Journal of Magnetics 2(2), 38-41(1997)

Magnetic Properties of $Fe_{3-x}Mn_xO_4$ Thin Films by FMR

Ki Hyeon Kim*, Young Ho Kim***,**
****Dept. of physics, Myongji University, Yongin 449-728, Korea*

Tae Wook Ha, Jeong Sik Lee**,**
***Dept. of physics, Kyungsung University, Pusan 608-736, Korea*

Mann Jang Park*
**Dept. of physics, Physics, Korea University, Seoul, 136-701, Korea*

(Received 13 May 1997)

Spinel ferrite thin films $Fe_{3-x}Mn_xO_4$(x=0.000, 0.006, 0.010, 0.015, 0.023) were prepared on the coverglass by ferrite plating technique. To investigate the uniaxial anisotrpy of the samples, the saturation and effective magnetization of the thin films were measured by VSM(vibrating sample magnetometer) and FMR(ferromagnetic resonance) measurements respectively. The spectroscopic splitting g factor were estimated from the ferromagnetic resonance curves. For x=0.000, 0.006, the effective magnetization was measured of temperatures form T=77 K to T=300 K. The results were analyzed in terms of Bloch's law $M_s(T) = M_s(0)\ (\ 1-BT^{3/2}-CT^{5/2}\ )$. The Bloch coefficient B, C were determined by fitting. $M_s(0)$ was obtained by extrapolating M_{eff} to 0 K. From this result, the spinwave stiffness constants D was also determined.

I. Introduction

Most of the polycrystalline ferrite films have been used in magnetic recording materials[1] and microwave device materials. In particular, when thin films are used in microwave monolithic integrated circuits(MMIcs)[2,3], it is necessary to investigate the temperature dependence of microwave properties.

In order to investigate the magnetic properties of spinel ferrite thin films, we fabricated $Fe_{3-x}Mn_xO_4$ thin films by the ferrite plating method[4~7]. The thickness of films were about 9000Å, measured by AAS(atomic absorption spectrometer) and SEM(scanning eletron microscopy). The especial merit of the ferrite plating method is that we can use non-heat-resistant materials, such as plastics or GaAs ICs, as the substrate. Though prepared at low temperatures, the ferrite plating films exhibits hard and soft magnetic properties as good as those prepared at high temperatures by conventional techniques[3,8,9]. Using these techniques, magnetic capsule toner and carrier was made for magnetography by coating polymer microsphere with a ferrite layer[10]. Therefore, we studied magnetic properties of $Fe_{3-x}Mn_xO_4$ thin films at the microwave range (J-band), and discussed the resonance frequencies, effective magnetic uniaxial anisotropy and spin wave stiffness of manganese ferrites.

II. Experiment

$Fe_{3-x}Mn_xO_4$ thin films(x=0.000, 0.006, 0.010, 0.015, 0.023) were prepared on the cover glass by ferrite plating method. Ferromagnetic resonance spectra were obtained by computer interfacing data at the J-band (7.24GHz)

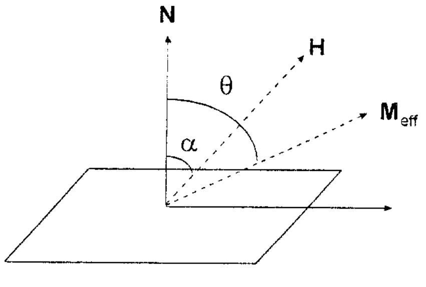

Fig. 1 Geometry of applied field (H) with respect to the film plane.

microwave frequency range, using conventional field modulation techniques with the reentrant coaxial cavity resonator of TE_{010} mode as described in Ref.[11]. In order to check validity of modeling the magnetic anisotrpy as a uniaxial anisotropy, FMR measurements were done in magnetic field is applied to the film ranging from α=0° (H_{app} $\perp$ film plane) to α=90° (H_{app} II film plane)[12] as shown in Fig. 1. The temperature dependence of the effective magnetization was measured from 77 K to a room temperature. Samples were mounted on a cubic quartz wall in the cylinderical cavity using vacuum grease.

