attributed to surface pits with an effective radius of $R=0.10\times10^{-4}$ cm. The results of this best fit for the data at 300°K are shown in Fig. 4. The sample and data are the same as those used in the comparison with the larger linewidth sample in Fig. 2. The comparison of the theoretical and experimental results for the sample at 4.2°K is shown in Fig. 5.

If F_{nmr} is assumed equal to unity, the variation observed in the mode linewidths cannot be fit to any of the Sparks-Loudon-Kittel results unless fractional micron pits are assumed responsible for the scattering. The observed variation can be fit fairly well for a pit radius of $R=0.125\times10^{-4}$ cm by assuming that the constant contributions ΔH_0 are the same as those in Figs. 4 and 5, and that the scattering is due to surface pits. Of course the same result will be obtained if the interaction is attributed to volume pits of the same radius and the effective number of pits scattering is taken to be 8.68×10^6. Since the abrasive often used for final polishing has a maximum size of about 0.30×10^{-4} cm and is undoubtedly broken down somewhat during the polishing process, it seems that the observed linewidth variation is more likely due to surface pit scattering rather than volume pit scattering. If F_{nmr} is allowed to take on the value appropriate to the individual modes, that is, a value of about 3, it is understandable that the best fit is now obtained with the choice of a somewhat smaller scattering pit radius. A value $R=0.10\times10^{-4}$ cm gives the best fit under these circumstances, and it is noted that this choice of effective surface pit size is about two-thirds that of the maximum polishing grit size.

The analysis of the data taken at 300°K shows that there is a linewidth contribution of $\Delta H_0=0.35$ Oe to all the modes due to processes other than the two-magnon process. The uniform precession mode (the 110 mode) therefore appears to have a contribution of 0.04–0.05 Oe from the two-magnon process. Fletcher, LeCraw, and Spencer[4] found that the two-magnon process contributed roughly 0.05 Oe to the linewidth of their sample. Since their experiment was performed at a lower frequency (6.2 Gc/sec) and on a smaller sample (0.34-mm radius), their figure should be adjusted for sample size and position of the resonance frequency with respect to the spin-wave manifold in order to compare it with the results of the present experiment. The Fletcher-LeCraw-Spencer two-magnon contribution when thus adjusted is found to be 0.03 Oe, a figure still within the experimental error of the measurements.

From Fig. 5 it is seen that at 4.2°K the constant contribution is $\Delta H_0=0.20$ Oe and if this is subtracted from the measured linewidth of the uniform precession mode, the two-magnon process is found to be contributing 0.09 Oe. The computed two-magnon contribution is somewhat lower, being 0.05 Oe. The measurements of

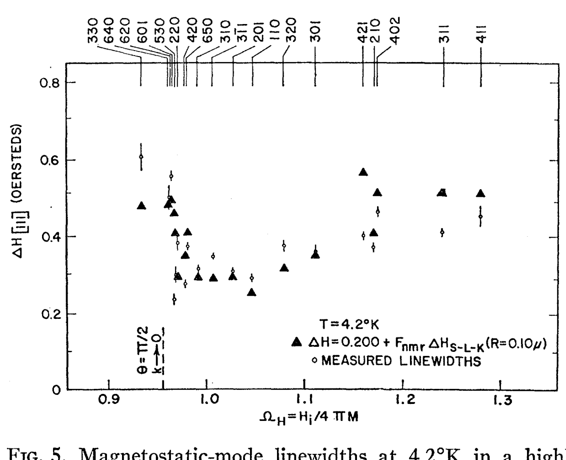

FIG. 5. Magnetostatic-mode linewidths at 4.2°K in a highly polished YIG sphere of radius 0.74 mm with the static magnetic field along the [111] direction. The mode indices according to Walker are listed at the top of the figure. Parameters used for obtaining Ω_H are $4\pi M=2457.5$ G, $H_{\rm ANIS}[111]=189.7$ Oe, $\gamma/2\pi=2.80$ (Mc/sec)/Oe, and $\Omega=\omega/\gamma 4\pi M=1.365$. Data for this sample at 300°K are shown in Fig. 4.

Spencer and LeCraw[5] at 9340 Mc/sec determined the contribution of this process in their sample to be 0.10 Oe. The size of the sample used in these measurements was not mentioned, but if the assumption is made that it was about the same as that used by these authors in the experiment at 300°K, the size-adjusted two-magnon contribution to be compared with our measurements is 0.05 Oe. The results obtained for the contribution of the two-magnon process using two different measurement techniques are therefore seen to be in good agreement.

With regard to the contribution of other than two-magnon process effects in the uniform precession at room temperature, Fletcher, LeCraw, and Spencer showed that in their sample at 6.2 Gc/sec this amounted to 0.42 Oe, whereas for our sample at 9.5 Gc/sec the contribution was 0.35 Oe. Since the spin-wave linewidth decreases with frequency,[19] adjustment for the frequency difference in the two measurements serves only to increase the observed differences. The contribution of other than the two-magnon process measured at 4.2°K also appears to be different in the two measurements. A value of $\Delta H_0 = 0.20$ Oe was determined for the sample used in this experiment, whereas Spencer and LeCraw appear to have obtained a value of 0.07 Oe. Since the sample used in this experiment was known to be made from starting material containing 1 part in 10^6 of paramagnetic impurities, and that of Spencer and LeCraw was made from material with one order-of-magnitude higher purity, the possibility of rare-earth impurities contributing the difference in the two measurements at 4.2°K should be considered. Since rare-earth impurities are known to increase the magnetic-resonance linewidth of YIG considerably in the temperature range of about 20°K to 100°K, the uniform precession linewidth of the sample used in this experiment was measured at 77°K and found to be 0.60 Oe. For samples prepared from the higher purity material, the linewidth at this temperature is 0.31 Oe.[20] In view of these results it is probably safe to conclude that the larger contribution at 4.2°K from other than the two-magnon process to the linewidth of the sample used in this experiment is due primarily to rare-earth impurities.

