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Microwave permeability of Y-type hexaferrites in zero field

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We have investigated the permeability at microwave frequencies of Y-type hexaferrites in zero-magnetic field. The permeability was measured from 0.045 to 10 GHz and it was correlated with multidomain and domain-wall resonances in zero-magnetic field. The basal plane anisotropy field, H_{ϕ}^{A}, magnetic remanence, $4\pi M$, domain-wall thickness δ, and length L of the domain wall were estimated from resonances and permeability data.
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I. INTRODUCTION

The rapid development of ferrite and electronic communication devices have placed a demand for high permeability, μ, materials at GHz frequencies. In the past ten years, a number of ferrite research groups have utilized Y- and Z-type hexaferrites for applications requiring high μ materials at low frequencies. For example, a number of papers[1–3] reported permeability behavior of (Y,Z)-type polycrystalline materials from 0 to 10 GHz frequencies. Some papers[1–7] have attributed the high μ values to spin rotation and domain-wall motions. In addition, others[1,5] reported on applications of Y-type polycrystalline materials as microwave absorbers, since the lossy component of μ can also be high. It was not clear in previous papers how spin rotation and wall motion were related to the measured, permeability, for example, at microwave frequencies. For example, by spin rotation, it was not clear whether it implied precessional motion or simply the displacement of magnetization. The purpose of this article is to provide a physical explanation for μ measured at microwave frequencies of oriented (Ref. 8) Y-type hexaferrites in zero-magnetic field and specifically, correlate μ with magnetic resonance and domain-wall motion.

The particles we prepared were oriented[8] with the *c*-axis normal to the slab plane. This means that the plane perpendicular to the *c* axis, or the slab plane, was the easy plane of magnetization. We measured the permeability from 1 to 6 GHz on oriented[9] particles of $Ba_2MnZnFe_{12}O_{22}$ in which an external magnetic field, *H*, was applied in the slab plane. In this article, we extend the previous[9] work by measuring μ in zero-magnetic field. In addition, we have developed a model which purports to explain the measurements of μ at microwave frequencies in zero as well as in an external magnetic field, *H*, for oriented or nonoriented hexaferrite materials of the Y type.

According to our model, magnetic losses at low frequencies ($f \leq 250$ MHz) are associated with domain-wall resonance. This conclusion was corroborated by our observation of domain-wall resonance in all of the samples. In addition, for $0.3 < f < 3$ GHz losses are associated with multidomain magnetic resonances. The multidomain resonances were observed in all samples and compared reasonably well with our calculations. We believe that permeability measurements on Y-type hexaferrites at microwave frequencies can be explained in terms domain-wall and multidomain resonances.

II. THEORY

Before we discuss ideas for multidomain and domain-wall resonances as related to Y-type hexaferrites, we wish to review some terminology that have been used in the past.[10] For example, the magnetic anisotropy field associated with rotation of the magnetization in the polar angle direction, θ, is designated as H_{θ}^{A} while with the azimuth rotation as H_{φ}^{A}.

A. Ferrimagnetic resonance

As in any magnetic process,[11] the starting point is the free energy of the sample and it is given as follows:

$$F = -M_s H \sin\vartheta \cos\varphi + \frac{1}{2}(N_x M_s^2 \sin^2\vartheta \cos^2\varphi + N_y M_s^2 \sin^2\vartheta \sin^2\varphi + N_z M_s^2 \cos^2\vartheta) - K_\vartheta \cos^2\vartheta - K_\varphi \cos 6\varphi. \quad (1)$$

With reference to of Eq. (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 rotation of the magnetization relative to the polar angle, θ, and the last term is the six-fold energy term associated with in plane rotation or basal plane energy. In our case, the basal plane coincided with the slab plane of our samples. The ferrimagnetic resonance (FMR) condition may be given as[11]

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

where $H = 0$. The anisotropy fields are defined as, respectively,

$$H_\vartheta^A = \frac{2K_\vartheta}{M_s},$$

and

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$$H_{\varphi}^{A}=\frac{36K_{\varphi}}{M_s}.$$

It is noted that for spherical samples $N_x=N_y=N_z=4\pi/3$. Thus, Eq. (2) reduces to the following:

