

Preface

The technological growth of the present world is mainly due to the invention of computers. From massive space-occupied computers with slow processing using cathode tubes, evolution has brought humanity to the era of laptops and mobile phones, where everything is one click away. All information is well distributed and accessible to the learner's desire through the Internet. These computers use electronics that utilize the charge of the electron. The charge current gives rise to the resistance and associated Joule heating. As a result, there is a limitation in the further compactness and fast functioning of processor units with current electronics.

The solution to the present limitation lies in the electron, which has two intrinsic properties: the charge on which electronics have been based and the spin. Since spin transport has minimal Joule heating, it can be an alternative to electronics for further technological enhancement. The dissipationless spin transport is a branch of condensed matter physics known as spintronics. Its functionality has been rooted in the spin current, which can have various origins, like the spin Hall effect. Likewise, one way to generate spin currents is through spin waves. Bloch predicted the spin wave in 1929, and as the waves propagate in the spin moment, they are called spin waves, and the quanta of spin waves are magnons. Therefore, the field of spintronics, which has a basis in magnons for information transport and processing, is known as magnonics. The additional advantage of magnonics is that

it is compatible with electronics and spintronics. There are both metallic and insulator hosts for the magnons, to name a few, $\text{Co}_x\text{Fe}_y\text{B}_z$, Permalloy, and Heusler alloys, and the insulators are yttrium iron garnets (YIG) and ferrites. The most popular insulating magnon host is YIG, which has the lowest reported Gilbert damping parameter. Because of this, magnons' free path length in μm thick YIG is a few tens of μm , making them the most promising for information transport. Furthermore, the power required to excite the spin wave in YIG is deficient, a few tens of dBm. Undoubtedly, ferromagnetic insulators are the conscious choice to explore magnonics as an alternative to electronics. Various sophisticated methods, like liquid phase epitaxy (LPE), pulsed laser deposition (PLD), and rf-sputtering, are utilized to deposit these ferromagnetic insulators. However, these methods require ultra-high vacuum to avoid contamination and sophisticated designed containers, which makes them expensive. This thesis aims to explore alternatives to these methods and study the iron garnet thin films deposited with a solution-processed method.

The present thesis focuses on the cost-effective deposition method of iron garnets ($\text{Y}_3\text{Fe}_5\text{O}_{12}$:YIG and $\text{Tm}_3\text{Fe}_5\text{O}_{12}$: Thulium Iron Garnet (TmIG)) using sol-gel-based spin coating method. The sol-gel-based spin coating is an all-solution-based method that uses nitrates and acetates as precursors, making it more approachable and economical. It uses a spin coater, tubular, and electric furnace, making it versatile and available for industrial use. This thesis presents the sample grown with a spin-coating rotation speed of 4000 rpm and an annealing temperature of 1173 K, comparable to the LPE. The magnetic characterization of these deposited samples, both polycrystalline and epitaxial thin films, have been investigated using in-house ferromagnetic resonance (FMR) and superconducting quantum interference device (SQUID)-vibrating sample magnetometry (VSM). YIG and TmIG thin films have shown the lowest Gilbert damping and high spin-orbit coupling with perpendicular magnetic anisotropy, respectively. These properties make both a potential host material for spintronic and magnonic applications. Therefore, these two have been deposited and

studied in the thesis. The polycrystalline YIG and TmIG samples have been deposited on thermally oxidized Si (100) substrates, and the epitaxial YIG and TmIG thin films on the $Gd_3Ga_5O_{12}$ (GGG) (111) substrates. The lab source and synchrotron X-ray diffraction (XRD) study were used to confirm the phase of the thin films. Thickness estimation is performed using X-ray reflectivity (XRR) and cross-sectional scanning electron microscopy (SEM). The topography and morphology of thin films have been analyzed using atomic force microscopy (AFM) and SEM. The stoichiometry of the constituent elements with their ionic state has been confirmed by utilizing X-ray photoelectron spectroscopy (XPS). Confirmation of the ferrimagnetic ordering and low-temperature compensation in the TmIG was performed using the SQUID-VSM. The estimation of the effective magnetization has been performed using FMR. The dissipation factors of the magnetic energy have also been estimated using the FMR.

