FMR and Magnetic Studies on Polycrystalline YIG Thin Films Deposited Using Pulsed Laser

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Ferromagnetic resonance (FMR) and magnetic studies have been carried out on polycrystalline YIG films deposited by pulsed laser deposition (at T_S : 750 °C, O_2 : 5×10^{-2} mbar) using a range of laser energies (240 to 350 mJ). The films were *ex situ* annealed (T_a : 700 °C, in air for 2 h). The film thickness increases from 100 to 290 nm for an hour of deposition with the increase in laser energy from 240 to 350 mJ. The FMR linewidth was found to reduce from 340 to 70 Oe for the same incremental variation in laser energy. Multiple resonance modes were observed in the perpendicular FMR spectra of our samples which are related to intrinsic as well as extrinsic mechanisms such as in homogeneities. Saturation magnetization measurement (M_s) is found to be dependent on the laser energy. The value of M_s is found to increase with laser energy and is close to 90% of the bulk value for the film with highest power. An attempt has been made to correlate the magnetization results with the microstructure.

***Index Terms*—Effective saturation magnetizations, Ferromagnetic resonance, linewidth, microwave.**

I. Introduction

THE study of yttrium iron garnet ($Y_3Fe_5O_{12}$ or YIG) films have been of interest due to its properties essential for microwave devices such as resonators, isolators, circulators, capacitors, wavelength accordable filters, etc., [1], [2]. Liquid phase epitaxy (LPE) method is commonly employed to deposit single crystal YIG thin films with several tens to hundreds micrometer thicknesses with low linewidth (~1 Oe) and saturation magnetization close to bulk value (1750 G) of YIG [2]. Work done by Dorsey *et al.* [3] and Buhay *et al.* [4] demonstrated that the pulsed laser deposition (PLD) technique can also be used to fabricate epitaxial YIG films on Gadolinium Gallium Garnet (GGG) for planar microwave devices. The connection between FMR linewidth and microstructure in YIG received considerable attention, starting with the early work on YIG sphere by Lecraw *et al.* [5]. One critical problem with the single crystal films prepared by PLD is that, the ferromagnetic resonance (FMR) linewidth (ΔH) of the samples is more than 10 times larger than expected for single crystals films prepared by LPE [6], [7].

The above discussed single crystal YIG film deposition always requires expensive GGG substrates due to its lattice matching with YIG. Making a cost effective low ΔH polycrystalline YIG films is always a difficult task due to the lattice mismatch with the substrate. There have been some studies on the polycrystalline YIG films [8]–[11] which are based on the effect of oxygen partial pressure and deposition temperature on magnetic properties. In case of polycrystalline YIG films, the change in magnetic and microwave properties could be due to several reasons, i.e., grain size, thickness of films, surface roughness, etc. Since the laser power plays a crucial role in modifying the microstructure or thickness, its consequences on microwave or magnetic properties need yet to be studied. In case one is interested in preparing a cost effective low line width YIG films, amorphous quartz substrate becomes a convenient choice because of its low cost and relatively high crystallization temperature (above 850 °C). In this paper we investigate the microstructure dependence of FMR in PLD polycrystalline YIG films on amorphous fused quartz substrates, with an aim to optimize the magnetic and microwave properties of the YIG films.

II. Experimental Details

YIG thin films were prepared by the PLD technique using a KrF excimer laser (Lamda physik, λ : 248 nm) of 10 ns pulse with in a vacuum chamber with a base pressure of 5×10^{-6} mbar. The YIG target was synthesized in our laboratory using a standard solid state reaction method with a sintering temperature of 1450 °C. The laser was focused on YIG target which was rotated (6 r/min) during the deposition process. The material ablated from the target was deposited onto 1 × 1 cm amorphous quartz substrates bonded to a resistive heater positioned 4 cm away from the target. Films were grown using laser energy varying from 240 to 350 mJ in oxygen atmosphere (5×10^{-2} mbar) at substrate temperatures of 750 °C. After deposition, the oxygen partial pressure in the vacuum chamber was raised to near atmosphere and the films were then cooled to room temperature. The as-grown layers were *ex situ* annealed in air atmosphere at 700 °C for 2 h.

