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Full Length Article

Effect of annealing temperature on surface morphology and ultralow ferromagnetic resonance linewidth of yttrium iron garnet thin film grown by rf sputtering

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ABSTRACT

We report high-quality yttrium–iron–garnet (YIG; $Y_3Fe_5O_{12}$) ultrathin films grown on {111} gadolinium–gallium–garnet (GGG; $Gd_3Ga_5O_{12}$) substrates using RF sputtering deposition on an off-stoichiometric target and optimized thermal treatments. We measured a narrow peak-to-peak ferromagnetic resonance linewidth (ΔH) whose minimum value was 1.9 Oe at 9.43 GHz for a 60-nm-thick YIG film. This value is comparable to the most recently published value for a YIG thin film grown by pulsed laser deposition. The temperature dependence of the ΔH was investigated systematically, the optimal annealing condition for our growing condition was 875 °C. Structural analysis revealed that surface roughness and crystallinity played an important role in the observed ΔH broadening. Furthermore, the thickness dependence of the ΔH, which indicated that 60 nm thickness was optimal to obtain narrow ΔH YIG films, was also investigated. The thickness dependence of ΔH was understood on the basis of contributions of surface-associated magnon scattering and magnetic inhomogeneities to the ΔH broadening. Other techniques such as transmission electron microscopy, scanning electron microscopy, and X-ray diffraction were used to study the crystalline structure of the YIG films. The high quality of the films in terms of their magnetic properties was expressed through a very low coercivity and high saturation magnetization measured using a vibration sample magnetometer.

1. Introduction

Yttrium–iron–garnet (YIG; $Y_3Fe_5O_{12}$) thin films are the most prominent candidates for spin-caloritronics and spintronics [1–9] due to their exceptionally low magnetization damping constant ($\alpha \sim 10^{-5}$) [10] that is one-to-two orders of magnitude smaller than those of metallic ferromagnets [11]. Additionally, the high Curie temperature (560 K) [12] and insulating property of YIG thin films are essential for various applications, especially those in high-frequency YIG-based electronic and spintronic microwave devices. Recently, emerging research on magnonics [13,14] and spin wave pumping [15–17] has created critical requirements for YIG thin films of ultralow damping constant and thickness in the nanometer range. Because the damping constant of YIG thin films, which is directly proportional to the peak-to-peak ferromagnetic resonance (FMR) linewidth (ΔH) [18], is strongly affected by structural and morphological properties of the films, high-quality and high-precision thin-film fabrication is critically required. Therefore, broad ranges of chemical and physical methods have been used to prepare such high-quality YIG thin films. Generally, physical methods are preferred for the preparation of low damping constant YIG thin films using vacuum-based processes to enhance quality and purity.

Among the various physical methods, both radio frequency (RF) sputtering and pulsed laser deposition (PLD) have been widely used for controlling thickness, stoichiometry, and magnetic properties of YIG films. The highest-quality YIG films have been prepared by PLD, which allows stoichiometric transfer of the target composition. The smallest ΔH for thin films reported to date is 1.3 Oe at 9.6 GHz for a 56-nm-thick PLD film [19]. Nevertheless, the sputtering method has several advantages over PLD, including higher uniformity films and a lower fabrication cost, features that are desirable for mass

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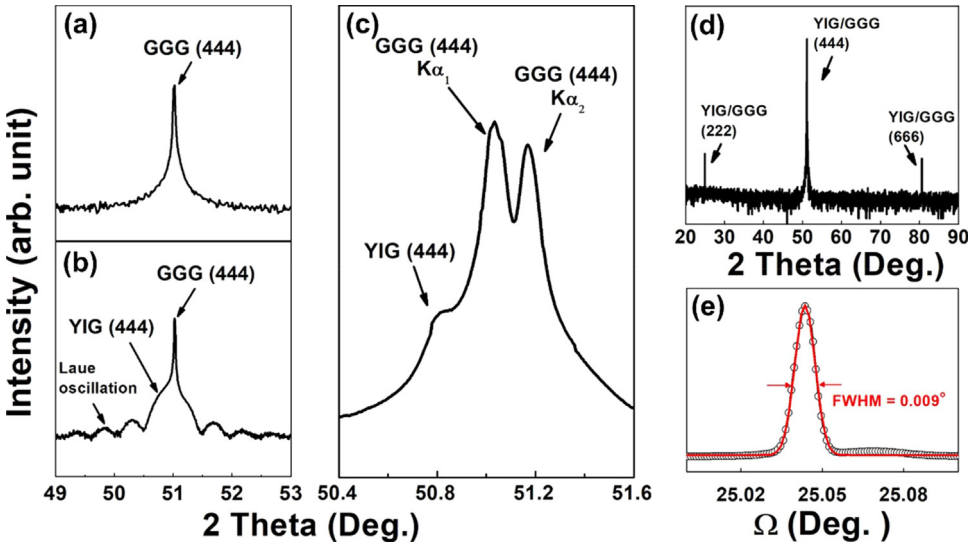