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

where $\gamma=2\pi\times1.4g\times10^{6}$ Hz/Oe, and $g\approx2$.[11] Equation (3) was previously derived by Smit and Wijin.[10] In our case, we needed to introduce demagnetizing factors N_x, N_y, and N_z, since we were using samples of a rectangular shape and approximated them as follows:

$$N_x=\frac{4\pi\frac{t}{\ell}}{1+\frac{t}{\ell}\left(1+\frac{\ell}{w}\right)},$$

and

$$N_y=\frac{4\pi\frac{t}{w}}{1+\frac{t}{w}\left(1+\frac{w}{\ell}\right)},$$

where t is the thickness, w is the width, l is the length of the slab, and $N_z=4\pi-N_x-N_y$.

As it is well known[12] in multidomain resonance that for $H<N_xM_s$ (unsaturated external fields), there is one resonance associated with the out of phase precessional motion of the magnetization vectors in two opposite magnetic domains. We estimate[12] this resonance to be at

$$\frac{\omega_2}{\gamma}\cong\frac{\omega_1}{\gamma}+2\pi M_s, \tag{4}$$

where ω_1 is the resonance frequency associated with inphase precessional motion, see Eq. (2). Thus, at zero-applied magnetic field, the two modes of resonance in a multidomain configuration are contained in Eqs. (2) and (4).

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

$$\mu_i=\frac{4\pi M}{H_{\varphi}^{A}}, \tag{5}$$

where $4\pi M$ is the net magnetization in zero-field or magnetic remanence. Our model applies specifically to hexaferrite materials. Other models for μ at microwave frequencies were developed earlier by Rado[13] and Globus[14] for cubic ferrites of the spinel and garnet materials. The basic difference between our model and theirs[13,14] is the fact that in cubic ferrites, $H_{\varphi}^{A}=0$ and $H_{\theta}^{A}=0$. We will compare these calculations with measured permeability from zero to 10 GHz, see Sec. III.

B. Domain-wall resonance

According to Dotsch *et al.*,[6] 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 represented by the magnetostatic energy. According to this theory,[6] the resonance was given as

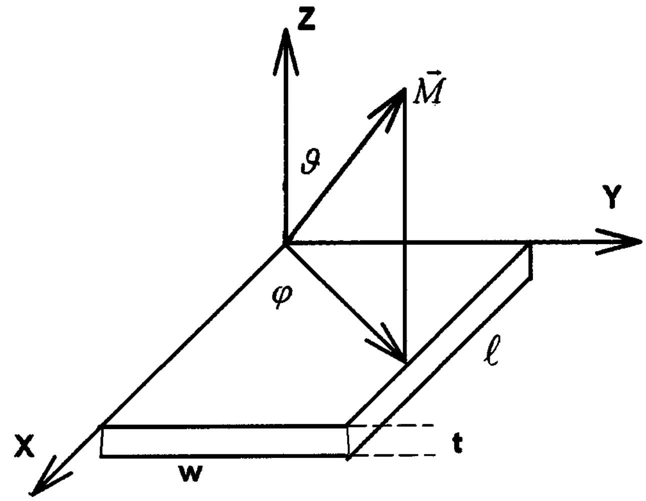

FIG. 1. Ferrite slab geometry.

$$\frac{\omega_d}{\gamma}\cong M_s\sqrt{\frac{4\pi\delta}{D}}, \tag{6}$$

where

$$\delta=\sqrt{\frac{A}{K_{\varphi}}},$$

and

$$D=\frac{\pi}{2M_s}\sqrt{\sigma L},$$

where A(exchange stiffness constant)[6] $\approx0.3\times10^{-6}$ erg/cm, M_s(saturation magnetization)[12] $=175$G, σ(domain-wall energy)[6] ≈2 ergs/cm^2, D is the spatial period of multidomains, and δ is the width of the domain wall.

In effect, there is only one adjustable parameter in Eq. (6), the length L of the domain wall in the slab plane. K_{φ} was determined from FMR experiments. We varied the parameter L between 1 mm and 4 mm which was the maximum value (the size of the samples were $4\times4\times0.254$ mm^3).

