Polycrystalline $Y_3Fe_5O_{12}$ Garnet Films Grown by a Pulsed Laser Ablation Technique

C. J. Yang and S. W. Kim
Electromagnetic Materials Laboratory, Research Institute of Industrial Science & Technology(RIST), 790-600 Pohang, Korea

(Received 12 June 1994, in final form 20 September 1994)

Abstract

$Y_3Fe_5O_{12}$ based garnet films (thin or thick) offer a great promise for the application of microwave communication components. We investigated the magnetic and crystallographic preperties of $Y_3Fe_5O_{12}$ thick films prepared by KrF eximer laser ablation of a stoichiometric garnet target. It was possible to obtain almost epitaxially oriented films on Al_2O_3 $(1\bar{1}02)$ plane. Although the crystalline quality depends on substrate temperature and O_2 partial pressure used(Po_2), 4.1 m thick films of $4\pi M_s = 1300$ Gauss and $H_c = 37.5$ Oe were obtained at the substrate temperature of 700℃ with the Po_2 of 100 mTorr after annealing the as-deposited films at 700℃ for 2 hours. These films are expected to be used for magnetostatic spin wave filters at narrow bandwidth frequency.

I. Introduction

The pulsed laser ablation technique is emerging as a viable method for preparing high quality films of magnetic ferrites as well as ferroelectric materials[1~7]. It has been already demonstrated that laser ablation has great advantages in preparing multicomponent oxide superconducting thin films [8~9]. The laser ablation technique is known to provide the most promising characteristics of the ferroelectric thin films due to the following reasons: Deposition in high oxygen pressure is possible because the energy source is outside of the system, and there is little difference in the composition between the target material and the deposited film. Although this technique appears to be promising based upon the earlier results[1~9], there are still obstacles to overcome before the magnetic ferrite such as garnet(YIG) based microwave films deposited by the above method become applicable.

Recently, the YIG based microwave ferrite films attract a great attention from the viewpoint of magnetic properties and applications for mobile communication components such as magnetostatic spin wave(MSW) filters or delay lines. For use in microwave components, the YIG films have been epitaxially grown on gadolinium gallium garnet (GGG) substrate by the liquid phase epitaxy (LPE) technique[12~13] or by the reactive ion beam sputtering(RIBS)[14]. Although the LPE technique provides a high quality films of YIG, both the LPE and RIBS processes have common disadvantages such as impurity admixture, low deposition rate which are very important factors determining the possibility of commercial applications.

In this respect, we studied the magnetic and structural characteristics of polycrystalline YIG films prepared by a laser ablation technique on various Al_2O_3 and MgO single crystal substrates. And we report the results of optimized conditions in terms of process parameters searching for economical applications.

II. Experimental

$Y_3Fe_5O_{12}$ thin and thick films were prepared by the pulsed laser ablation using a KrF excimer laser ($\lambda = 248$ nm) with a 20 ns pulse width at 10 Hz pulse rate in a vacuum chamber starting with a base pressure under 5x10^{-6} Torr. The laser beam was focussed to an energy density of 2~3 J/cm^2 onto a polycrystalline YIG target which was rotated during deposition. The $Y_3Fe_5O_{12}$ films were deposited onto 1cmx1cm Al_2O_3 single crystal substrates of (0001), (1$\bar{1}$02) planes and MgO (100) plane as well. The substrate holder was a resistive heater positioned 5 cm away from the target, and was rotated during laser deposition. Films were grown at 600~800℃ under oxygen partial pressure ranging 50~800 mTorr.

Structural characterization of the YIG films was made by using X-ray diffraction from CuKα source. Scanning electron microscopy was used to determine the film morphology, and the composition was identified by inductively coupled plasma spectroscopy. The ferromagnetic resonance(FMR) measurement was carried out using a conventional Varian E-line spectromenter at 9.5 GHz frequency. Magnetic measurements were made using a vibrating sample magnetometer.

