

Chapter 1

Introduction

1.1 Introduction

The twentieth century has been an era of technological evolution, and at the end of this century, every household was equipped with personal computers. This century has seen the development of computers of the size of a room and their evolution to be as compact as fit in a lap [1]. The twenty-first century is moving towards more compact and faster personal computers, advanced supercomputers, and futuristic quantum computers [2]. The development of current computers has been based on electronics but the main drawback of it is Joule heating [3]. Electronics require a significant current (for example, the maximum load current of the TPS54332 chip is 0.55 A) that increases temperature due to the Joule heating [4]. If further current density increases with present electronics, it can even cause metal migration that can cause short circuits in compact ICs. As the size reduces, these challenges enhance, and a less heat-producing way of transport needs to be studied. One way of doing this is using the electron's spin component and phenomena based on the spin current, which comes fundamentally under spintronics and magnonics. Spintronics is the

field that deals with spin transport. Many advanced scientific studies focus on developing spintronics because the scientific community sees their future in it for e.g. developing spintronic based information storage and processing in terms of designing the spin transfer torque magnetic random access memory [5], magnetic tunnel junctions [6], and tunneling magnetic resistance sensors [6]. Therefore, to faster and denser information transport and memory, it is essential to understand and enhance our knowledge about spintronics [7, 8, 9]. The discovery of giant magnetoresistance (GMR) in magnetic/nonmagnetic superlattice by A. Fert and P. Grünberg [10, 11] has been considered as the origin of the spintronics. The collective work of GMR was awarded the Nobel Prize in 2007 for its contribution to Physics. With the discovery of the spin valve-based GMR, memory storage densification has improved. Spintronics has many applications, a few of them has been discussed in Figure (1.1). GMR and Tunneling magnetoresistance (TMR) have made hard disk drives (HDD) more compact [12].

Understanding the presence of the spin-based electronic phenomenon has been advanced by presenting the idea of spin injection by Datta and Das in 1990, [13]. They proposed the replacement of electro-optical modulation with an electric spin-oriented field effect transistor (spin FET). A successful demonstration of the spin-polarized Light Emitting Diode (Spin LED) was done in 2001 by Zhu *et al.* [14]. Along with the electron-based spin current, the more dynamic spin wave-based spin current has also been experimentally observed [15]. Bloch predicted a spin wave in 1929 [16]. The excitation of the magnetic ordering behaved as the wave propagating and is quantized as magnons. Magnons has opened a new area of research to get Joule heating free information transfer [17, 18]. The field of data processing and information transfer using magnons is known as magnonics. There are various advantages of using magnons in the information processing; they are as follows:

conducting and insulating magnonic materials for, e.g., $\text{Co}_x\text{Fe}_y\text{B}_z$, permalloy (NiFe), Yttrium iron garnet (YIG), rare-earth iron garnets, and Heusler alloys. This thesis deals with the magnonic crystals YIG and thulium iron garnet (TmIG). Both insulating ferromagnets have long spin wave propagation distances and low damping parameters. There are various methods to synthesize these. This thesis focuses on a cost-effective solution-based spin coating method, as it has not been explored as much as the other synthesis methods when discussing iron garnets. For a basic understanding of the properties of these magnonic crystals, ferromagnetic resonance (FMR) has been utilized. The following section elaborates on choosing iron garnets, sol-gel-based spin-coating, and FMR as systems to study, the method of deposition, and the method to use, respectively.

