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Xin Guo, Ying Chen, Genshui Wang, Jun Ge, Yuanyuan Zhang, et al.. Improved Performance of YIG (Y3Fe5O12) Films Grown on Pt-Buffered Si Substrates by Chemical Solution Deposition Technique. Journal of the American Ceramic Society, 2016, 99 (7), pp.2217-2220. 10.1111/jace.14293 . hal-03525733

HAL Id: hal-03525733 https://hal.science/hal-03525733v1

Submitted on 3 Oct 2024

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Improved Performance of YIG (Y3Fe5O12) Films Grown on Pt-Buffered Si Substrates by Chemical Solution Deposition Technique

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High-quality Yttrium iron garnet (YIG) films with crack-free surface and improved magnetic performance were grown on platinum (Pt)-buffered Si substrates by chemical solution deposition technique. The saturation magnetization of obtained YIG films can reach 124 emu/cm³, which was the 88% theoretical value of YIG single crystal. The effects of annealing condition were also discussed. When annealed at 750°C for 1 h, YIG films showed a very small coercive field of 12 Oe and the peak-to-peak ferromagnetic resonance linewidth can be as low as 95 Oe at 9.10 GHz. The results demonstrated that YIG films prepared on Pt-buffered Si substrates can be beneficial to the application of YIG films to integrated devices.

Keywords: yttrium/yttrium compounds; films; ferromagnetism/ferromagnetic materials; magnetic materials/properties

I. Introduction

Y TTRIUM iron garnet (YIG) Y₃Fe₅O₁₂ has been one of the most studied garnet ferrites since its discovery.^{1,2} YIG is an important magnetic compound with many applications to microwave tunable devices, such as microwave generators and analyzers. YIG-based phase shifters and tuners are vital components in phased array radar systems.³⁻⁶ The applications are attributed to lower losses of YIG in microwave band, such as high resistivity,⁷ small magnetic uniaxial anisotropy,⁸ and extremely narrow ferromagnetic resonance (FMR) linewidth $\Delta H < 1$ Oe.⁹

Normally, YIG films are grown on gadolinium gallium garnet Gd₃Ga₅O₁₂ (GGG) substrate which has the same crystal structure, similar thermal expansion coefficient (GGG: 9.3×10^{-6} /°C, YIG: 9.2×10^{-6} /°C) and good lattice match (GGG: a = 12.376 Å, YIG: a = 12.380 Å) with YIG. Up to now, epitaxial YIG films are only obtained on GGG substrates.^{10–13} However, the integration of YIG films to semiconductors such as Si, quartz, GaAs, Al₂O₃, and so on are necessary for integrated devices application.^{14–17} Si-based substrates are widely used for the excellent integrated performance and large-scale production. However, because of the lattice mismatch and thermal expansion coefficient, the difference between Si $(4.7 \times 10^{-6})^{\circ}$ C) and YIG $(9.2 \times 10^{-6})^{\circ}$ C),¹⁴ cracks always appear in films when prepared on Si. It is necessary to introduce a buffer layer on Si substrates to reduce interface mismatch. Some efforts have been made to avoid cracks, such as using oxidized Si or Si/CeO₂ substrates, but there is no significant improvement.^{13,14,18} Platinum (Pt) is mostly used as an electrode material for the perfect conductivity, few are focused on its compatibility. Pt has a thermal expansion coefficient of 9.0×10^{-6} /°C, which is very close to YIG (9.2×10^{-6} /°C), for this reason that Pt maybe a good buffer layer to avoid YIG cracks when annealing. Therefore, we tried YIG preparation on Pt-buffered Si substrates for the purpose to obtain better performance.

Chemical solution deposition (CSD) technique was utilized to prepare YIG films. Compared with PLD and sputtering, some pores can be formed in YIG films with CSD method, which will leave space for reducing stress and lowering cracks formation. Furthermore, CSD technique can have large-scale production, excellent chemical homogeneity, and lower cost.

In this work, YIG thin films were deposited on Pt-buffered Si/SiO_2 substrates by CSD method, which showed good performances with crack-free and improved magnetic values. The effect of annealing conditions was also discussed.



Fig. 1. XRD patterns of YIG films in four annealing conditions: (1) 750°C for 30 min, (2) 750°C for 60 min, (3) 800°C for 30 min, (4) 850°C for 30 min.

