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Microwave properties of pulsed laser deposited Sc-doped barium hexaferrite films

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The properties of thick $BaSc_xFe_{12-x}O_{19}$ ($0.3<x<0.6$) scandium hexaferrite films were measured by static and microwave field techniques. Films were deposited by pulsed laser ablation onto *c*-plane sapphire at oxygen pressures of 20 and 50 mTorr. Vibrating sample magnetometer measurements as corroborated by x-ray data showed that the films below 3 μm had easy axis of magnetization (*c*-axis) normal to the film plane with saturation magnetization values of 3.0–3.8 kG. From the ferrimagnetic resonance frequency versus external magnetic field, we deduced a *g* value of 1.96 ± 0.03 and uniaxial anisotropy field of $\sim$10 kOe. The ferrimagnetic resonance linewidth for the film thicker than 5 μm was maximum at 32 GHz and decreased with increasing frequency, indicating evidence for nonuniform magnetic field scattering internally. However, the linewidths were lower for films having thickness below 3 μm.
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I. INTRODUCTION

Hexagonal ferrites are playing more and more important roles in microwave and millimeter wave applications. Because of their strong internal anisotropy field (H_A), microwave and millimeter wave devices that use hexagonal ferrites will require minimum external magnetic biasing fields. This effect can greatly reduce the overall device size compared to those made from spinel or garnet ferrites operating in the same frequency range.[1] In particular, *M*-type hexagonal ferrites $MeO{\cdot}6Fe_2O_3$ ($Me=Sr, Pb, Ba$) are emerging as important materials in microwave and millimeter wave devices, such as circulators. *M*-type hexagonal ferrites have saturation magnetization ($4\pi M_S$) values around 4 kG,[2] which are much higher than the garnets and comparable to spinel ferrites. This high value of $4\pi M_S$ allows for wideband microwave and millimeter wave devices.

One advantage of the hexaferrites is that the magnetic properties can be systematically varied by substituting for the Fe cations. In particular, the substitution of scandium for iron has shown to provide interesting magnetic properties, since increases in the scandium substitutions lead to reduction in the value of H_A.[3] The design[1] of the wideband circulators working at 35 GHz requires, for example, H_A to be about 11 kOe. The H_A values for $BaSc_xFe_{12-x}O_{19}$ (ScM) ($1.8>x>0.3$) range from 0 to 14 kOe. Other important properties of ScM include its low sensitivity to temperature near room temperature, high Neel temperature, and low microwave loss tangent.[4] The low loss tangent is a result of fact that the Sc substitution stabilizes the valence state of Fe ions to 3+ rather than a combination of 2+ and 3+, where otherwise the coexistence of 2+ and 3+ valence states would induce valence hopping and, therefore, high loss tangents.[4]

Last year we reported on the structural and dc magnetic properties of Sc-doped barium hexaferrite films deposited by laser ablation techniques.[5] The thickness of the films that we reported on was less than 1 μm. This year, we reported on ScM ferrite films having thickness ranging from 1 to 5 μm. In comparison to bulk single crystals of similar compositions, the ferrimagnetic resonance (FMR) linewidth of these films are much broader. This increase in linewidth may be attributed to internal scattering of spinwaves from defects.[6,7]

II. FILM PREPARATION

The scandium-doped barium hexaferrite films were deposited onto *c*-plane sapphire (Al_2O_3) substrates by pulsed laser ablation deposition (PLD). The light source was a Lambda Physik ComPex 205 excimer laser (248 nm) operating at a frequency from 10 to 50 Hz. The laser beam was focused onto the ablation target to yield an energy density greater than 5 J/cm^2. Films were deposited in a Neocera PLD chamber, which was pumped down to a base pressure below 2×10^{-6} Torr before the introduction of high purity (99.996%) oxygen gas. The targets were 1 in. diameter pressed powder puck of $BaSc_xFe_{12-x}O_{19}$. The target-substrate distance was 4 cm. The sapphire substrates were heated to a fixed temperature of 900 °C[8,9] for film A, B, D, and to 925 °C for film C. The growth conditions for the four Sc-doped barium hexaferrite films were as follows: Film A was deposited at an oxygen pressure of 20 mTorr for 168 min, film B at 20 mTorr for 45 min, and film C at 50 mTorr for 60 min. Finally, film D was deposited at 50 mTorr for 70 min. The thickness of these films were measured by scanning electron micrographs or Dektak profilometer traces, and are listed in Table I.

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TABLE I. The properties of $BaFe_{12-x}Sc_xO_{19}$.

