Ferromagnetic resonance studies of strain tuned Bi:YIG films

Ravinder Kumar, B Samantaray and Z Hossain

Condensed Matter—Low Dimensional Systems Laboratory, Department of Physics, Indian Institute of Technology (IIT) Kanpur—208016, India

E-mail: zakir@iitk.ac.in

Received 8 April 2019, revised 24 June 2019
Accepted for publication 2 July 2019
Published 26 July 2019

Abstract
Bismuth-doped Yttrium iron garnet (Bi:YIG) thin films known for large magneto-optical activity with low losses still need to get probed for its magnetization dynamics. We demonstrate a controlled tuning of magnetocrystalline anisotropy in Bi-doped Y₃Fe₅O₁₂ (Bi:YIG) films of high crystalline quality using growth induced epitaxial strain on [1 1 1]-oriented Gd₃Ga₅O₁₂ (GGG) substrate. We optimize a growth protocol to get thick highly-strained epitaxial films showing large magneto-crystalline anisotropy, compare to thin films prepared using a different protocol. Ferromagnetic resonance measurements establish a linear dependence of the out-of-plane uniaxial anisotropy on the strain induced rhombohedral distortion of Bi:YIG lattice. Interestingly, the enhancement in the magnetoelastic constant due to an optimum substitution of Bi³⁺ ions with strong spin orbit coupling does not strongly affect the precessional damping (∼2 × 10⁻³). Large magneto-optical activity, reasonably low damping, large magnetocrystalline anisotropy and large magnetoelastic coupling in Bi:YIG are the properties that may help Bi:YIG emerge as a possible material for photo-magnonics and other spintronics applications.

Keywords: epitaxial strain, magnetocrystalline anisotropy, spin dynamics, spintronics, ferromagnetic resonance, pulsed laser deposition

(Some figures may appear in colour only in the online journal)
of spin–orbit torque in heavy metals (HM) [25–29] and topological insulators (TI) [30–32] capped ferrimagnetic garnet heterostructures show potential to improve the efficiency of magnetic manipulations as it will not shunt a charge current applied to the capping conducting layer [33]. Being an insulating material, only electron’s spin degrees of freedom is allowed, resulting in pure spin current, which is not the case with conducting-oxides (Region III), metals and metal-alloys (Region IV). Besides having the ability to generate pure spin current, the magneto-optical properties of YIG enhances in proportion to Bismuth (Bi) concentration at Yttrium site [34–37]. Due to enhanced magneto-optical activity in the UV, visible and IR regions along with low propagation loss, Bi-YIG is a potential material for light based magnonics applications, motivated the studies reported here. In this study, we grow high quality Bi:YIG films on GGG(1 1 1) crystals using two different growth protocols which allow us to achieve different strain-states induced by rhombohedral distortion due to film-substrate lattice mismatch. We prepared two sets of samples, set-A and set-B. Set-A consists of thin Bi:YIG films with large magnetocrystalline anisotropy due to the large magnitude of strain, and, set-B consists of thick Bi:YIG films with reasonably large strain. Despite being thick, the films from set-B show large magnitude of strain that leads to large value of magnetocrystalline anisotropy, for an example; the magnitude of uniaxial magnetocrystalline anisotropy field. The importance of Bismuth substituted YIG as a potential material for light based magnonics applications, motivated the studies reported here. In this study, we grow high quality Bi:YIG films on GGG(1 1 1) crystals using two different growth protocols which allow us to achieve different strain-states induced by rhombohedral distortion due to film-substrate lattice mismatch. We prepared two sets of samples, set-A and set-B. Set-A consists of thin Bi:YIG films with large magnetocrystalline anisotropy due to the large magnitude of strain, and, set-B consists of thick Bi:YIG films with reasonably large strain. Despite being thick, the films from set-B show large magnitude of strain that leads to large value of magnetocrystalline anisotropy, for an example; the magnitude of uniaxial magnetocrystalline anisotropy field. The importance of Bismuth substituted YIG as a potential material for light based magnonics applications, motivated the studies reported here. In this study, we grow high quality Bi:YIG films on GGG(1 1 1) crystals using two different growth protocols which allow us to achieve different strain-states induced by rhombohedral distortion due to film-substrate lattice mismatch. We prepared two sets of samples, set-A and set-B. Set-A consists of thin Bi:YIG films with large magnetocrystalline anisotropy due to the large magnitude of strain, and, set-B consists of thick Bi:YIG films with reasonably large strain. Despite being thick, the films from set-B show large magnitude of strain that leads to large value of magnetocrystalline anisotropy, for an example; the magnitude of uniaxial magnetocrystalline anisotropy field. The importance of Bismuth substituted YIG as a potential material for light based magnonics applications, motivated the studies reported here. In this study, we grow high quality Bi:YIG films on GGG(1 1 1) crystals using two different growth protocols which allow us to achieve different strain-states induced by rhombohedral distortion due to film-substrate lattice mismatch. We prepared two sets of samples, set-A and set-B. Set-A consists of thin Bi:YIG films with large magnetocrystalline anisotropy due to the large magnitude of strain, and, set-B consists of thick Bi:YIG films with reasonably large strain. Despite being thick, the films from set-B show large magnitude of strain that leads to large value of magnetocrystalline anisotropy, for an example; the magnitude of uniaxial magnetocrystalline anisotropy field.