The thickness of the films was measured by profile-meter. The structures of the as-prepared and annealed YIG films were studied using X'pert Pro (PW 3040) X-ray diffraction (XRD) using Cu $K\alpha_1$ ($\lambda = 0.15406$ nm) line. The surface morphology of the films were investigated with atomic force microscopy (AFM, SEIKO SPA-300HV) and field emission scanning electron microscope (FESEM, JSM-7600F) respectively. The dc magnetic measurements were carried out using a physical property measurement system (PPMS, Quantum Design). The

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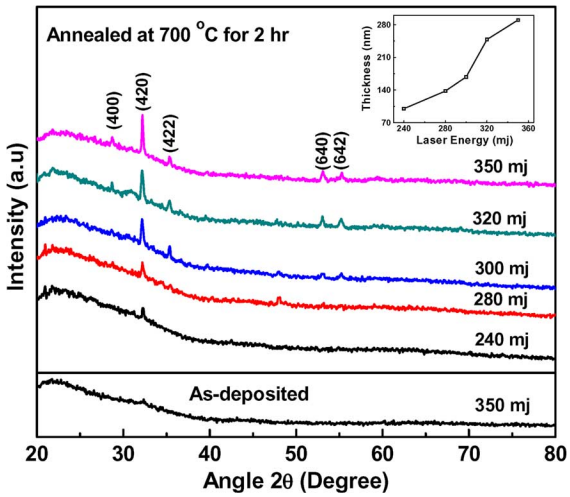

Fig. 1. XRD patterns of as-deposited (only 350 mj) and annealed (at 700 °C for 2 h in air) YIG thin films deposited at different laser energy. Inset: Variation thickness as a function of laser energy.

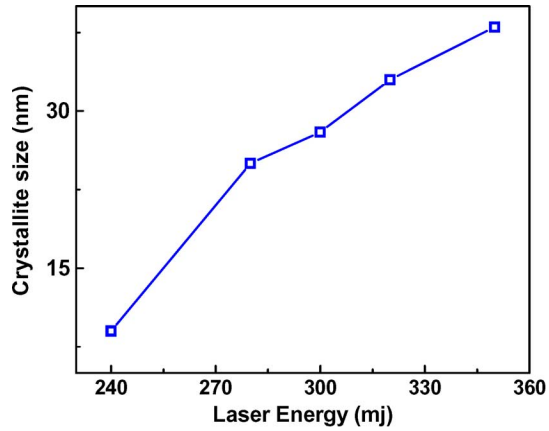

Fig. 2. Variation of crystallite size of annealed YIG thin films as a function of laser energy.

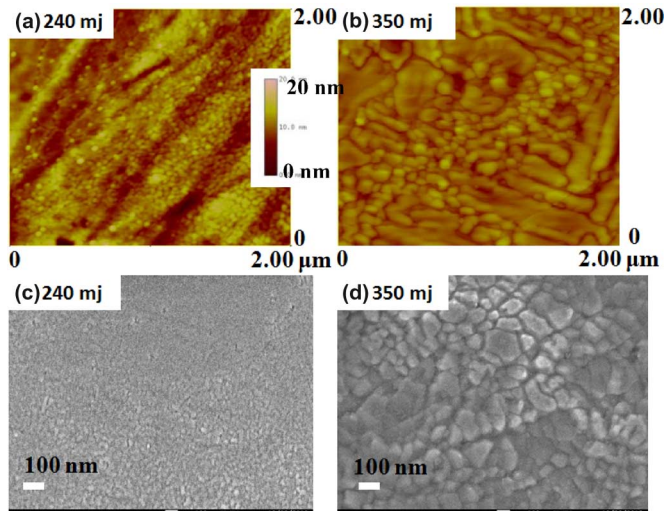

Fig. 3. (a) and (b) AFM and (c) and (d) FEGSEM images of annealed YIG films grown at 240 and 350 mj, respectively.

