Pulsed laser deposition of epitaxial yttrium iron garnet films with low Gilbert damping and bulk-like magnetization

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Pulsed laser deposition of epitaxial yttrium iron garnet films with low Gilbert damping and bulk-like magnetization

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Yttrium iron garnet (YIG, Y3Fe5O12) films have been epitaxially grown on Gadolinium Gallium Garnet (GGG, Gd3Ga5O12) substrates with (100) orientation using pulsed laser deposition. The films were single-phase, epitaxial with the GGG substrate, and the root-mean-square surface roughness varied between 0.14 nm and 0.2 nm. Films with thicknesses ranging from 17 to 200 nm exhibited low coercivity (< 2 Oe), near-bulk room temperature saturation moments (~135 emu cm−3), in-plane easy axis, and damping parameters as low as 2.2 × 10−4. These high quality YIG thin films are useful in the investigation of the origins of novel magnetic phenomena and magnetization dynamics.

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Magnetic garnets are important materials in a range of both bulk and thin film devices requiring magnetic insulators. Early experiments in magnonics were motivated by radar and microwave applications and were made using bulk yttrium iron garnet (YIG) crystals, largely due to YIG’s intrinsically low magnetic (spin-wave) damping.1–3 Historically, bulk YIG crystals were used for studying magnonics at the micrometer to millimeter length scales needed for microwave devices.4 However, fabrication of integrated spintronic, magnonic, or magnetooptical devices instead requires thin films with high structural and magnetic quality. In particular, achieving very low damping in thin film YIG is a critical enabler for fabrication of magnonic logic devices to transport, store, and process microwave and digital information for the post-CMOS (complementary metal-oxide-semiconductor) era.5 Applications include integrated multi-modal spin-wave devices, delay lines, filters, resonators, generators, multi-channel receivers, directional couplers, and Y-junctions. In addition, low damping (high quality-factor of 200–30 000) can be utilized for spin-based resonant sensing applications.6

Both sputtering and pulsed laser deposition (PLD) of complex oxides such as YIG has enabled excellent control of film thickness, stoichiometry, surface roughness, and magnetic properties. Previous reports on YIG films grown using liquid phase epitaxy, PLD or sputtering have found damping parameters ranging from 1.6 × 10−4 to 4.3 × 10−3, summarized in Table I. These damping parameters were extracted from ferromagnetic resonance measurements at comparable resonance frequencies.7–16 The intrinsic damping in bulk YIG has been reported to be $\alpha = 3 \times 10^{-5}$.17 In this study, we present PLD growth and characterization of epitaxial YIG thin films grown on Gadolinium Gallium Garnet (Gd3Ga5O12, GGG) substrates with saturation magnetization close to bulk and low damping parameters. The processes developed for ceramic YIG target preparation

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and film growth by PLD are presented. We characterize the crystalline structure, phase purity, and lattice parameters of YIG films with different thicknesses using high-resolution x-ray diffraction (HRXRD). The thickness dependence of magnetic properties (saturation moment and Gilbert damping) and structural properties (out-of-plane lattice parameter and surface roughness) has been investigated.

One-inch diameter stoichiometric YIG targets were prepared by mixing Y$_2$O$_3$ and iron oxide, followed by ball milling, calcination, and sintering at 1400 $^\circ$C. GGG substrates (Supplier: MTI Crystals, Inc.) were ultrasonicated first in acetone and then in isopropanol for surface cleaning. YIG films were pulsed laser deposited on 10 mm × 10 mm GGG (100) substrates held at 650 $^\circ$C under 20 mTorr oxygen pressure (5 × 10$^{-6}$ Torr base pressure) using a KrF coherent excimer laser ($\lambda = 248$ nm) with 400 mJ pulses at 10 Hz pulse rate, yielding about 3 nm/min growth rate. After deposition, the chamber was cooled in oxygen ambient at 5 $^\circ$C/min until the substrate reached 200 $^\circ$C, when the heater was switched off. After growth, the films were annealed at 800 $^\circ$C for 120 s ex situ using a rapid thermal anneal (RTA). YIG films with 17, 34, 49, 64, 79, 92, 97, and 190 nm thicknesses were grown on GGG substrates in separate deposition runs using the recipe described above. The target-substrate distance was kept at 85 mm for all samples. Among the deposition parameters, the correct target stoichiometry, oxygen pressure, temperature, and a low deposition rate were critical for achieving bulk-like magnetization and high crystal quality YIG films on GGG substrates. The structural, magnetic, and damping properties of YIG films were characterized after RTA and are given in Tables II and III.