Figure 1. Relationship between effective magnetization and Gilbert damping coefficient. Here, we compare some of the interesting work from existing literature; region I and II: ferromagnetic insulators in the form of bulk, thin films (polycrystalline and epitaxial); region III: conducting-oxides and; region IV: pure metals and metal-alloys. Different regions of interest have been shaded with different colors. Note: references are provided at the end of this paper in figure 8.
120 min to get atomically flat surfaces and then cooled down to 500 °C in 4.0 × 10^{-2} mbar oxygen pressure to deposit the films. The target was sufficiently preablated before actual deposition to get a steady state target surface. We incorporated two routes to deposit these epitaxial films to obtain different strain states by changing the laser fluence at a fixed oxygen ambient and growth temperature. For set-A, the fluence was ∼1 J cm^{-2} with a spot size of ∼10.0 mm^2 and hence the realized growth rate was ∼0.25 Å s^{-1}. For set-B, we almost doubled the fluence (∼1.9 J cm^{-2}) by reducing the spot size (∼5.4 mm^2) to achieve an enhanced growth rate of ∼0.45 Å s^{-1}. We deposited five films of thicknesses 10.2, 18.1, 37.0, 92.5 and 200 nm using set-A growth parameters, hereafter denoted as A1, A2, A3, A4 and A5, respectively. Another five films of thicknesses 18.7, 39.8, 100, 150 and 200 nm were grown using growth protocol-B, hereafter denoted as B1, B2, B3, B4, and B5 respectively. The growth rate and hence the thicknesses of different samples were pre-calibrated using Dektak stylus profilometer. PANalytical X’Pert PRO four circle diffractometer equipped with Cu-Kα1 source (λ = 1.54059 Å) was used to characterize the crystallinity and to quantify the state-of-strain. Room temperature vibrating sample magnetometry (VSM) measurement was performed using a quantum design physical property measurement system (PPMS). For the dynamic magnetization measurements, we used both commercial and a custom-made FMR setup. Angular dependent FMR measurements were performed using bruker EMX EPR spectrometer with cavity mode frequency f ≈ 9.60 GHz. Frequency dependent FMR measurements were performed by using a broadband coplanar waveguide (CPW). The CPW assembly was housed in an external homogeneous DC magnetic field along with the superposition of a small and low frequency AC field. This small modulation of magnetic field is required to get differential of absorbed radio frequency (RF) power which is measured by a Schottky diode detector and a lock-in amplifier.

Figure 2 summarizes the x-ray measurements on Bi:YIG films grown on (1 1 1) oriented GGG substrates. Panel (a) shows reflectivity measurements on all the samples except 100, 150 and 200 nm (pre-calibrated using profilometry), as there were no visible thickness fringes due to larger thickness. Reflectivity data was fitted to calculate and standardize the profilometric pre-calibrated thickness and gives very low roughness ranging from 0.25 to 0.39 nm. The panel (b) of figure 2 shows intensity normalized ω scan profiles with low values of full width half maximum (FWHM) ranging between 0.0448 to 0.0072°, signifies high crystallinity. The panel (c) of figure 2 shows x-ray diffraction patterns of all the Bi:YIG samples where the pronounced trail of Laue oscillations characterizes smooth surfaces and sharp interfaces. The bulk lattice constant for Bi_{0.25}Y_{2.75}Fe_{5}O_{12} comes out to be 12.389 Å and the corresponding 2θ peak position is shown by a vertical bar beneath substrate peak. Thin film lattice constant (a_{∥}) differs due to lattice mismatch between substrate and film (shown by vertical up arrows). This lattice mismatch causes rhombohedral distortion in the films and hence contributes to diagonally stretched unit cells along the [1 1 1] growth direction. The strain induced rhombohedral distortion in these epitaxial Bi:YIG films can be quantified using the parameter σ = (a_{∥} - a_{b})/a_{b} = Δa/a_{b} , where, a_{b} is the bulk Bi:YIG lattice parameter and a_{∥} is the stretched film lattice parameter along the [1 1 1] direction [2, 3, 64]. For set-A samples, XRD patterns show strain relaxation as the thickness increases from 10.2 to 200 nm (2θ value approaches the bulk value), the strain-induced lattice distortion decreases from 1.162% to almost ∼0.0%. Surprisingly, set-B samples...
having thicknesses 18.7, 39.8, 100, 150 and 200 nm, show relatively high strain (1.122% for 18.7 nm thin film and 0.171% for 200 nm thick film). The variation of \(a_{\perp}\) and the lattice strain (\(\sigma\)) w.r.t. to Bi:YIG film thickness from both the sets are shown in figure 2 panel (c). It can be seen that the value of \(a_{\perp}\) approaches bulk value for a film of thickness 200 nm from set-A, whereas, a 200 nm thick film from set-B possess elongated \(a_{\perp}\). Similarly, a 200 nm thick film from set-A show negligible lattice strain but a 200 nm thick film from set-B possesses reasonably large lattice strain. The 2-axis \(\omega\) versus \(2\theta - \omega\) maps are shown in figure 3. The top panel shows symmetric maps in the 444 direction of Bi:YIG films. Whereas, the bottom panel shows the 642 asymmetric direction maps. We show \((444)\) symmetric and \((642)\) asymmetric 2-axis maps for 10.2, 37.0 and 92.5 nm films from set-A, and, 100 and 200 nm films from set-B. It can be clearly seen that the \(2\theta - \omega\) value for film (represented by +; red colored) shifts toward higher value as the film thickness increases and approaches to the GGG substrate spot (represented by \(\times\); black colored). The map of a 92.5 nm thick film from set-A shows large relaxation compare to a 100 nm thick film from set-B. Which confirms the inference drawn from \(\theta - 2\theta\) XRD measurement. The laser ablation conditions greatly impact the lattice constant of deposited films irrespective of oxygen pressure and growth temperature. We observe that the laser fluence plays an important role in tuning the lattice constant of the films. The set-A films prepared using slow growth rate (\(\sim 0.25 \text{ A s}^{-1}\)) with a lower laser fluence (\(\sim 1 \text{ J cm}^{-2}\)) show less lattice expansion and complete relaxation with thickness increment. Whereas, the set-B films prepared using almost doubled growth rate (\(\sim 0.45 \text{ A s}^{-1}\)) due to higher laser fluence (\(\sim 1.9 \text{ J cm}^{-2}\)) show tendency to possess reasonably large lattice expansion even for higher thicknesses (panel (c) and (d) of figure 2). The laser fluence (growth rate) is low in the case of set-A Bi:YIG films, which gives sufficient settle down time to the ablated plasma species and hence lead to strain relaxation. In contrast, the higher laser fluence (growth rate) in the case of Bi:YIG films from set-B, does not allow the ablated plasma species to settle down and get relaxed. Table 1 contains XRD, magnetization and FMR derived parameters for both the sets of samples. The negative sign of \(\sigma\) indicates the presence of compressive strain which relaxes with increment in film thickness [2, 64–66].