	Thickness (μm)	x	FWHM (008)	H_A (Oe)	$4\pi M_s$ (G)	g	ΔH (Oe)	H_c (Oe)
Film A	5.2	0.6	(Poly)	9200	3850	⋯	1720–2740	517
Film B	2.3	0.6	0.857	9210	2950	⋯	1580–1930	715
Film C	2.5	0.3	0.944	12 940	3350	1.96	890–1100	631
Film D	2.2	0.4	⋯	11 520	3690	1.96	310	281
Single crystal	Bulk	⋯	⋯	11 250	3450	2.0[a]	80	7.0

[a]Assumed value.

III. EXPERIMENTS

Crystallographic data were obtained on the films from reflection x-ray diffraction (XRD) measurements. Figure 1 shows a Θ–2Θ XRD pattern for the film C. Using the rocking curve results for the (008) diffraction peak; we can see that the films below 3 μm are highly c-axis oriented, the full width at half maximum (FWHM) results are listed in Table I. Meanwhile, the x-ray data showed polycrystalline structure for film above 5 μm. The fractions of scandium cations in the films were determined from energy dispersive x-ray spectroscopy measurements and are listed in Table I.

The magnetometry measurements were taken by using vibrating sample magnetometer in the presence of an external magnetic field (H_{ext}) as high as 14 kOe. The applied field is greater than H_A of the scandium-doped barium ferrite. The saturation magnetization ($4\pi M_S$) was deduced from the ''knee'' of the magnetization curves when we apply a magnetic field normal to film plane and is listed in Table I. The value of $4\pi M_S$ obtained by this method agrees reasonably well with the saturated value of the moment divided by its volume. Figure 2 shows hysteresis loops for film D where the magnetic field is applied both normal to (solid line) and parallel to (dashed line) the film plane. Values for the coercive field (H_c) were also obtained from the normal hysteresis loop measurements and are also listed in Table I. Table I also lists values obtained for a bulk single crystal of $BaSc_xFe_{12-x}O_{19}$ ($x=0.5$).

FMR measurements were performed by applying a swept dc magnetic field normal to the film plane, i.e., perpendicular FMR configuration. The film was placed into a waveguide such that the microwave magnetic field was maximum and in the film plane and H_{ext} was perpendicular to the film plane. The frequency was held fixed during each field sweep, but measurements were taken over the frequency range from 30 to 55 GHz. When H_{ext} is normal to the film, the FMR condition is given as follows:[10]

$$f=g\times 1.4\times 10^6\times(H_{ext}+H_A-4\pi M_S), \qquad (1)$$

where g is the Lande g factor.

Figure 3 shows the frequency versus measured resonant field value (H_r) for films C and D. The accuracy of the frequency was less than 0.1 GHz, while that of the dc field was 10 G. The magnetic field alignment was accurate to $\pm 1°$. Altogether, this translated into an accuracy of the g value in the order of ± 0.03. The g factor was deduced from Fig. 3 as the slope of the FMR frequency versus H_r. The results deduced for these films are listed in Table I. From these results, a g-factor value of $g=1.96\pm 0.03$ was obtained. It is interesting that the g values reported in the literature for barium hexaferrite vary between 1.87 and 2.05.[11] Our data fall in between these two values even for the thick films, although there are no data in the literature for the scandium-doped ferrite material. We are not reporting the g value for film A, since the film is polycrystalline. In addition, Fig. 3 was also used in deducing the value of H_A from FMR measurements, by utilizing the fact that $H_r=4\pi M_S$ when the film is biased to saturation. This means that Eq. (1) simplifies to

$$f=g\times 1.4\times 10^6\times H_A. \qquad (2)$$

The resulting values for H_A are listed in Table I.

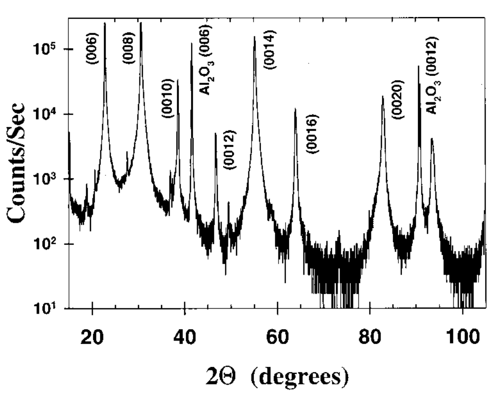

FIG. 1. X-ray results of film C. Θ–2Θ XRD pattern.

