

# Microwave Permeability Measurement of Unsaturated Hexaferrites of Y-type

Mahmut Obol and Carmine Vittoria

Electrical and Computer Engineering Department, Northeastern University,  
Boston, MA02115

**Abstract** - We have investigated the zero field permeability behavior of Y-type Hexaferrites. The measured permeability from 0.045 to 10 GHz was theoretically explained in term of zero magnetic field multi-domain and domain wall resonance. The basal plane anisotropy field,  $H_A^A$ , domain wall thickness  $\delta$  and length  $L$  of the domain wall were estimated from resonance and permeability data.

## 1. INTRODUCTION

The rapid development of ferrite and electronic communication devices have placed a huge demand for high permeability materials at GHz frequencies. In the past, a number of ferrite research groups have utilized Y and Z-type hexaferrites for application requiring high  $\mu$  materials at low frequencies. For example, a number of papers<sup>[1,2,5]</sup> reported permeability behavior of (Y,Z)-type polycrystal materials at GHz frequencies. Some papers<sup>[1,2,3,4,5,6,7]</sup> have attributed the high  $\mu$  to spin rotational and domain wall motions. It was not clear what was meant by spin rotation or wall motion as it related to permeability specifically. For example, by spin rotation it was clear whether it implied precessional motion or simply displacement of magnetization. In addition, others<sup>[1,5]</sup> reported on absorber applications of Y-type polycrystal materials, since the lossy component of  $\mu$  can also be high. The purpose of this paper is to provide a physical explanations for the  $\mu$  values of Y-type hexaferrites in zero magnetic field and correlate them with measurements of samples produced by us. explained in terms of material parameters.

## 2. THEORY

Before we develop the ideas for multi-domain and domain wall resonance as related to Y-type hexaferrites we wish to introduce some terminologies that have been used in the past<sup>[11]</sup>. For example, the magnetic anisotropy field associated with rotation of the magnetization in the polar angle direction,  $\theta$ , is designated as  $H_\theta^A$  while with the azimuth rotation as  $H_\phi^A$ .

We believe that oriented particles of Y-type hexaferrites can provide high permeability in the GHz frequency regime. The particles prepared by us are oriented<sup>[8]</sup> with the c-axis normal to the slab plane. This means that the plane perpendicular to the c-axis or the slab plane is the easy plane of magnetization. Permeability,  $\mu$ , measurements from 1 to 6 GHz on oriented particles of  $\text{Ba}_2\text{MnZnFe}_{12}\text{O}_{22}$  were reported<sup>[9]</sup> in which an external magnetic field,  $H$ , was applied in the slab plane. In this paper we measure  $\mu$  in a zero bias external magnetic field and we compare the measurements with a model proposed by us. Besides oriented polycrystalline particles we include single crystal slabs of the Y-type hexaferrite as well. The purpose of the measurements in zero field is to (1) correlate all the measurements ( including polycrystalline, single crystal, in an external field, zero field and by others<sup>[1,2,3,4,5,9]</sup>) (2) formulate a single theory consistent with the measurements.

We find that magnetic losses at low frequency ( $0 \leq 250$  MHz) are associated with domain wall resonance. We were able to observe domain wall resonance in all the samples and it compared well with calculations. For  $0.3 < f < 3$  GHz losses are associated with multi-domain resonance. Again, we observed that this type of resonance in all samples and compared reasonably well with our calculations. Clearly, in an external magnetic field both type of resonances are not excited, since domains are "wiped" out. As a result of this work permeability measurements in zero field can now be

### a) Ferrimagnetic Resonance

As in any magnetic processes<sup>[13]</sup> the starting point is the free energy of the sample and it is given below as:

$$\begin{aligned}
 F = & -M_s H \sin \vartheta \cos \varphi + \frac{1}{2} (N_x M_s^2 \sin^2 \vartheta \cos^2 \varphi \\
 & + N_y M_s^2 \sin^2 \vartheta \sin^2 \varphi + N_z M_s^2 \cos^2 \vartheta) \\
 & - K_1 \cos^2 \vartheta - K_3 \cos 6\varphi
 \end{aligned} \quad (1)$$