Room temperature in-plane magnetic hysteresis loops are measured using VSM on Quantum Design PPMS. In-plane and out-of-plane magnetization loops for a 37.0 nm thick film from set-A is shown in figure 4(b), where the paramagnetic background from GGG was subtracted. The values of saturation magnetization \((4\pi M_s)\) for samples from set-A and set-B ranges between 1720 ± 100 to 1407 ± 25 Oe and 1608 ± 17 to 1457 ± 12 Oe, respectively. The coercivity \((H_C)\) of these samples are in the range of \(\sim 13\) to 23 Oe. These values fall in the range of reported YIG magnetization data [20] [64–68]. To probe the static and dynamic magnetic properties of Bi:YIG epitaxial films, we performed angular and frequency dependent FMR measurements on both the sets of samples. Generally, the magnetic garnet thin films with a hard axis in the [111] direction (i.e. In-plane easy axis), possesses extrinsic uniaxial magnetic and intrinsic magnetocrystalline

![Figure 3. The \(\omega\) versus \(2\theta - \omega\), 2-axis maps in (444) symmetric and (642) asymmetric directions: left panel shows maps of 10.2 nm, 30 nm and 92.5 nm Bi:YIG films from set-A. Right Panel shows maps of 100 nm and 200 nm Bi:YIG films from set-B.](image-url)
cubic anisotropies. FMR can directly deduce the magnetic anisotropies in a precise manner. The coordinate system used for FMR study on (111) oriented epitaxial Bi:YIG films is shown in figure 4(a). The orientations of static magnetic field $H$ and magnetization vector $M$ with reference to coordinates $x:[2T T], y:[01T]$ and $z:[111]$ are described by the angles $\phi_H$, $\theta_H$ and $\phi_M$, $\theta_M$, respectively. The total free energy per unit volume of the media for (111) oriented cubic garnet system has the form [69, 70],

$$F = -HM_s\left[\sin \theta_H \sin \theta_M \cos (\phi_H - \phi_M) + \cos \theta_H \cos \theta_M\right] + 2\pi M_s^2 \cos^2 \theta_M$$

$$-K_1 \cos^2 \theta_M + \frac{K_1}{12} \left(7\sin^2 \theta_M - 8\sin^2 \theta_M + 4 - 4\sqrt{3}\sin^2 \theta_M \cos \theta_M\right)$$

$$+ \frac{K_1}{108} \left(-24\sin^2 \theta_M + 45\sin^4 \theta_M - 24\sin^2 \theta_M + 4 - 2\sqrt{3}\sin^2 \theta_M \cos \theta_M + (5\sin^2 \theta_M - 2) \cos 3\phi_M + \sin^2 \theta_M \cos 6\phi_M\right) .$$

(1)

The first term in equation (1) corresponds to the Zeeman energy, the second term to the demagnetization energy, the third term to the out-of-plane uniaxial magneto-crystalline anisotropy energy $K_a$ and the last two terms are due to first and second order cubic magneto-crystalline anisotropy energies, $K_1$ and $K_2$, respectively. The total free energy was minimized ($\partial F / \partial \phi_M = \partial F / \partial \theta_M = 0$) to obtain the equilibrium orientation of the magnetization vector $M(H)$. The evaluation of resonance frequency ($\omega_{\text{res}}$) of uniform magnetization precessional mode at equilibrium condition can be made using total free energy and is expressed as:[70–72]

$$\omega_{\text{res}} = \frac{\gamma}{M_s \sin \theta_M} \left[\frac{\partial^2 F}{\partial \phi_M^2} \frac{\partial^2 F}{\partial \theta_M^2} \left(\frac{\partial^2 F}{\partial \phi_M \partial \theta_M}\right)\right]^{1/2}$$

(2)

here $\gamma$ and $M_s$ denote gyromagnetic ratio and saturation magnetization, respectively. These coupled and indirectly defined functional equations were solved numerically to obtain the equilibrium angles at resonance condition and fit the angular dependent resonance data ($H_{\text{res}}$ versus $\theta_H$) to determine $g$-factor, $K_a$, $H_u$, $H_1$, $H_2$ and $E_{\text{ani}}$ (see table 1). Figure 4(c) shows representative angular-FMR spectra of a 39.8 nm thick film from set-B at a microwave frequency of ~9.6 GHz. The peak-to-peak difference of FMR derivative gives linewidth ($\Delta H$) which decreases as the film thickness increases. The measured in-plane $\Delta H$ values for set-A samples $A_1$, $A_2$, $A_3$, $A_4$, and $A_5$ at ~9.6 GHz are 154, 120, 93, 39, and 14 Oe, respectively. Similarly, for set-B samples $B_1$, $B_2$, $B_3$, $B_4$, and $B_5$ the in-plane $\Delta H$ values are 150, 105, 50, 44, and 23 Oe, respectively. The energy minimization governed by the correspondence between $\theta_H$ and $\theta_M$ is shown in the inset of figures 4(d) and (e), where the equilibrium magnetization angle $\theta_H$ was estimated numerically. It can be seen that energy minimization attains large curvature for thin Bi:YIG film from both the sets and hence large anisotropy compare to thick film from the respective sets. Figures 4(d) and (e) show $\theta_H$ dependence of $H_{\text{res}}$ for set-A and set-B samples, respectively.

