

## Measurement of magnetic properties of single crystal YIG by non-resonant method

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**Abstract.** A non-resonant microwave technique has been employed for the determination of saturation magnetisation ( $4\pi M_s$ ) and  $g$ -factor of pure and Gd-doped single crystal YIG spheres yielding accurate values. It was found that  $4\pi M_s = 1720-1751$  g for undoped and 1595 g for 10% Gd-doped YIG spheres while  $g_{\text{eff}} = 2.00-2.0039$ . The advantages of this method are simplicity and complete absence of propagation corrections necessary in cavity resonance experiments.

### 1. Introduction

The important parameters of single crystal yttrium iron garnet ( $\text{Y}_3\text{Fe}_5\text{O}_{12}$  or YIG) spheres are (i) the saturation magnetisation  $4\pi M_s$ , (ii) the  $g$ -factor  $g_{\text{eff}}$ , (iii) linewidth  $\Delta H$  and anisotropy field  $K_1/M_s$ . Rapid and accurate measurement of these parameters is often required for characterisation of spheres required for applications in tunable devices such as filters and oscillators in the microwave frequency range. The manner in which important device characteristics are determined by these parameters has been reviewed in detail (Matthaei *et al* 1964). This paper describes the measurement of these parameters by a simple non-resonant waveguide technique and discusses the results obtained in comparison with other established techniques. The main advantage is the absence of propagation corrections which must be used in all cavity methods for accurate determination of  $4\pi M_s$  and  $g_{\text{eff}}$ .

### 2. Measurement of $4\pi M_s$

Measurement of saturation magnetisation  $4\pi M_s$  is carried out by (i) ballistic methods, (ii) methods using vibrating coil or vibrating sample magnetometer, (iii) force methods and (iv) microwave methods. The microwave methods consist of determination of (a) demagnetisation effect of sample on resonance (Narayan *et al* 1969), (b) cavity absorption characteristics (Artmann and Tannenwald 1955), (c) cut-off frequency (Helsajn 1972) and (d) frequencies of higher-order magnetostatic modes (Fletcher *et al* 1959; Lam 1965; Magid 1964).

For narrow line-width material such as single crystal YIG the last method is convenient because the same experimental arrangement can be used for measuring line-width and magnetic anisotropy. Also the measured quantities, which are the differences between magnetic field values can be accurately determined by using a proton resonance magnetometer.

### 3. Theory

$\text{Y}_3\text{Fe}_5\text{O}_{12}$  has a cubic garnet structure ( $a = 12.376 \text{ \AA}$ ) with 3 types of cation sites. The 3 non-magnetic  $\text{Y}^{3+}$  ions occupy  $c$  sites which have dodecahedral co-ordination,  $2\text{Fe}^{3+}$  ions occupy octahedral  $a$  sites and the remaining  $3\text{Fe}^{3+}$  ions are in tetrahedral  $d$  sites. Thus the net magnetisation is due to one  $\text{Fe}^{3+}$  ion which has a spin magnetic moment of  $5\mu_B$ . There are 8 formula units per unit cell and the saturation magnetisation is  $4\pi M_s = 2460 \text{ g (0}^\circ\text{ K)}$ , in good agreement with theory, decreasing to 1750 g at  $300^\circ\text{ K}$ .  $\text{Gd}^{3+}$  enters the lattice in the  $c$  sites with its spin magnetic moment  $7\mu_B$  aligned anti-parallel to that in the  $d$  sites thus decreasing  $4\pi M_s$  at low concentrations.

When a YIG sphere is placed in a static magnetic field  $H_0$ , the application of an orthogonal rf magnetic field causes the spins to precess at a frequency

$$f_0 = \gamma (H_0 + kH_a) \simeq \gamma H_0, \quad (1)$$

where  $\gamma$  = gyromagnetic ratio  $2.8 \text{ MHz/Oe}$ ,  $k$  = constant dependent on sphere orientation and  $H_a$  = anisotropy field. Due to inhomogeneities in the static and rf magnetic fields or relatively large sphere diameter, higher-order magnetostatic modes are also generated whose frequencies differ from  $H_0$  by amounts determined by  $4\pi M_s$ .