Microstructure, lattice parameter, thickness, and surface roughness of the YIG films were analyzed using HRXRD (Bruker D8 Discover), X-ray reflectivity (HRXRR), and atomic force microscope (AFM Nanoscope IV, tapping mode operation at 2 Hz scan rate). Figure 1(a) shows the HRXRD 2$\theta$–$\theta$ plot near the YIG (800) and GGG (800) peaks for the 97 nm thick YIG film. A Ge-monochromator was installed to filter out the K$_{\alpha2}$ and to only observe the K$_{\alpha1}$ (1.5406 Å) peaks. The HRXRD peak with the highest intensity at 2$\theta$ = 59.70$^\circ$ corresponds to the (800) substrate peak of GGG giving a lattice parameter of 12.381 Å. The second peak on the left shoulder of the substrate peak corresponds to the (800) peak of the YIG film at 2$\theta$ = 59.56$^\circ$. Bulk YIG has a unit cell with lattice parameter $a = 12.376$ Å and space group Ia$3d$.

The oscillations to left and right of the substrate peak are Laue oscillations, or thickness fringes, of the YIG film, which indicate well-defined crystallographic layers and high ordering. The oscillations were present for each sample and could be fitted to derive the film thickness, lattice parameter, and strain even for films with weak intensity YIG peaks. For the fit to the Laue oscillations, we assumed an in-plane coherent interface but allowed a variation of the out-of-plane lattice constant as function of thickness, which can occur to relax residual stress at the interface. The mean out-of-plane lattice parameter resulting from this fit of the thickness fringes is shown in
Table II, column 3 and, as expected, is the same as that determined from the (800) peak maximum (Table II, column 2) within the error estimation. Based on this, the in plane and out of plane lattice parameters of the YIG differed by less than 0.2%. HRXRR data measured at grazing incidence were also fitted to extract film thickness from the period of the low angle oscillations, seen in Figure 1(b), which yielded values slightly larger than those from the Laue oscillations.

The rocking curves of the YIG (800) peak were also measured for the thicker films, which showed a narrow single symmetric peak (typically 0.014°–0.025°; see Table II, column 7), indicating a good crystallographic texture and no significant mosaicity within the films. FWHM of the substrate (800) rocking curve is around 0.008°. Reciprocal space map measurements of the 79 nm thick film indicate that the film is lattice matched to the substrate (see supplementary material for the 79 nm-thick YIG film).

There was no x-ray evidence of the growth of other crystalline phases, based on measurements of the thickest (190 nm) film. In this measurement, the angular range covers 2θ = 15°–61° to survey the presence or absence of Fe$_2$O$_3$ peaks near 33.5° and 41° and the strong Y$_2$O$_3$ peak near 2θ = 38°. This range covers the most intense peaks for YIG and for any potential secondary phases.

Figure 2 shows AFM surface profiles of (a) 17 and (b) 79 nm thick YIG films. The root-mean-square (RMS) surface roughness of the films was 0.14 nm and 0.20 nm, respectively. The surface roughness increased slightly for the thicker YIG films, but RMS roughness was smaller than 0.5 nm for all samples, which is less than the lattice parameter. The target-substrate distance of around 85 mm led to a slow growth rate of YIG (3 nm/min) increasing adatom diffusion distances at the high growth temperature.