With reference to fig 1, the first term is the magnetizing energy, the second term contains the demagnetizing energy, the third term is the usual uniaxial magnetic anisotropy energy associated with changes in  $\theta$  and the last term is the six-fold energy term associated with in plane rotations or basal plane energy. In our case the basal plane coincides with the slab plane of our samples. The FMR condition may be given as<sup>[13]</sup>

$$\left(\frac{\omega_1}{\gamma}\right)^2 = [(N_z - N_x)M_s + H_\vartheta^A] + [(N_y - N_x)M_s + H_\varphi^A], \quad (2)$$

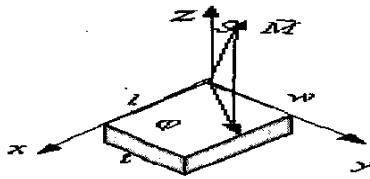


Fig. 1 slab geometry

where Smit-Beljers condition<sup>[13]</sup> was used, and  $H=0$ . The anisotropy fields are defined as, respectively,

$$H_\vartheta^A = \frac{2K_1}{M_s} \quad \text{and} \quad H_\varphi^A = \frac{36K_3}{M_s}$$

It is noted that for spherical particles  $N_x = N_y = N_z = \frac{4\pi}{3}$ .

Eq. (2) reduces to the following:

$$\frac{\omega}{\gamma} = \sqrt{H_\varphi^A H_\vartheta^A}, \quad (3)$$

where  $\gamma = 2\pi g 10^6$  Hz/Oe, and  $g = 2$ <sup>[13]</sup>.

Eq. (3) was previously derived by Smit and Wijin<sup>[11]</sup>. In our case, we need to introduce demagnetizing factors  $N_x$ ,  $N_y$ , and  $N_z$ , since we are using samples of rectangular shape and they are approximated by us as follows

$$N_x = \frac{4\pi \frac{t}{l}}{1 + \frac{t}{l}(1 + \frac{l}{w})}, \quad \text{and} \quad N_y = \frac{4\pi \frac{t}{w}}{1 + \frac{t}{w}(1 + \frac{w}{l})}$$

where  $t$  is thickness,  $w$  the width, and  $l$  the length of the slab, and  $N_z = 4\pi N_x N_y$ . As it is well known<sup>[12]</sup> in multi-domain resonance that for  $H < N_x M$  (unsaturated external fields), there is another resonance associated with the out of phase precessional motion of magnetization vectors in two opposite magnetic domains. We estimate<sup>[12]</sup> this resonance to be at

$$\frac{\omega_2}{\gamma} \equiv \frac{\omega_1}{\gamma} + 2\pi M_s \quad (4)$$

where  $\frac{\omega_1}{\gamma}$  is the resonance condition associated with in-phase precessional motion, see equation (2). Thus, at zero applied magnetic field, the two modes of resonance in a multi-domain configuration would be equations (2) and (4).

The corresponding initial permeability for  $H=0$  of oriented Y-type materials is given as

$$\mu_i = \frac{4\pi M}{H_\vartheta^A} \quad (5)$$

where  $4\pi M$  is the net magnetization in zero field or remanence. We will compare above calculations with measured permeability from zero to 10 GHz, see section 3.