Fletcher and Bell (1959) developed a general theory to predict the resonant frequency of any mode for a sphere at given  $H_0$  which gives

$$f_{nmr} = \gamma \cdot 4\pi M_s (\Omega_i + F_{nmr}), \quad (2)$$

where  $F_{nmr}$ , the form-factor of each mode, has been calculated theoretically and given in table 1. The index (nmr) describes each magnetostatic mode, where  $n$  describes the periodicity of the magnetic potential in  $\theta$ , the polar angle;  $m$  describes the periodicity in the co-ordinate  $\phi$  and  $r$  is the order of the root of the transcendental equation which is to be satisfied by the boundary condition. In (2)

$$\Omega_i = H_t / 4\pi M_s = (H_0 - N_t \cdot 4\pi M_s) / 4\pi M_s. \quad (3)$$

For a sphere the demagnetising factor  $N_t = 1/3$  and hence

$$f_{nmr} = \gamma [H_0 - 4\pi M_s (1/3 - F_{nmr})]. \quad (4)$$

Table 1. Form factor of magnetostatic modes.

Mode	210	320	110	540	220	330	440	550
$F_{nmr}$	0.20	0.286	0.333	0.363	0.40	0.420	0.445	0.455

Table 1 gives  $F_{nmr}$  for modes with  $n - m = 0$  or 1 and  $r = 0$ . It is only for these modes that  $F_{nmr}$  is independent of  $\Omega_i$  and hence these modes tune linearly with  $H_0$ . The uniform precession mode is the (110) mode for which  $F_{nmr} = 1/3$  and hence  $f_{nmr} \simeq \gamma H_0$ , if  $H_a$  is neglected for the moment since  $H_a \ll H_0$ .

For a fixed measurement frequency  $f$ , the static field  $H_{nmr}$  required for (nmr) mode is given in terms of the field required for the (110) mode  $H_{110}$  by

$$H_{nmr} - H_{110} = -(F_{nmr} - F_{110}) 4\pi M_s, \quad (5)$$

and hence

$$4\pi M_s = (H_{nmr} - H_{110}) / (F_{110} - F_{nmr}). \quad (6)$$

Hence by accurately measuring the magnetic field difference given by the numerator and identifying the (nmr) mode and thus knowing  $F_{nmr}$ ,  $4\pi M_s$  can be calculated. Equation (4) can be re-written as

$$F_{nmr} = 1/3 + \frac{(f_{nmr}/\gamma) - H_0}{4\pi M_s}. \quad (7)$$

A plot of the rhs of equation (7) vs  $H_0/4\pi M_s$  for different modes given by Fletcher *et al* (1959) is shown in figure 1. It is seen that for modes for which  $n - m = 0$