### Table II. YIG lattice constants and thickness extracted from XRD. OP indicates out of plane.

<table>
<thead>
<tr>
<th>YIG thickness from XRR (nm)</th>
<th>OP lattice parameter from (800) peak position (Å)</th>
<th>OP lattice parameter from Laue oscillations (Å)</th>
<th>Estimated error (Å)</th>
<th>Thickness derived from Laue oscillations (nm)</th>
<th>Rocking curve FWHM (°)</th>
<th>Error in FWHM (°)</th>
</tr>
</thead>
<tbody>
<tr>
<td>17 ± 1</td>
<td>· · ·</td>
<td>12.395</td>
<td>0.01</td>
<td>19</td>
<td>3</td>
<td>· · ·</td>
</tr>
<tr>
<td>34 ± 1</td>
<td>· · ·</td>
<td>12.400</td>
<td>0.005</td>
<td>34</td>
<td>3</td>
<td>· · ·</td>
</tr>
<tr>
<td>49 ± 2</td>
<td>· · ·</td>
<td>12.400</td>
<td>0.005</td>
<td>47</td>
<td>3</td>
<td>· · ·</td>
</tr>
<tr>
<td>64 ± 2</td>
<td>12.391</td>
<td>12.395</td>
<td>0.005</td>
<td>63</td>
<td></td>
<td>· · ·</td>
</tr>
<tr>
<td>79 ± 2</td>
<td>12.406</td>
<td>12.402</td>
<td>0.005</td>
<td>78</td>
<td>3</td>
<td>0.02595</td>
</tr>
<tr>
<td>92 ± 2</td>
<td>12.408</td>
<td>12.405</td>
<td>0.005</td>
<td>89</td>
<td>4</td>
<td>0.02552</td>
</tr>
<tr>
<td>97 ± 2</td>
<td>12.407</td>
<td>12.403</td>
<td>0.005</td>
<td>96</td>
<td>5</td>
<td>0.02364</td>
</tr>
<tr>
<td>190 ± 2 (from profilometer)</td>
<td>12.399</td>
<td>12.398</td>
<td>0.005</td>
<td>186</td>
<td>6</td>
<td>0.01810</td>
</tr>
</tbody>
</table>

### Table III. Damping values derived from the VNA-FMR measurements of the YIG (100) samples.

<table>
<thead>
<tr>
<th>YIG thickness (nm)</th>
<th>Gilbert damping error (Oe)</th>
<th>FWHM at 10 GHz (Oe)</th>
<th>FWHM at 10 GHz error (Oe)</th>
<th>$\Delta H_0$ (Oe)</th>
<th>$\Delta H_0$ error (Oe)</th>
<th>$M_s$ (emu cm$^{-3}$)</th>
<th>$M_s$ error (emu cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>17</td>
<td>$7.0 \times 10^{-4}$</td>
<td>0.7 $\times 10^{-4}$</td>
<td>9.0</td>
<td>0.8</td>
<td>4.0</td>
<td>0.4</td>
<td>126</td>
</tr>
<tr>
<td>34</td>
<td>$5.8 \times 10^{-4}$</td>
<td>0.5 $\times 10^{-4}$</td>
<td>5.3</td>
<td>0.5</td>
<td>1.2</td>
<td>0.3</td>
<td>135</td>
</tr>
<tr>
<td>49</td>
<td>$4.1 \times 10^{-4}$</td>
<td>0.5 $\times 10^{-4}$</td>
<td>4.8</td>
<td>0.5</td>
<td>1.8</td>
<td>0.3</td>
<td>135</td>
</tr>
<tr>
<td>64</td>
<td>$3.8 \times 10^{-4}$</td>
<td>0.2 $\times 10^{-4}$</td>
<td>4.4</td>
<td>0.3</td>
<td>1.6</td>
<td>0.1</td>
<td>133</td>
</tr>
<tr>
<td>79</td>
<td>$2.2 \times 10^{-4}$</td>
<td>0.2 $\times 10^{-4}$</td>
<td>3.0</td>
<td>0.2</td>
<td>1.4</td>
<td>0.1</td>
<td>137</td>
</tr>
<tr>
<td>92</td>
<td>$3.4 \times 10^{-4}$</td>
<td>0.3 $\times 10^{-4}$</td>
<td>3.7</td>
<td>0.4</td>
<td>1.2</td>
<td>0.2</td>
<td>137</td>
</tr>
<tr>
<td>97</td>
<td>$2.6 \times 10^{-4}$</td>
<td>0.3 $\times 10^{-4}$</td>
<td>4.2</td>
<td>0.3</td>
<td>2.4</td>
<td>0.1</td>
<td>135</td>
</tr>
<tr>
<td>190</td>
<td>$4.1 \times 10^{-4}$</td>
<td>0.5 $\times 10^{-4}$</td>
<td>4.4</td>
<td>0.6</td>
<td>1.5</td>
<td>0.3</td>
<td>133</td>
</tr>
</tbody>
</table>
FIG. 1. (a) HRXRD $2\theta/\theta$ plot around YIG (800) and GGG (800) peaks. Dashed lines indicate (800) peaks of bulk YIG and GGG. (b) HRXRR plot for 34, 97, and 190 nm thick YIG films. HRXRD and HRXRR plots for other thicknesses were similar except for the period of the oscillations, which was shorter for thicker films.