III. EXPERIMENTAL RESULTS

A. Single crystal slabs

The length of the microstripline was 4 mm long which was the same as the ℓ dimension in Fig. 1. The microstripline device was tested using a TRL calibration technique.[15] In Fig. 2, we plot the measured permeability of a single crystal slab of $Ba_2Zn_2Fe_{12}O_{22}$ as a function of frequency. As in a previous paper by us,[9] from the measurement of S_{21} we determined both the real, μ', and imaginary, μ'', parts of the permeability of the ferrite slab. In Fig. 3, the measurements were extended to 10 GHz. At frequencies below 0.1 GHz, the TRL calibration[15] was inaccurate and that explained the erratic measurements of μ.

Let us now analyze the data in terms of the theory modeled in Sec. II. We see in Fig. 2 that there are two magnetic

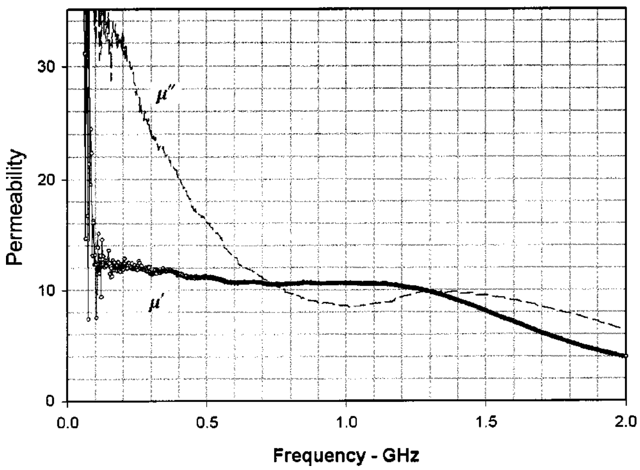

FIG. 2. Zero-field permeability data for single crystal Y-type hexaferrite from 0 to 2 GHz.

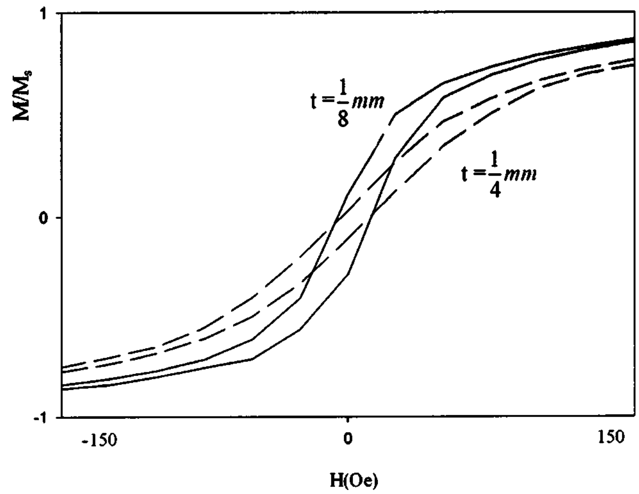

FIG. 4. Normalized magnetization is plotted as a function of external field. The parameter t is the slab thickness (solid line: $t=0.125$ mm, dashed line: $t=0.25$ mm) and $4\pi M_s$ is the saturation magnetization.

resonances, as μ'' exhibits maxima at these two frequencies. The first resonance occurred at about 200 MHz. We attribute this to the domain-wall resonance and the other one at 1.5 GHz to the zero-field FMR (in our designation ω_1). If we assume $L=1$ mm (length of the 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 μ'' is maximum at 170 MHz, μ' asymptotically reaches a value of 14.2 as $f\rightarrow 0$. We now wish to explain this result. As Eq. (5) predicts, we needed knowledge of both the remanance and H_φ^A to estimate μ'. The remanance $4\pi M$ was measured via a vibrating sample magnetometer (VSM) technique and we obtained a value of 88 G, see dashed line in Fig. 4. H_φ^A was measured from FMR measurements. By plotting the magnetic resonant field, H_r, versus the in-plane angle of the external field, φ, we were able to deduce the following $(N_y\text{-}N_x)M_s=20$ Oe. We utilized the fact that the demagnetizing energy is uniaxial in symmetry and the H_φ^A term field is six-fold symmetric, see Fig. 5.