P. Gomez-contributing editor

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II. Experimental Procedure

YIG films were deposited on $Pt/SiO_2/Si$ substrates by CSD method as follows: Yttrium oxide and ferric nitrate were firstly dissolved in acetic acid and deionized water with a concentration of 0.1 M/L, then the solution was deposited on $Pt/SiO_2/Si$ substrates with a spinning speed of 5000 rpm for 30 s. After, the obtained gel coatings were dried at 200°C and then pyrolyzed at 400°C. At last, films were crystallized under four annealing conditions in air: (1) 750°C for 30 min, (2) 750°C for 60 min, (3) 800°C for 30 min, (4) 850°C for 30 min.

The structure and morphology of films were checked by Xray diffraction with CuK_1 radiation (Model RAX-10; Rigaku, Tokyo, Japan) and atomic force microscopy, AFM (Bruker, Karlsruhe, Germany). An impedance analyzer Agilent 4294A (Agilent Technology Japan, Ltd., Kobe-shi, Japan) was employed to show the dielectric properties. The hysteresis loops (M-H) were obtained in a vibrating sample magnetometer of PPMS (PPMS-9, Quantum Design, San Diego, CA). The room FMR was carried out using an electron spin resonance (JEOL-FA, Tokyo, Japan) spectrometer working at 9.10 GHz.

III. Results and Discussions

XRD patterns of YIG films are shown in Fig. 1 with different annealing conditions: (1) 750°C for 30 min, (2) 750°C for 60 min, (3) 800°C for 30 min, (4) 850°C for 30 min. As shown, pure poly-crystallization YIG garnet structures can be formed without any second phase. From calculation, the lattice constants of YIG films were about 1.2372 nm, which were much closer to the bulk YIG value (1.2380 nm). It meant that YIG films on Pt/Si substrate can maintain good garnet structure with a small strain, for the reason that Pt can buffer the lattice constant mismatch between YIG and Si substrates.



Fig. 2. 3D AFM images of YIG films crystallized in (a) 750°C for 30 min, (b) 750°C for 60 min, (c) 800°C for 30 min, (d) 850°C for 30 min.



Fig. 3. Magnetic and dielectric properties of YIG films in different annealing conditions: (A) 750°C for 30 min, (B) 750°C for 60 min, (C) 800°C for 30 min, (D) 850°C for 30 min. (a) M-H loops of YIG films. The coercive field (Hc), saturation magnetization (Ms and 4π Ms) can be read from curves and are shown in Table I. (b) Dielectric constants and dielectric loss (tan δ) of YIG films upon frequency at room temperature.

The surface morphology was displayed by AFM with an area of 10 μ m × 10 μ m in Fig. 2. No cracks can be seen on the surface, which was excellent performance compared with other reports of cracks.^{16,19} Pt has a thermal expansion of 9.0×10^{-6} /°C, which is much closer to YIG (9.2×10^{-6} /°C), in this condition, when YIG grown on Pt-buffered Si substrate, there was little thermal strain in films, resulting in better surface without cracks. Film roughness was lower than 10 nm, with increasing annealing time and temperature, RMS was increased, which comes from the high energy for grain growth when higher temperature and longer time.

Figure 3 and Table I gave the magnetic and dielectric properties of YIG films: (A) 750°C for 30 min, (B) 750°C for 60 min, (C) 800°C for 30 min, (D) 850°C for 30 min. As shown in Fig. 3(a), all films showed typically soft

Table I. Magnetic Data of YIG Films in Different Annealing Conditions

		Magnetic performance	e
Annealing conditions	Hc (Oe)	Ms (emu/cm ³)	4πMs(G)
750°C for 30 min 750°C for 60 min 800°C for 30 min 850°C for 30 min	14 13 12 12	103 110 120 124	1293 1382 1507 1552

(a) (b) **Derivative of FMR absorption ∆H=97.2Oe Derivative of FMR absorption** -o- Data Lorentzian fit 9.10GHz 750°C for 30mins 1600 2000 1600 2000 2400 2800 3200 3600 4000 Magnetic field(Oe) (c) (d) **∆H=105.5Oe** Derivative of FMR absorption **Derivative of FMR absorption** –o– Data Lorentzian fit 9.10GHz 800°C for 30mins 2800 1600 2000 2400 3200 3600 4000 1600 2000

ferromagnetism with narrow loops. The dependence of saturation magnetization (Ms) and coercive field (Hc) on annealing conditions are shown in Table I. With increasing annealing temperature and time, Ms was increased for better crystallization. In 850°C for 30 min, Ms was 124 emu/cm³, resulting a magnetic induction $4\pi Ms$ of 1.55 kG, which can the 88% value of single-crystal reach YIG $(4\pi Ms = 1.75 \text{ kG}^8)$. The little decrease in saturation magnetization maybe due to the interface between YIG and substrate, which would reduce the effective density of YIG films. Furthermore, the strain-induced anisotropy in poly-The magcrystalline YIG films could lower magnetization.^I netic value we got was close to the (111) orientation YIG films prepared by PLD on GGG with Hc = 3.8 Oe and 4π Ms = 1.58 kG.²⁰ The average coercive field of YIG films was about 13 Oe, which was much smaller than YIG films deposited on Si, Si/CeO₂, or porous SiO₂ substrates.^{14,15,19,21}

