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Oriented barium hexaferrite thick films with narrow ferromagnetic resonance linewidth

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Hexagonal BaFe₁₂O₁₉ ferrite films, having thicknesses ranging from 200–500 μm, were prepared by a screen printing process followed by sintering heat treatments. Structural, magnetic, and microwave measurements confirmed that the polycrystalline films were suitable for applications in self-biasing microwave devices in that they exhibited a large remanence ($4\pi M_r=3800$ G), high hysteresis loop squareness ($M_r/M_s=0.96$) and low microwave loss. A derivative linewidth ΔH of 310 Oe was measured at 55.6 GHz. This represents the *lowest* ΔH measured in polycrystalline hexaferrite materials. ΔH can be further improved by reducing porosity and improving the *c*-axis orientation of grains in polycrystalline ferrite. © 2006 American Institute of Physics. [DOI: 10.1063/1.2173240]

Hexagonal barium ferrites are a class of magnetic materials that potentially can play an important role in the propagation of electromagnetic waves in microwave and millimeter wave devices. These components are an important part of a diverse range of commercial and military applications from radar to wireless communications.¹ In particular, a key component of radar electronics is the circulator. Traditionally, circulator designs have included ferrites pressed as compacts with biasing magnets placed above and/or below to provide the necessary biasing magnetic field for operation. These designs result in three-dimensional constructs that are often bulky and costly to fabricate. Special circuitry and packaging features of the permanent magnet need to be considered in all microwave devices.²

One important challenge in the development of next-generation nonreciprocal millimeter wave devices for monolithic microwave integrated circuits applications is the development of devices that are planar, self-biased and low loss. Since the concept of self-biased devices was first proposed in the 1970's,³ fabrication of thick ferrite films by pulsed laser deposition,⁴ liquid phase epitaxy,⁵ and screen printing⁶ has been explored. To this day, the development of oriented ferrite thick films with low microwave losses has been a long-standing problem whose solution will have a wide ranging impact on the field of microwave magnetic devices.

The present work focuses on the refinement of the screen-printing process that can lead to the development of thick ferrite films with self-biased and low microwave loss properties. We have developed a processing scheme in which Ba-hexaferrite (M-type, henceforth BaM) films have been produced having thicknesses up to 500 microns, high remanent magnetization (>95% of saturation values), perpendicular magnetic anisotropy axis, and low microwave losses [ferromagnetic resonance (FMR) linewidths of less than 500 Oe] at U band. These are all relevant magnetic and/or electrical properties which are necessary to allow the development of planar microwave circulators.

The processing technique made use of a screen printing technology where a paste, consisting of BaM particles suspended in a binder (B-75000, Ferro), was spread over a template onto a suitable microwave substrate, e.g., 0.25 mm thick alumina (supplied by Accumet Engineering Corp.). The starting powders were prepared by conventional ceramic processing. After repeated solid state reaction of the initial oxide reagents (BaCO₃:Fe₂O₃=1:6) at 1250 °C for 15 h, the powders were reduced to 1.0–1.5 μm diameter particles by ball milling. The screen printing paste typically consisted of 25.5 wt % binder, 2.5 wt % glass frit, and 72 wt % barium hexaferrite powder.

The resulting film was subjected to a magnetic field (~8000 Oe) aligned perpendicular to the film plane during the first heat treatment (150–250 °C, 1–20 min). The magnetic field acting upon the still “wet” film aligned the BaM particles with respect to the magnetic field direction. This alignment forced the *c* axes of the hexaferrite particles, and subsequently the magnetization, to align along the direction perpendicular to the film plane. This low-temperature heat treatment acts to vaporize the binder and fix the orientation of the particles. The film then underwent a “hot-pressing”⁷ during the second sintering heat treatment in air to temperatures ranging from 900–1300 °C at times ranging from 1–15 h. This hot-pressing process is an essential step in improving the film density and reducing the microwave losses of the film. A third heat treatment was sometimes required to complete the sintering of the film and to reduce strain. After the high-temperature anneals, the film density was improved to 85–90% and loop squareness (i.e., M_r/M_s) to 93–96%. The films had a diameter of 8 mm and thickness of 200–500 μm.