Magnetic hysteresis loops of YIG films measured at room temperature (22 $^\circ$C) using a vibrating sample magnetometer (Digital Measurement Systems Torque/Vibrating Sample Magnetometer Model 1660 Signal Processor) are shown in Figure 3(a) for magnetic fields applied parallel to the sample plane (in-plane measurement with H-field along [100]). The paramagnetic contribution of

FIG. 2. AFM surface profiles of (a) 17 nm and (b) 79 nm thick YIG films. RMS surface roughness was 0.14 nm and 0.20 nm, respectively.
the GGG substrates was removed by subtracting a linear function from the raw magnetic hysteresis loop data. Saturation moments were also extracted from ferromagnetic resonance measurements described below and were used to calibrate the VSM data. SQUID (superconducting quantum interference device) magnetometry confirmed the saturation moments determined from FMR (ferromagnetic resonance). The coercivity of the samples was below 2 Oe, with the 17 nm sample at 5 Oe, but the accuracy of these values was limited by the step size of 2 Oe used to acquire data below 100 Oe.

The YIG had an in-plane easy axis for all thicknesses due to its shape, and saturation moments \( M_s = 137 \text{ emu cm}^{-3} \), with less than 5% measurement error) were close to bulk room temperature YIG values \( M_s = 140 \text{ emu cm}^{-3} \)\(^{21} \) as shown in Table III and in Figure 3(b). The measurement error is mostly attributed to the error in film thickness measurement. Apart from the thinnest sample, the saturation moments of the films were very consistent. The thinnest sample yielded a slightly lower value, but considering the error estimate, this difference may not be significant. The growth time for the thinnest films was <4 min and the target or substrate may not have reached a steady state temperature during the deposition. Several of the ~17 nm thick films were grown and they showed more variability in magnetization than the thicker films. The value reported here is the \( M_s \) averaged over multiple samples grown for each thickness. Hard-axis (out of plane) saturation fields near 2 kOe were consistent with shape anisotropy being the dominant anisotropy.

The rapid thermal annealing step after film growth had the following effects: (i) YIG coercivity was unchanged (to within the 2 Oe measurement error), (ii) the RMS surface roughness decreased...
by ~0.03 nm on average, and (iii) the saturation magnetic moment increased by up to 10 emu cm$^{-3}$.

The annealing step therefore improves the saturation magnetic moment but can be omitted if a lower thermal budget is required.

To determine the suitability of the epitaxial YIG films for spintronics applications, we measured Gilbert damping parameter $\alpha$ by FMR using a Vector Network Analyzer (VNA) attached to a grounded coplanar waveguide (GCPW). The sample is placed with the film facing towards the signal stripe on the GCPW. Signal stripe and external magnetic field are oriented parallel to the in-plane [100] direction of the sample. The measurements were performed by recording absorption of the $S_{21}$ signal emitted by the VNA in the frequency range of 7 GHz–13 GHz. For one measurement, the frequency was swept over a suitable range, while the applied magnetic field was kept constant. The inset of Figure 4(a) shows an example of one of these measurements for the 92 nm thick film. A Lorentz fit model is applied to determine the frequency linewidth $\Delta f$ from these signals. Assuming no pronounced magnetic anisotropy other than shape anisotropy, as indicated by the hard-axis saturation field, we converted the obtained frequency linewidth $\Delta f$ into the corresponding $\Delta H$ H-field linewidth by using the Kittel formula

