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## ADVERTISEMENT



# Pulsed laser deposition of epitaxial BaFe<sub>12</sub>O<sub>19</sub> thin films

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Epitaxial thin films of barium hexaferrite (BaFe<sub>12</sub>O<sub>19</sub>) have been fabricated by the pulsed laser deposition technique on basal plane sapphire. Structural studies reveal the films to be predominantly single phase and crystalline, with the *c* axis oriented perpendicular to the film plane. The magnetic parameters deduced from vibrating sample magnetometer and ferromagnetic resonance (FMR) measurements are close to the parameters associated with bulk materials. Post annealing of the films reduced the FMR linewidth by more than a factor of 3 so that it compares reasonably well with single-crystal films. The derivative FMR linewidth was measured to be 66 Oe at 58 GHz and 54 Oe at 86 GHz. Spin-wave-like modes have been observed for the first time in barium ferrite films. The deduced exchange stiffness constant of  $0.5 \times 10^{-6}$  ergs/cm is in reasonable agreement with recent calculations.

## INTRODUCTION

Flux melt,<sup>1</sup> liquid phase epitaxy (LPE),<sup>2</sup> and sputtering<sup>3,4</sup> techniques have been used in the past to prepare barium hexaferrite in bulk<sup>1</sup> and thin-film form.<sup>2-4</sup> The narrowest ferromagnetic resonance (FMR) linewidth was obtained with single-crystal spheres of barium hexaferrite prepared by the flux melt technique and was about 30 Oe at 80 GHz.<sup>5</sup> To date, thin-film techniques have not produced materials that match the quality of the single-crystal spheres. Although bulk spheres of barium hexaferrite have some use in microwave applications,<sup>6</sup> there are many more applications that require thin films, such as in microwave monolithic integrated circuitry (MMIC).

The LPE technique has produced moderately good films, but the technique has a number of practical limitations, including development of unique flux mixtures and substrates for each type of ferrite. Sputtering is somewhat more versatile because of the easy fabrication of targets and substrates; it also lends itself to the production of large area films. However, stoichiometry is not preserved from target to film.<sup>7</sup> Furthermore, the FMR linewidth in films of various hexaferrites prepared by sputtering<sup>8</sup> is invariably about 200 Oe at 80–90 GHz compared to a 62 Oe linewidth as measured for single-crystal films of barium hexaferrite prepared by the LPE technique.<sup>9</sup>

In this paper, we report fabrication of barium hexaferrite films by the pulsed laser deposition (PLD) technique.<sup>10</sup> PLD produces mostly single phase, single-crystal films of barium hexaferrite on basal plane sapphire (Al<sub>2</sub>O<sub>3</sub>) in which the *c* axis was aligned perpendicular to the film plane. FMR derivative linewidths were as narrow as 54 Oe at 86 GHz. As a result of the narrow FMR linewidth, spin-wavelike resonances were observed, which under some conditions obeyed an  $n^2$  law, where  $n$  is the spinwave order number. The deduced exchange stiffness constant,  $A$ , was in agreement with recent calculations

based upon molecular field approximations of the sublattice magnetic interactions.<sup>11</sup>

## FILM PREPARATION AND CHARACTERIZATION

Thin films of nominal composition BaFe<sub>12</sub>O<sub>19</sub> were fabricated by PLD on (0001) (basal plane) sapphire substrates. Although there is a 7% room-temperature lattice misfit of oxygen layers (with sapphire smaller) in the two crystals, it has been shown that BaFe<sub>12</sub>O<sub>19</sub> can grow epitaxially on basal plane Al<sub>2</sub>O<sub>3</sub>.<sup>4</sup> The PLD apparatus is shown schematically in Fig. 1. The output of the KrF excimer laser (248 nm, 300 mJ/pulse, 10 Hz) was focused off-center onto a rotating target with a 50 cm focal length lens to produce an energy density of about 2 J/cm<sup>2</sup>. The ceramic target was a commercially purchased 5 cm disk of stoichiometric barium hexaferrite. The target was mounted at 45° with respect to the laser beam. Laser-ejected material, whose distribution is centered normal to the target surface, is deposited onto the sapphire substrate that is 4 cm away. The substrate is heat-sunk with silver paste to a radiatively heated stainless-steel block. The block was heated to temperatures between 600 and 935 °C. It is estimated that the actual substrate surface temperature is 25 to 50 °C cooler. The depositions took place in an oxygen ambient at a chamber pressure between 10 and 400 mTorr. Typically, 20 000 laser shots yielded ferrite films that were about 0.5 μm thick.