1.2 Why Iron Garnets?

Garnet is an English word meaning "dark red," as the abundant garnets are dark red. Natural garnets are available worldwide. Iron garnets were synthesized in the laboratory by Bertaut and Forrat in 1956 [22]. YIG was the very first garnet invented. Later on, various single-crystal and polycrystalline rare-earth garnets were synthesized [23]. YIG has been utilized extensively to study magnon properties, as it has the lowest FMR linewidth and lowest reported Lorentz damping parameter 4.2×10^{-4} [24]. The smaller the linewidth, the lesser the dissipation of the spin wave energy. There are broadly two dissipation contributions: 1) extrinsic and 2) intrinsic. Spin-orbit coupling, electron-electron interactions, and spin relaxation in the direction of the applied field contribute to intrinsic dissipation. The magnon-magnon scattering and the presence of defects contribute to the inhomogeneous extrinsic dissipation. YIG has been utilized to understand these contributions extensively [25, 26, 27, 28]. Gadolinium gallium garnet (GGG) substrate was used as a substrate to get the epitaxial YIG as a thin film. However, The lattice mismatch between YIG and GGG is 0.61% [29], making a less strained thin film and avoiding the strain-induced perpendicular

magnetic anisotropy (PMA). Substituted GGG is used to generate PMA in the thin films [30]. It is essential to have PMA in the present day to densify further the information, spin polarization, magneto-optical devices, spin-torque oscillators, and magnon valves [31, 32, 33].