This thesis presents an optimization of the annealing duration of polycrystalline YIG thin film deposited using the sol-gel-based spin coating on the thermally oxidized Si (100) substrate at the annealing temperature of 1173 K. The reason to study further the 5 hrs sample is the XPS study which confirms the oxygen deficiency in the YIG annealed for 10 hrs, which supports the SQUID-VSM data for the lower magnetization in the 10 hrs sample. Further gyromagnetic ratio, effective magnetization, and Gilbert damping parameters have been studied in the YIG 5 hrs sample. The cross-sectional SEM shows the thickness of the deposited thin film is 20 ± 3 nm. The Gilbert damping parameter estimated is $\alpha = 4.7 \pm 0.4 \times 10^{-3}$, which is an order higher than PLD-grown samples along with the higher extrinsic dissipation linewidth of 5.0 ± 0.3 mT. This work concludes that the sol-gel-based spin coating has higher Gilbert damping and higher saturation magnetization with lower coercivity due to the high crystalline nature of the nanoparticles of the uniform thin film. Therefore, the epitaxial thin film of the YIG has been deposited on the GGG (111) substrate. This epitaxial thin film has been studied using synchrotron XRD, confirming

its excellent crystallinity. XPS has been utilized to verify the elemental composition to observe stoichiometry is maintained in the solution processes. This results in concluding the presence of Fe^{3+} in the sample confirms that stoichiometry is maintained with the desired ionic state of Fe, i.e., 3+. The thickness of the epitaxially deposited thin film is estimated to be 18 ± 1 nm using XRR with the topographical average roughness of 0.4 nm. The FMR study suggests that the effective magnetization is lower than the saturation magnetization with the strong presence of anisotropy in the epitaxial YIG. The stress calculation shows the significant contribution to the magnetic anisotropy in the YIG was caused by stress-induced anisotropy. The epitaxial sample has a higher damping parameter $7.6 \pm 0.3 \times 10^{-3}$ compared to the polycrystalline sample with a less inhomogeneous contribution to the linewidth 2.1 ± 0.7 mT. The gadolinium iron garnet interface formation between the GGG and YIG causes spin inhomogeneity, contributing to Gilbert damping.

Similarly, the thesis presents the study of the polycrystalline TmIG sample. It discusses the single-phase formation of the TmIG with the deposition method at the annealing temperature of 1223 K and the duration of 3 hrs. The XPS study of the TmIG is done for the first time in this thesis. Confirming the Tm^{3+} state and the Fe^{3+} cationic state. The theoretically predicted low-temperature magnetization of the TmIG is established with the experimental determination of the compensation temperature ≈ 15 K. After that, the room temperature ferrimagnetic ordering along with the low-temperature modulated ferromagnetic ordering is confirmed experimentally with the magnetic hysteresis loops and followed by the deposition of an epitaxial thin film of the TmIG on the GGG(111) substrate utilizing the sol-gel-based spin-coating method. It has also been studied using synchrotron XRD, confirming the deposited thin film's excellent crystallinity and low interfacial roughness. The deposited epitaxial TmIG thin film is of the thickness 12 ± 1 nm as estimated using the XRR. For more compact processing and memory, the alignment of the spins in the thin film should be an out-of-plane easy axis, and it is

known as perpendicular magnetic anisotropy. The presence of perpendicular magnetic anisotropy (PMA) in the deposited epitaxial TmIG thin film is confirmed by polar magneto-optical microscopy. The origin of this PMA is similar to the literature, i.e., stress-induced anisotropy. The thesis also discusses that thin films have a lower g-factor of 1.391 ± 0.006 compared to the literature because of the high spin-orbit coupling of the f-orbital of thulium cation and higher strain in the deposited thin film. The Gilbert damping parameter is estimated $\alpha = 0.018 \pm 0.004$. The quality of the deposited TmIG is equivalent to the PLD-grown samples of TmIG, making this method explore the inverse spin Hall effect study to estimate the spin Hall angle of the rare-earth iron garnets. This sol-gel-based spin-coating deposition method has the potential to be researched further and can be a cost-effective alternative to sophisticated methods.