Magnetic Properties of Heteroepitaxial $Y_3Fe_5O_{12}$ Films Grown by a Pulsed Laser Ablation Technique

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Yttrium Iron Garnet($Y_3Fe_5O_{12}$) films have been succsssfully grown on(111)GGG wafer by KrF excimer laser ablation of stoichiometric garnet target at the oxygen partial pressure, $P(O_2)$, ranging 20 to 500 mTorr. During the deposition of the films the substrate temperature was maintained at 700 ℃ and the laser beam energy density at 7.75 J /cm^2. Microstructure, composition and magnetic properties of the films obtained were investigated as a function of oxygen pressure and thickness of the films. Epitaxial films with a dense and a smooth surface were reproducible at a low oxygen pressure. The films of 2.75 μm in thickness deposited at 20 mTorr of $P(O_2)$ showed $4\pi M_s$ of 1500 Gauss and H_c of 3 Oe after annealing at 800 ℃ for 20 minutes. As-deposited films of 0.8 μm in thickness exhibited the $4\pi M_s$ of 1730 Gauss and H_c of 7 Oe. The magnetic properties of the films obtained were almost identical to those of a single crystal YIG.

I. Introduction

Ever since the laser ablation technique has been successfully utilized in making thin films of good quality superconductors[1, 2], the technique has been extended in preparing ferrites as well as ferroelectric thin films(3~8). It is known that the laser ablation technique renders special advantages on depositing ferroelectric thin-films : The external energy source allows to deposit such oxide targets under high oxygen pressure without altering its film composition[1~9]. Since it has been attemped to deposit garnet(YIG) films, it attracted great interests to demonstrate its application in the MSW devices. In recent years special interest of microwave devices has risen in telecommunication system such as YIG film for magnetostatic wave filter of delay lines. It is also known that YIG single crystal shows the lowest insertion loss which is generally expressed by ferromagnetic resonance linewidth(△H). YIG single crystal shows the resonance linewidth of 1 Oe which would be a milestone value for the applications of YIG films. Thus many attempts have been made to prepare YIG epitaxial films on GGG substrate by LPE technique[12, 13]. The present study was attempted to demonstrate feasibility of producing a high quality YIG films by controlling oxygen partial pressure and lasing energy density utilizing a KrF excimer laser.

II. Experimental

Stoichiometric composition($Y_3Fe_5O_{12}$) of polycrystalline materials with 99.9 % purity(MetVac Technology) was used for target and a GGG single crystal of (111) substrate was selected to achieve epitaxial YIG films. The deposition was carried out using KrF excimer laser($\lambda = 248$ nm) with a 20 nsec pulse duration and at 10 Hz repitition rate in a vacuum chamber starting with a basis pressure at 4×10^{-6} Torr. The laser beam was focussed to an energy density of 7.75 J /cm^2 onto the target which was rotated at 3 rpm during deposition. Substrate temperature was controlled between 600 ~ 800 ℃ during deposition and cooling rate of 3 ℃ /min down to 300 ℃ after deposition was controlled by programming.

Film thickness was measured by an "Alpha step" (Tencor Instrument), topography was checked by

SEM and TEM, crystal structure was indentified by XRD, and chemical composition was analyzed by EDS technique. Magnetic properties were characterized by a vibrating sample magnetometer(VSM). Fig. 1 illustrates the experimental set-up for film deopsition.

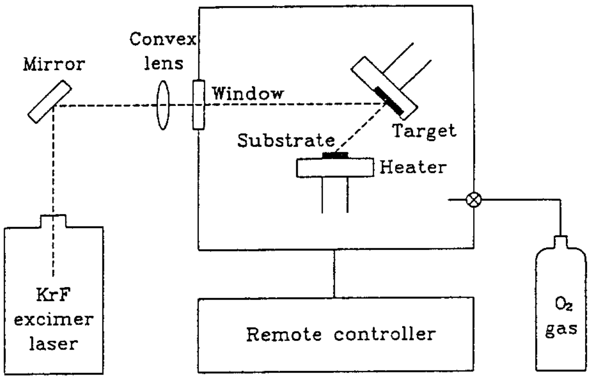

Fig. 1. Experimental set up of KrF excimer laser deposition set-up used in the present study.