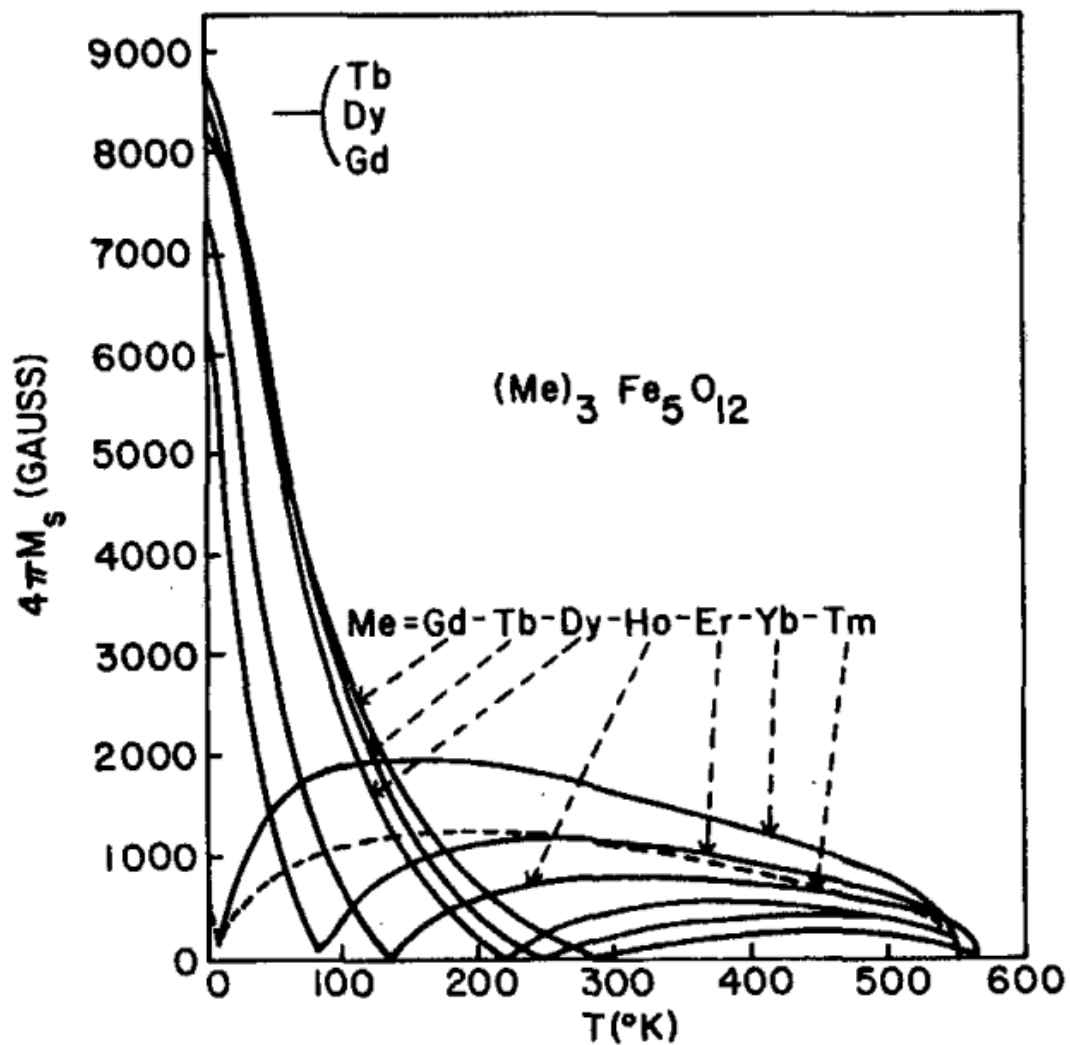


Fig. 1.2 Magnetization as a function of temperature for rare-earth iron garnets (adapted from [23]).

The substitution of the yttrium site in YIG with the rare-earth elements results in PMA and high spin-orbit coupling (SOC) in the thin films. This substitution changes the magnetic

behavior of the iron garnet at low temperatures [23]. Figure (1.2) depicts the magnetic behavior of the various rare-earth iron garnets. A compensation temperature is present due to the competing thulium and iron magnetic moments. The lowest compensation temperature belongs to TmIG and ytterbium iron garnet. This thesis discusses the TmIG due to its low compensation temperature and presence of high SOC and PMA in thin film on GGG [34, 35]. The opportunity for spin-orbit torque opens wide magnonics applications for the TmIG/Pt heterostructures.

1.3 Why Sol-Gel-based Spin Coating?

Sol-gel-based spin coating is an all-solution-processed method, used for both industry and scientific purposes. It has industrial applications in depositing the protective layers, organic and inorganic layers, and anti-reflective coating on glass, namely, lenses, showrooms, solar cells, and TV screens [36, 37]. For scientific purposes, it has been used, for example, in the organic coating of polymers in the biomedical field, titanium oxide layer deposition for the buffer layer deposition, and organic light emitting diodes [38, 39, 40].

The term sol-gel was used by Graham in the mid-1800s, and in fact, the first synthesized material from the sol-gel processing was silica. It was synthesized when the hydrolysis of the tetraethyl orthosilicate was performed under an acidic environment [41]. In the past 30 years, the sol-gel method has been further improved by combining it with other deposition methods, like spin coating, dip coating, spray coating, and casting [42, 43, 44]. However, the objective of the thesis is to deposit a thin film of nm thickness; therefore, the sol-gel-based spin coating was opted for. The rotation speed can optimize the thickness and achieve a uniform thin film. The sol-gel-based spin coating is the amalgamation of the sol-gel method and the spin coating, where the condensate of precursors' nitrates or alkoxides colloidal solution in aqueous or organic solvent is deposited after treating the substrate over

the spin coater, followed by the annealing treatment. The preceding procedure produces a smooth, thin film with a few nm thickness to μm . The advantages of its application in the industries and scientific community make it a promising method of deposition that should be used for the YIG and the TmIG thin film growth. The Peña-Garcia et al. have prepared YIG with it earlier [45]. However, the TmIG has been deposited for the first time in the present work.

1.4 Significance of Ferromagnetic Resonance

Knowing the materials analyzed in the thesis is essential, but knowing how to characterize these materials is also crucial. The prominent method used is FMR, which was discovered in 1946 by Griffith [46], and Kittel did with its detail description in 1947 [47]. Consequently, it is relevant in spintronics because its straightforward analysis gives the Gilbert damping parameter, effective magnetization (a combination of saturation magnetization and the effective anisotropic field), and the gyromagnetic ratio, which are essential to deciding their application properties. Broadband FMR can measure the gyromagnetic ratio, saturation magnetization, and Gilbert damping parameter[48].