Ⅲ. Results and Discussion

By matching the X-ray diffraction spectrums of the YIG films with the standard $Y_3Fe_5O_{12}$ polycrystal powders, it was confirmed that all the films were single phase $Y_3Fe_5O_{12}$ regardless of the orientation of the substrate used. However, a perfect crystallization of the $Y_3Fe_5O_{12}$ stoichiometry was only obtained after annealing the films at 700℃ for 2 hours in air. Fig. 1 shows the X-ray diffraction patterns comparing the crystallization of the as-deposited and annealed $Y_3Fe_5O_{12}$ films on MgO(100), Al_2O_3(0001) and Al_2O_3(1$\bar{1}$02) planes at 600℃ under a O_2 partial pressure of 100 mTorr, respectively. The spectrum of dotted line denotes the as-5deposited films and the solid line is of annealed films. The patterns of Fig. 1 was a typical examples obtained from the $Y_3Fe_5O_{12}$ films deposited at 600℃ for 1.5 hours to have 4~6 m thickness. When the substrate temperature of 700℃ is employed, the deposition aspect of the films was not changed before and after the annealing comparing with that of 600℃. However, the deposition rate decreased to 300~500 Å/min which was 400~600 Å/min when the substrate temperature was 600℃. Fig. 2 shows the aspect of $Y_3Fe_5O_{12}$ films deposited at 600℃ and 700℃, respectively. As a lower substrate temperature(600℃) was used, $Y_3Fe_5O_{12}$ crystals of coarse texture can be seen in Fig. 2(b). The film deposited at 700℃ in Fig. 2(a) shows a finer texture which definately result in a smooth surface. The smooth surface of the YIG films is essential for employing those films in the microwave filters.

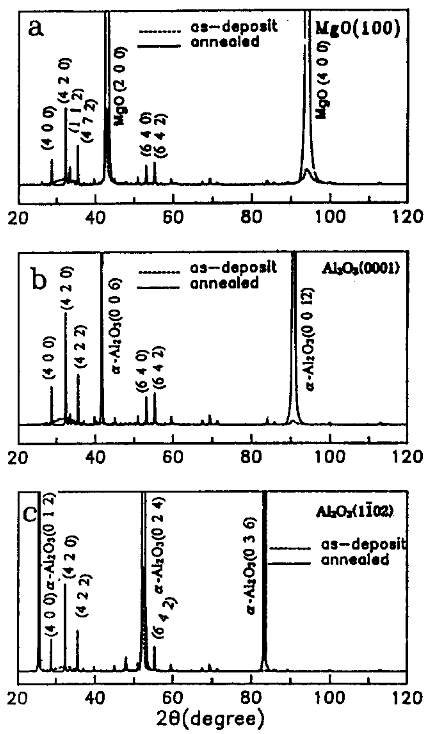

Fig. 1. X-ray diffraction patterns comparing the as-deposited YIG films on (a) MgO(100), (b) Al_2O_3 (0001) and (c) Al_2O_3(1$\bar{1}$02) planes, respectively.

The influence of O_2 partial pressure on YIG films was found to be very important. Fig. 3(a)~(d) shows the effect of O_2 partial pressure on the epitaxial condition of the $Y_3Fe_5O_{12}$ films and its surface aspect as well. Fig. 3(a) and (b) is as-deposited film on $Al_2O_3(0001)$ and $Al_2O_3(1\bar{1}02)$ planes at 700℃, respectively, under O_2 partial pressure as shown in figures. The X-ray specturms (a) and (b) of $Y_3Fe_5O_{12}$ films of as-deposited at 700℃ for 1.5 hours indicate that the near epitaxial crystallites can be formed under a lower O_2 partial pressure(100 mTorr) rather than under a higher pressure. It was subsequently found that the $Y_3Fe_5O_{12}$ films deposited under O_2 partial pressure of 100 mTorr shows a smooth surface in Fig. 3(c), while the films under O_2 partial presure of 800 mTorr form a rough surface in Fig. 3(d). Consequently, $Y_3Fe_5O_{12}$ films deposited at a high substrate temperature(700℃) under a low O_2 partial pressure(100 mTorr) exhibited a qualified surface considering that the smooth surface of garnet film allows the microwave propagation without a scattering of signal. The typical magnetic and microwave properties of the $Y_3Fe_5O_{12}$ films are summerized in Table I. The magnetic properties were measured along the in-plane direction of each sample.

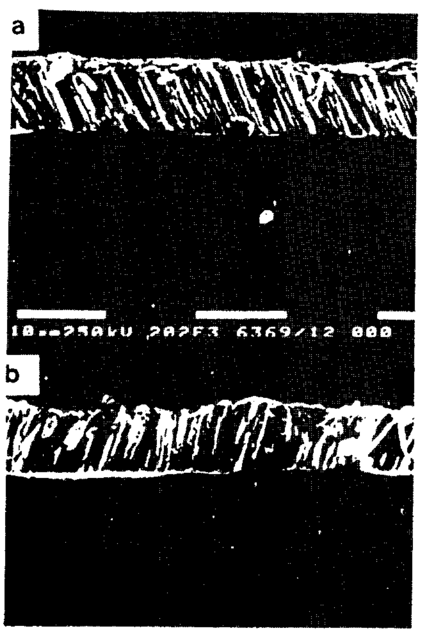

Fig. 2. SEM micrographs showing the aspect of YIG films deposited at (a) 600℃ and (b) 700℃ on $Al_2O_3(1\bar{1}02)$ planes under a Po_2 of 100 mTorr