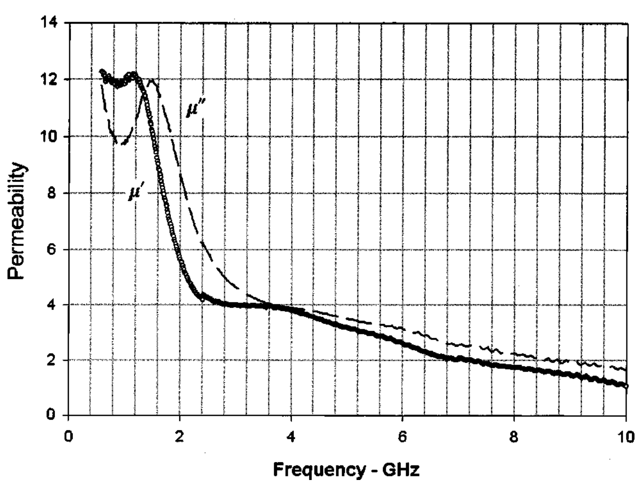

FIG. 3. Zero-field permeability data for single crystal Y-type hexaferrite from 0 to 10 GHz.

By applying Eq. (2) directly to the multidomain resonance occurring at 1.42 GHz, see Fig. 2, we deduced a value of H_φ^A of 6.2 Oe. Hence, we estimated μ' at zero frequency as 14.2, see Eq. (5), compared to experimental value of 15. According to Eq. (4), the out-of-phase multidomain resonance is predicted at ~3.3 GHz. From Fig. 3, resonance occurred between 3.5 and 5 GHz. However, the observed resonance was rather weak. In fact, if we take the absorption bandwidth, Δf, Fig. 3 as approximately 2 GHz, we deduce[11] a permeability of

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

compared to 4.0 in Fig. 3.

B. Oriented polycrystalline slabs

The same microstripline device and measurement technique were applied to measure the permeability of oriented

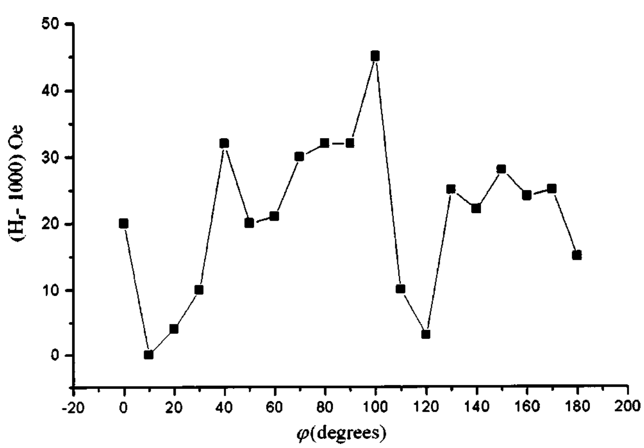

FIG. 5. In-plane FMR at 9.56 GHz, φ is the in-plane angle of rotation.

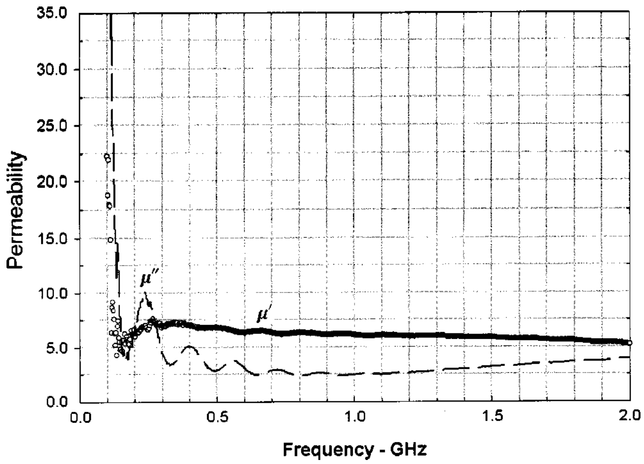

FIG. 6. Zero-field permeability data for oriented Y-type hexaferrite from 0 to 2 GHz.

