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Magnetic and microwave properties of basal-plane oriented $BaFe_{11}In_1O_{19}$ ferrite thick films processed by screen printing

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In-doped $BaFe_{11}In_1O_{19}$ particles were prepared by a modified ceramic reaction using In_2O_3, $BaCo_3$, and Fe_2O_3 followed by mechanical dispersion. X-ray diffraction analysis confirmed the formation of pure $BaFe_{11}In_1O_{19}$ phase and scanning electron micrographs showed platelet particles of about 1 μm in diameter. This powder was subsequently screen printed on alumina substrate using a suitable binder and oriented under a dc magnetic field of 15 kOe. The screen printed films were annealed at different durations to produce dense and thick ferrite materials. The hysteresis loops for the as-prepared and annealed screen printed, in-plane oriented films show a hysteresis squareness ratio (M_r/M_s) of 0.93, saturation magnetic moment of 4000 G, and coercivity of 634 Oe. The ferrimagnetic resonance measurements showed a linewidth (ΔH) of $\sim$860 Oe. The g (Lande spectroscopic splitting factor) value deduced from the relation between resonant frequencies versus resonance field for the screen printed films was found to be 1.91. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2829905]

I. INTRODUCTION

In recent years, microwave magnetic materials have attracted considerable attention due to their important role in microwave and millimeter wave applications (isolators, phase shifters, circulators, and related components).[1–4] Hexagonal ferrites have been proposed as one of the possible material solutions to make microwave devices since the large uniaxial magnetocrystalline anisotropy field (H_A $\sim$17 000 Oe) can be used as an advantage by reducing the need for external high magnetic biasing fields and the low microwave losses [low ferromagnetic resonance (FMR) linewidth]. One advantage of the hexaferrite is that the high anisotropy field can be adjusted by appropriate substitution for Fe and Ba ions, allowing for tuning the resonance frequencies from 1 to 100 GHz. This degree of freedom makes the hexagonal ferrite a choice material for many monolithic microwave integrated circuit applications.[5] Magnetic properties ($4\pi M$ and H_A) can be systematically varied by substituting for the Fe cations. Elements such as scandium (Sc), aluminum (Al), and indium (In) are used to replace the Fe^{3+} ion in $BaFe_{12}O_{19}$. The substitution of Sc or In for Fe^{3+} has shown to reduce the anisotropy field (H_A) of the hexaferrite.[6–8] The design of X-band circulators and phase shifters requires the low anisotropy compared with the pure barium ferrite, high remanent and saturation magnetization, high Nèel temperature, and low FMR linewidth. Also, the Sc or In doping will stabilize the valence state of Fe^{3+} and avoid high loss tangents.[9,10] The anisotropic field can vary over a wide range as a function of substitution level.[6] In addition to the composition, shape, and size, the orientation of the particles has a significant effect on its microwave absorption properties. When the particle size is at or near the single domain size, the domain-wall resonance mechanism no longer exists. Most of the publications in pure and doped hexaferrite materials dealt with the preparation of thin films by pulsed laser deposition and liquid phase epitaxy methods. In this paper, preparation and magnetic and microwave characterizations of In-substituted Ba ferrite ($BaFe_{11}In_1O_{19}$) are presented using a modified ceramic technique. The main goal of this work is to prepare materials with magnetically aligned grains along the in-plane direction, while maintaining a moderate coercivity, and high remanent magnetization for self-biased microwave device applications at low frequencies.

II. EXPERIMENTAL

High purity $BaCO_3$, Fe_2O_3, and In_2O_3 were used as raw materials. They were weighed stoichiometrically and then mixed by using a low energy ball mill. The pressed compact powders were preheated at 1100 °C, crushed, and sintered. After it is crushed again and sieved, it finally goes through an annealing process. In order to obtain single phase crystallographic characteristics and magnetic properties, a post-thermal-annealing was required. Samples were heated in air at a rate of 5 °C/min. The temperature was then maintained between 1000 and 1150 °C for 5 h and then cooled to room temperature. The sintered compact samples were milled using a planetary ball mill. Since the In_2O_3 has a relatively low melting point, it will easily accelerate grain growth through

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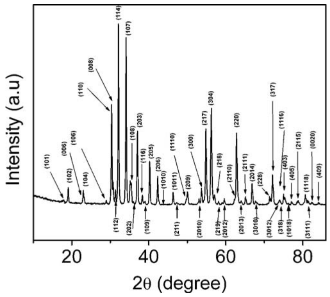

FIG. 1. X-ray diffractograms of the $BaFe_{11}In_1O_{19}$.

its own fluxing action. The milled particles were suspended in an epoxy and hardener and screen printed onto a dielectric (Al_2O_3) substrate with a thickness of about 1–2 mm. Particle orientation was performed under an external dc magnetic field of 15 kOe. The screen printed and in-plane oriented green compact were annealed at different temperatures to make highly dense thick films for further characterization.