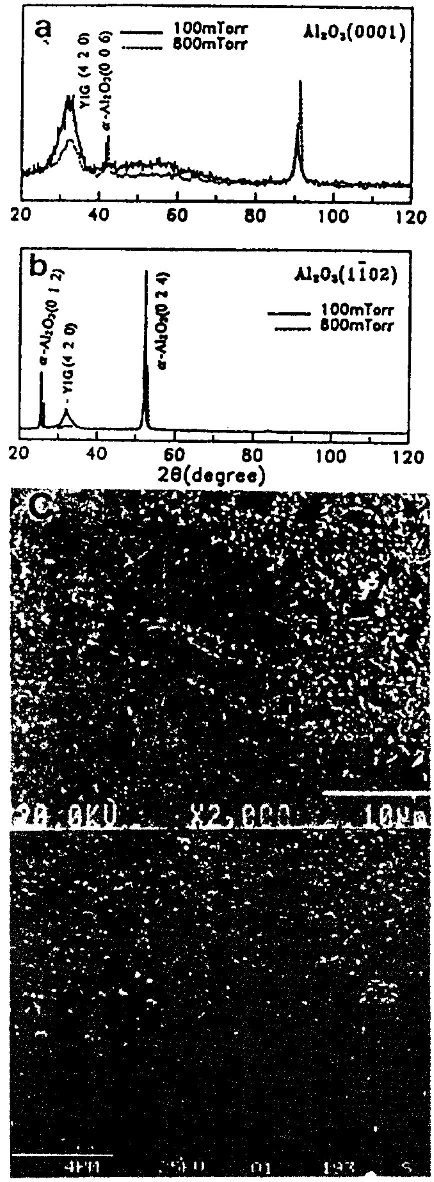

Fig. 3. X-ray diffraction patterns comparing the effect of O_2 partial pressure on YIG films on (a) $Al_2O_3(0001)$ and (b) $Al_2O_3(1\bar{1}02)$ and (c) is a SEM micrograph showing the surface aspect of the film on (a), and (d) is of (b) plane.

Taking into account not only the X-ray diffraction patterns in Fig. 3(a)~(b) but also the results of surface roughness in Fig. 3(c)~(d) as well as the magnetic measurements, it is concluded that the optimized substrate for deposition of $Y_3Fe_5O_{12}$ films is of $Al_2O_3(1\bar{1}02)$ plane. And the best films can be obtained at 700℃ under the O_2 partial pressure of 100 mTorr followed by annealing at 600 ℃ in air. Magnetic losses of microwave devices are caused largely by the ferrimagnetic resonance. If the ferrimagnetic resonance line width is wider, the resonance becomes broader and resonance losses can manifest themselves in the operating frequency range of the microwave devices such as circulator, isolators or filters. The FMR line width measured from $Y_3Fe_5O_{12}$ films in Table I seems to be quite narrow compared with those of standard single crystal $Y_3Fe_5O_{12}$ having $\Delta H=45$ Oe[15]. The YIG films deposited on $Al_2O_3(1\bar{1}02)$ plane exhibit quite small value($\Delta H=20$ Oe), which indicates that the $Y_3Fe_5O_{12}$ films deposited on $Al_2O_3(1\bar{1}02)$ plane have textured structure approcaching epitaxy. The effective saturation magnetization($4\pi M_{eff}$) in Table I is very important. Because the saturation magnetization of the microwave devices should be chosen such that the YIG film is fully saturated by externally applied magnetic field in the device. To meet the resonant condition, one need[15]:

$$\frac{\omega}{\gamma} > 4\pi M_s + H_a$$

where ω is the resonant frequency, γ is the gyromagnetic ratio an H_a is the anisotropy field of YIG films, respectively. In either perpendicular or plane direction of the YIG film, the relationship of $4\pi M_s = 4\pi M_{eff} + H_a$ can be used. Therefore the following equation is made:

$$\frac{\omega}{\gamma} > 4\pi M_{eff} + 2H_a$$

Since the $4\pi M_s$ for YIG in the literature is 1750 Gauss[15], the Ha of $Y_3Fe_5O_{12}$ films obtained in this sutdy is calculated as $H_a=225$ Oe.