Figure 1 is an x-ray diffraction (XRD) pattern acquired using Cu *K*α radiation from a BaM film after alignment and heat treatment procedures. In this figure, the diffraction peaks, indexed to (1, 0, 2*n*) and (2, 0, 2*n*), have been identified. The enhanced intensity of these reflections is consistent with the preferential alignment of *c*-axis grains perpendicular to the sample plane. After the signals corresponding to the substrate and sample holder were extracted from the XRD pattern, the film's diffraction pattern was fully consistent

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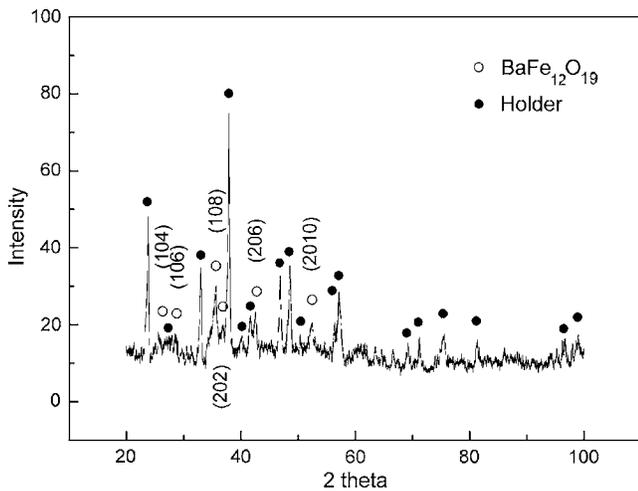


FIG. 1. XRD pattern acquired using Cu $K\alpha$ radiation from a BaM film after alignment and heat treatment procedures.

with a pure phase hexagonal M-type structure.

Scanning electron microscopy (SEM) images of the screen printed film after alignment and heat treatments are displayed in Fig. 2. In Fig. 2(a), one sees the morphology of the surface of a 200 μm thick film having a density of 88%, i.e., porosity of 12%. Although the distribution of grain size is broad, most of the grains ranged in size from 1.5 to 4.0 μm , the average grain diameter was about 2.2 μm . For a block of uniaxial crystal, domain spacing can be estimated from Kittel's approximation:⁸ $D_0 = M_s^{-1}(\sigma_w/1.7)^{1/2}L^{1/2}$, where $\sigma_w = 4\sqrt{AK_u}$ represents density energy of 180° wall, and L is thickness of sample. Highly grain-oriented polycrystals can approximately simulate single crystals since coupling between grains is relatively strong. As such, our sample domain spacing, D_0 is 6.7 μm , assuming exchange stiffness constant

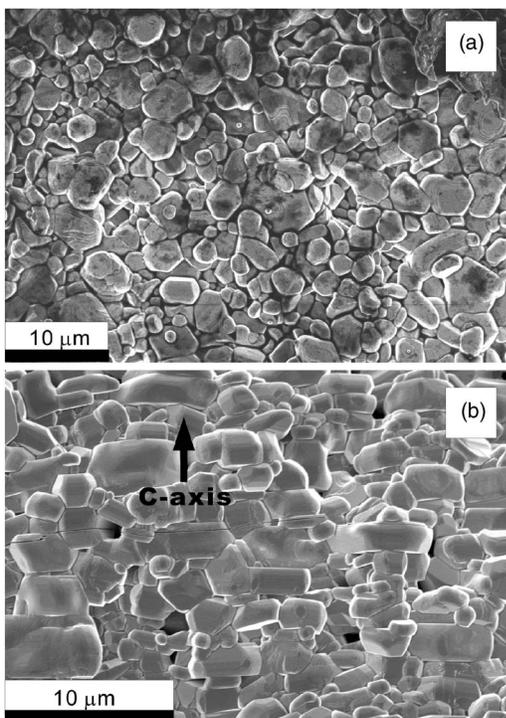


FIG. 2. SEM images of a 200 μm screen printed film after alignment and heat treatment procedures: (a) Surface and (b) cross section.