substrate contribution to the magnetization was subtracted while plotting the M-H curve. The spontaneous magnetization $(4\pi M_S)$ of the films was evaluated by extrapolating the high field part of the M–H loop to zero magnetic fields [12]. FMR was performed using a conventional Varian E-line spectrometer operating at 9.1 GHz with dc magnetic field (H) applied in parallel and perpendicular (normal) directions to the film plane. The FMR was recorded in the conventional form in which dP/dH, the first field (H) derivative of the absorbed RF power (P) was plotted as a function of the applied magnetic field up to 5.5 kOe. The resonance field was determined at the point of intersection of the dP/dH curve with the zero line. The perpendicular and parallel FMR ΔH was taken as the difference of fields between the maximum and minimum of dP/dH curve, whenever the curve was symmetric. In case of asymmetric FMR shapes and for an overlap of other small secondary modes, the ΔH was estimated by doubling the half width from the portion where line shapes were good.

III. Result and Discussion

The films were deposited at five different laser energies (240, 280, 300, 320, and 350 mJ). The thickness was found to increase from 100 to 290 nm for an hour of deposition with the increase in laser energy from 240 to 350 mJ (inset: Fig. 1). The XRD patterns of all the as-deposited films show no clear and distinct peaks even though the films were deposited at substrate temperatures of 750 °C. A representative pattern for the film deposited at 350 mj is also shown in Fig. 1 along with the XRD patterns of the annealed films grown at different laser energy. We thus see that annealing leads to sharper and stronger XRD peaks. This demonstrates the need for post-deposition annealing to induce proper crystallization of YIG films [10], [11]. Such difference in post deposition annealing and substrate heating has also been seen in other ferrite systems [12]. This can be understood as the deposition is a nonequilibrium process and its dynamics is likely to be different from *ex situ* annealing. During deposition the ablated particles continuously come from the target and hit the substrate. The atoms may not get sufficient time to rearrange themselves and form a proper crystalline structure even though the substrate is at 750 °C. On the other hand, during annealing, atoms may diffuse in a more controlled manner, in the absence of any incoming bombardment of particles, to form a crystalline structure even if the temperature may be only 700 °C.

From XRD patterns it is clear that a larger number of peaks are seen for the samples deposited at high energy. Peaks seen for all the samples irrespective to energy can be identified as coming from YIG phase and no peak from any other phase is seen. From the full-width at half maximum of the most intense peaks (420), the crystallite size for the annealed films was estimated using the Scherrer's formula. These values increase from ∼9 to ∼38 nm with the increase in laser energy as shown in Fig. 2.

Fig. 3 shows the typical AFM and FEGSEM images of annealed YIG films grown at two extreme laser energies. We observed a crack free YIG films despite of mismatch in thermal expansion coefficients between the substrate and film. The figures show a surface exhibiting granular like features, the size of which increases from ∼30 to ∼250 nm as the laser energy increased from 240 to 350 mj. These numbers are very different

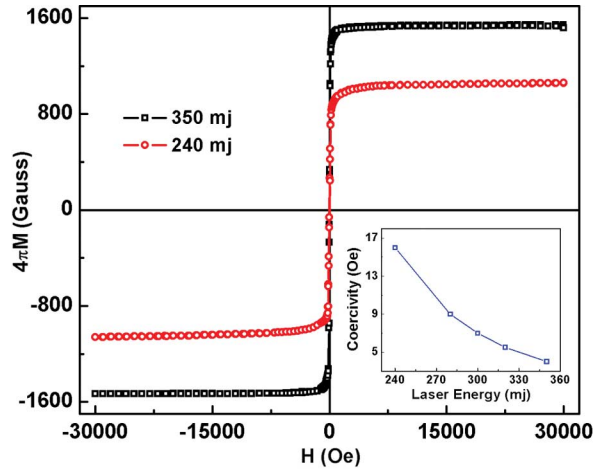

Fig. 4. Room temperature *M–H* loop of YIG thin films deposited at two extreme laser energies. Inset: Variation of H_C YIG thin films a function of Laser energy.

from the one obtained using Scherrer's formula. This is because AFM is a surface technique and each observed granular structures may the combination of multiple crystallites.