The fit using equations (1) and (2) agrees well with the measured data. All the extracted parameters for both the sets of samples are shown in table 1, separated by a solid line. We mainly focus on the out-of-plane uniaxial anisotropy field ($H_u$) due to its large contribution to total magnetic anisotropy and systematic variation with film thickness or lattice strain. In contrast, we could not witness a systematic thickness or strain dependence of cubic first and second order anisotropy which are weak in magnitude. Interestingly, $H_u$ for 10.2 nm thin and 200 nm thick films from set-A comes out to be ~1831 ± 227 Oe and ~70 ± 25 Oe, respectively, which provides a strain tuning over a range of more than 1700 Oe. It suggests that the rhombohedral distortion induces substantial out-of-plane uniaxial anisotropy via the magnetostriction, which decreases systematically with increase in the film thickness. The g-factor for thin films is as large as 2.13, greater than the spin-only value 2.0. This corroborates the existence of spin-orbit coupling that lead to strain-induced anisotropy. However, the g-factor for thick films are smaller (~2.0). This variation in ‘g’ possibly arises due to different strain state, which may change the occupation of orbitals and hence the magnitude of orbital angular momentum and spin–orbit coupling. The strain induced variation of $H_u$ and $E_{\text{ani}}$ is picturized in figures 5(a) and (b), respectively. It is clear from figures 5(a) and (b) that the magnitudes of $H_u$ and $E_{\text{ani}}$ increases almost linearly as the magnitude of rhombohedral distortion increases. The enhancement in uniaxial anisotropy field is due to the larger magnitude of growth induced strain in the samples from set-B as compare to set-A. The substrate-film lattice mismatch causes lattice-distortion in deposited films which results in a definite strain-state. The lattice distortion influences the magnetic properties. This magnetization-lattice coupling gives rise to strain-induced out-of-plane uniaxial anisotropy field, $H_u$. The strain induced by rhombohedral distortion in a cubic lattice relaxes as the film thickness increases and hence results in very low or almost negligible strain, which ultimately makes the film isotropic, having properties similar to bulk. The value of $H_u$ for Bi:YIG films $B_1$, $B_2$, $B_3$, $B_4$, and $B_5$ from set-B are found to be ~1292 ± 137, ~1010 ± 103, ~648 ± 82, ~246 ± 50, and ~108 ± 17 Oe, respectively. It is important to note that the values of $H_u$ for thicker films from set-B are larger compare to respective film thicknesses from set-A. If we compare the uniaxial anisotropy field of Bi:YIG films from both the sets of almost equal thicknesses, i.e. $A_2$ (18.1 nm) and $B_1$ (18.7 nm), comes out to be ~977 ± 117 Oe, and ~1292 ± 137 Oe, respectively. The uniaxial anisotropy field magnitude for set-B Bi:YIG film is almost 300 Oe larger compare to the value of set-A Bi:YIG film.

The magnetoelastic energy density for a strain dependent FMR measurement is given by $F_{\text{ME}} = -\sigma_b [\cos \Theta]^3$, where $b$ is magnetoelastic constant and $\Theta$ is the angle between $M$ and strain direction [2, 3]. For $M$ pointing in the [111] direction, the magnetoelastic energy density has the form, $F_{\text{ME}} = -\sigma b$. Figure 5(b) shows the linear dependence and least-square fit of anisotropy energy $E_{\text{ani}} = -1/2\{M_H H_u\}$ with different strain
Table 1. XRD, $M - H$ and FMR derived parameters of Bi:YIG epitaxial films grown by two protocols. Set-A ($A_1$, $A_2$, $A_3$, $A_4$, and $A_5$) and set-B ($B_1$, $B_2$, $B_3$, $B_4$, and $B_5$) are separated by a horizontal line.