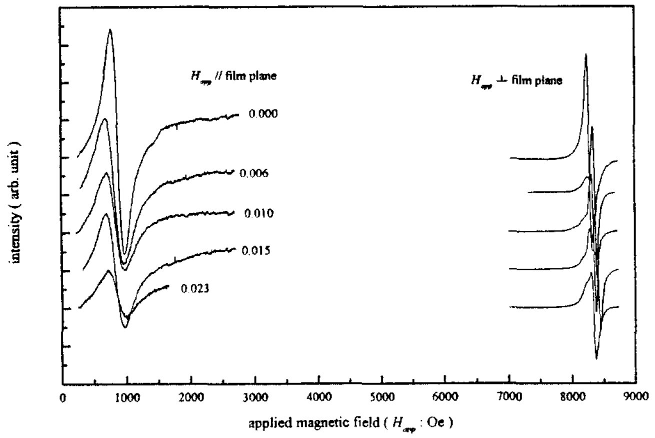

Fig. 2 Derivative absorption of $Fe_{3-x}Mn_xO_4$ as applied parallel and perpendicular to the plane of the films.

III. Results and Discussion

FMR spectra obtained at a room temperature T=300 K showed single resonance line for the $Fe_{3-x}Mn_xO_4$ thin films(x=0.000~ 0.023) as shown in Fig. 2. Here, the line widths in the applied field $H_{//}$ parallel to the film plane are larger than those in the field $H_{\perp}$perpendicular to the plane. The narrow lines in the present films clearly support the idea of well aligned 'cluster'. The large values are attributed to 'anisotropy broadening attendant upon having a random distribution of cluster orientations[13]. The Kittel equations for parallel and perpendicular fields to the film plane are given by[14]

$$(\omega/\gamma)^2 = H_{//}(H_{//} + 4\pi M_{eff}), \qquad (1)$$

$$(\omega/\gamma) = H_{\perp} - 4\pi M_{eff}, \qquad (2)$$

$$4\pi M_{eff} = 4\pi M_s - H_A \qquad (3)$$

Here, $H_A = 2K_u/M_s$ is the uniaxial anisotropy field with the symmetry axis normal to the film. where K_u is the uniaxial anisotropy constant. M_s is the saturation magnetization. $\gamma = g_{eff}(e/2mc)$ is gyromagnetic ratio, ω is the frequency. The g values, obtained for $H_{//}$ and $H_{\perp}$ at 7.24 GHz, increases from 2.12 to 2.16 with increasing Mn content as indicated in Fig. 3.

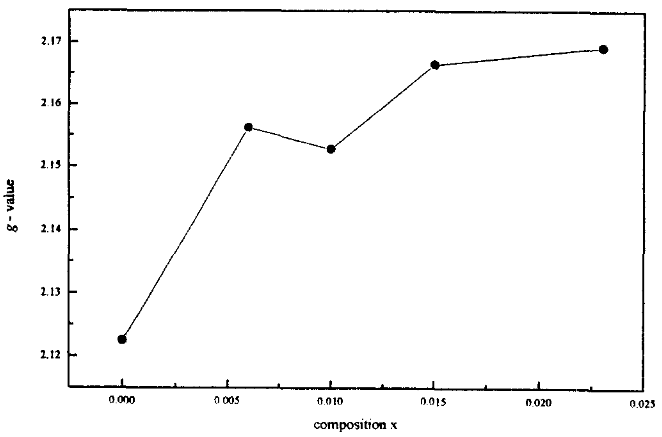

Fig. 3 The spectroscopic splitting g factor as composition x at 300 K.

