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► To cite this version:

Xin Guo, Ying Chen, Genshui Wang, Yuanyuan Zhang, Jun Ge, et al.. Growth and characterization of yttrium iron garnet films on Si substrates by chemical solution deposition (CSD) technique. *Journal of Alloys and Compounds*, 2016, 671, pp.234-237. 10.1016/j.jallcom.2016.02.072 . hal-03525725

HAL Id: hal-03525725

<https://hal.science/hal-03525725v1>

Submitted on 3 Oct 2024

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Growth and characterization of yttrium iron garnet films on Si substrates by Chemical Solution Deposition (CSD) technique

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Article info

Abstract

Keywords:

YIG films
Chemical Solution Deposition
Si substrates
Magnetic performance

Yttrium Iron Garnet (YIG) films were prepared on Si substrates by Chemical Solution Deposition (CSD) technique using acetic acid and deionized water as solvents. Well-crystallized and crack-free YIG films were obtained when annealed at 750 °C and 850 °C respectively, showing a low surface roughness of several nanometers. When annealed at 750 °C for 30 min, the saturated magnetization (M_s) and coercive field (H_c) of YIG films were 0.121 emu/mm² ($4\pi M_s = 1.52$ kGs) and 7 Oe respectively, which were similar to that prepared by PLD technique. The peak-to-peak linewidth of ferromagnetic resonance (FMR) was 220 Oe at 9.10 GHz. The results demonstrated that CSD was an excellent technique to prepare high quality yttrium iron garnet (YIG) films on silicon, which could provide a lower-cost way for large-scale production on Si-based integrated devices.

1. Introduction

Yttrium iron garnet (YIG) $Y_3Fe_5O_{12}$ is a hot candidate for magnetic devices and gained much attention since its discovery [1,2]. Because of the high resistivity [3], small magnetic uniaxial anisotropy [4] and extremely narrow ferromagnetic resonance (FMR) line width $\Delta H < 1$ Oe [5], YIG with lower losses has been vital component in microwave tunable devices such as phase shifters and tuners [6–9]. The Curie temperature of YIG is about 550 K, which makes it a wider application above room temperature.

For the application of microelectronic devices, YIG in the form of films is necessary. Mostly, YIG films are grown on gadolinium gallium garnet, $Gd_3Ga_5O_{12}$ (GGG) substrates which have the similar crystal structure with YIG. Moreover, the difference of lattice constants between GGG and YIG is extremely small, on the order of 0.001 Å. These facts make GGG materials the best substrate for high-quality epitaxial YIG films [10–12]. However, it is hard to obtain large size films on GGG substrates, and the application of YIG films to integrated devices is limited. Therefore, it's essential to deposit YIG films on Si substrates, which is not only beneficial

towards large scale production, but also favorable to Si integrated device applications. However, cracks are easily formed in YIG films if using Si as substrates, because of the lattice mismatch and large thermal expansion coefficient difference (Si: $4.7 \times 10^{-6}/^\circ\text{C}$, YIG: $9.2 \times 10^{-6}/^\circ\text{C}$) [13].

Compared with PLD and sputtering, Chemical Solution Deposition (CSD) technique can offer considerable advantages to prepare films such as better mixing of starting materials, more excellent chemical homogeneity and lower cost. But very few articles were reported about YIG films prepared by CSD technique on Si substrate, for the reason that it is not easy to get pure cubic YIG phase without cracks on the surface [13,14].

In this article, we tried CSD method to prepare YIG films on Si substrates. Pure cubic phase YIG films were grown on Si substrates, using oxide and nitrates as starting materials, acetic acid and deionized water as solvents. Crack-free YIG films with low surface roughness and good magnetic properties were obtained.

2. Experimental section

2.1. Films preparation

YIG thin films with thickness ~300 nm were deposited on Si

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substrates by CSD method as follows: Yttrium oxide and ferric nitrate were firstly dissolved in acetic acid and deionized water to form a chelate complex with a concentration of 0.1 M/L, then the solution was spin-coated on Si substrates at a speed of 5000 r/min for 20 s. After spin-coating, the obtained gel coatings were dried at 200 °C and pyrolyzed at 400 °C. At last, films were crystallized under two annealing conditions: (1) 750 °C for 30 min and (2) 850 °C for 30 min.

2.2. Characterization

TG/DTA measurements were carried out to figure out the thermal behavior of the YIG precursor. The crystalline phases of the samples were checked by X-ray diffraction with $\text{CuK}\alpha_1$ radiation. The surface morphologies and thicknesses of films were shown with an atomic force microscopy (AFM) and cross-section scanning electron microscopy (SEM). The hysteresis loops ($M-H$) were obtained in a vibrating sample magnetometer (VSM) of PPMS. An electron spin resonance (ESR) spectrometer working at 9.10 GHz was used to test the room ferromagnetic resonance. Static magnetic field H was applied parallel to film plane.

