

Microwave and magnetic properties of self-biased barium hexaferrite screen printed thick films

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The interest in barium hexaferrite thick films, particularly with high remanent magnetization, is driven by the development of small planar ferrite microwave devices. We report here processing and microwave characterization of BaFe₁₂O₁₉ ferrite thick films (100–400 μm). The films were deposited on silicon and alumina substrates by screen printing, oriented under a magnetic field of 8 kOe, then annealed at 250 °C and sintered at temperatures ranging from 850 to 1300 °C. Scanning electron microscopy and x-ray diffraction exhibited strong crystallographic alignment of *c*-axis crystals perpendicular to the film plane. The magnetization measurement indicated that a typical dense film with 270 μm thickness yielded a high squareness (M_r/M_s) of 0.93. Ferrimagnetic resonance (FMR) measurements were performed in the frequency range of 40–55 GHz. From the linear dependence of FMR frequency on the external field, a *g* factor of 2.03 ± 0.08 was deduced, while the smallest linewidth was obtained to be 1.2 kOe at 40 GHz. The broadening of the FMR linewidth arises predominantly from magnetic inhomogeneities in the polycrystalline films. The thick films have great potential for use in future microwave and millimeter wave monolithic integrated circuits. © 2006 American Institute of Physics. [DOI: 10.1063/1.2163288]

I. INTRODUCTION

Polycrystalline hexagonal ferrites of the *M* type, BaFe₁₂O₁₉, have traditionally been used as permanent magnets in various electronic and microwave applications due to its large uniaxial magnetic anisotropy. Over the past several decades, it has been shown that microwave ferrite materials can be used effectively in circulators, filters, isolators, inductors, and phase shifters, which are usually built using bulk ferrite materials.¹ In an age when communications systems are to be highly portable and fully integrated into a small chip, this approach is no longer satisfactory. The trend today is toward replacing bulk ferrite materials with thick and thin ferrite films integrated into a single semiconductor package. The fabrication and utilization of such ferrite thick films for microwave communication devices are of significance in the development of modern microwave technology.^{2–4}

In recent years, several thin film deposition techniques, such as pulsed laser deposition (PLD)⁵ and liquid phase epitaxy (LPE), have been used to grow high quality yttrium iron garnet (YIG) and barium hexaferrite films with thicknesses ranging from 1 to 100 μm.^{6,7} Any attempts to produce thicker films (>100 μm) resulted in stressed or cracked films with extremely large linewidths.

Screen printing technology is capable of producing films up to 1 mm thick. In the modern electronics industry, the technique is usually applied to produce thick film circuits and sensors.⁸ Remarkably, this technique has not been extended to fabricate textured ferrite thick films for microwave applications. In this paper, we have investigated this preparation technique for the purpose of producing ferrite thick

films of good quality to be used in microwave devices. Preliminary results demonstrate that we can produce ferrite thick films (100–400 μm) with microwave losses lower than those found in polycrystalline bulk materials. The details of screen printing preparation, and microwave and magnetic characterization are presented for *c*-axis oriented Ba ferrite at 40–55 GHz.

II. EXPERIMENT

Thick films of *M*-type barium ferrite were processed on silicon and alumina substrates using a screen printing technique.⁹ The powders for the screen printing are prepared by conventional ceramic processing. After solid state reaction of the initial oxide reagents (BaCO₃:Fe₂O₃=1:6) at 1250 °C, 15 h, the powder was reduced to 1 μm diam particles by ball milling. X-ray diffraction (XRD) analysis indicates that the starting powders are a pure hexagonal BaM phase.