Film crystal structure and orientation was determined by x-ray diffraction (XRD) using a 12 kW Cu target, rotating-anode generator operated at 50 kV and 200 mA. Scans were collected using a standard  $\theta/2\theta$  diffractometer. The goniometer has a 250 mm radius, 1/6° divergence and scatter slits, and a receiving slit 0.15 mm wide. Rocking curves ( $\theta$  scans) about the (0008) plane of the film were used to measure *c*-axis dispersion perpendicular to the film plane.

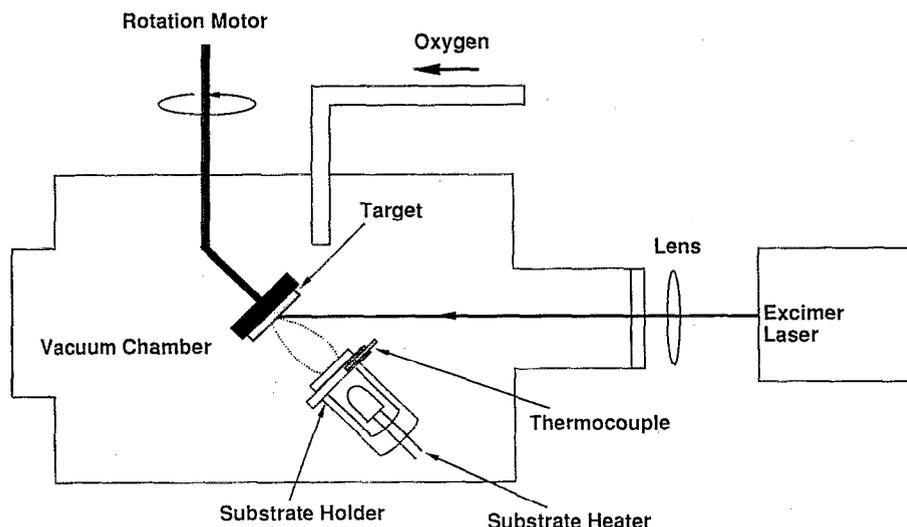


FIG. 1. Schematic of the pulsed laser deposition (PLD) apparatus. The laser parameters are: wavelength, 248 nm; energy, 300 mJ/pulse; and frequency 10 Hz.

Figure 2 displays the XRD data for an epitaxial film of  $\text{BaFe}_{12}\text{O}_{19}$  on a (0001) sapphire surface that was grown by PLD. The film was deposited at a temperature of 920 °C and an ambient pressure of 400 mTorr of  $\text{O}_2$ . The prominent diffraction peaks, except for the labeled substrate peak, are from the various orders of the (000L) planes of  $\text{BaFe}_{12}\text{O}_{19}$ . The full-width at half-maximum (FWHM) of the rocking curve about the (0008) plane of the  $\text{BaFe}_{12}\text{O}_{19}$  was 0.251°, which is comparable to the FWHM for the rocking curve of the sapphire substrate (0.227°). The most prominent peak seen in randomly oriented  $\text{BaFe}_{12}\text{O}_{19}$ , the (10 $\bar{1}$ 7) peak, is not present. The weak peak at  $2\theta = 27.480^\circ$  corresponds to the most prominent diffraction planes of  $\alpha\text{-BaFe}_2\text{O}_4$ , and indicates the presence of a small amount of second phase of that compound in the film. Films grown at comparable temperatures but progressively lower  $\text{O}_2$  pressures (300 to 10 mTorr) are oriented but show increasing FWHM of the rocking curve; e.g., at 10 mTorr the FWHM is 0.546°. A film grown at 800 °C and 30 mTorr shows the presence of nonoriented  $\text{BaFe}_{12}\text{O}_{19}$ ; the

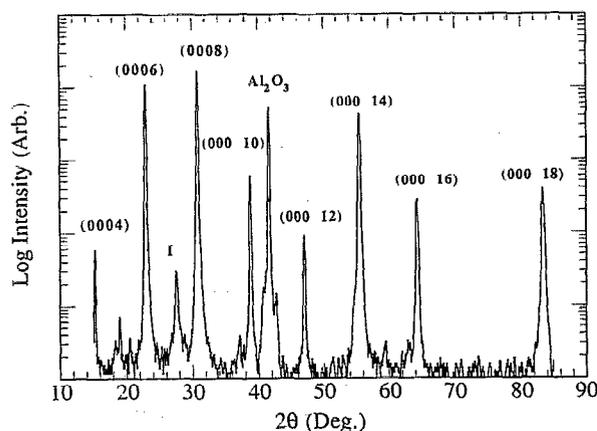


FIG. 2. X-ray diffraction data for barium hexaferrite film grown *in situ* on basal plane sapphire by PLD. The (000L) peaks are from the oriented hexaferrite; the peak labeled "I" may be from  $\alpha\text{-BaFe}_2\text{O}_4$ .