3. Results and discussions

The TG/DTA curves and XRD patterns were demonstrated in Fig. 1. A slow endothermic peak appeared before 150 °C, which came from the evaporation of water and organic solvents. The following big weight losses in TG curve and corresponding sharp exothermic peaks in DTA curve at 239 °C, 308 °C and 386 °C were attributed to the decomposition of iron and yttrium complexes. During this process, precursor was pyrolysis to an amorphous YIG state. While after 400 °C, there was only a weak exothermic trend without obvious weight losses, which indicated that amorphous oxide gradually crystallized into a stable garnet structure. When temperature was up to 750 °C, there was no exothermic change which demonstrated that stable crystalline phase was formed by this temperature. According to the TG/DTA curves, YIG films were crystallized in: (a) 750 °C for 30 min and (b) 850 °C for 30 min. As shown in Fig. 1 (b), pure poly-crystallization YIG garnet cubic structures were formed without any second phase. The annealing temperature was much lower than that in 900 °C for 2 h [14]. Through calculation, the out-plane lattice constant of YIG films was about 1.237 nm, which was similar to bulk YIG (1.238 nm). The slight contraction of out-plane lattice may be derived from the thermal tensile stress in YIG films, which could be attributed to the bigger thermal expansion of YIG ($9.2 \times 10^{-6}/^\circ\text{C}$) than Si ($4.7 \times 10^{-6}/^\circ\text{C}$).

Fig. 2 displayed the in-plane AFM images (a, b) with an area of $10 \times 10 \mu\text{m}^2$ and cross-section SEM images (c, d) of YIG films under

two annealing conditions, which showed the surface morphologies and thicknesses of YIG films. No cracks could be seen on the uniform surface and the cross-section, which was an excellent performance compared with the research reported before [13,15]. As shown in enlarged AFM images, YIG film annealed at 850 °C was formed with bigger grain size because the high temperature can offer high energy for grain growth, and the average grain diameter was increased from 80 nm (750 °C for 30 min) to 120 nm (850 °C for 30 min). Consequently, the root-mean-square (RMS) surface roughness ($10 \mu\text{m} \times 10 \mu\text{m}$) were 4.076 nm and 6.554 nm respectively. Furthermore, according to the cross-sectional SEM images, the thicknesses of YIG films were about 300 nm, and both films exhibited porous morphologies. During the process of solvents evaporation, the formation of pores and empty space in films could reduce the effect of thermal expansion and lattice mismatch, which could be beneficial to avoid cracks in films.

To further confirm the quality of YIG films on Si substrates, magnetic properties of YIG films were demonstrated in Fig. 3. Hysteresis loops ($M-H$) in Fig. 3 (a) showed typically soft ferromagnetism with narrow loops. In order to quantify the H_c value of YIG films more correctly, the enlarged images near the coercive field range were especially shown inset. In 750 °C for 30 min, the coercive field (H_c) was about 7 Oe, and saturated magnetization (M_s) was 0.121 emu/mm^3 , resulting in a magnetic induction $4\pi M_s$ of 1.52 kGs. Meanwhile, under the condition of 850 °C for 30 min, YIG films presented the value of $H_c = 13$ Oe, $M_s = 0.122 \text{ emu}/\text{mm}^3$ ($4\pi M_s = 1.53$ kGs). These results were similar to the data ($4\pi M_s = 1.57$ kGs, $H_c = 30$ Oe), of which YIG films were prepared by PLD technique on porous silica [16]. YIG single crystal has a theoretical saturation magnetization of $4\pi M_s = 1.75$ kGs [17] and a coercivity of $H_c < 15$ Oe¹⁸. The decrease of saturated magnetization maybe due to the pores in YIG films and the interface between YIG and Si which could reduce the effective density of YIG films. Furthermore, the strain-induced anisotropy in poly-crystalline YIG films could lower magnetization [19]. However, the magnetic performance of YIG films on Si prepared by CSD were superior to other poly-crystallization films prepared by PLD and sputtering [18,24], and close to the (111) orientation YIG films on GGG with $H_c = 3.8$ Oe and $4\pi M_s = 1.58$ kGs [20]. The temperature dependence of magnetization ($M-T$) were shown in Fig. 3 (b). It can be seen that the Curie point (T_c) of YIG films was about 545 K, which was close to 550 K as reported [8].