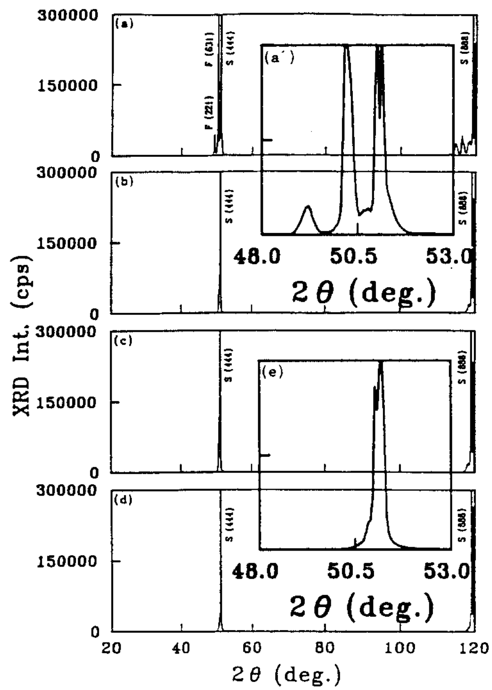

Fig. 2. X-ray diffraction patterns of the YIG films on (111)GGG substrate deposited at 700 ℃ with 7.75 J/cm^2 of laser energy density at oxygen pressure of (a) 20 mTorr, (a′) magnifeid peak of (a), (b) 100 mTorr, (c) 300 mTorr, (d) 500 mTorr, and (e) corresponding (444) peak from GGG substrate, respectively.

Ⅲ. Results and discussion

1. The effect of oxygen partial pressure on the microstructure, composition and magnetic properties of YIG films

X-ray diffraction patterns of the films deposited on a (111) GGG under different oxygen pressures revealed a strong partial pressure dependency of crystal orientation as shown in Fig. 2. In this study as-deposited films at P(O_2) of 20 mTorr exhibited a textured structure of (631) orientation on (444) GGG plane. Epitaxial films of YIG were reported to be obtained only after annealing the as-deposited films at 700~800 ℃[15, 17]. Fig. 2(a′) shows the magnified peaks of (631)YIG and corresponding (444)GGG substrate. At the same time (e) shows the corresponding (444)GGG peak which clearly indicates the heteroepitaxial films of (631)YIG on (444)GGG. The splitted peak from (444)GGG is due to the presence of twins in GGG single crystal. Above P(O_2) of 20 mTorr all the as-deposited films revealed the presence of (420), (332), (422), (840), (221) and/or (640) although it is not clearly shown in Fig. 2. But the magnified X-ray diffraction pattern obtained from a small scale chart showed the peaks mentioned above. This fact was also true for the films after annealing. It is an indication of poor crystal growth or presence of amorphous phase which will be discussed in the latter section. Fig. 3 is SEM topography showing the film surface and cross sectional view of films prepared at P(O_2) of 20 mTorr and 500 mTorr, respectively. The influence of oxygen partial pressure on the microstructure is generally known that depositing with increasing oxygen pressure causes ejection and splash of submicron particles from porous target body due to the expansion of oxygen in the pores[8, 9]. However, in the present study such particles were not evidenced on the surface of

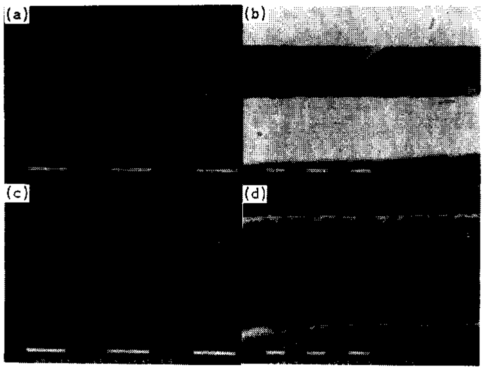

Fig. 3. SEM topographs showing (a) film surface, and (b) cross sectional view of the films deposited at oxygen pressure of 20 mTorr, and (c) surface, and (d) cross section of the films deposited at oxygen pressure of 500 mTorr, respectively.

films prepared even at 500 mTorr. On the other hand with increasing $P(O_2)$ from 20 to 500 mTorr, the cross section of films shows a columnar structure of clusterd crystals inducing rough surface. It is suggested that as increasing oxygen pressure, the oxygen molecules or radical near the film surface cause an active gas scattering which leads the ablated particles to lose their high energy, surface migration may become slow down. Eventually it results in non-uniform structure.