| Thickness (nm) | $2\theta$ (Degree) | $a_\perp$ (Å) | $\Delta a/a_\parallel$ (%) | $4\pi M_s$ (VSM) (Oe) | g-factor | $K_u$ ($\times 10^3$ erg cc$^{-1}$) | $H_u$ (Oe) | $H_1$ (Oe) | $H_2$ (Oe) | $E_{ani}$ ($\times 10^3$ erg cc$^{-1}$) |
|----------------|---------------------|----------------|---------------------------|------------------------|---------|-----------------|---------|---------|---------|-----------------|
| 10.2 ($A_1$)   | 50.406              | 12.533         | -1.162                    | 1720 ± 100             | 2.12    | -125.40 ± 8.23  | -1831 ± 227 | -15.3 ± 1.9 | 3.1 ± 1.1 | -126.24 ± 8.24   |
| 18.1 ($A_2$)   | 50.766              | 12.450         | -0.492                    | 1432 ± 63              | 2.09    | -55.68 ± 4.21   | -977 ± 117  | -29.7 ± 2.2  | 13.8 ± 1.6 | -56.59 ± 4.21   |
| 37.0 ($A_3$)   | 50.919              | 12.415         | -0.210                    | 1482 ± 37              | 2.03    | -27.68 ± 3.02   | -469 ± 63   | -40.9 ± 1.8  | 30.9 ± 1.7 | -28.27 ± 3.01   |
| 92.5 ($A_4$)   | 51.011              | 12.394         | -0.048                    | 1507 ± 38              | 2.01    | -7.43 ± 2.71    | -124 ± 48   | -12.1 ± 0.9  | 52.1 ± 2.0 | -5.03 ± 2.70    |
| 200 ($A_5$)    | 51.033              | 12.389         | 0.0                       | 1407 ± 25              | 2.00    | -3.91 ± 1.32    | -70 ± 25    | -3.2 ± 0.6   | 3.2 ± 0.7 | -3.91 ± 1.31    |
| 18.7 ($B_1$)   | 50.425              | 12.528         | -1.122                    | 1582 ± 38              | 2.13    | -81.42 ± 6.72   | -1292 ± 137 | -57.3 ± 1.9 | 6.7 ± 1.0 | -84.61 ± 6.81   |
| 39.8 ($B_2$)   | 50.530              | 12.504         | -0.928                    | 1545 ± 25              | 2.05    | -62.12 ± 5.33   | -1010 ± 103 | -14.4 ± 0.8 | 160.2 ± 3.4 | -53.16 ± 5.42   |
| 100 ($B_3$)    | 50.747              | 12.454         | -0.525                    | 1520 ± 25              | 2.04    | -39.23 ± 4.28   | -648 ± 82   | -40.0 ± 1.2  | 65.1 ± 1.9 | -37.71 ± 4.37   |
| 150 ($B_4$)    | 50.807              | 12.440         | -0.414                    | 1608 ± 17              | 2.03    | -15.78 ± 3.04   | -246 ± 50   | -19.6 ± 0.6  | 118.5 ± 1.4 | -9.45 ± 3.11    |
| 200 ($B_5$)    | 50.939              | 12.410         | -0.171                    | 1457 ± 12              | 2.01    | -6.25 ± 0.91    | -108 ± 17   | -23.9 ± 0.7  | 113.5 ± 1.2 | -1.06 ± 0.96    |
R Kumar et al.

states of Bi:YIG films from both the sets. The derived expressions from least-square fit in figure 5(b) for set-A and set-B are

\[ E_{\text{ani}} = (12.58 \pm 3.59) \times 10^3 + (7.71 \pm 1.18) \times 10^6 \left( \frac{a_b - a_{\perp}}{a_b} \right) \] (erg cc\(^{-1}\)) and \[ E_{\text{ani}} = (10.74 \pm 0.51) \times 10^6 \] (erg cc\(^{-1}\)), where the slope of the lines give \( -b = (10.74 \pm 0.51) \times 10^6 \) (erg cc\(^{-1}\)) and \( -b = (7.71 \pm 1.18) \times 10^6 \) (erg cc\(^{-1}\)), respectively. The negative sign of \( b \) implies that the magnetic easy axis is parallel to the compressed lattice plane; [1 1 1]. The magnetoelastic constant of Bi:YIG comes out to be larger than in pure-YIG film [2].

Pure YIG exhibits almost quenched orbital momentum of half-filled \( d \) shell in \( \text{Fe}^{3+} \) electron configuration, leads to weak SOC and shows low magnetoelastic coupling constant. The substitution of strong SOC ions such as \( \text{Bi}^{3+}, \text{Dy}^{3+} \) and \( \text{Tm}^{3+} \) etc enhances the spin–orbit coupling which results in improved magnetoelastic coupling. It suggests that the

**Figure 4.** Room temperature magnetization and out-of-plane angular dependence of resonance field for Bi:YIG films from both the sets. (a) Typical schematic of spherical coordinate system for FMR measurements and analysis of [1 1 1] oriented epitaxial Bi:YIG/GGG(111) samples. (b) Magnetic hysteresis loops measured in in-Plane (red) and out-of-plane (Green) configuration of a 37.0 nm thin film from set-A using VSM. (c) Representative FMR derivative spectra for a 39.8 nm Bi:YIG film from set-B. Panel (d) picturizes out-of-plane angular variation (\( \theta_{H} \)) of the resonance fields (\( H_{\text{res}} \)) and fitted curves for set-A. Inset: energy minimization comparison for 10.2 nm and 200 nm thick Bi:YIG films from set-A. Panel (e) picturizes out-of-plane angular variation (\( \theta_{H} \)) of the resonance fields (\( H_{\text{res}} \)) and fitted curves for set-B. Inset: energy minimization comparison of 18.7 nm and 200 nm thick Bi:YIG films from set-B.

**Figure 5.** (a) Out-of-plane uniaxial anisotropy field \( H_u \) and (b) total anisotropy energy \( E_{\text{ani}} \) as a function of the rhombohedral distortion \((a_b - a_{\perp})/a_b\)% of the Bi:YIG films on GGG(111). Blue solid lines are the least-square fit to obtain magnetoelastic coupling constant. Dashed curves serve as a guide to the eye.
strain-tuning could be very crucial to obtain large magneto-
crystalline anisotropy even in thick ferrimagnetic-insulating
films.

Gilbert damping coefficient $\alpha$ for our Bi:YIG films has
been calculated from frequency-dependent FMR measure-
ment between 7 and 12 GHz. The external magnetic field is
swept at various fixed frequencies. Figures 6(a) and (b) show
the frequency versus $H_{\text{res}}$ data and its fit (corresponding
colored solid curves) for set-A and set-B, respectively. Black solid lines represent fit to the experimental data.