(10 $\bar{1}$ 7) peak is present, but not as prominent as it would be in completely randomly oriented material.

Films were analyzed for thickness, uniformity and composition by elastic backscattering spectrometry (EBS) with 6.2 MeV He ions and a scattering angle of 165°. The thickness of the film described in Fig. 2 was  $0.44 \pm 0.05 \mu\text{m}$ , as determined by the concentration of the barium and iron, where the film density is assumed to be  $5.28 \text{ gm/cm}^3$ . The films were uniform in composition through the thickness to  $\pm 5\%$ , but were substoichiometric in iron. The ratio of Fe/Ba for the films grown in 400 mTorr of  $\text{O}_2$  at 920 °C was  $11.1 \pm 0.2$ . The presence of a second phase of  $\text{BaFe}_2\text{O}_4$ , in the ratio of 1  $\text{BaFe}_2\text{O}_4$  molecule per 11.1  $\text{BaFe}_{12}\text{O}_{19}$  molecules, would give this composition.

Hexaferrite films grown by PLD were also postannealed in oxygen at temperatures between 900 and 1000 °C for two hours. A film, grown by PLD at 920 °C and 400 mTorr of  $\text{O}_2$ , and annealed as described showed no major structural changes. The weak x-ray diffraction peak from the  $\alpha\text{-BaFe}_2\text{O}_4$  did disappear, but the peaks from the epitaxial hexaferrite remained unchanged, with slightly narrower rocking curve FWHM of 0.227°. EBS detected no diffusion of the aluminum from the sapphire substrate into the film during growth or after annealing. The topography of the film was examined by scanning electron microscopy (SEM). The surface showed aligned hexagonal surface facets, about  $0.7 \mu\text{m}$  across, superimposed every few microns with hexagonal hillocks that protruded about  $0.1 \mu\text{m}$  out of the surface. The film appeared uniform in cross section on an intentionally fractured surface, with no evidence of grain structure or porosity.

## MAGNETIC CHARACTERIZATION

The magnetization,  $M$ , of the hexaferrite film, grown by PLD at 920 °C and 400 mTorr, was measured along the magnetic field,  $H$ , direction using a vibrating sample magnetometer (VSM). In Figs. 3(a) and 3(b),  $H$  was applied perpendicular to the film plane and parallel to the  $c$ -axis direction. Figure 3(a) was obtained for the as-grown sam-

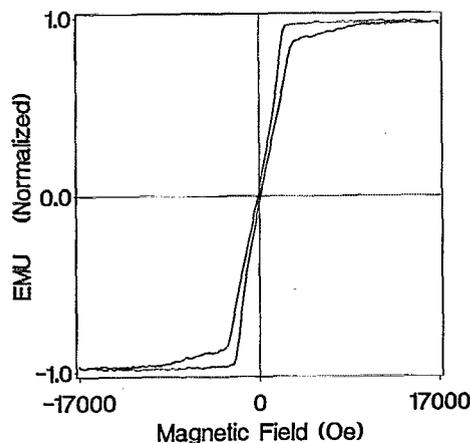
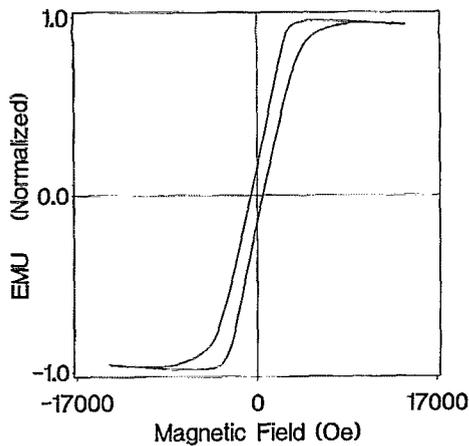


FIG. 3. (a) Hysteresis curve for barium hexaferrite grown *in situ* on basal plane sapphire by PLD.  $H$  is applied parallel to the  $c$  axis. (b) Hysteresis curve for barium hexaferrite film postannealed in oxygen at  $950^\circ\text{C}$ .  $H$  is applied parallel to the  $c$  axis.