The ferromagnetic resonance linewidth($\triangle H$), a milestone of insertion loss of microwave characteristics, is expressed below[17] :

$$\triangle H = \triangle H_{single} + \left| \frac{K_1}{M_s} \right| + 1.5\left(\frac{M_s}{\mu}\right) P$$

where P is fraction of porosity, $\triangle H_{single}$ is the linewidth of single crystal YIG, K_1 is anisotropy constant, M_s is the intrinsic saturated magnetization, and μ is permeability of the corresponding composition. Magnetic ferrite films for microwave application require low $\triangle H$ which is dependent on both crystal anisotropy and porosity as shown above. Accordingly microstructure-dependent properties such as a low anisotropy, $Ha(=K_1/M_s)$, and a dense structure are essential when μ is fixed. In this respect the films prepared at $P(O_2)$ of 20 mTorr in the present study was found to be suitable.

Fig. 4 shows chemical analysis results of the de-

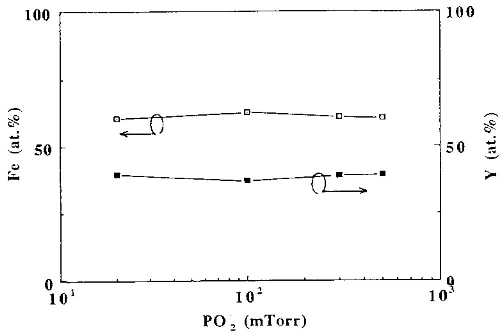

Fig. 4. Oxygen partial pressure dependency of composition of the YIG films on (111)GGG substrate deposited with 7.75 J /cm^2 laser energy density at 700 ℃ substrate temperature.

posited YIG films in atomic percent of Y and Fe. It is encouraging to note that Y and Fe contents show almost invariant as a function of oxygen pressure, however, with approximately ± 2 % variation.

In microwave region it is desirable for the magnetic ferrite films to have a high saturation moment($4\pi M_s$) in order to increase resonance frequency and high attenuation of signals. Also a low anisotropy field(H_a) is required for a small $\triangle H$ as shown in the equation above. Fig. 5 shows hysteresis curves measured along in-plane direction of the films prepared at $P(O_2)$ of 20, 100, 300 and 500 mTorr, respectively. The substrate temperature used was 700 ℃. As the $P(O_2)$ increases squareness of the hysteresis loops appeared to be poor with a decreased $4\pi M_s$, while H_c increases. Of the hysteresis loops the films deposited in 20 mTorr showed a loop with good squareness to have a high $4\pi M_s$ and a low H_c as well. The measurements were made applying the external magnetic field of 200 Oe. Fig. 6 shows the variation of both $4\pi M_s$ (plot

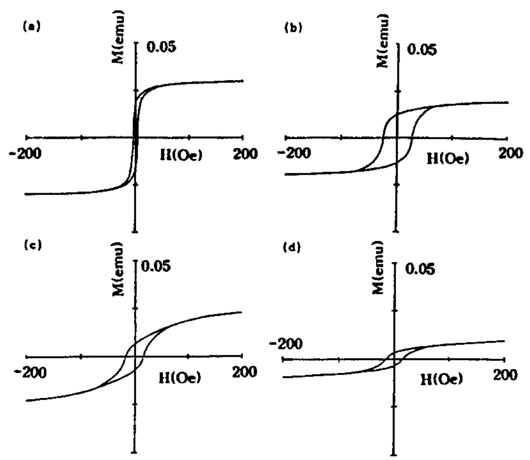

Fig. 5. Hysteresis curves of the YIG films on (111) GGG substrate deposited with 7.75 J/cm^2 laser energy density under oxygen pressure of (a) 20 mTorr, (b) 100 mTorr, (c) 300 mTorr, and (d) 500 mTorr, respectively. The curves were measured along the in-plane direction.