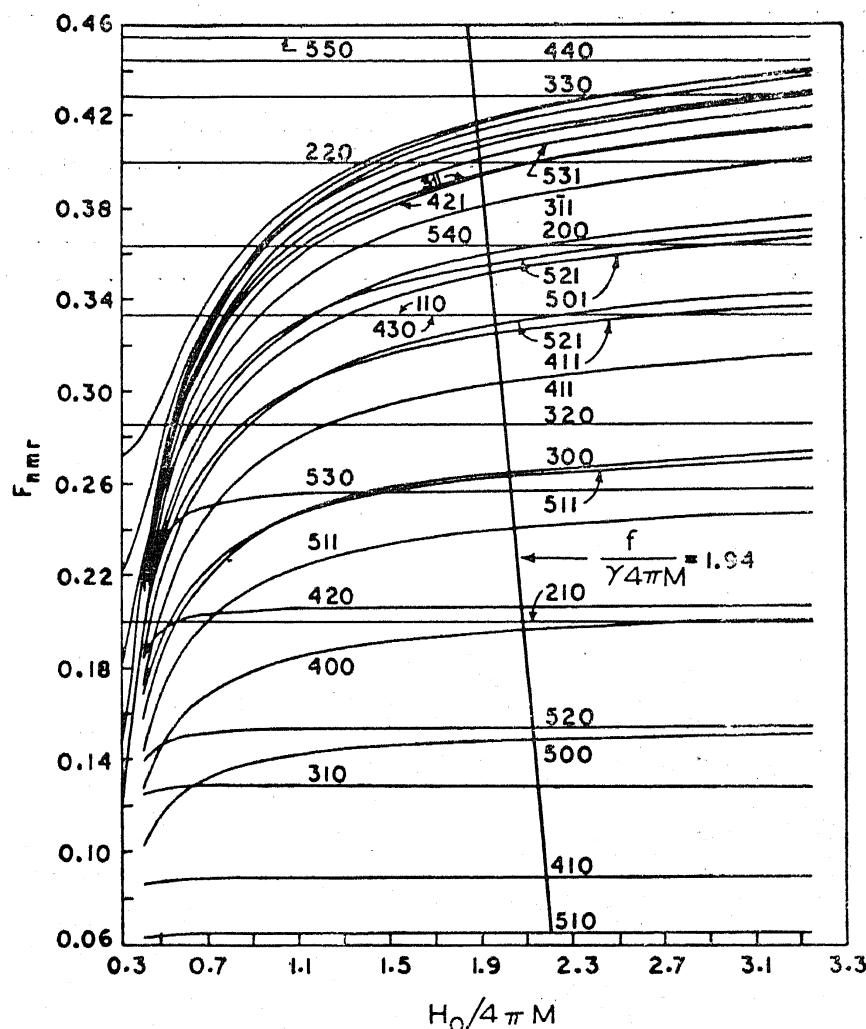


Figure 1. Mode-chart of magnetostatic modes giving form-factor  $F_{nmr}$  vs.  $H_0/4\pi M_s$  for modes with  $n$  upto 5. The straight line represents a frequency  $f = 9.709$  GHz.

or 1 and  $r = 0$ ,  $F_{nmr}$  is constant and  $f$  varies linearly with  $H_0$ . These modes are used for the determination of  $4\pi M_s$ . Intersecting the mode curves is a straight line with  $f/(y \cdot 4\pi M) = 1.94$  corresponding to a fixed frequency  $f = 9.709$  GHz. The figure thus indicates that as the static field  $H_0$  is increased the modes excited successively are (550), (440), (330), (220), (540), (110), (320), (210) etc.

The uniform precession or (110) mode has the highest intensity. The relative intensities of the modes have been tabulated by Fletcher *et al* (1959) and are in the order (110) > (540) > (220) > (330) > (320) > (210). This provides a guide to the identification of modes although the field  $H_0$  at which the modes are excited is a more precise identification factor.

In the first method of determining  $4\pi M_s$  (table 2) eq. (6) has been used in identifying the (110) and (*nmr*) modes and measuring the magnetic field separation  $H_{nmr} - H_{110}$ . It was however found that in some cases mode identification was not unambiguous leading to a large spread in the values of  $4\pi M_s$ , e.g., for the sphere IGD - 1, the values were 1559 g, 1436 g and 1438 g.

A new method of mode identification was thus evolved using the phenomenon of mode-crossing. Figure 1 shows that the linearly tuning or flat (200) mode intersects the non-flat (540) when  $F_{540} = F_{200} = 0.364$ . From the  $x$ -axis of the mode-chart at this frequency  $H_{540}/4\pi M_s = H_{200}/4\pi M_s = 2.18$ . Substituting in (7),

$$f_o / (\gamma \cdot 4\pi M_s) = 2.211, \quad (8)$$

where  $f_c$  = mode intersection frequency. The iterative method to find  $4\pi M_s$  is as follows. A value of  $4\pi M_s$  is assumed. Using (8) or equivalent expression for the expected mode,  $f_c$  is computed.