Fig. 4 manifested the FMR data at 9.10 GHz with field parallel to film plane: (a) 750 °C for 30 min, (b) 850 °C for 30 min. Resonance frequency in single crystal can be described by the Kittel formula, $f_r = \gamma[(H_0 + H_a)(H_0 + H_a + 4\pi M_s)]^{1/2}$, where γ is the gyromagnetic ratio determined by $g\mu_B/h$. H_a is an effective anisotropy field, and H_0 is the applied magnetic field [21]. Accordingly, films annealed at

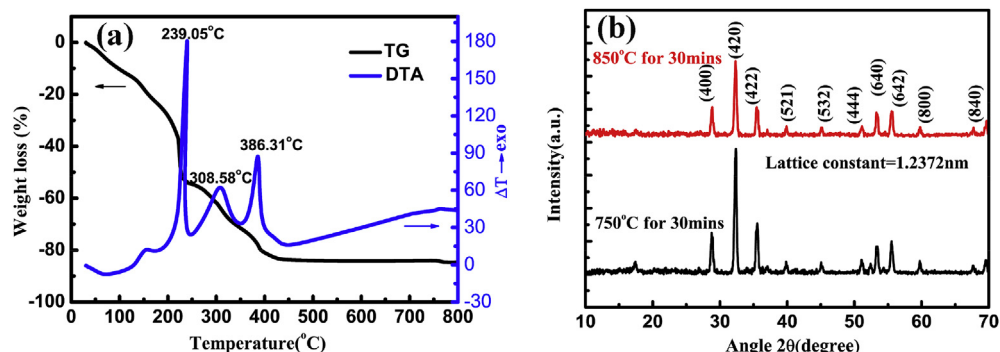


Fig. 1. (a) TG/DTA curves of powders dried from precursor solution and (b) XRD patterns of YIG films under two annealing conditions: (A) 750 °C for 30 min, (B) 850 °C for 30 min.

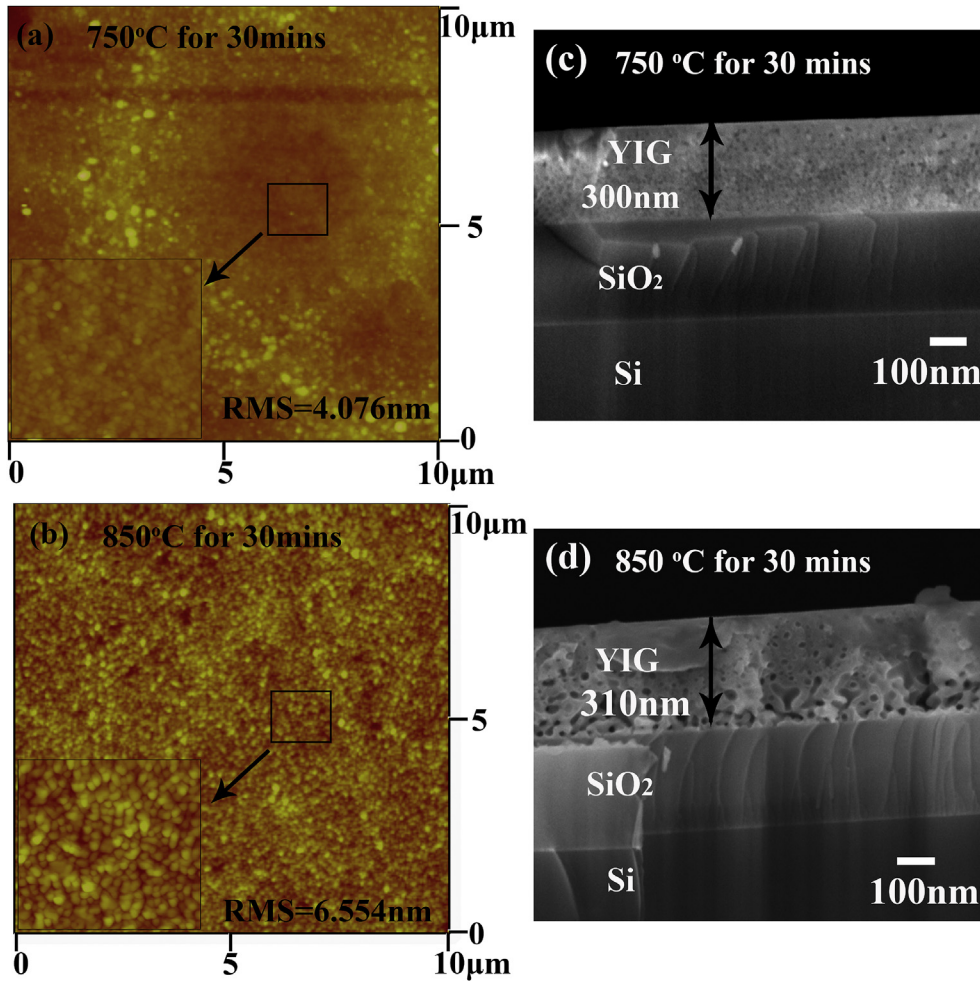


Fig. 2. In-plane AFM and cross-sectional SEM images of YIG films crystallized in (a) (c) 750 °C for 30 min, (b) (d) 850 °C for 30 min.