ple, and Fig. 3(b) for the sample annealed at  $950^\circ\text{C}$ . The total moment of the film, measured at magnetic saturation ( $H = 10^4$  Oe) is  $1.95 \times 10^{-3}$  emu ( $4\pi M_s = 4400$  G) and is unchanged before and after annealing. However, there is a noticeable shift in the "knee" of the magnetization curve from  $4400$  G before annealing to  $3200$  G after annealing. This reduction may be attributed to the change in the surface topography with annealing, and the subsequent effect of that growth on the demagnetization in the perpendicular direction.<sup>12</sup> The coercive field,  $H_c$ , was reduced from  $700$  to  $100$  Oe upon annealing the film.

Finally, for the same film and  $H$  applied parallel to the film plane, magnetic saturation is reached for  $H = 16.5 \pm 0.4$  KOe (Fig. 4). This is close to the value for  $H_A$  obtained on single-crystal films of barium ferrite grown by the LPE technique.<sup>9</sup> Annealing the films did not change  $H_A$  significantly. In Fig. 4 we also note that  $M$  does not scale with  $H$  at low fields implying another magnetic phase besides barium ferrite, that has the easy axis of magnetization in the film plane.

The ferromagnetic resonance (FMR) measurements are summarized in Table I.  $H$  was applied along the  $c$  axis.

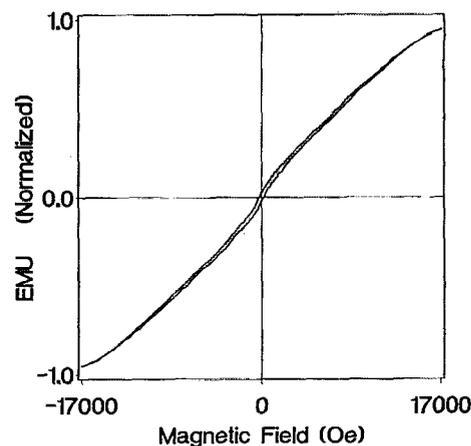


FIG. 4. Hysteresis curve for barium hexaferrite film grown *in situ* on basal plane sapphire by PLD.  $H$  is applied perpendicular to the  $c$  axis.

The FMR condition for this field direction is simply

$$\frac{\omega}{\gamma} = H - 4\pi M_s + H_A = H_i \quad (1)$$

where  $\omega = 2\pi f$ ,  $f$  is the operating frequency,  $\gamma = 2\pi(e/2mc)g$ , and  $H_i$  is the internal field. In determining the  $g$  factor, uncertainty in  $4\pi M_s$  and  $H_A$  is removed by taking the difference in resonance frequency and  $H$ , or simply by using the following equation:

$$g = \frac{(\delta f / \delta H)}{1.4 \times 10^6} \quad (2)$$

The data for the annealed film in Table I, taken between  $58$  and  $86$  GHz, and between  $50$  and  $86$  GHz, are used to find  $g = 1.99 \pm 0.01$ . If one were to deduce  $4\pi M_s$  from FMR measurements, using, e.g., (1), and assuming  $g = 1.99$  and  $H_A = 16.5 \pm 0.4$  kOe, one obtains  $4\pi M_s = 4400 \pm 400$  G. This value is in good agreement with literature values<sup>1,9</sup> and with the total moment found from the vibrating-sample magnetometer (VSM) data of Fig. 3.

The FMR spectrum for the annealed film is shown in Fig. 5.  $H$  is perpendicular to the film, in the  $c$ -axis direction, and the frequency is  $86$  GHz. There are three important features. Firstly, the main line (most intense) is tabulated in Table I. Secondly, for the FMR field above the main line there appears to be another FMR excitation.

TABLE I. Summary of ferromagnetic resonance experiments.

Frequency (GHz)	Magnetic Field (kOe)	Differential linewidth (Oe)
50	5.84	280
	5.82 <sup>a</sup>	88 <sup>a</sup>
58	8.72	220
	8.56 <sup>a</sup>	66 <sup>a</sup>
86	---	---
	18.66 <sup>a</sup>	54 <sup>a</sup>

<sup>a</sup>Film was annealed at  $950^\circ\text{C}$  for 2 h.