### b) Domain wall Resonance,

According to H. Dotsch<sup>[6]</sup> et. al, domain wall resonance is very much analogous to the motion of mass in a restoring force in which the mass is represented by the size of the domain wall and the restoring force is provided by the magnetostatic energy. According to this theory<sup>[6]</sup> the resonance is given as

$$\frac{\omega_d}{\gamma} \equiv M_s \sqrt{\frac{4\pi\delta}{D}}, \quad (6)$$

$$\text{where} \quad \delta = \sqrt{\frac{A}{K_3}}, \quad \text{and} \quad D = \frac{\pi}{2M_s} \sqrt{\sigma L}$$

$A$  (exchange stiffness constant)<sup>[6]</sup>  $\approx 0.3 \times 10^{-6}$  erg/cm,  $M_s$  (saturation magnetization)<sup>[13]</sup>  $= 175$  G, and  $\sigma$  (domain wall energy)<sup>[6]</sup>  $\approx 2$  ergs/cm<sup>2</sup>,  $D$  is the spatial period of multi-domains,  $\delta$  is domain wall width.

In effect there is only one adjustable parameter in equation (6), the length  $L$  of the domain wall in the slab plane.  $K_3$  will be determined from FMR experiments. We vary the parameter  $L$  between 1mm and 4mm which is the maximum value (the size of the samples are 4x4x0.254mm<sup>3</sup>).

## 3. EXPERIMENTAL RESULTS

### Permeability measurements in zero magnetic field

#### a) single crystal slab

The length of the microstripline was 4mm long which was the same as the  $l$  dimension in fig. 1. The stripline

device was tested by TRL calibration technique<sup>[9]</sup>. In fig. 2, we plot the measured permeability of a single crystal slab of  $\text{Ba}_2\text{Zn}_2\text{Fe}_{12}\text{O}_{22}$  as a function of frequency. As in the previous<sup>[9]</sup> paper by us, from the measurement of  $S_{21}$  we determined both the real part,  $\mu'$ , and imaginary,  $\mu''$ , parts of the permeability of the ferrite slab. In fig. 3 the measurements were extend to 10 GHz. At frequencies below 0.1 GHz the TRL calibration is inaccurate and that explains the enhanced measurements of  $\mu$ .

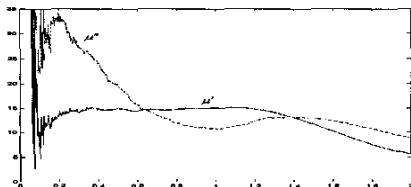


Fig. 2, Dashed line is imaginary and solid line is real parts of permeability. Vertical axis is relative permeability and horizontal axis is in GHz.

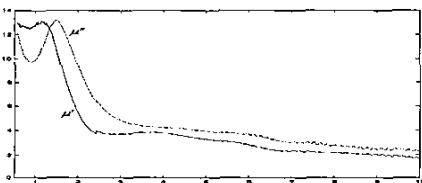


Fig. 3, Dashed line is imaginary and solid line is real parts of permeability. Vertical axis is relative permeability and horizontal axis is in GHz.

Let us now analyze the data in terms of the theory modeled in previous section. We see in fig. 2 that there are two magnetic resonances, as  $\mu'$  exhibits maxima at these two frequencies. The first resonance occurred at about 200 MHz. We attribute this resonance to the domain wall resonance and the other one at 1.5 GHz to the zero field FMR (in our designation  $\omega_0/\gamma$ ). If we assume  $L = 1\text{mm}$  (length of domain wall in the slab plane), we obtain resonance (eq. 6) at 175 MHz. However, if we assume a maximum value of 4 mm (lateral dimension of the sample) resonance occurs at 90 MHz. Given the amount of uncertainty in the parameter  $L$ , eq (6), the agreement is remarkable.

We note that although  $\mu'$  is maximum at 170 MHz,  $\mu'$  asymptotically reaches a value of 14 as  $f \rightarrow 0$ . We wish now to explain this result. As eq. (5) predicts we needed knowledge of both remanence and  $H_\phi^A$  to estimate  $\mu'$ . The remanence  $4\pi M$  was measured via VSM techniques and we obtained a value of 88 Gauss, see solid line in the fig. 4.  $H_\phi^A$  was measured from FMR measurements. By plotting the magnetic resonant field,