The static field is adjusted to give  $f_0 = f_{nmr}$  and  $f_{nmr} - f_{11.0}$  is measured. With the assumed value of  $4\pi M_s$ ,  $R = (f_{nmr} - f_{11.0})/\gamma$ .  $4\pi M_s$  is calculated. In the example

**Table 2.**  $4\pi M_s$  determination from magnetostatic modes

Table 3.  $4\pi M_s$  using mode-crossing technique.

Step No.	Assumed $4\pi M_s$ (g)	$f_a$ (GHz)	$f_a - f_{110}$ (MHz) expt.	$R$
1.	1635	10.1	134.39	0.0290
2.	1735	10.7	145.5	0.0299
3.	1835	11.35	164.2	0.0319
4.	1785	11.04	156.79	0.0313
5.	1760	10.89	153.06	0.0310

given if  $R$  is found to be  $> 0.031$ , the assumed value of  $4\pi M_s$  is more than the actual value. Depending on whether or not  $R > 0.031$  a new value of  $4\pi M_s$  is assumed and the procedure repeated till  $R = 0.031$ . The simple bisection algorithm is used to modify the  $4\pi M_s$  values in successive iterations and gives the correct value. The results are given in table 3.

#### 4. Measurement of $g_{\text{eff}}$

In the 2-sublattice model, the effective  $g$ -factor is given by

$$g_{\text{eff}} = g_A g_B (M_A - M_B) / (g_B M_A - g_A M_B), \quad (9)$$

for the case of negligible damping. In pure YIG the only magnetic ion present is  $\text{Fe}^{3+}$  which has a ground state  $^6\text{S}_{5/2}$  ( $L = 0$ ) and is distributed in the  $a$  and  $d$  sites for each of which it is expected that  $g_A = g_B = 2.00$ , giving  $g_{\text{eff}} = 2.00$ . The experimental values of  $g_{\text{eff}}$  given by von Aulock (1965) are: 2.0025 (150° K), 2.005 (300° K) and 2.0068 (400° K).

Important dopants in YIG are  $\text{Gd}^{3+}$  used for reducing  $4\pi M_s$  and  $H_0^{3+}$ ,  $\text{Dy}^{3+}$  used for increasing  $h_{\text{crit}}$  and hence power handling capacity.  $\text{Gd}^{3+}$  like  $\text{Fe}^{3+}$  has a  $L = 0$  ground state ( $^8\text{S}_{7/2}$ ) and thus  $g = 2.00$ . For  $\text{Dy}^{3+}$ ,  $g = 1.33$  (theor) and 1.36 (exp) while for  $H_0^{3+}$ ,  $g = 1.25$  (theor).  $\text{Fe}^{2+}$  which is an undesired presence since it introduces losses due to electron hopping between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  has a  $g_{\text{eff}}$  value of 1.89. Thus accurate determination of  $g_{\text{eff}}$  is important to obtain information on both desired and undesired dopants in YIG.

From the point of device characterisation  $g_{\text{eff}}$  is directly related to  $\gamma$ , the gyro-magnetic ratio since

$$\gamma = (ge/4\pi M_s) \text{ Hz/Oe.} \quad (10)$$

For  $g = 2.00$ , it is found that  $\gamma = 2.7992 \text{ MHz/Oe}$ . For doped crystals  $g$  may not be ideally 2.00 and hence the slope of the tuning curve given by (1) may have a different value.