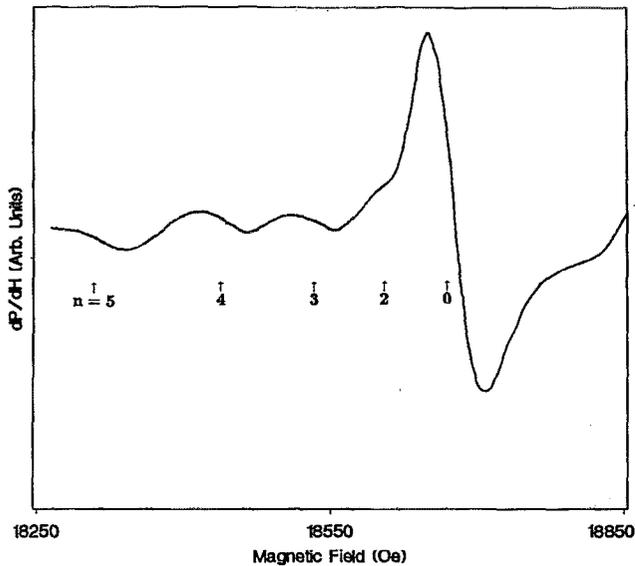


FIG. 5. Derivative FMR spectrum at 86 GHz for  $H$  applied parallel to the  $c$  axis. The spin-wave order number,  $n$  is assigned to each of the spin-wavelike excitations.

This line may be localized at the interface due to variations in the internal field,  $H_i$  at the interface.<sup>13</sup> Assuming the width of the interfacial region to be 50–60 nm,<sup>14</sup> and measuring the shift above the main line to be 200 Oe, the variation in  $H_i$  is of the order of 1000 Oe.<sup>13</sup> Further investigation is needed to clarify whether the variation in  $H_i$  is due to changes in  $H_A$  and/or  $4\pi M_s$ . Thirdly, below the main line there appear subsidiary excitations. These excitations are consistent with standing spin-wave mode excitations obeying the following dispersion relation:

$$\frac{\omega}{\gamma} = H_i + \frac{2A}{M} \left( \frac{\pi}{d} \right)^2 n^2, \quad (3)$$

where  $A$  is the exchange stiffness constant,  $d$  is film thickness, and  $n$  is the integer spin-wave order number. By assigning an  $n$  for each excitation and plotting  $H$  vs  $n^2$  (Fig. 6)  $A$  is found to be  $0.5 \pm 0.1 \times 10^{-6}$  ergs/cm. For this film we have assumed  $d = 0.44 \mu\text{m}$  and  $M_s = 350$  G. Our measured value of  $A$  is in reasonable agreement with calculated values of  $A = 0.36 \times 10^{-6}$  ergs/cm for  $k$  parallel to the  $c$  axis and  $0.5 \times 10^{-6}$  ergs/cm for  $k$  perpendicular to the  $c$  axis, where  $k$  is the spin-wave propagation vector.<sup>11</sup>

## CONCLUSIONS

The PLD technique has been demonstrated as a viable method for producing nearly perfect single crystals of barium hexaferrite, grown epitaxially on basal plane sapphire. The saturation magnetization, magnetic anisotropy field, and the  $g$  factor are in good agreement with literature values obtained on bulk samples. Although ours was the first attempt in producing films of barium hexaferrite by

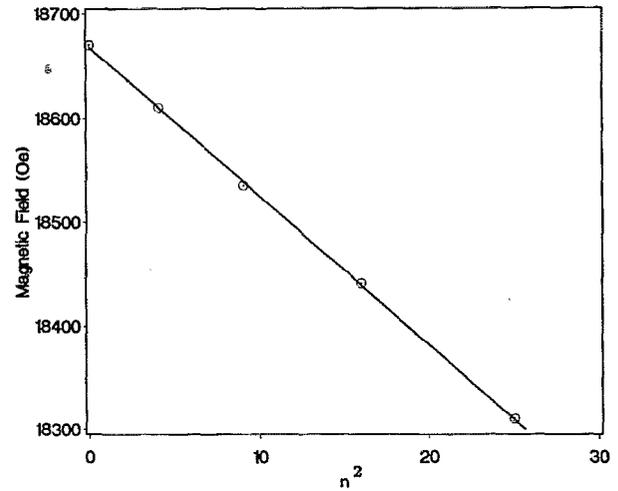


FIG. 6. Applied magnetic field versus square of the spin-wave order number. The slope,  $-14.5$  Oe, is used to calculate the exchange stiffness constant.

this technique, the measured FMR linewidth compares well with single-crystal films of barium hexaferrite prepared by the LPE technique. Annealing studies indicate that the ultimate lower limit in the FMR linewidth may not have been reached. It is believed that a linewidth of 30–40 Oe at 70 GHz can be achieved with the PLD technique.

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