A paste suitable for screen printing was obtained by combining the barium hexaferrite powders with a binder (e.g., an epoxy and hardener). The paste consisted of 70–75 wt. % BaM powder, a 20–25 wt. % binder, and a 2–5 wt. % glass frit. After the thick films were printed onto the silicon or alumina substrate, the most critical step was to dry the wet printed film at a temperature of 250 °C in a magnetic field of 8 kOe aligned perpendicular to the film. The magnetic field was applied for the whole “dry” cycle. Then, the dried sample was sintered in air for a time of 2–10 h at temperatures ranging from 850 to 1300 °C. The sintering temperature was chosen based upon the initial loading factor and ultimately the desired magnetic and micro-

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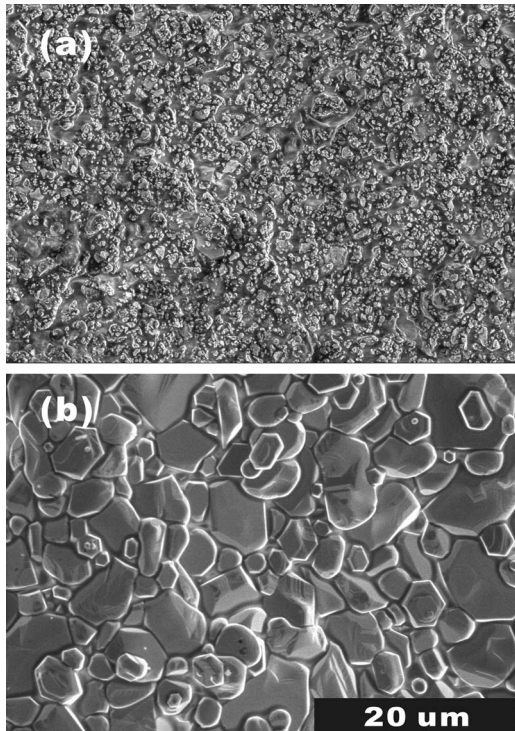


FIG. 1. SEM micrographs of the surface of the screen printed BaM thick films (a) before sintering and (b) after sintering at 1200 °C for 3 h.

wave properties of films. The sample was shaped into a disk with a diameter of 8 mm and thicknesses ranging from 100 to 400 μm .

The screen printed films were characterized in terms of their structural, magnetic, and microwave properties. XRD provided information on the crystal structure and orientation of the films. The morphology of the films was observed using a scanning electron microscope (SEM). In order to measure the millimeter-wave properties of the films, ferrimagnetic resonance (FMR) measurements were performed between 40 and 55 GHz using a field-swept waveguide technique with the dc magnetic field parallel to the c axis. The FMR linewidth, ΔH , the Landé g factor, and H_A can be obtained from the FMR data. Saturation magnetization $4\pi M_s$ was measured using a standard vibrating sample magnetometer (VSM) with a maximum field of 12.5 kOe.

III. RESULTS AND DISCUSSION

Figure 1(a) shows a morphology for the printed film after the “burnout” of the binder and before sintering at the elevated temperature. Since the binder cannot be burned out completely at 250 °C, the film contained a lot of pores and residual binders. Small particles of $\sim 1 \mu\text{m}$ appeared to be loosely arranged on the substrate. After sintering, the film displayed a distinctly different morphology. The sintering of the samples resulted in a dense polycrystalline structure with grains oriented with the c axis perpendicular to the film plane. It was noted that a recrystallization was sensitively dependent upon the sintering temperature and time. As shown in Fig. 1(b), it is easy to find many hexagonal BaM crystals that are aligned with the c axis perpendicular to the film plane having an average grain size of 3.2 μm . XRD

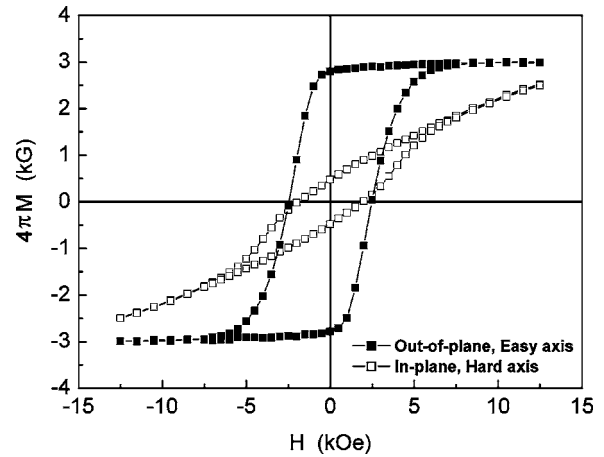


FIG. 2. Hysteresis loops of a typical screen printed BaM ferrite film with thickness of 270 μm .

results reveal peaks having enhanced intensity corresponding to (006), (008), and (0014) planes in this sample.