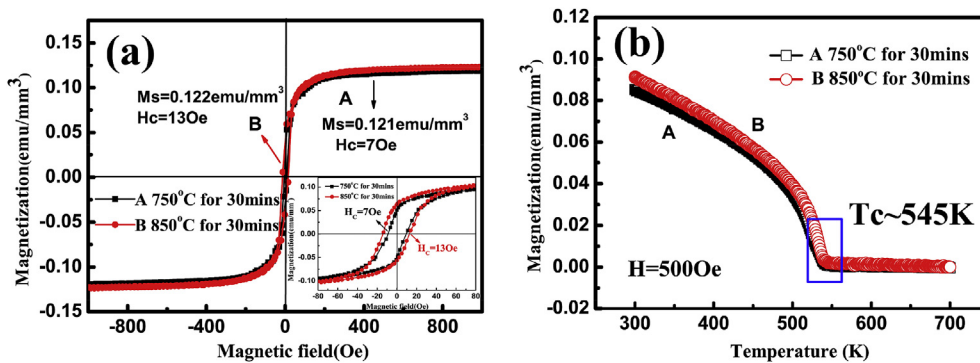


Fig. 3. Magnetic properties of YIG films under different annealing conditions: (A) 750 °C for 30 min, (B) 850 °C for 30 min (a) M–H loops of YIG films (Inset: The enlarged image around the near coercive field range). (b) Temperature dependence of magnetization of YIG films. The coercive field (H_c), saturated magnetization ($4\pi M_s$) and Curie temperature (T_c) can be read from the curves.

850 °C had a smaller Landé g factor ($g = 2.0$) and higher resonance field ($H_r = 3200$ Oe) than that annealed at 750 °C ($g = 2.3$, $H_r = 2800$ Oe) for the different anisotropy and demagnetization effects. Normally, bigger Landé g factor means stronger spin electron–lattice interaction, thus a larger FMR resonance linewidth. The g value of YIG films annealed at 850 °C was equal to standard data because of good crystallization. FMR curves showed spin relaxation process which can be described by the interaction of

magnons. For polycrystalline ferrites, two-magnons scattering, the dominant loss mechanism is extrinsic which arises from the inhomogeneities such as randomly oriented crystallization, impurities, defects, surface roughness and so on [8,20–23]. Therefore, ΔH can be much larger in polycrystalline materials than single crystalline YIG [20]. Fig. 4 showed that FMR resonance linewidth (ΔH) of YIG films were 220 Oe and 206 Oe at 9.10 GHz, corresponding to the Landé g factor respectively. These values were close

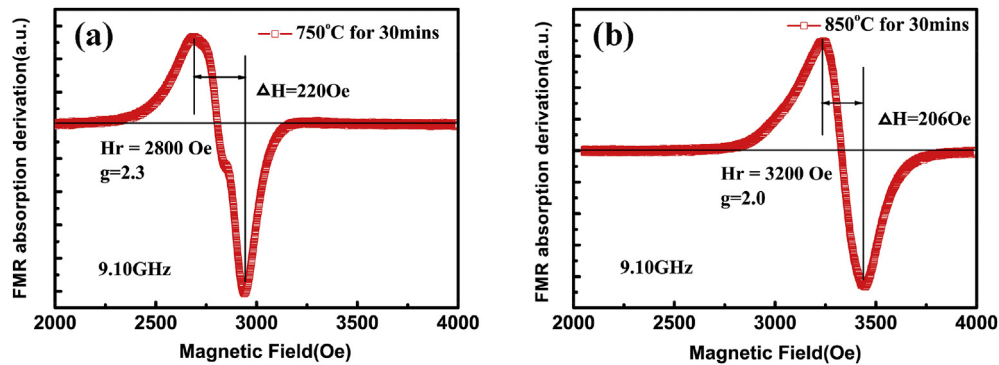


Fig. 4. FMR curves at 9.10 GHz of YIG films in: (a) 750 °C for 30 min (b) 850 °C for 30 min.

to 198 Oe reported by Scooter D. Johnson [24], whose YIG films were prepared on sapphire by aerosol deposition sintered at 1280 °C for 2 h.

4. Conclusions

In summary, this work demonstrated the feasibility of CSD technique for YIG films preparation on Si substrates using acetic acid and deionized water as solvents, which could effectively reduce cracks. The extrinsic two-magnons scattering is the dominant loss mechanism in poly-crystallization YIG films. YIG films with crack-free surface exhibited good performance on Si substrate which could be comparable with that prepared by PLD technique. According to the results and discussions above, CSD technique could offer a simple method with lower cost to fabricate high quality YIG films on Si and be potential for magnetic integrated device applications.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (No. 61371059), Shanghai Natural Science Foundation (No. 13ZR1445600) and international partnership project of Chinese Academy of Science.

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