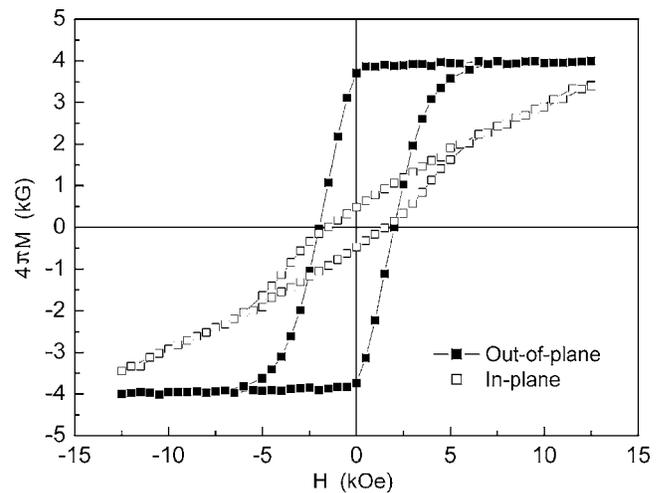


FIG. 3. Magnetic hysteresis loops acquired with the applied magnetic field aligned along the in-plane sample direction (open squares) and perpendicular to the sample plane (solid squares).

of $A = 0.5 \times 10^{-6} \text{ erg cm}^{-1}$ (Ref. 9) and $K_u = 3.3 \times 10^6 \text{ erg cm}^{-3}$.⁸ In addition, we estimate the domain spacing by minimum of magnetostatic energy to be 6.1 μm , which is consistent with the above approximation. The estimates indicate that most of the grains in the film are single domain, since our measured grain size of 2.2 μm is much smaller than the estimated domain spacing.

However, fine grains lead to a broadening of the FMR linewidth, as a result of magnetoelastic scattering.¹⁰ Therefore, optimization of grain size and its distribution will dominate the microwave losses in self-biased ferrites. In Fig. 2(b), the cross section of the film shows highly oriented grains with an average distribution angle of $< 5.8^\circ$ of the c -axes. We clearly see oriented platelets aligned perpendicular to the film plane, though one can also see isolated pores between grains. The average height of the hexagonal grains was estimated to be 1.4 μm .

Figure 3 is a plot of the magnetic hysteresis loops recorded with the applied magnetic field aligned along the in-plane sample direction and perpendicular to the sample plane. We measured $4\pi M_s$ of 3960 G, which was lower than the reported magnetization value of 4500 G.⁵ The discrepancy in $4\pi M_s$ was due to the fact in our measurements we included porosity and defects in the volume of the sample. In this figure, the square loop with high remanent magnetization (solid squares) corresponds to the out of plane orientation. This is very important in that it demonstrates two important properties of the screen printed films. First, the magnetization prefers the direction normal to the sample plane, and second, that upon removal of the applied field the sample retains up to $\sim 96\%$ of the saturation magnetization. Only the film consisting of single-domain grains can yield such a high loop squareness or remanence, which is further evidence to support the discussion above on single-domain grains. Otherwise, formation of multidomain grains destroys the self-biasing of the film character. This remanence enhancement may be also attributed to exchange and dipolar coupling between grains. The tendency of neighboring grains to align M_s parallel to each other results in a magnetic texture which minimizes the exchange energy between interacting grains. High remanence and saturation magnetization, as well as low