Hysteresis curves were measured with the dc magnetic field applied normal to the film plane and in the film plane. Figure 2 shows a typical hysteresis curve for a 270 μm thick film sintered at 1200 °C for a time of 3 h. For H_0 , the external field, applied normal to the film plane and parallel to the c axis the squareness ratio, M_r/M_s , was measured to be 0.93 (where M_r and M_s are the remanence and the saturation magnetization, respectively.) The c -axis orientation was confirmed by the SEM and XRD measurements. The saturation magnetization of the film was measured to be $4\pi M_s = 3 \pm 0.25 \text{ kG}$ with a coercive field $H_c = 2.468 \text{ kOe}$. Again for the H_0 external field perpendicular to the film plane, an external field of 12.5 kOe was sufficient to saturate the film. For H_0 in the film plane, we measured a squareness of 0.15 and a coercive field of 1.927 kOe.

In Fig. 3(a), we have plotted the FMR frequency as a function of external field for H_0 applied perpendicular to the film plane. For fields above 2 kOe, the measured resonance dispersion obeyed the following relation:

$$f = \gamma'(H_0 + H_A - NM_s), \quad (1)$$

where f is the resonance frequency, H_0 is the resonance external field, and $\gamma' = 2.85 \text{ GHz/kOe}$. Clearly, a 2 kOe field can almost magnetize the sample to saturation [see the inset to Fig. 3(a)]. We obtained a linear fit to the data having the equation $f = 2.85H_0 + 39.16$; therefore, the Landé g factor was deduced to be $g = 2.03 \pm 0.08$. An extrapolated zero field resonant (ZFR) frequency was 39.16 GHz, which is less than the reported ZFR frequency of 47 GHz.¹⁰ The difference is attributed to the fact that in the presence of multidomains the resonance condition is given as $f = \gamma H_A$ rather than Eq. (1). Thus, the difference is the factor, $\gamma 4\pi M_s$, or about 8 GHz. Furthermore, fitting the experimental data to Eq. (1), we obtained $H_A - 4\pi M_s = 13.75 \text{ kOe}$, where $4\pi M_s = 3 \text{ kG}$, as deduced from the VSM measurement. Thus, H_A was extracted to be 16.7 kOe, which is in reasonable agreement with the anisotropic field $H_A = 16.4 \text{ kOe}$ obtained from the VSM measurement. The results are very close to reported value (17.4 kOe) for epitaxial BaM ferrite films.⁷