Fig. 4 shows the *M–H* loop of annealed YIG films grown at two extreme laser energies. From these measurements it is seen that $4\pi M_S$ steadily increases from 1030 to 1520 G while coercivity (H_C) decreases from 16 to 4Oe, as the laser energy is varied from 240 to 350 mj (inset: Fig. 4). The value of $4\pi M_S$ is found to increase with the laser energy and is close to 90% of the bulk (1750 G) value for the film deposited at highest energy. Several authors have mentioned that layers with lower grain sizes may manifest inferior magnetic properties due to the presence of a greater amount of grain boundary volumes [10]–[12]. At these boundaries, the material is expected to be having poorer magnetic ordering than the material within the grain. During annealing, the grain growth diminishes point defects/dislocations and grain boundary volume leading to the improvement of saturation magnetization of the films. The decrease in coercivity with the increase in crystallite size/thickness is quite reasonable. It is easier for the domain walls to move when the film thickness and the grain size are larger [13].

The parallel and perpendicular FMR spectra of YIG thin films of different thicknesses are recorded at 300 K and are shown in Fig. 5. It is clear from the figures that all the parallel FMR spectra consist of single resonance mode while the signals split into multiple resonance modes for perpendicular position. In FMR of thin films, the multiple resonance modes can be observed either because of standing spin wave resonance (SSWR) or because of inhomogeneity present in the film. If SSWR is responsible for the multiple resonance modes, then the mode positions and intensities are very sensitive to the thickness of the film. Hence changing of the thickness of the film is expected to change the position of peaks in the resonance spectra.

In order to understand the origin of multiple resonance modes, we performed acid etching experiment. We decreased the thickness of the films (deposited at 350 mj) by etching it in concentrated HCl for different period of time (30, 60, and 90 min) and after that FMR spectra were again recorded in

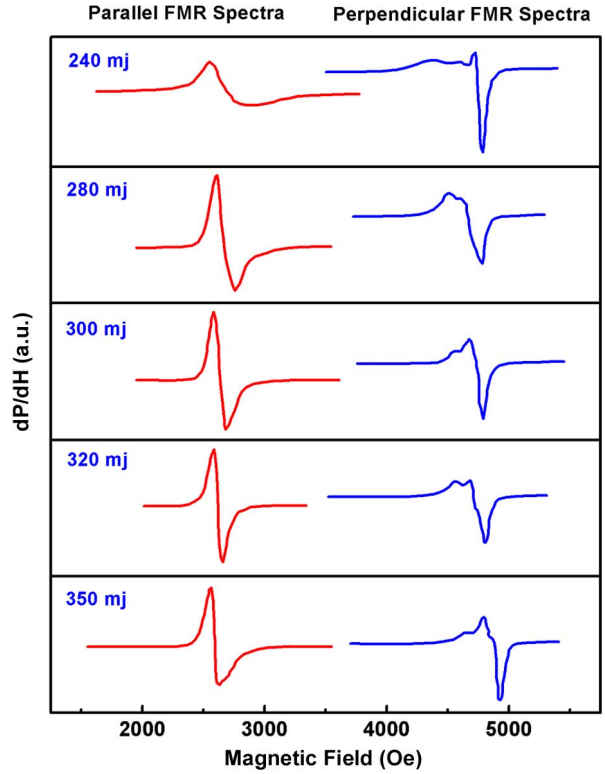

Fig. 5. Perpendicular and parallel FMR spectra of YIG thin films deposited at different laser energy.

perpendicular position (Fig. 6). We note that after etching the FMR spectrum gets cleaner and the main peak shifts towards the lower field accompanied by an increase in ΔH. This implies the inhomogeneous regions present on the surface of the film have been etched out by etching. The shifting of resonance field implies a reduction in effective saturation magnetization. This could be because of change in saturation magnetization or of any uniaxial anisotropy, if at all present in the films. It is possible that due to acid etching, the high magnetic moment regions present on the surface are etched out causing the shifting of resonance mode toward lower magnetic field. In any case, this experiment confirms that the multiple modes seen in nonetched film are not standing spin wave resonance modes but arise because of randomly distributed inhomogeneous regions [14], [15].