IV. CONCLUSION

Careful measurements of the magnetostatic-mode linewidths in highly polished YIG spheres at 300°K have verified the conjecture based on the results of Fletcher, LeCraw, and Spencer that the appreciable variations observed in magnetostatic-mode linewidths in a larger linewidth sphere would be much reduced in highly polished samples. Measurements made on one of the highly polished samples at 4.2°K indicate that the variation in the magnetostatic-mode linewidths is also considerably less than that observed by White but increases somewhat over that observed in the same sample at 300°K. The pit-scattering theory of Sparks, Loudon, and Kittel was considered for explaining the observed variation of mode linewidths in highly polished samples. The results of these authors were recast into a form suitable for calculation, and the behavior of the linewidth versus position of the resonance with respect to the spin-wave manifold was examined for fractional-micron-size pits. The shape of the curve showing linewidth versus reduced field for resonance changes considerably as the pit size is varied in the fractional micron range. When the pit-scattering process is occurring at the surface of the sphere, it was reasoned that the Sparks-Loudon-Kittel result should be multiplied by a factor F_{nmr}. This enhancement factor was calculated for many of the low-order modes and found to be equal to $(2n+1)/3$ for most modes. The indices of the modes whose linewidths were measured were obtained by comparing the observed mode spectrum with the theoretical spectrum. The essential features of the variation in the magnetostatic mode linewidths at both 4.2 and 300°K could be explained by the enhanced Sparks-Loudon-Kittel result for scattering by surface pits, if the diameter of the surface pits was assumed to be about two-thirds the maximum diameter of commonly used polishing material. The magnetostatic-mode linewidths were also assumed to have a contribution that did not depend on the position of the mode with respect to the spin-wave manifold. A best fit for this constant term was chosen at each temperature.

The extent to which the two-magnon process contributes to a magnetostatic-mode linewidth is seen to be dependent on mode number as well as on various other factors. For the purpose of discussing the breakdown of two-magnon versus other processes contributing to the linewidth, the results obtained in this experiment for the uniform precession mode were compared to those obtained by Fletcher, LeCraw, and Spencer using a modulation technique. It was found that the contribution from other than two-magnon process deduced by the two methods of measurement differed by about 0.07 Oe at room temperature. No explanations can be offered to account for this difference. At 4.2°K the contribution from processes other than the two-magnon process in the sample used in this experiment was about 0.13 Oe larger than in the sample used by Spencer and LeCraw. The conjecture was made that the observed difference is due to a higher concentration of paramagnetic impurities in the sample used for this experiment, since this is consistent with the impurity levels of the materials used in the growth of the crystals and with the relative uniform precession linewidths at 77°K. When adjusted for sample size and frequency, the results of the two

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methods of measurement of the two-magnon-process contribution to the uniform precession linewidth are in good agreement. At 300 and 4.2°K both methods of measurement showed the two-magnon-process contribution to be less than 0.10 Oe, and the differences between the results of the two methods of measurement to be not far from the experimental error of the measurements. In the sample used here, relaxation of the uniform precession mode at either 300 or 4.2°K was not predominantly via the two-magnon process, but at the latter temperature this is probably due to broadening of the linewidth by paramagnetic impurities.

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Measurements of Thermal Expansion and Thermal Equilibrium Defects in Silver Chloride*

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Linear thermal expansion of silver chloride has been measured by simultaneous macroscopic and x-ray methods on the same sample and the same temperature scale (±0.1°C). The two expansions agree within the experimental error of about 3×10^{-5} throughout the measurement range, from −62°C to within 4° of the melting point. This implies that the equilibrium concentrations of Schottky defects are less than 9×10^{-5} and that their enthalpy of formation is greater than 1.45 eV. This limit is consistent with results of Compton for chlorine diffusion. Results of other workers for high-temperature variations in ionic conductivity, thermal expansivity, silver diffusivity, and heat capacity can be interpreted in terms of a single species of defect, the cation Frenkel defect. An empirical Mie-Grüneisen equation of state represents, by a suitable choice of fitting parameters, the measured expansion of silver chloride when defect concentrations are small. However, the values of the fitting parameters appear to be inconsistent with those to be expected from the Grüneisen phenomenological theory. Because the concentrations of Frenkel defects are relatively large, an extrapolation method applied to the thermal expansion can be used to estimate their formation enthalpy as 1.4 eV. Further analysis is consistent with the suggestion that the defects make an explicit contribution to the thermal expansion coefficient of the crystal.

I. INTRODUCTION

SPECULATIONS on the equilibrium-defect structure of silver halide crystals at high temperatures[1–5] mention the possibility of appreciable concentrations of Schottky defects. Experimenters who measured heat capacity,[6] thermal expansion,[7] and ionic conductivity[8] have also given interpretations requiring appreciable Schottky-defect concentrations. Unfortunately, these arguments either (1) contain rather arbitrary assumptions concerning the behavior of the hypothetical defect-free crystal, (2) depend upon an uncertain separation of the presumed effects of the Schottky defects from those of the larger concentrations of cation Frenkel defects known to be present,[9,10] or (3) do not permit quantitative estimates of the equilibrium-defect concentrations to be made.

There is a type of experiment in which such arbitrariness is not present. It has been shown[11] for a cubic

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