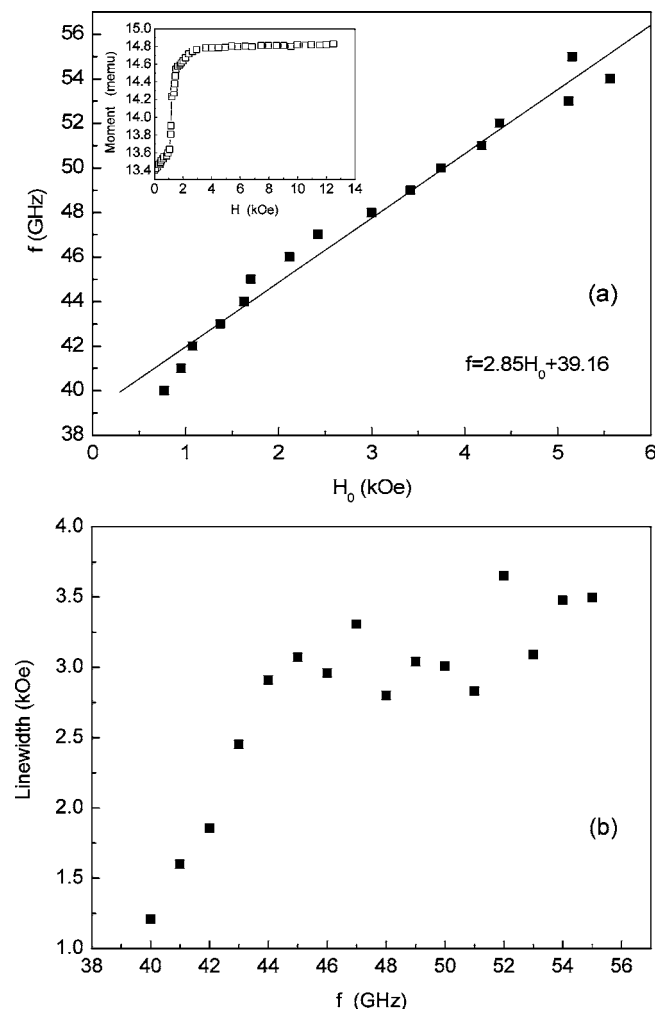


FIG. 3. (a) Dependence of FMR frequency on the external field for a screen printed ferrite 270 μm thick film. The inset shows a magnetization curve starting at the remanent magnetization M_r . (b) FMR linewidth vs frequency at the frequency range of 40–55 GHz.

Figure 3(b) is a plot of FMR linewidth versus frequency (40–55 GHz). The plot shows that the linewidth, ΔH , varies weakly with frequency for the range of 45–55 GHz. It is worth noting that the linewidth depends strongly on frequency below 45 GHz. For example, a minimum linewidth ($\Delta H = 1.2$ kOe) is measured at a frequency of 40 GHz. The value of the linewidth was less than the previously observed ($\Delta H > 2$ kOe) in polycrystalline BaM ferrite.^{11,12} There are various mechanisms that broaden the linewidth and they include

$$\Delta H = \Delta H_i + \Delta H_a + \Delta H_p, \quad (2)$$

where ΔH_i is the intrinsic linewidth, ΔH_a is the broadening due to the random anisotropy axes of different grains, and ΔH_p is the broadening due to porosity. The contribution from ($\Delta H_a + \Delta H_p$) is referred to as the extrinsic mechanism, e.g., grain boundaries and structural imperfections. In polycrystalline ferrites, the pores give rise to an inhomogeneous field that causes different parts of the sample to resonate under different external fields. Therefore, the pores will be the primary contribution to the broadening linewidth. Based on a

linewidth broadening model,¹³ we have calculated the contribution of porosity to linewidth in our sample with 33% porosity. The calculation indicated that the porosity gives rise to 0.750 kOe linewidth broadening in BaM ferrite. Obviously, inhomogeneity plays a dominant role in broadening linewidth in thick film.

Another source of line broadening in the polycrystalline films can arise from the random orientation of anisotropy energy axes from grain to grain. Although this film has a high squareness of 0.93, it is believed that the broadening linewidth arises partially from nonaligned grains. We estimated this contribution to be approximately $\Delta H_a = 0.385$ kOe based on our experimental value ($\Delta H = 1.2$ kOe), and calculated $\Delta H_p = 0.750$ kOe and $\Delta H_i = 0.065$ kOe. Our estimate of ΔH_a is comparable to values in the literature.¹⁴

IV. CONCLUSION

This article reports on the processing of thick, low loss, self-biased hexaferrite films on a silicon and alumina substrates by screen printing. The method allows for the growth of thicker ferrite films in comparison to other techniques, including pulsed laser deposition and liquid phase epitaxy deposition. The films exhibited superior magnetic and microwave characteristics for the *c*-axis self-biased hexaferrite thick films when compared to polycrystalline compacts. The textured polycrystalline thick films have a thickness ranging from 100 to 400 μm , anisotropy field of the order of 16.7 kOe, saturation magnetization of 3 kG, coercive field of 2.4 kOe (in easy axis), and FMR linewidths of 1.2 kOe at 40 GHz. Our estimates for the extrinsic contribution to the linewidth are in reasonable agreement with the measured data.

ACKNOWLEDGMENTS

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