![Figure 6](image)

**Figure 6.** In-plane, frequency and thickness dependent room
temperature FMR measurements. Panel (a) and (b) represent
frequency versus resonance field plots for set-A and set-B,
respectively. The fit to experimental data has been shown by
the corresponding colored solid curves. Panel (c) and (d) represent
frequency dependent linewidth variation for set-A and set-B,
respectively. Black solid lines represent fit to the experimental data.

| Thickness (nm) | $4\pi M_{\text{eff}}$ (Oe) | $\alpha \times 10^{-3}$ | $\Delta H_0$ (Oe) |
|----------------|------------------|-----------------|-----------------|
| 10.2 ($A_1$)   | 3482 ± 65        | 18.3 ± 1.3      | 84              |
| 18.1 ($A_2$)   | 2441 ± 27        | 12.7 ± 0.9      | 72              |
| 37.0 ($A_3$)   | 1970 ± 8         | 6.9 ± 0.7       | 67              |
| 92.5 ($A_4$)   | 1673 ± 46        | 2.4 ± 0.3       | 30              |
| 200 ($A_5$)    | 1510 ± 2         | 2.0 ± 0.1       | 5               |
| 18.7 ($B_1$)   | 2928 ± 15        | 16.1 ± 1.5      | 92              |
| 39.8 ($B_2$)   | 2437 ± 2         | 9.6 ± 0.6       | 68              |
| 100 ($B_3$)    | 2125 ± 3         | 3.4 ± 0.1       | 37              |
| 150 ($B_4$)    | 1787 ± 4         | 3.2 ± 0.1       | 31              |
| 200 ($B_5$)    | 1399 ± 2         | 2.9 ± 0.2       | 10              |

**Table 2.** Frequency and thickness dependent FMR derived effective
magnetization, Gilbert damping coefficient and inhomogeneous
broadening of Bi:YIG epitaxial films grown by two different
protocols. Set-A and set-B are separated by a solid horizontal line.
reduced form of equations (1) and (2) in a limiting in-plane magnetic field geometry ($\theta_H = 90^\circ, \phi_H = 0^\circ$). The derived compact expression in asymptotic limit has the form (in-plane Kittel equation),
\[
\omega_{\text{res}} = \gamma \sqrt{H_{\text{res}} (H_{\text{res}} + 4\pi M_{\text{eff}})}
\]
with the effective magnetization $4\pi M_{\text{eff}} = 4\pi M_S - H_{\text{ani}}$, where, $H_{\text{ani}}$ is the anisotropy field parameterizes out-of-plane uniaxial and cubic anisotropies. It is clear from figures 6(a) and (b) that the data fits perfectly without even considering additional in-plane anisotropy contributions. In equation (3) we do not consider a renormalization shift in the resonance frequency and a small shift in resonance field which can arise by two-magnon scattering and a static dipole interaction between the ferrimagnetic film and the paramagnetic substrate, respectively, due to negligibly small contributions. Figures 6(c) and (d) show in-plane frequency dependencies of linewidth ($\Delta H$) for set-A and set-B films, respectively. The standard Landau–Lifshitz–Gilbert equation justifies the linear dependence of $\Delta H$ with frequency and used for straightforward determination of the intrinsic Gilbert damping coefficient ($\alpha$): $\Delta H = \Delta H_0 + (4\pi \alpha/\sqrt{3} \gamma) f_{\text{res}}$, where $\Delta H_0$ is the intrinsic linewidth broadening due to magnetic inhomogeneities within the material. The extracted values of $4\pi M_{\text{eff}}$, $\alpha$ and $\Delta H_0$ for films from both the sets are shown in table 2.

Figure 7(a) shows strain dependent variations of $4\pi M_{\text{eff}}$ and $4\pi M_S$. The values of $4\pi M_{\text{eff}}$ for both the sets systematically decreases with the increase in film thickness but the values strongly depend on the state-of-the-strain in the films. It can be seen that $4\pi M_{\text{eff}}$ is significantly larger than the Bi:YIG saturation magnetization generated simple shape anisotropy i.e. $4\pi M_S$, revealing the presence of a negative uniaxial anisotropy, signature of easy in-plane magnetization. The gap between $4\pi M_{\text{eff}}$ and $4\pi M_S$ represents magnitude of anisotropy field $H_{\text{ani}} = 4\pi M_S - 4\pi M_{\text{eff}}$ which decreases with increment in film thickness. The magnitude of $H_{\text{ani}}$ for ~100 nm thick Bi:YIG film from set-B is larger than that expected and comparable to ~37 nm thin film from set-A, which is due to growth induced large strain. Figure 7(b) shows magnetization ($4\pi M_{\text{eff}}, 4\pi M_S$) dependence on uniaxial anisotropy field, where, the magnetization decreases in proportion with the magnitude of uniaxial anisotropy field. Figure 7(c) shows the variation of $\alpha$ with respect to the film thickness from both the sets. Whereas, inset shows induced strain dependency of $\alpha$. We notice that the value of $\alpha$ decreases nonlinearly as film thickness increases (or strain relaxes) and vice-versa. We include effective magnetization, uniaxial anisotropy field and damping data of YIG/GGG(1 1 1) films from literature by Bhoi et al [64] which also follow the same trend. The lowest damping possessed by a 200 nm thick film from set-A is (2.0 $\pm$ 0.1) $\times$ 10$^{-3}$ with an inhomogeneous broadening of ~5 Oe, whereas, a 200 nm thick film from set-B shows slightly larger damping (2.9 $\pm$ 0.2) $\times$ 10$^{-3}$ with an inhomogeneous broadening of ~10 Oe but inherit reasonably large uniaxial anisotropy field (~108 $\pm$ 17 Oe) which is almost two times larger compare to former. Although, the damping in Bi doped YIG enhances due to strong spin orbit coupling, still it’s passably small compare to metallic systems [59–61]. As the values of $\alpha$ and $|H_0|$ increases as a function of the induced strain, we therefore plot $\alpha$ versus $H_0$ graph (see figure 7(d)) to see the correlation between the precessional damping and magnetic anisotropy. In our Bi:YIG thin film system, we observe a nonlinear relationship between $\alpha$ and $H_0$, similar to YIG and can be attributed to spin wave damping induced by increment in strain [64]. Rhombohedral distortion arising due to lattice mismatch between the film and the substrate leads to change in magnetic properties through spin orbit coupling [3]. The inclusion of lattice distorted SOC along with phonon-magnon scattering, two-magnon scattering or charge transfer relaxation may explain the thickness dependent enhancement of uniaxial anisotropy and reduction of magnetic damping [2, 3, 33, 60, 61, 64, 73].