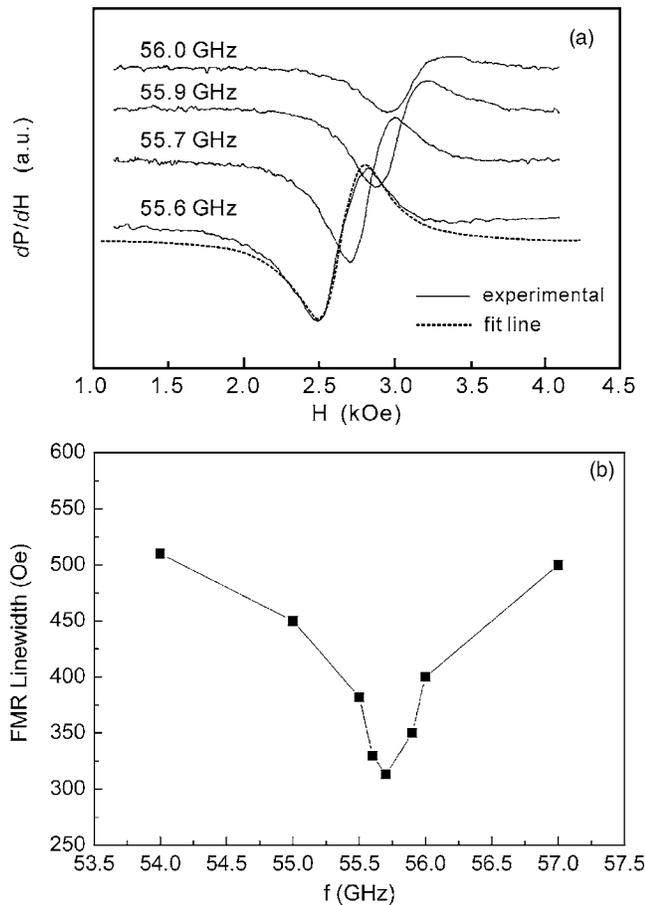


FIG. 4. (a) Variation of the derivative of power absorption with external field perpendicular to the ferrite film plane. (b) Variation of FMR derivative linewidth with external frequency.

coercive field are highly desirable properties for applications in microwave magnetic devices.

Figure 4(a) shows a plot of the derivative of the absorbed power versus the external field H . The solid line traces experimental data at different frequencies: 55.6, 55.7, 55.9, and 56.0 GHz, whereas the dash line shows a fit based on a Lorentzian absorption response with a center field position of 2647 Oe and a half power linewidth of 533 Oe.

Figure 4(b) shows the variation of the FMR derivative linewidth ΔH with frequency over a range of 54–56 GHz. One can clearly see a V-shaped curve with a minimum FMR linewidth of 310 Oe corresponding to a frequency of 55.6 GHz. This value is much less than values previously reported for polycrystalline materials that typically range above 2000 Oe.³ Obviously, a linewidth measurement of 310 Oe represents the narrowest linewidth measured in polycrystalline hexaferrite materials. From Fig. 4(b), clearly, ΔH does not scale linearly with frequency, which is usually observed for single crystals and epitaxial films.¹¹ This implies that the inhomogeneous broadening of the linewidth is not linearly proportional to frequency. Clearly, the materials prepared by a screen printing technique are of a sufficiently high quality

in FMR linewidth to be useful for many planar microwave applications.

In order to analyze qualitatively the nonuniform contributions to the FMR linewidth in our samples, we express the contribution to linewidth as follows^{12,13}

$$\Delta H = \Delta H_{anis} + \Delta H_{poros} = 1.08J \left(\frac{H_A^2}{4\pi M} \right) + 0.502J4\pi Mp, \quad (1)$$

where H_A is the anisotropy field equal to $2|K_u|/M$, p is the porosity in the material, and J is a shape factor.¹² We estimate $\Delta H_{anis} = 64$ Oe and $\Delta H_{poros} = 210$ Oe from which $\Delta H = 274$ Oe; this is assuming a porosity $p = 0.12$, $4\pi M = 3960$ G, and $J = 0.87$ (demagnetizing factor of $N_Z = 0.9$). The measured linewidth was 310 Oe. Hence, the remaining or residual linewidth may be interpreted in terms of an intrinsic linewidth of 36 Oe. This value compares well with measured values of ΔH in LPE films of 27 Oe at 56 GHz.⁶ In the thick film, about 70% of the total linewidth is attributed to porosity. Polycrystalline materials are almost always porous, giving rise to a line broadening which is very significant as compared with the effects of anisotropy, even for 99% dense ferrites.¹⁴ Experimental results presented here imply that a further reduction in linewidth can be achieved by increasing the film density and by enhancing the alignment of grains with the c -axes perpendicular to the film plane.

In summary, we have developed a screen printing processing scheme for the production of several hundred microns thick films of Ba-hexaferrite having high remanent magnetization, low FMR linewidth (~ 310 Oe), and magnetic anisotropy perpendicular to the film plane. The work has demonstrated that the screen printing technique is capable of processing thick, self-biased, low-loss BaM films; an integral step in the processing of planar microwave magnetic devices.

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