The crystallographic phase of the particles was analyzed using θ-2θ x-ray powder diffraction (XRD) (Rigaku, Cu $K\alpha$ radiation, $\lambda = 1.545\,06$ Å). The surface morphology of the particles was examined by scanning electron microscopy (SEM) (Hitachi S-4100). Chemical analyses have been carried out using an induction coupled plasma spectrophotometer (ICP 20P VG Elemental Plasma Quad2) and as well as SEM-EDAX facility. The magnetic properties were measured using a vibrating sample magnetometer (ADE Technologies). FMR measurements were performed in both out-of-plane and in-plane FMR conditions by using a TE_{01} rectangular waveguide at room temperature in K_a-band frequency. The FMR data allow us to calculate the effective magnetization, anisotropy field, and FMR linewidth (ΔH_{FMR}).

III. RESULTS AND DISCUSSION

XRD patterns in Fig. 1 confirmed the formation of single phase In-doped Ba-hexaferrite with the magnetoplumbite structure.[11] Chemical and SEM-EDAX analyses showed that the samples had the appropriate stochiometric ratios of Ba, Fe, and In. The scanning electron micrograph [Fig. 2(a)] of the $BaFe_{11}In_1O_{19}$ sample sintered at 1150 °C shows the formation of larger grains, which are about a few microns and have better intergrain connectivity. Figure 2(b) shows the $BaFe_{11}In_1O_{19}$ particles after ball milling with an elongated thin platelet shape. The average size of the particles will be about 1–1.5 μm. The ball milled particles were screen printed onto alumina (Al_2O_3) substrate using a suitable binder and hardener. The loading factor, i.e., the binder and particle ratio, is 70:30. A maximum dc magnetic field of 15 kOe was employed to orient the particles along the basal-plane direction. The screen printed films were then annealed at different temperatures to produce a dense and thick film. Figures 3(a) and 3(b) showed the cross section of the $BaFe_{11}In_1O_{19}$ film sintered at 1000 and 1100 °C for 1 h, respectively. The structure of the film is revealed to contain elongated grains with the short axis parallel to the c axis. Some pores remain visible. Figure 4 represents the hysteresis loops for the in-plane oriented and sintered $BaFe_{11}In_1O_{19}$ film at 1000 and 1100 °C for 1 h. These films have a coercivity of 1210 Oe but also a very high hysteresis loop squareness ratio of 0.93, providing these thick films with self-bias properties. When the sintering temperature was increased from 1000 to 1100 °C, the coercivity decreases to 1067 Oe and the squareness was greater than 0.9. The saturation magnetic moment for both the films was about 4000 G.

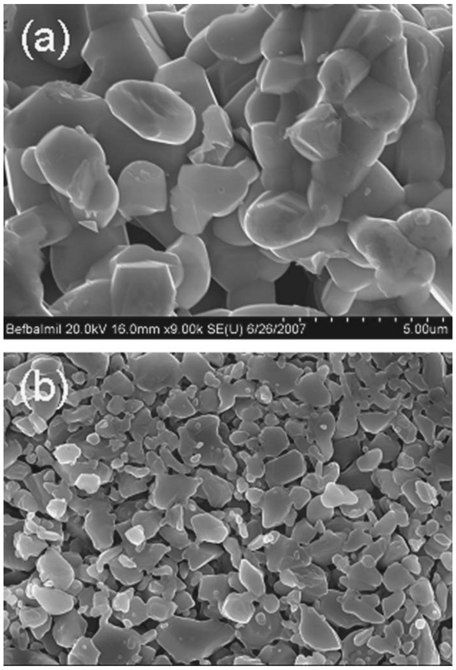

FIG. 2. Scanning electron micrograph of the $BaFe_{11}In_1O_{19}$ (a) bulk and (b) after ball milling.

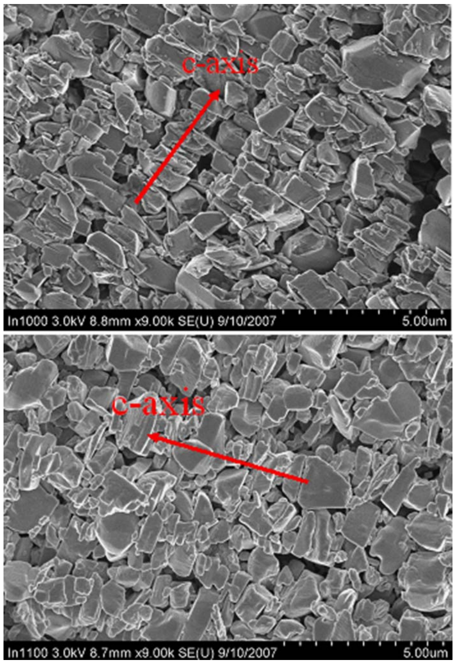

FIG. 3. (Color online) Scanning electron micrograph of the screen printed, in-plane oriented $BaFe_{11}In_1O_{19}$ thick films sintered at (a) 1000 °C and (b) 1100 °C for 1 h.