In summary, we have been able to grow high quality epitaxial Bi:YIG thin films on GGG(1 1 1) crystals as evidenced by prominent Laue oscillations in x-ray diffraction pattern. A usual trend of the film lattice relaxation and decrease in magnetic anisotropies as the film thickness increases has been observed. Our study shows that strain can be a crucial parameter to tune the magnetocrystalline anisotropy. We optimize a growth protocol to get thick epitaxial films with large lattice strain which allows us to achieve large magneto-crystalline anisotropy. The Bi:YIG films grown using higher laser fluence show large magneto-crystalline anisotropy compare to films of respective thicknesses grown using lower laser fluence. We show that the incorporation of growth induced large strain in thick Bi:YIG films can be helpful to improve the magnetic properties. Out-of-plane uniaxial anisotropy varies linearly with strain induced rhombohedral distortion of Bi:YIG lattice. Still, we are able to achieve fairly low Gilbert damping $\sim 2 \times 10^{-3}$ with enhanced magnetoelastic coupling. Further, as Bismuth substitution enhances the magneto-optical responses enormously, the coupling of large magneto-crystalline anisotropy, improved magnetoelastic coupling and low damping with strong magneto-optical activity in Bismuth substituted YIG may provide unique opportunities for photon-based-magnonics to develop efficient and low loss spintronics and caloritronics devices.
Figure 8. Region–I: Hauser et al [23], Chang et al [21], Chang et al [1], Bhoi et al [64], Lucas et al [74], Le et al [75], Onbasli et al [20], Liu et al [73], Yang et al [27], Gallagher et al [76], Sun et al [77], Jungfleisch et al [78], Howe et al [22], Patti et al [79], Wu et al [80], Jermain et al [33], Budhani et al [81], Nosach et al [82], Yoshimoto et al [83], Dubes et al [84], Pirro et al [85], Heinrich et al [86], Haertinger et al [87], Fang et al [88], Harii et al [89], Chang et al [1].

Region–II: Iguchi et al [90], Kehlberger et al [91], Vasili et al [92], Siu et al [93].

Region–III: Lee et al [94], Emori et al [95], Qin et al [96], Luo et al [97].

Region–IV: Ando et al [98], Tu et al [99], Gong et al [100], Lu et al [101], Gong et al [102], Fujimoto et al [103], Nosach et al [104], Kehlberger et al [105], Belmeguenai et al [106], Belmeguenai et al [107], He et al [108], Kurebayashi et al [109], Zhao et al [110], Yoshimoto et al [111], Kobayashi et al [112]. Reference details of the relationship between effective magnetization and Gilbert damping coefficient shown in figure 1. It was constructed using the effective magnetization (saturation magnetization in few cases) and Gilbert damping coefficient values from various (Region I and II) ferro- and ferrimagnetic insulators, (Region III) conducting oxides and (Region IV) pure metals and metal-alloys, as reported in previous studies.

Acknowledgments

We thank Prof R C Budhani for fruitful discussion and Dr Veena Singh for technical assistance during FMR measurements. We gratefully acknowledge financial support from Indian Institute of Technology Kanpur (IIT Kanpur).