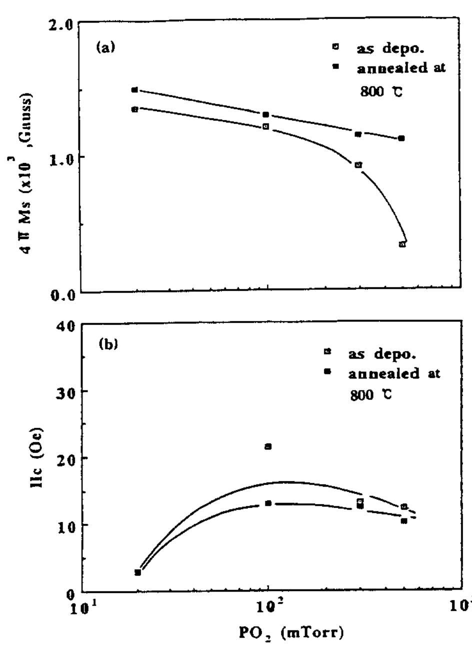

Fig. 6. Oxygen pressure dependency of (a)saturation magnetization($4\pi M_s$) and (b) coercivity(H_c) of the YIG films.

a) and H_c(plot b) of the films prepared at $P(O_2)$ of 20, 100, 300 and 500 mTorr. The comparison was made before and after annealing the films at 800 ℃ for 20 minutes. It is shown that magnetic saturation of the films has been improved after annealing. The magnetic saturation varies in a linear trend such that $4\pi M_s$ decreases from 1500 to 1100 Gauss as $P(O_2)$ increases from 20 to 500 mTorr, while at higher $P(O_2)$ the as-deposited films decrease sharply from 1350 to 330 Gauss. The decreasing trend of $4\pi M_s$ with increasing in $P(O_2)$ is possibly attributed to the fact that excess oxygen gas scattering causes a rapid cooling effect on the formation of ablated garnet particles on the substrate. Such a reaction forms a off-stoichiomertic composition in lack of oxygen of the film material that reduces $4\pi M_s$ value. During annealing treatment, however, access oxygen could be supplied to approach stoichiometric composition which resulted in an enhanced $4\pi M_s$. Regarding the trend of H_c it shows an increasing trend up to 100 mTorr oxygen partial pressure and beyond it slightly decreases. It seems that beyond 100 mTorr a little effect on H_c, while the effect is considerable on $4\pi M_s$. It is interesting to note that H_c values were measured to be low after annealing. However, H_c of the films deposited at $P(O_2)$ of 20 mTorr was 3 Oe, which is still higher than that of single crystal YIG(1 Oe).

2. Thickness dependency of YIG film structure and magnetic properties

Magnetic thin film application in microwave filter is known to require its minimum thickness of several μm[16]. To observe the effect of film thickness on its structure and magnetic properties, thin films at $P(O_2)$ of 20 mTorr, in which the best quality was obtained, were fabricated with a lasing energy of 7.75 J/cm^2 in a range of its thickness from 0.8 to 2.75 μm. X-ray diffraction analysis of the films in Fig. 7 revealed that the films with its thickness of 1.53, 1.92 and 2.7 μm indicate two strong peaks, (221)/(631) for YIG phase and (444)GGG substrate. It is evidenced that (631)

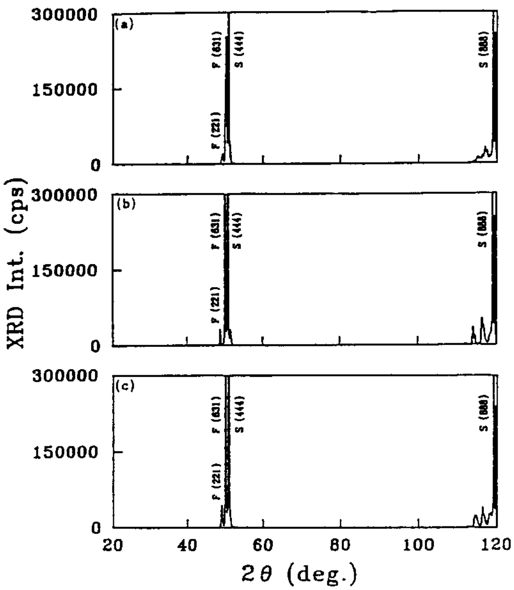

Fig. 7 X-ray diffraction patterns obtained from the films under oxygen pressure of 20 mTorr at 700 ℃ substrate temperature. (a) for the films of 1.53 μm thick, (b) for 1.92 μm, and (c) for 2.7 μm thick.