The FMR is a spectroscopy technique that probes the Larmor precession of the sample's magnetic moment along the external magnetic field, as explained in Chapter 3. The frequency of the Larmor precession in ferromagnetic materials is in the microwave range [49]. When the microwave and Larmor frequencies are identical, resonance happens. This resonance has been recorded using the transmission spectra of the input microwave exposed through the coplanar waveguide. Therefore, in FMR used in this thesis, samples at varying external magnetic fields exposed to the microwaves in the range of 2 - 10 GHz to observe the transmission of the microwave signal across the 2 - 800 mT external magnetic field [50]. Knowing the dynamics by Landau-Lifshitz- Gilbert equation and the Kittel equation

of the relation of net magnetization, magnetic anisotropy, resonance field, and frequency of excitation make this technique a non-destructive method to analyze the materials constants [47, 51].

This thesis utilized the broadband FMR of 2 - 10 GHz frequency to explore the ferromagnetic insulators' static and dynamic magnetic parameters.

1.5 Outline of the thesis

The present thesis is organized into eight chapters as follows

Chapter 1 describes the motivation, introduction, and background behind selecting iron garnets as ferrimagnetic insulators for magnonics-related studies. The use of sol-gel-based spin coating and FMR has been discussed.

Chapter 2 elaborates on the theoretical background of the exchange, super-exchange, anisotropy, the ferromagnetic resonance conditions, and the magnetic damping factors of the magnetic energy.

Chapter 3 provides a detailed synthesis method, i.e., sol-gel-based spin coating. The working principles and the setup of the characterization methods are explained in a very concise manner. The characterization methods discussed are X-ray diffraction (XRD), X-ray reflectivity (XRR), atomic force microscopy (AFM), scanning electron microscopy (SEM), X-ray photoelectron spectroscopy (XPS), magnetic property measurement system (MPMS), and the FMR.

Chapter 4 discusses the optimization of the annealing duration for the single-phase polycrystalline YIG deposition using sol-gel-based spin-coating on a thermally-oxidized Si (100) substrate. The stoichiometry of the element is confirmed using XPS. The compre-

hensive FMR study of a 20 ± 3 nm thick YIG thin film has been discussed. The lowest solution-based Gilbert damping $\alpha = 4.7 \pm 0.4 \times 10^{-3}$ is presented.

Chapter 5 discusses the epitaxial growth of the YIG on the GGG (111) single-crystal. The 18 ± 1 nm thick YIG epitaxial thin film of excellent crystallinity is comprehensively studied to estimate the Gilbert damping parameter $\alpha = 7.6 \pm 0.3 \times 10^{-3}$ and the inhomogeneous damping. The presence of the stressed-induced anisotropy as a significant contributor to the uniaxial-anisotropy has been presented.

Chapter 6 discusses the polycrystalline 22 ± 1 nm thick TmIG thin film synthesized with all solution-processed methods. The elemental constituents' atomic percentages are estimated using the XPS, with the results confirming that the stoichiometry is maintained compared to the other methods, namely, PLD and rf-sputtering—the experimental confirmation of the low-temperature compensation temperature in the thin films. The study of the low temperature and room temperature magnetic behavior confirms the room temperature ferrimagnetic and the low temperature modulated ferromagnetic magnetic behavior.

Chapter 7 discusses the epitaxial 12 ± 1 nm thick TmIG thin films. Epitaxy of the thin film is confirmed with the synchrotron-based grazing incident ϕ - scan XRD. The excellent crystalline TmIG thin film has been confirmed with the Laue oscillations in the XRD. XRD-based strain-induced anisotropy is estimated, which has confirmed that the major contribution to the uniaxial anisotropy is stress-induced anisotropy, which results in PMA. The Gilbert damping parameter observed is equivalent to the pulsed laser-deposited thin films.

Chapter 8 summarises the final experimental findings of the present thesis and discusses related prospects.