ORCID iDs

Ravinder Kumar https://orcid.org/0000-0001-8548-8487

References

[1] Chang H, Praveen Jananatha P A, Ding J, Liu T, Cline K, Gelfand J N, Li W, Marconi M C and Wu M 2017 Sci. Adv. 3 e1601614
[2] Wang H, Du C, Hammel P C and Yang F 2014 Phys. Rev. B 89 134404
[3] Du C, Adur R, Wang H, Hauser A J, Yang F and Hammel P C 2015 Phys. Rev. Lett. 110 147204
[4] Wang C T, Liang X F, Zhang Y, Liang X, Zhu Y P, Qin J, Gao Y, Peng B, Sun N X and Bi L 2017 Phys. Rev. B 96 224403
[5] Parkin S S P 2004 IEDM Technical Digest. IEEE Int. Electron Devices Meeting, 2004 pp 903–906
[6] Fukami S, Sato H, Yamanouchi M, Ikeda S, Matsukura F and Ohno H 2014 19th Asia and South Pacific Design Automation Conf. p 684
[7] Joshi V K 2016 Eng. Sci. Technol. Int. J. 19 1503
[8] Klingler S, Pirro P, Bröicher T, Leuen B, Hillebrands B and Chumak A V 2014 Appl. Phys. Lett. 105 152410
[9] Klingler S, Pirro P, Bröicher T, Leuen B, Hillebrands B and Chumak A V 2015 Appl. Phys. Lett. 106 212406
[10] Grundler D 2015 Nat. Phys. 11 438
[11] Ganzhorn K, Klingler S, Wimmer T, Geprägs S, Gross R, Huebl H and Goennenwein S T B 2016 Appl. Phys. Lett. 109 022405
[12] Chumak A V, Vasyuchka V I, Serga A A and Hillebrands B 2015 Nat. Phys. 11 453
[13] Makarov A, Windbacher T, Sverdlov V and Selberherr S 2016 Semicond. Sci. Technol. 31 113006
[14] Egel E, Csaba G, Dietz A, Breitkreutz-von Gamm S, Russer J, Russer P, Kreupl F and Becherer M 2018 AIP Adv. 8 056001
[15] Uchida K, Takahashi S, Harii K, Ieda J, Koshibae W, Ando K, Maekawa S and Saitoh E 2008 Nature 455 778
[16] Uchida K et al 2010 Nat. Mater. 9 606
[17] Bauer G E W, Saitoh E and Ohno H 2008 2010 Nat. Mater. 9 606
[18] Kirihara A, Uchida K I, Kajiwara Y, Ishida M, Nakamura Y, Manako T, Saitoh E and Yorozu S 2012 Nat. Mater. 11 391
[19] Heremans J P and Boona S R 2014 Physics 71 71
[20] Onbasli M C, Kehlberger A, Kim D H, Jakob G, Kläui M, Chumak A V, Hillebrands B and Ross C A 2014 Appl. Mater. 2 106102
[21] Chang H, Li P, Zhang W, Liu T, Hofmann A, Deng L and Wu M 2014 IEEE Magn. Lett. 5 1
[22] Howe B M, Emori S, Jeon H, Oxtom T M, Jones J G, Mahalingam K, Zhuang Y, Sun N X and Brown G J 2015 IEEE Magn. Lett. 6 1
[23] Liu J, Yang et al [64], Lucas et al [74], Le et al [75], Onbasli et al [20], Liu et al [73], Yang et al [27], Gallagher et al [76], Sun et al [77], Jungfleisch et al [78], Howe et al [22], Patti et al [79], Wu et al [80], Jermain et al [33], Budhani et al [81], Nosach et al [82], Yoshimoto et al [83], Dubes et al [84], Pirro et al [85], Heinrich et al [86], Haertinger et al [87], Fang et al [88], Harii et al [89], Chang et al [1].
[91] Kehlberger A et al 2015 Phys. Rev. Appl. 4 014008
[92] Vasili H B et al 2017 Phys. Rev. B 96 014433
[93] Siu G G, Lee C M and Liu Y 2001 Phys. Rev. B 64 094421
[94] Lee H K, Barsukov I, Swartz A G, Kim B, Yang L, Hwang H Y and Krivorotov I N 2016 AIP Adv. 6 055212
[95] Emori S, Alaun U S, Gray M T, Sluka V, Chen Y, Kent A D and Suzuki Y 2016 Phys. Rev. B 94 224423
[96] Qin Q, He S, Song W, Yang P, Wu Q, Feng Y P and Chen J 2017 Appl. Phys. Lett. 110 112401
[97] Qing Qin S H, Wu H, Yang P, Liu L, Song W, Pennycook S J and Chen J 2018 (arXiv:1804.00554v1)
[98] Luo G Y, Belmeguenai M, Roussigné Y, Chang C R, Lin J G and Chérif S M 2015 AIP Adv. 5 097148
[99] Ando K et al 2011 J. Appl. Phys. 109 103913
[100] Lee A J, Brangham J T, Cheng Y, White S P, Ruane W T, Esse B D, McComb D W, Hammel P C and Yang F 2017 Nat. Commun. 8 234
[101] Tu H Q et al 2017 Sci. Rep. 7 43971
[102] Lu L, Wang Z, Mead G, Kaiser C, Leng Q and Wu M 2014 Appl. Phys. Lett. 105 012405
[103] Fermin J R, Avevedo A, Aguiar F M D, Li B and Resende S M 1999 J. Appl. Phys. 85 7316
[104] Gong Y, Cevher Z, Ebrahimi M, Lou J, Pettiford C, Sun N X and Ren Y H 2009 J. Appl. Phys. 106 063916
[105] Ikeda S, Miura K, Yamamoto H, Mizunuma K, Gan H D, Endo M, Kanai S, Hayakawa J, Matsukura F and Ohno H 2010 Nat. Mater. 9 721
[106] Lindner J, Barsukov I, Raeder C, Hassel C, Posth O, Meckenstock R, Landeros P and Mills D L 2009 Phys. Rev. B 80 224421
[107] Belmeguenai M, Tuzcuoglu H, Gabor M S, Petrisor T, Tiusan C, Berling D, Zighem F, Chauveau T, Chérif S M and Moch P 2013 Phys. Rev. B 87 184431
[108] He P, Ma X, Zhang J W, Zhao H B, Lüpike G, Shi Z and Zhou S M 2013 Phys. Rev. Lett. 110 077203
[109] Kurebayashi H et al 2013 Appl. Phys. Lett. 102 062415
[110] Zhao Y, Song Q, Yang S H, Su T, Yuan W, Parkin S S P, Shi J and Han W 2016 Sci. Rep. 6 22890
[111] Samantaray B, Singh A K, Perumal A, Ranganathan R and Mandal P 2015 AIP Adv. 5 067157
[112] Koçbay A N, Yılgın R, Topkaya R, Ahsen A Ş, Öztürk O and Aktaş B 2012 J. Supercond. Nov. Magn. 25 2813