The frequency dependence of YIG films permittivity are shown in Fig. 3(b). The dielectric constants and losses were almost independent of frequency, showing stable values in a broad frequency from 10 kHz to 20 MHz. As shown, the dielectric constants were kept at 6-8 and the dielectric loss of tand can be as low as 0.008@20 MHz. Though the test frequency was much lower than microwave range, the results were still significant because it showed the better stability and lower losses of YIG films, which could be the proof of CSD technique feasibility to prepare YIG films on Si.



Fig. 4. FMR curves at 9.10 GHz of YIG films in: (a) 750°C for 30 min, (b) 750°C for 60 min, (c) 800°C for 30 min, (d) 850°C for 30 min.

Table II. Comparison of YIG Films Grown on Pt/SiO₂/Si in this Work with Those Reported in Literatures

Samples	YIG/Pt/SiO ₂ /Si(100)	YIG/SiO ₂ /Si(100) ¹³	YIG/Sapphire ¹⁵	YIG/Quartz ¹⁹	YIG/Porous Silicon ²⁵
Method	CSD (Chemical solution deposition)	Sputtering	Aerosol deposition	PLD	PLD
Annealing temperature	750°C	900°C	1280°C	750°C	650°C
Coercive field (Oe)	13	16	6	25	30
Peak-to-peak FMR width (Oe)	95	150	198	200	96

In Fig. 4, FMR curves and linewidth ΔH_{FMR} were shown in different annealing conditions. All curves showed a good Lorentzian fit (red line). A second peak appeared besides the main resonance absorption, which was also reported in others' work.^{13,21,22} According to G. Woltersdorf and Vladimir L. Safonov's theory, 23,24 this behavior can be attributed to defects in films. The confluence process can occur in the presence of defects, impurities, or fluctuations which can induce an additional absorption. The measured linewidth versus frequency is often interpreted with a simple relationship:²

$$\Delta H_{\rm FMR(f)} = \Delta H(0) + \alpha \frac{2\pi f}{\gamma} \tag{1}$$

where f is the radio-frequency, α is the Gilbert damping constants ($\alpha = (10.3 \pm 0.5) \times 10^{-4}$), γ (=2.8 MHz/Oe) is the gyromagnetic ratio, and $\Delta H(0)$ is the linewidth at zero frequency. ΔH (0) is a long-range inhomogeneity-caused line broadening, which is extrinsic and depends on the film quality. The narrower ΔH , the better is the crystal quality and lower losses. The lowest FMR width was obtained with $\Delta H = 95.2$ Oe at 9.10 GHz when annealed at 750°C for 60 min in air, which was smaller than films on other nongar-net substrates.^{13,15,19,25} The comparisons of specific values are shown in Table II. We can see clearly that YIG films in this work showed a smaller coercive field (Hc = 13 Oe) and lower resonance width ($\Delta H = 95.2$ Oe) with 750°C annealing. The data suggested that YIG prepared on Pt-buffered Si/ SiO₂ substrate by CSD can show an improved performance including the surface quality and magnetic property with a relative lower annealing temperature. Our results proved that Pt can successfully buffer the lattice structure mismatch and large thermal expansion difference between YIG and Si as predicted, improving the performance of YIG films with less cracks and defects.

IV. Conclusions

In summary, this work showed high-quality YIG films growth on Pt-buffered Si substrates. Films with crack-free and improved magnetic performance were obtained, showing a small coercive field (Hc = 13 Oe) and low FMR width $(\Delta H = 95 \text{ Oe})$. Furthermore, we proved the feasibility of CSD technique for YIG films on Si substrates with a relative low annealing temperature. This is an important proof that high-quality YIG films can be fabricated on Si-based substrate when introducing Pt as a buffer layer. It would be a promising way for better application of YIG films to silicon integrated devices with lower loss.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (Nos. 61371059 and 51302295), Shanghai Natural Science Foundation (No. 13ZR1445600), the Visiting Scholar Foundation of Key Lab. of Micronano Devices and System Technology in Chongqing University, and international partnership project of Chinese Academy of Science.

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