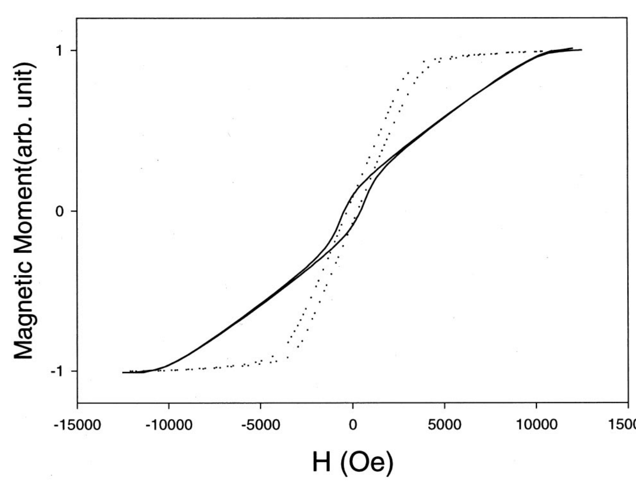

FIG. 2. Hysteresis loops are shown for film D. H parallel to film normal (dashed line), H parallel to film plane (solid line).

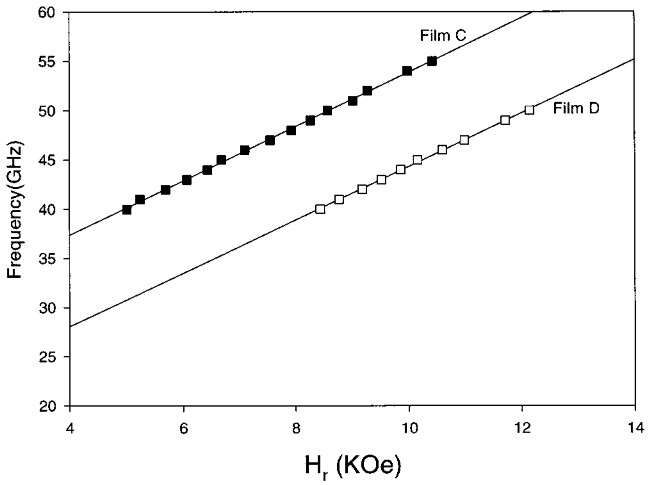

FIG. 3. FMR frequency vs H_r for films C and D.

From the FMR measurements, we obtain the FMR linewidth of the hexaferrite films. The FMR linewidth is important for the applications of ferrites in microwave and millimeter wave devices. The wider the linewidth, the bigger the magnetic loss. We investigated the relationship between the applied microwave frequency and the linewidth. We found that for the more lossy films, for example films A and B, the FMR linewidth was a maximum at 32 GHz, the FMR linewidth decreased with increasing frequency above the 32 GHz. Meanwhile, for film D, which exhibited the smallest microwave linewidth ($\Delta H \sim 310\,\mathrm{Oe}$), linewidth increases slightly with frequency from 30 to 50 GHz.

IV. CONCLUSION

We have investigated the microwave properties of thick scandium-doped barium hexaferrite films deposited by PLD. Based upon the magnetic and microwave magnetic parameters measured for film D and bulk single crystal results, scandium hexaferrite films appear to be good candidates for use in self-biased devices at microwave and millimeter frequencies. Ideally or practically one would like to realize an increase in H_c to $4\pi M_S$ while maintaining a narrow FMR linewidth. Is this possible? Increased values of H_c can only be obtained through increased pinning in the films, either through crystallite grain size, surface roughness, or the presence of defects. Unfortunately, some of these material properties may be detrimental to the microwave linewidth. Our data is inconclusive, for the films less than 3 μm, at frequency 34 GHz, ΔH varies from 310 to 1600 Oe with H_c from 281 to 715 Oe, whereas for the thicker film (>5 μm), $H_c \sim 517\,\mathrm{Oe}$, and $\Delta H \sim 2000\,\mathrm{Oe}$. The fact that ΔH is rather broad for thicker film implies that some form of scattering of the uniform mode energy occurs into particles of critical size.[7] For film D, which shows the lowest coercive field, and improved epitaxial growth, the FMR linewidth is also the smallest of the set of films. Clearly, the randomness of the *c* axis in thick films is the main cause of the increase in FMR linewidth. A temperature gradient during the growth process may induce this type of randomness as the film grows thicker.[11] We need to be able in the future to produce thick films with *c*-axis orientation perpendicular to the film plane. Preliminary results by us indicate that front surface heating[12] of substrate should improve the quality of thick films.

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