The values of effective saturation magnetization $(4\pi M_{\text{eff}})$ and the Lande's factor (g) were calculated for all the films from the perpendicular (the fields of the highest intensity modes) and parallel field positions of the FMR spectra using Kittel's equations (1) and (2)

$$H = \frac{\omega}{\gamma} + 4\pi M_{\text{eff}} \tag{1}$$

$$\left(\frac{\omega}{\gamma}\right)^2 = H(H + 4\pi M_{\text{eff}}). \tag{2}$$

Fig. 7(a) shows the variation of $4\pi M_{\text{eff}}$ as a function of laser energy along with $4\pi M_S$ values obtained from PPMS for the sake of comparison. It is observed that the values of $4\pi M_{\text{eff}}$ follow the same trend as the evaluated from PPMS. However the values of $4\pi M_S$ obtained from PPMS are always smaller

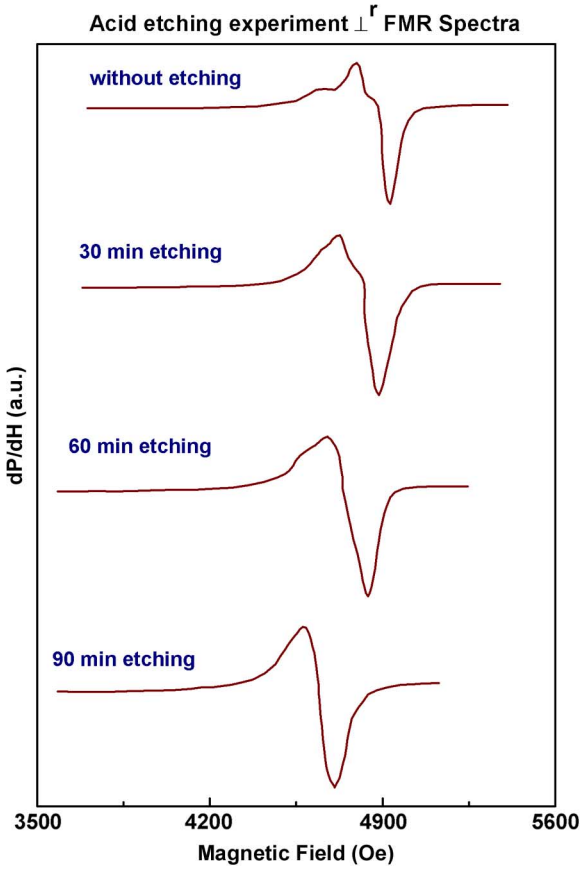

Fig. 6. Perpendicular FMR spectra YIG thin films (deposited at 350 mj) for different time of acid etching.