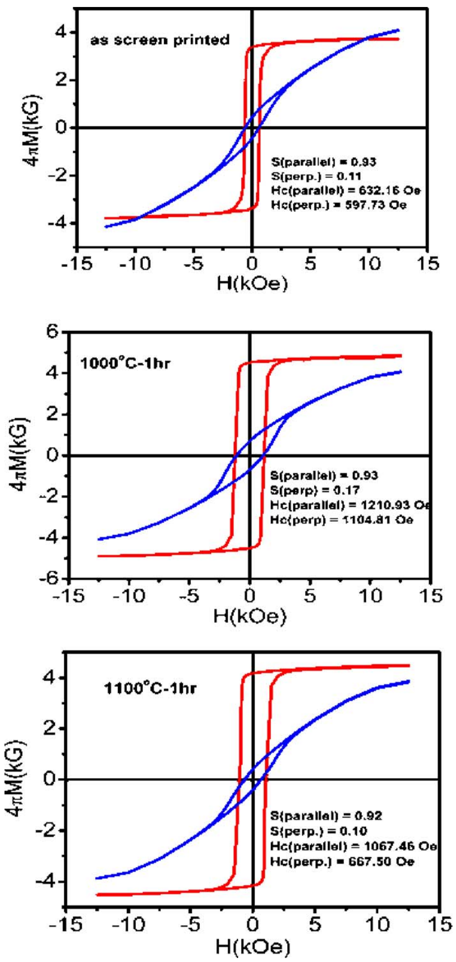

FIG. 4. (Color online) The hysteresis loops for the as-prepared and screen printed, in-plane oriented $BaFe_{11}In_1O_{19}$ film (a) before sintering, (b) after sintering for 1 h at 1000 °C, and (c) after sintering at 1100 °C for 1 h.

FMR measurements were performed by applying a swept dc magnetic field parallel to the film plane, i.e., parallel FMR configuration. The frequency was fixed during each field sweep and the measurements were taken for a frequency range from 27 to 40 GHz. When H_{ext} is parallel to the film, the FMR condition is given as follows:[12]

$$\frac{\omega}{\gamma} = \sqrt{(H_{\mathrm{ext}} + H_A)(H_{\mathrm{ext}} + H_A + 4\pi M_S)},$$

where $\omega=2\pi f$ and $\gamma=2\pi(g\times 1.4\times 10^6)$ Hz/Oe. Figure 5 shows the variation of the FMR derivative linewidth (ΔH) with frequency over a range of 27–40 GHz for the $BaFe_{11}In_1O_{19}$ thick films. A minimum linewidth of 860 Oe was realized. These values are small compared to the polycrystalline compacts (typically >2000 Oe) acceptable for many microwave applications. The linewidth can be further reduced by improving the density of the film. An experimental value for the Lande spectroscopic splitting factor (g), deduced, from the relation between resonant frequencies and the resonance field, was 1.91 which is in good agreement with $g=2$ reported for bulk samples.[13,14]

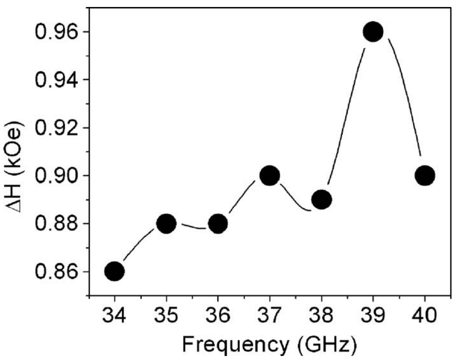

FIG. 5. FMR derivative linewidth (ΔH) as a function of frequency. Continuous line is a guide to the eyes.

IV. CONCLUSIONS

In-plane oriented $BaFe_{11}In_1O_{19}$ thick films have been prepared using a modified ceramic method followed by ball milling and screen printing. These films have low coercivity of 1210 Oe but also a high hysteresis loop squareness ratio of 0.93, with self-bias properties. The high squareness ratio, coercivity, and narrow linewidth depend strongly on the sintering temperature. A minimum FMR linewidth of 860 Oe has been achieved for the screen printed films. We conclude that these in-plane oriented films are possible materials for microwave devices such as phase shifters.

ACKNOWLEDGMENTS

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