of YIG phase has preferentially increased as decrease in its thickness. This certainly suggest that an epitaxial film has been successfully grown. The lattice mismatch between (444)GGG and (631)YIG film was calculated to be only 5.42 %. The effect of thickness on magnetic properties was also observed as shown in Fig. 8. The magnetic properties were characterized on the basis of hysteresis curves shown in Fig. 5, In Fig. 6 the squareness (figure of merit) of the curves has been improved as increase in the film thickness. Both $4\pi M_s$ and H_c were measured to decrease with increasing the film thickness. As compared with $4\pi M_s$ for bulk YIG (1750 Gauss), that of the thin film(0.8 μm) reached 1730 Gauss and H_c for bulk single crystal YIG(1 Oe) compared with 2.5 7 Oe for film YIG, the possibility for an use of microwave devices may be a compromse between $4\pi M_s$ and H_c. It is speculated that the thinner films at 700 ℃ substrate can be annealed in the chamber during deposition, and result in magnetic quality as good as that of the intentionally annealed films. H_c in Fig. 8 increases monotonically with decreasing the thickness. Since the preferred orientation of (631) diminishes as the film thickness increases, the coercivity measured along in-plane direction showed decreasing values as increase in the film thickness. Besides, the magnetostatic energy acting through the film thickness may also induce an increased coercivity along the plane direction[19].

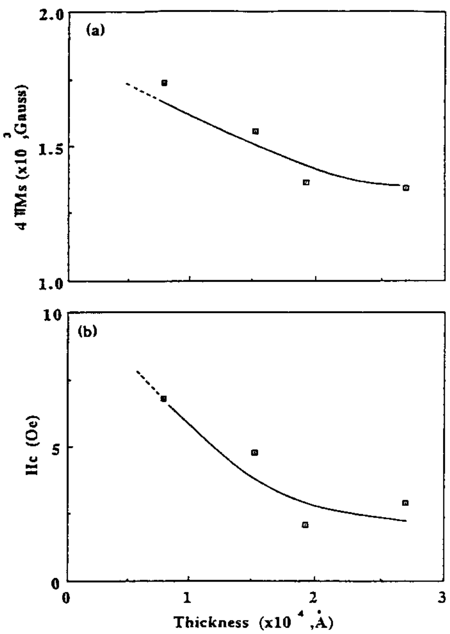

Fig. 8 Variation of magnetic properties as a function of the films deposited at the substrate temperature of 700 ℃ under oxygen pressure of 20 mTorr.

Ⅳ. Conclusion

Heteroepitaxial YIG thin films on (111)GGG substrate have been successfully deposited using KrF excimer laser ablation technique. Appropriate depositing conditions were determined by controlling laser energy density, oxygen partial pressure and substrate temperature. The deposition were performed at 7.75 J /cm^2 of lasing energy density, 700 ℃ of substrate temperature, in a range of oxygen partial pressure from 20 to 500 mTorr. This deposition technique has produced a potential quality of YIG films with uniform stoichiometry and little variation as a function of oxygen pressure. The effect of oxygen pressure and the film thickness on magnetic properties was observed. It was found that annealing the as-deposited films at 800 ℃ for 20 minutes enchanced $4\pi M_s$ further. The films with 0.8 μm of thickness, which were deposited at 20 mTorr, showed the best magnetic properties.

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펄스 레이저 증착기술에 의한 $Y_3Fe_5O_{12}$ 에피택셜 박막제조

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YIG 단결정 소재가 주로 circulator, isolator, filter 및 resonator 등의 고주파 협대역 이동통신용 소자에 사용되고 있으나, 최근 소형/경량화에 따른 YIG 박막의 소요가 예상되면서 에피택셜 박막성장 기술이 많이 연구되고 있다. 본 연구에서 KrF excimer laser를 사용하고, 산소분압을 20~500 mTorr로 조절하면서 박막을 제조하여 산소분압과 박막두께가 박막의 조직, 조성 및 자기특성에 미치는 영향을 조사하였다. 기판으로는 GGG(111)을 사용하였고 700 ℃에서 증착을 실시하였다. 사용한 laser beam의 에너지 밀도는 7.75 J/cm^2였다. 20 mTorr 산소분압 하에서 증착한 박막은 항상 에피택셜 성장을 보이고, 2.75 μm 두께의 시편은 800 ℃/20분 열처리 후에 $4\pi M_s$ 가 1500 Gauss, H_c는 3 Oe의 값을 보였고, 특히 0.8 μm 두께의 시편은 열처리 없이 $4\pi M_s$ 값이 1730 Gauss, H_s는 7 Oe의 우수한 특성을 보여, YIG 단결정의 자기특성에 육박하는 값을 보였다.