$$\Delta H = (\partial f_{\text{Kittel}}/\partial H)^{-1} \Delta f = \Delta f \left(2\sqrt{H^2 + H M_S}\right) / ((\gamma \mu_0/2\pi)(2H + M_S)), \quad (1)$$

FIG. 4. (a) $\Delta H$ as a function of the resonance frequency. The inset shows one example of the measurement for a resonance frequency of 10.704 GHz. All these data were recorded for the 92 nm thick YIG film. Damping ($\alpha$) is $3.4 \times 10^{-4}$ and $\Delta H_0 = 1.2$ Oe for this sample. (b) Damping parameter of YIG films as a function of film thickness.
where $H$ is the magnetic field during the measurement and $\gamma/2\pi \approx 28$ (GHz/10 kOe) is the literature value of the gyromagnetic ratio.\textsuperscript{25} The value for $M_S$ was extracted from the resonance frequency as function of the applied field by fitting the Kittel equation\textsuperscript{23,24} neglecting magnetostrictive or magnetoelastic anisotropy terms, which are much smaller than shape anisotropy, and is given in Table III. Using the $\Delta H$ obtained from those measurements, we extracted the Gilbert damping parameter $\alpha$ as the slope of $\Delta H$ vs. the applied driving frequency. In the Landau-Lifshitz model, the relation between $\Delta H$ and excitation frequency is given by $\Delta H = \Delta H_0 + \frac{\Delta \alpha f}{\pi}$, where $\Delta H_0$ describes the inhomogeneous broadening for zero frequency.\textsuperscript{23} The derived values are given in Table III.

The damping ($\alpha$) of the YIG films grown for this study is shown in Figure 4(b). The damping parameters are low compared to other thin film YIG,\textsuperscript{7-16} though they exceed the value reported for bulk YIG.\textsuperscript{17} Magnetic moment and damping parameters are independent of film thickness (within the error bars), except for the thinnest sample. The damping is presumably independent of film thickness because of similar growth conditions and microstructures of the films. The bulk YIG damping, $\alpha = 3 \times 10^{-5}$,\textsuperscript{17} is an order of magnitude lower than damping values found in these thin films. The difference can arise from thin film effects (i.e., confinement along the through-thickness dimension) as well as possible contributions from nanoscale inhomogeneities in the stoichiometry.

In summary, pulsed laser deposition of epitaxial, low damping YIG ($Y_3Fe_5O_{12}$) films with magnetic saturation close to bulk has been presented. The YIG films were single-phase with low roughness. The slow growth rate and large target-substrate distance enabled the growth of smooth YIG films (i.e., measured RMS roughness < YIG lattice parameter), therefore reducing magnon scattering due to roughness. YIG films with thicknesses ranging from 17 nm to 200 nm had saturation magnetization 137 emu cm\textsuperscript{-3} (5% error) with in-plane easy axis and coercivity ~2 Oe. Damping parameters extracted from FMR measurements of YIG films were $2.2 \times 10^{-4}$ to $8 \times 10^{-4}$ in all samples, which is lower than other reports. In addition, the resonances obtained near 10 GHz for a range of film thicknesses have linewidths narrower than those commonly reported in the literature. The film properties presented here represent significant improvements in microstructural and magnetic properties in YIG with respect to earlier studies.\textsuperscript{26,27} The results of this study will help in the development of magnonic logic devices, sensors, and other devices to transport, store, and magnetic properties in YIG with respect to earlier studies.\textsuperscript{26,27} The results of this study will help in the development of magnonic logic devices, sensors, and other devices to transport, store, and...
20 See supplementary material at http://dx.doi.org/10.1063/1.4896936 for 79 nm-thick YIG film.