#### 5. Experimental

The experimental arrangement, shown in figure 2, has been previously used by Desormiere (1965) and Masters *et al* (1960) to measure the narrow line-width of

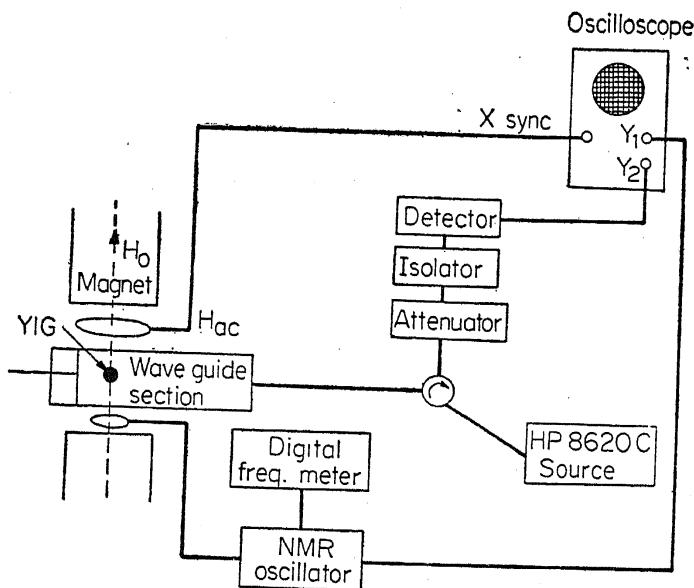


Figure 2. Experimental arrangement for non-resonant measurement of  $4\pi M_s$  and  $g_{\text{eff}}$  of single crystal YIG spheres.

materials such as single crystal YIG. This technique allows variation of sphere-circuit interaction by changing the position of the wave-guide short. The YIG sphere is mounted on a perspex rod and is free to rotate and align itself with a  $\langle 111 \rangle$  easy axis along  $H_0$ . Microwave power is supplied from an HP 8620C source and the reflected power detected and displayed on one channel of an oscilloscope. Power absorbed by the YIG sphere at resonance appears as a dip in the reflected power.

The static magnetic field  $H_0$  is measured using a proton resonance magnetometer. The oscillator frequency is measured using a 6-digit frequency meter and its output, with the NMR absorption pip, displayed on another channel of the oscilloscope. A 50 Hz ac field produced by Helmholtz coils is used to modulate  $H_0$  and sweep it through resonance. The accuracy of measurement of  $H_0$  is  $\pm 0.1$  Oe.

For the determination of  $4\pi M_s$ ,  $f$  is kept constant and  $H_0$  varied to first obtain the  $\langle 110 \rangle$  mode and then the higher order modes, for each of which  $H_0$  is measured. The mode-chart figure 1 is used for mode-identification.

For the measurement of  $g_{\text{eff}}$ , the value of  $\gamma$  was determined using (1) and  $g_{\text{eff}}$  using eq. (8). However since for  $\langle 111 \rangle // H_0$

$$H_{\text{eff}} = H_0 - 4/3 K_1/M_s \quad (9)$$

the anisotropy field  $H_0 = -K_1/M_s$  need be determined. This was done using the standard method (Matthaei *et al* 1964) of fixing the sphere and rotating it about a  $\langle 110 \rangle$  axis and measuring the maximum and minimum  $H_0$  values for resonance which occur when the  $\langle 100 \rangle$  and  $\langle 111 \rangle$  axes respectively lie along  $H_0$  since:

$$f = \gamma (H_0 + 2 K_1/mc) \text{ for } \langle 100 \rangle // H_0, \quad (10)$$

$$\text{and } f = \gamma (H_0 - 4/3 K_1/M_s) \text{ for } \langle 111 \rangle // H_0. \quad (11)$$

Thus for constant  $f$ ,

$$H_0|_{100} - H_0|_{111} = -10/3 K_1/M_s.$$

Table 4. Magnetic properties of YIG spheres.