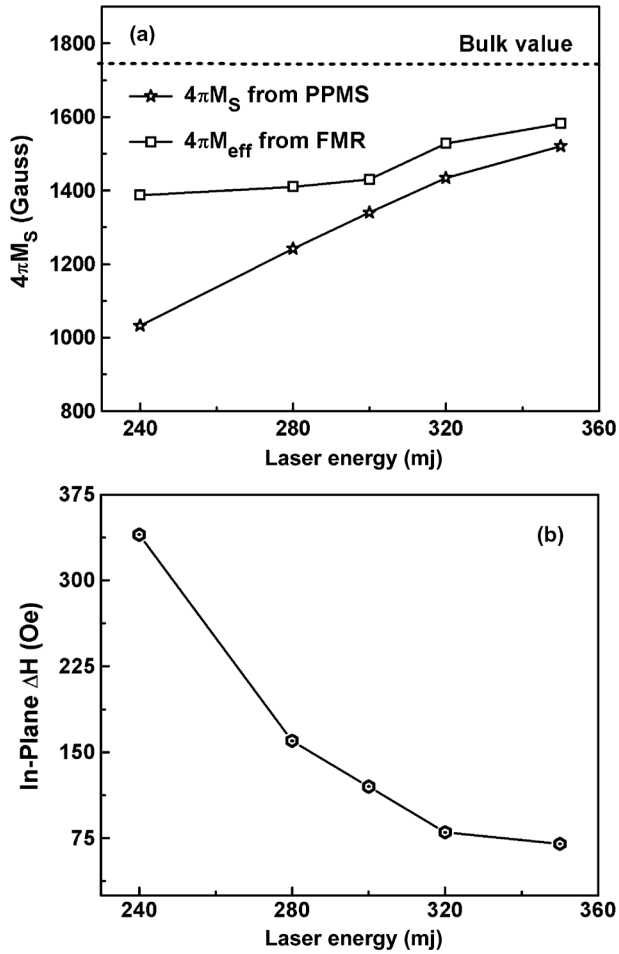

Fig. 7. (a) Comparison of saturation magnetization of YIG thin films (deposited at different laser energy) obtained from PPMS and FMR measurement. (b) Variation of in-plane line width of YIG thin films as a function of laser energy.

than $4\pi M_{eff}$ obtained by FMR. This discrepancy could arise from the fact that we are using only the highest intensity mode to evaluate $4\pi M_{eff}$ assuming it to be uniform precession. The presences of other modes from inhomogeneous region of the sample are, therefore, not taken into account. FMR being a microscopic technique, it shall yield magnetization corresponding to the region from where a mode is arising. On the other hand the $4\pi M_S$ obtained from PPMS shall depend on the average moment over the entire sample. This is also confirmed from earlier carried out acid etching experiment, where the perpendicular FMR spectrum was found to shift towards lower field due to decrease in magnetization as a result of etching. Further if the samples possess some uniaxial anisotropy that may also make $4\pi M_{eff}$ differ from $4\pi M_S$. The "g" values of YIG films calculated from Kittel's equations are found to vary between 1.94 and 2.03.

One of the most interesting results obtained from the study pertains to the FMR parallel linewidth $(\Delta H_{||})$ of the samples. We note from Fig. 7(b) FMR $\Delta H_{||}$ reduces from 340 to 70 Oe as laser energy increases from 240 to 350 mj. This value observed for 290-nm-thick film is very small (by a factor $\sim$2.5) compared to previously reported results on polycrystalline YIG films [11], [12]. The variation of line width as a function of energy can be due to the change in the porosity which is likely to be smaller for higher energy deposition. The lower porosity in turn may reduce the line width. In addition to porosity, larger thickness of sample for high power may also be a cause. In the film of lower thickness, contribution towards the damping processes may be dominated by surface properties [16]. But with the increased thickness the relative contribution from surface would be smaller [17].

From the present investigations it is evident that the laser energy increases the crystallite size as well as thickness resulting in an increase of magnetization. At the same time there is always a chance of increase in density and decrease in porosity in the films with the same increment in energy. So a decrease in line width is found with the increase in laser energy.

The results reported in the present study are interesting from the view point of the possibility of application in devices which require YIG films where very low line width is not essential. There these low cost polycrystalline YIG films can be used instead of high cost epitaxial YIG films. From this study, we conclude that by controlling the power, thickness, microstructure and the microwave properties can be controlled.

IV. Conclusion

In conclusion, we have investigated the phase formation, microstructure, magnetic and microwave properties of YIG thin films prepared by PLD varying the laser energy. The value of $4\pi M_S$ is found to increase with the laser energy and is close

to 90% of the bulk value for the film deposited at highest laser energy. This film also has the highest thickness with lowest line width of 70 Oe. From detailed analysis a correlation between the microstructure and magnetic/microwave properties has been established.

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