$H_r$ , versus in plane angle of the external field,  $\phi$ , we were able to deduce the following  $(N_y-N_x)M_s = 20$  Oe. We utilized the fact that the demagnetizing energy is uniaxial symmetry and the  $H_\phi^A$  term field is six fold symmetric, see fig. 5. By applying eq.(2) directly to the multidomain resonance occurring at 1.43 GHz, see fig. 2, we deduced a value of  $H_\phi^A$  of 6.2 Oe. Hence, we estimate  $\mu'$  at zero frequency as 14.2, see eq.(5) compared to experimental value of 15. According to eq.(4) the out of phase multi-domain resonance is predicted at ~3.3 GHz. From fig.3 resonance occurred between 3.5 and 5 GHz. However, the resonance is rather weak. In fact, if we take the absorption bandwidth,  $\Delta f$ , fig. 3 as approximately 2 GHz, we deduce a permeability of

$$\mu'' \approx \frac{\gamma^4 \pi M}{\Delta \omega} \approx 3.1$$

compared to 4 in fig. 3.

### b) Oriented polycrystalline slab

The same device and measurement technique was applied to measure the permeability of oriented Y-type hexaferrite slab. The measured domain wall resonance occurred at 200MHz compared to 175 MHz in the single crystal slab, see fig. 4. There appears other domain wall resonances at higher frequencies. This may be due to non-uniformity in particle orientation. Multi-domain resonances are shown in fig. 4.

As in previous procedure for single crystal analysis, we deduce  $H_\phi^A \approx 23.6$  Oe and  $(N_y-N_x)M = 20$  Oe. However, remanence magnetization of 7% was measured by VSM for oriented polycrystalline sample. This corresponds to an actual remanence of 154 Gauss. The deduced value of  $H_\phi^A$  of oriented Y-type as deduced from in plane FMR is roughly four times higher than the single crystal sample. This is reasonable since the actual chemical composition of the oriented particles was  $\text{Ba}_2\text{Mn}_2\text{Zn}_2\text{Fe}_{12}\text{O}_{22}$ . The values of  $4\pi M_s$ ,  $H_\phi^A$  and  $\gamma$  are the same as that of single crystal. However, there is no prior measurement of  $H_\phi^A$  in the literature<sup>[14]</sup> for this composition of oriented hexaferrites.

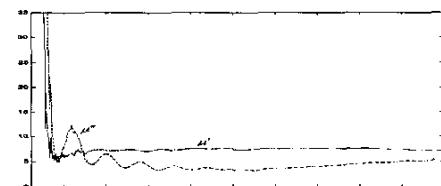


Fig. 4 Dashed line is imaginary and solid line is real parts of permeability. Vertical axis is relative permeability and horizontal axis is in GHz.

Using  $H_\phi^A$  and the other magnetic parameters as before ( see previous section) we predict multi-domain resonance at 1.89 GHz which compares very well with experimental value of 2 GHz, see fig. 5. Resonance corresponds to the frequency where  $\mu''$  is maximum. Using eq.(5) we estimate the permeability,  $\mu'$ , as  $f \rightarrow 0$  to be 6.5 compared to experimental value 6.2. In this estimate and that for the single crystal we used  $4\pi M_s = 2200$  Gauss. Remanence at  $H=0$ ,  $4\pi M_r$ , was measured by VSM to be 7% or 154 Gauss. There were no adjustable parameters in the estimate of  $\mu'$ .

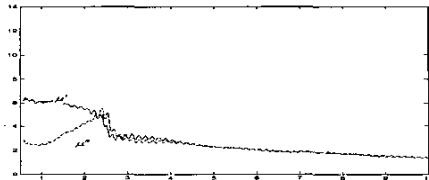


Fig. 5 Dashed line is imaginary and solid line is real parts of permeability. Vertical axis is relative permeability and horizontal axis is in GHz.

#### 4. DISCUSSION AND CONCLUSION

1) From our estimates the single domain wall thickness of the Y-type hexaferrites was about 0.47 micron. In the past, we have attempted to orient Y-type hexaferrites particles of 0.5 micron unsuccessfully. Based on this present results, particle size need to be greater than half-micron in size to be oriented.