Sphere	$f$ (GHz)	$-\frac{K_1}{M_s}$ (Oe)	$H_{eff}$ (k Oe)	$\gamma$	$g$	$\Delta H$ (Oe)
WJ-1	10.858	43.0	3.8788	2.7993	2.00007	0.33
I-2	10.9	43.8	3.8864	2.8047	2.0039	0.42
	11.15	43.8	3.9777	2.8031	2.0028	
IGD-1	10.88	46.8	3.8861	2.7997	2.0036	0.85
	11.02	46.8	3.9367	2.7992	2.0000	

## 6. Results

The results of the first method of determining  $4\pi M_s$  using (6) is given in table 2.

The mode-crossing technique is illustrated by results given in table 3.

Thus  $4\pi M_s = 1760$  g for the sphere which was undoped YIG.

A similar procedure used for the 10% Gd-doped sphere IGD-1 gave  $4\pi M_s = 1581$  g compared with the values obtained in table 1 and the expected value of  $4\pi M_s = 1595$  g (Fletcher and Bell 1959).

The results of measurement of  $K_1/M_s$  and hence  $g_{eff}$  are given in table 4.

## 7. Discussion

The determination of  $4\pi M_s$  from higher order magnetostatic modes reported by Fletcher *et al* (1959) and used by Lam (1965) was carried out in cavities at a fixed frequency. Large propagation shifts upto 36.6 Oe for the (110) mode, 20 Oe for the (220) mode, etc., occurred, and it was necessary to correct this to obtain agreement between theory and experiment. Electromagnetic propagation effects are neglected in the magnetostatic approximation which assumes  $\lambda \gg a$ , where  $\lambda$  = incident wavelength and  $a$  = sphere diameter. Spin-wave propagation is also neglected since it is assumed that  $\lambda^2 \gg H_{ex} \cdot l/M_s$ , where  $H_{ex}$  = equivalent exchange field and  $l$  = magnetic lattice constant. Propagation shifts have been calculated theoretically by Hurd (1958) for the (110) mode and found experimentally for the other modes. However this theory which makes certain assumptions, like resonance shift being less than  $\Delta H/2$ , are not valid for low line-width materials.

Magid (1964) employed non-resonant broad band strip-line transmission and reflection structures for measurement of  $4\pi M_s$ . He used the frequency difference between the (110) and (210) modes which was 700 MHz approximately for undoped YIG. However his measured values were 5-7% higher than static values for spheres 0.5 mm dia and as much as 20% higher for spheres 0.75 mm dia.

The great advantage of non-resonant waveguide structure is that propagation corrections are not necessary for any modes and the  $4\pi M_s$  values are accurately determined by the mode-crossing technique. This is borne out by the measure-

ment on pure YIG spheres (obtained from Watkins-Johnson (WJ) and prepared at the Indian Institute of Science, Bangalore (I) and also on a Gd-doped sphere. The absence of propagation shifts may be attributed to the use of non-resonant structure because the rf magnetic fields at the sphere are much lower than those in high  $Q$  cavities and hence, for a given wavelength and sphere size, the inhomogeneities in the fields are also much less. The interaction between crossing modes was also negligible. However, the fact that the number of higher order modes excited is reduced and the weak modes are not detected acts as a disadvantage. An adequate number of modes are however available for the measurement of  $4\pi M_s$  of low line-width spheres.

The validity of the method is also borne out by the accurate values of  $g_{eff}$  obtained without incorporating any propagation corrections. Determination of the frequency of the microwave source which is  $\pm 5$  MHz gives an error in  $g_{eff}$  of  $\pm 0.0011$ . This can obviously be reduced using a microwave frequency counter. The results may be compared with  $g$  values found by Rodrigue *et al* (1960) which are  $g = 2.003$  (*a* site) and  $2.0047$  (*d* site) for pure YIG;  $g = 1.994 \pm 0.005$  at  $480^\circ\text{K}$  for GdIG.

The line-widths of the spheres have also been measured in the same set-up and found to vary between  $0.23$ - $0.34$  Oe for the Watkins-Johnson spheres and  $0.42$ - $0.85$  Oe for spheres grown by us.

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