In summary, the polycrystalline $Y_3Fe_5O_{12}$ films deposited on $Al_2O_3(1\bar{1}02)$ plane at 700℃ under O_2 partial pressure of 100 mTorr exhibit a textured crystal structure. The YIG films showed the $4\pi M_{eff}=1300$ Gauss and $H_a=225$ Oe. This medium value of $4\pi M_{eff}$ is in working limit for a narrow bandwidth frequency.

Table I. Magnetic properties of $Y_3Fe_5O_{12}$ films deposited on various substrates

deposition condition (℃/O_2 pressure)	substrate	film thichness (μm)	magnetic properties $4\pi M_{eff}$ (Gauss)	$4\pi M_r$ (Gauss)	H_c (Oe)	Resonance Linewidth (ΔH)
700℃	MgO(100)	4.7	780	600	41.0	50
/100mTorr	$Al_2O_3(0001)$	6.2	900	600	38.0	35
(annealed)	$Al_2O_3(1102)$	4.1	1300	700	37.5	20

References

[1] R. Nawathey, R. D. Vispute, S. M. Chaudhari and S. B. Ogate, Solid state Commun. 71, 9(1989) .
[2] G. M. Davis and M. C. Gowe, Appl. Phys. Lett. 55, 112(1989).
[3] R. Ramesh, K. Luther, B. Wilkens and J. M. Tarascon, Appl. Phys. Lett. 57, 1505(1990).
[4] S. Otsubo, T. Maeda, T. Minamikawa and T. Shinizu, Jpn. J. Appl. Phys. 29, L133(1989).
[5] A. Morimoto, S. Otsubo, T. Shimizu and T. Ogawa, Mater. Res. Soc. Symp. Proc. 191, 31 (1990).
[6] H. Kidoh, A. Morimoto and T. Shinizu, Appl. Phys. Lett. 59(2), 237(1991).
[7] P. C. Dorsey, S. E. Bushnell, R. G. Seed and C. Vittoria, J. Appl. Phys. 74(2), 1242(1993).
[8] S. Otsubo, T. Minamikawa, Y. Yonezawa, T. Maeda and T. Shinizu, J. Appl. Phys. 27, L1999(1988).
[9] S. Otsubo, T. Minakawa, Y. Yonezawa and T. Shimiza, Jpn. J. Appl. Phys. 28, 2211 (1989).
[10] H. L. Glass, Proc. IEEE. 76(2), 151(1988).
[11] W. S. Ishak, Proc. IEEE. 76(2), 171(1988).
[12] R. C. Linares, R. B. McGraw and J. B.

Schroeder, J. Appl. Phys. 36, 2884(1965).
[13] C. Vittoria, P. Lubitz, P. Hansen and W. Tolksdorf, J. Appl. Phys. 57, 3699(1985).
[14] T. Okuda, N. Koshizuka, K. Hayashi and H. Yamamata, IEEE Trans. Magn. 23, 3491 (1987).
[15] D. K. Linkhart, Microwave Circulator Design (ArTech House, Inc., Norwood, MA, 1989) pp. 67 Captions of figure.

엑시머 레이저 증착기술에 의한 $Y_3Fe_5O_{12}$ 다결정 박막 제조

김상원 · 양충진

산업과학기술연구소, 전자소재연구실, P. O. Box 135, 790-600 Pohang

(1994년 6월 12일 받음, 1994년 9월 20일 최종수정본 받음)

페리마그넷(Ferrimagnetic) $Y_3Fe_5O_{12}$(Garnet) 박막 또는 후막은 초고주파 대역에서 사용하는 통신부품의 소자로서 핵심 역할을 하고 있다. 본 연구에서는 요즘 신기술로 소개된 펄스 레이저 증착기술(Laser Ablation Technique)에 의하여 가넷의 표준조성인 $Y_3Fe_5O_{12}$ 후막을 에피성장 시키는데 성공하였다. KrF 가스를 사용한 Eximer 레이저를 10 Hz의 펄스주파수로 $Al_2O_3(1\bar{1}02)$ 면에서 거의 집합조직의 에피후막을 성장시켰다. 후막의 자기특성 및 성장 양상은 사용한 기판 및 기판온도와 산소분압에 따라 결정되지만 본 연구에서 얻어진 최적의 자기특성은 가넷두께 4.1 μm에서 $4\pi M_s$=1300 Gauss, H_c=37.5 Oe 의 값을 산소분압 100 mTorr 및 기판온도 600℃에서 증착한 후 700℃에서 2시간 소둔처리하여 최적값을 얻을 수가 있었다. 이러한 가넷후막은 협대역 주파수 범위에서 Magnetostatic Spin Wave 원리를 이용한 Filter로 사용 가능하다.