2) The permeability analysis showed that the initial permeabilities are correlated with  $H_\phi^A$  and magnetization remanence,  $4\pi M_r$ . It may be possible to obtain very high permeability in the order of 100, if either magnetization remanence is increased or  $H_\phi^A$  is reduced.

3) One method of increasing  $4\pi M_r$  or remanance is to reduce the in plane demagnetizing factors. For example, from VSM measurements on ten mills slab remanance was measured to be 6.7% , but for five mills slab, remanance was measured to be 19.7 %. Clearly, the coercive field is the same for both slabs .

4). We believe that the proposed models by us for domain wall resonances, multi-domain resonances and initial permeability in oriented as well as single crystal Y-type hexaferrites has validity, since the comparison between theory and experiment is reasonable in view of the fact that there were no adjustable parameters in the model.

5) In zero or in an applied external field Y-type hexaferrite materials should form the bases of high  $\mu$  materials in the future.

#### References

- 1) H.J.Kwon et all, "The microwave absorbing and resonance phenomena of Y-type hexagonal microwave absorbers", *J. Appl. Phys.*, Vol. 75, No.10, 15 May 1994.
- 2) Tatsuya Nakamura and Ken-ichi Hatakeyama, "Complex permeability of polycrystalline" hexagonal ferrites", *IEEE TRANSACTIONS ON MAGNETICS*, VOL. 36, NO. 5, SEPTEMBER 2000.
- 3) David Bariou, Patrick Queffelec, Philippe Gelin, and Marcel Le Floc'h, "Extension of the effective medium approximation for determination of the permeability tensor of unsaturated polycrystalline ferrites" *IEEE TRANSACTION ON MAGNETICS*, VOL. 37, NO. 6 NOVEMBER 2001.
- 4) Ernst Schlomann, "Microwave behavior of partially magnetized ferrites", *J. Appl. Phys.*, Vol. 41, No. 1, 1970.
- 5) Tetsuji Inui and Kouichi Konishi, "Fabrication of broad-band RF -absorber composed of planar hexagonal ferrites", *IEEE TRANSACTIONS ON MAGNETICS*, VOL. 35, NO. 5, SEPTEMBER 1999.
- 6) H. Dotsch, W. Tolsdorf, and F. Welz, "Domain wall oscillation of bubble and stripe lattices in hexagonal ferrites", *J. Appl. Phys.*, 51(5), July, 1980.
- 7) M. A. EL Hiti et all, "Semiconductivity in  $Ba_2Ni_{2-x}Zn_xFe_{12}O_{22}$  Y-type hexaferrites", *JMMM* 195(1999) 667-678.
- 8) M. Obol, X. Zuo, and C. Vittoria, "Oriented Y-type hexaferrites for ferrite device", *J. Appl. Phys.*, VOL. 91, NO. 10.
- 9) M. Obol and C. Vittoria, "Measurement permeability of oriented Y-type hexaferrites", submitted to *JMMM*.
- 10) J. Aarts and Abu Shiekah, "Domain structure in polycrystalline MnZn ferrite imaged by magnetic force microscopy", *J. Appl. Phys.*, Vol. 85, No. 10.
- 11) Smit and Wjin, "FERRITES", N.V. Philips' Gloeilampenfabrieken, Eindhoven, Holland, 1959.
- 12) F. J. Rachford, P. Lubitz, and C. Vittoria, "Microwave resonance and propagation in nonsaturated ferromagnetic media. 1. Magnetic resonance in single crystal ferrite platelets", *J. Appl. Phys.* 53(12), December 1982.
- 13) Carmine Vittoria, "Microwave properties of magnetic films", 1993 by World Scientific Publishing Co. Pte Ltd.
- 14) Landolt-Bornstein, "Numerical Data and Functional Relationships in Science and Technology", 1970 New York.