Fig. 1. X-ray diffraction patterns. (a) Gadolinium–gallium–garnet (GGG; $Gd_3Ga_5O_{12}$) substrate. (b) 20 nm-thick yttrium–iron–garnet (YIG; $Y_3Fe_5O_{12}$) film annealed at 875 °C grown on GGG with monochromator measurement. (c) 20 nm-thick YIG film annealed at 875 °C grown on GGG without monochromator measurement. (d) Wide-scan diffractogram of sample (b); the solid line shows the Gaussian fit to the peak. (e) Rocking curve of the YIG peak.

production. Many researchers have investigated the synthesis and ΔH of sputtered YIG thin films. In 2014, Wang et al. presented the results of a 20-nm-thick YIG film with a peak-to-peak ΔH of 11.7 Oe using an off-axis sputtering method [20]. Chang et al. reported a 22-nm-thick YIG film with a ΔH of 6.8 Oe using on-axis sputtering [21]. Liu et al. also reported on a series of YIG nano-thick films that showed the lowest ΔH of 6.6 Oe for a 100-nm-thick film [22]. Lustikova et al. synthesized a 40-nm-thick YIG film with a ΔH of only 3.8 Oe [23]. Most recently, Du et al. studied spin transport and the spin Hall effect using a 20-nm-thick YIG film having a ΔH of 7.4 Oe [24].

It is well-known that the ΔH of YIG films is dominated by intrinsic and extrinsic inhomogeneities [25–27]. Therefore, precise controls of crystallinity, stoichiometry, and surface morphology are the main issues for the production of low-damping YIG films. In this study, we investigated the growth of ultralow FMR linewidth YIG nano-thick films based on RF sputtering deposition using an off-stoichiometric target and optimized thermal treatments with precise control of surface morphology. We report a YIG thin film exhibiting the lowest ΔH of 1.9 Oe at 9.43 GHz which is lower than that of recently reported sputtering films and comparable to YIG films grown by PLD [19]. The dependence of ΔH on thickness was also investigated.

2. Experiments

YIG thin films were grown on 1-cm^2 gadolinium–gallium –garnet (GGG; $Gd_3Ga_5O_{12}$) (111) single-crystal substrates, sliced from a 3-in.-diameter wafer. A 2-in.-diameter oxide target was synthesized in our lab using a conventional solid-state method as follows. High-purity Y_2O_3 (99.99%) and Fe_2O_3 (99.99%) powders were mixed at the molar ratio of 3:7 and ball milled with acetone and various sizes of Al_2O_3 balls for 24 h. The off-stoichiometric ratio was optimized using X-ray photoelectron spectroscopy (XPS) measurements. The composition of the as-deposited YIG film was Y:Fe:O = 3:5.07:12.1. Then, the compounds were dried in a vacuum oven at 75 °C for 12 h, sieved through 500- and then 106-μm mesh screens and finally calcined at 1200 °C over a period of 6 h to obtain fine grains. These grains were sieved and then pelletized into cylinders using a steel mold under an applied pressure of 6 tons/m^2 for 5 min. The pellets were then sintered at 1350 °C for 6 h to form polycrystalline YIG targets. YIG thin films were grown at a base pressure of less than 1×10^{-6} Torr and the off-stoichiometric target was used to adjust the YIG film composition. The depositions were carried out in an Ar/O_2 gas mixture at a flow rate of 22.5 sccm/2.5 sccm, a 15-mTorr working pressure, and a source power of 100 W at an elevated substrate temperature of 600 °C. The deposition rate was estimated at 0.67 nm/min from Alpha-Step profilometer and scanning electron microscopy (SEM) measurements. After deposition, the samples were cooled in high vacuum at an average rate of 4 °C /min. The samples were subsequently annealed in a tube furnace in an oxygen environment at temperatures ranging from 750 to 1000 °C. The ramping and cooling rates were 4 and 2 °C/min, respectively. The thickness of the samples varied from 20 to 100 nm.

The morphology and structural properties of the YIG films were analyzed by field-emission scanning electron microscopy (FE-SEM; model S-4800; Hitachi), atomic force microscopy (AFM; model MEP-3D-BIO; Asylum Research), X-ray diffraction (XRD; model D8 HR-XRD; Bruker) and transmission electron microscopy (TEM; model TECNAI G2 F30; ThermoFisher Scientific) at an acceleration voltage of 300 kV. The ΔH measurements were carried out at room temperature by electron spin resonance spectrometry (ESR; model JES-TE300; JEOL) while sweeping the direct current (dc) magnetic field at a constant RF frequency of 9.43 GHz (X-band). Frequency-sweep (6–12 GHz) FMR measurements were made using a homemade embedded waveguide spectrometer. Magnetization measurements of the YIG films were performed using a vibration sample magnetometer (VSM; model US/7407; Lake Shore Cryotronics) with an in-plane configuration. Finally, the valence state of the Fe ions was determined by high-performance XPS (model MultiLab 2000; ThermoFisher Scientific).

3. Results and discussion

First, we examined the crystallinity of the YIG thin films to investigate the FMR linewidth. The annealing temperature was optimized to obtain a single garnet phase. XRD was performed by ω/2θ scanning of the YIG and GGG (444) reflections. Fig. 1a shows the XRD spectrum of the GGG substrate. Only a peak corresponding to single-crystalline GGG (444) was evident. Measurements after annealing at various temperatures revealed a single garnet phase above 850 °C. Fig. 1b shows the ω/2θ scan of a 20-nm-thick YIG