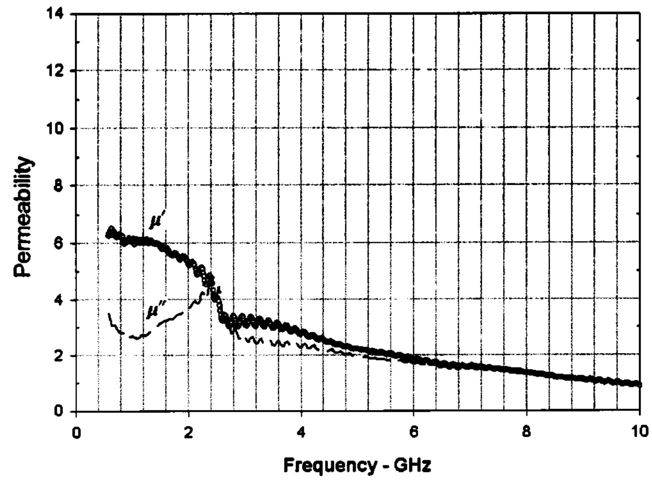

FIG. 7. Zero-field permeability data for oriented Y-type hexaferrite from 0 to 10 GHz.

Y-type hexaferrite slabs. The measured domain-wall resonance occurred at 200 MHz compared to 175 MHz in the single crystal slab, see Fig. 6. Other domain-wall resonances appeared at higher frequencies, which may be due to nonuniformities in orienting particles.

As in a previous analysis of our data on single crystal slabs, from FMR experiments, we deduced $H_\varphi^A \approx 23.6$ Oe and $(N_y - N_x)M_s = 20$ Oe. However, the remanence magnetization was measured to be 154 G by VSM for oriented polycrystalline slab samples. The deduced value of H_φ^A of oriented Y-type (as deduced from in-plane FMR) was roughly four times higher than the single crystal sample. This is reasonable since the actual chemical composition of the oriented particles was $Ba_2MnZnFe_{12}O_{22}$ and that of the single crystal composition was $BaZn_2Fe_{12}O_{22}$. The values of $4\pi M_s$, H_θ^A, and γ are the same as those of the single crystal samples.

From our measurements, we were able to deduce H_φ^A and other magnetic parameters (see Sec. II) and from these parameters, we predicted multidomain resonance at 1.89 GHz which compared very well with the experimental value of 2 GHz, see Fig. 7. Multidomain resonance was recognized as the frequency where μ'' was maximum, see Fig. 7. Using Eq. (5), we estimated the permeability, μ', as $f \to 0$ to be 6.5 compared to the experimental value of 6.2, see Eq. (5). There were no adjustable parameters in the estimate of μ'. Summary of the results are given in Table I.

IV. DISCUSSION AND CONCLUSION

In fitting Eq. (6) to the domain-wall resonance, we conclude that the thickness of the domain-wall thickness was about 0.47 μm. In the past, we have unsuccessfully attempted to orient Y-type hexaferrites particles of 0.5 μm in diameter. Based on the present results, the particle size needs to be greater than 0.5 μm in order to orient particles of Y-type hexaferrites. The permeability analysis and data showed that the initial permeabilities are correlated with H_φ^A and magnetization remanence, $4\pi M$, see Eq. (5). It may be possible to obtain very high permeabilities in the order of 100, if the magnetization remanence is increased or H_φ^A is reduced. This implies the use of Y-type hexaferrite materials whose compositions consist of *S*-state magnetic ions. One method of increasing $4\pi M$ or remanence is to reduce the in-plane demagnetizing field. For example, from VSM measurements on a 10 ml slab, remanence was measured to be 6.7%, but for 5 ml slab remanence was measured to be 19.7%. The coercive field was the same for both slabs (see the solid line in Fig. 4).

We believe that the proposed model by us for domain-wall resonances, multidomain 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 our model. We believe that

TABLE I. Summary of magnetic parameters as deduced from data.

Parameters	H_φ^A Oe	H_θ^A kOe	$4\pi M_s$ kG	$(N_y\text{-}N_x)M_s$ Oe	f_0(MHz)	f_1(GHz)	f_2(GHz)	$\mu'(f\to 0)$
Zn_2Y[a]	···	···	···	30	90–175	1.42	3.3	14.2
An_2Y[b]	6.2	9.5	2.0	20	100–200	1.5	3.0–5.0	13.0
MnZnY[a]	···	···	···	30	90–175	1.89	3.3	6.5
MnZnY[b]	23.6	9.5	2.3	20	150–230	2.0	3.0–5.0	6.2

[a]Predicted parameters.
[b]Measured parameters.

Y-type hexaferrite materials should form the base for high μ materials at microwave frequencies.

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