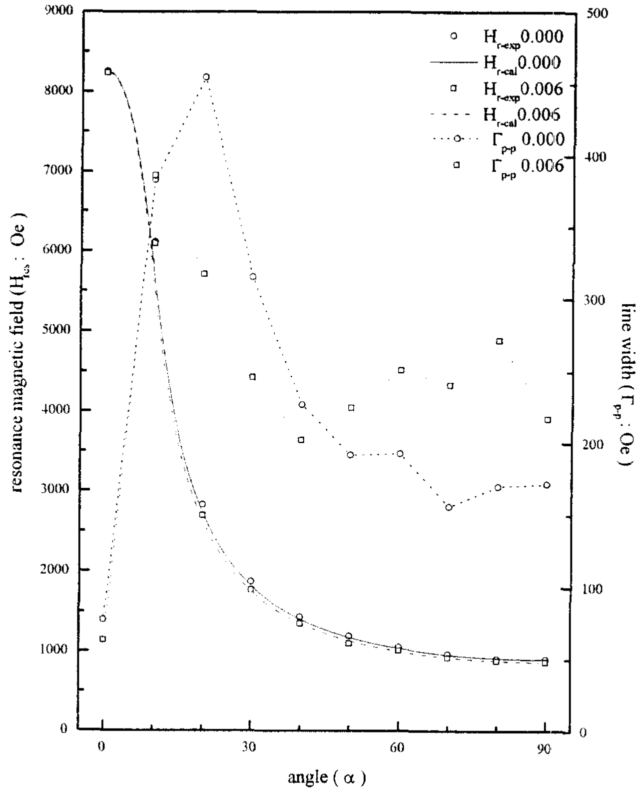

Fig. 4 Resonance magnetic field and line width as function of applied field angle α.

For a thin film with negligible plane anisotropy, one expects that the resonance field obeys the equation

$$(\omega/\gamma)^2 = (H\cos\alpha - 4\pi M_{eff}\cos\theta)^2 + H\sin\alpha(H\sin\alpha + 4\pi M_{eff}\sin\theta) \qquad (4)$$

where θ and α measure the inclinations of M_{eff} and H_{app}, respectively, with respect to the axis normal to the plane of film and is given by

$$\frac{sin(\theta-\alpha)}{sin\theta\ cos\theta} = \frac{4\pi M_{eff}}{H} \qquad (5)$$

Fig. 4 shows resonance field and line width(peak-to-peak) as functions of the tilted angle α of applied field H_{app}. The experimental values are in well coincidence with theoritical lines. Obtaining γ and $4\pi M_{eff}$ from Eqs. (1) and (2), one uses (4) and (5) to obtain the full lines. The agreement is quite remarkable and exhibits a clear indication that if the FMR arises from clusters embedded in the matrix, such clusters must be strongly coupled with one another so that the entire film behaves like a single magnetic entity with a common normal. The effective magnetization ($4\pi M_{eff}$) and saturation magnetizations ($4\pi\ Ms_{\perp}$, $Ms_{//}$), measured with FMR and VSM, are shown in Fig. 5. The values of the uniaxial anisotropy field and the anisotropy constant change the sign from negative to positive, as exhibited in Fig. 6. The deviation due to spin wave excitations is determined from Bloch's law

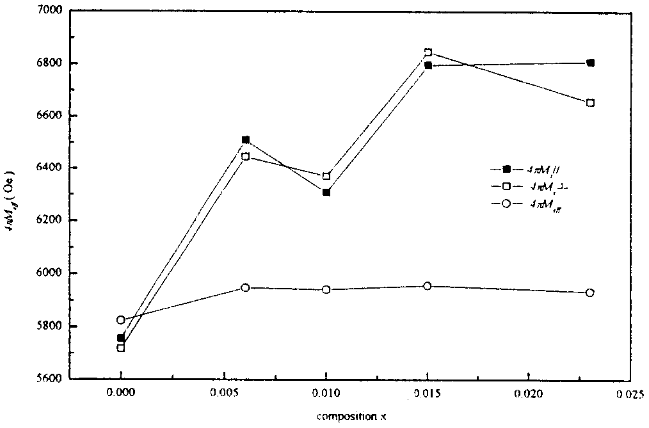

Fig. 5 Effective magnetization and saturation magnetization of $Fe_{3-x}Mn_xO_4$ thin films as composition x at 300K.

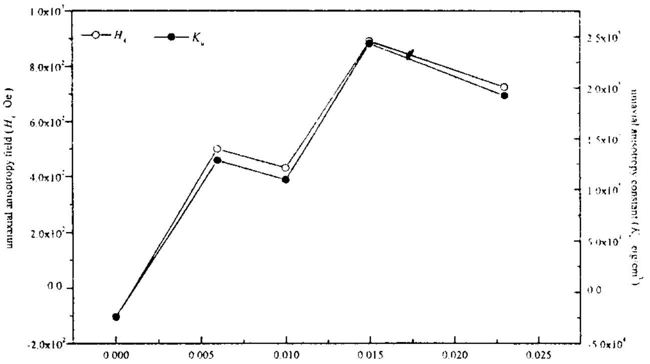

Fig. 6 The uniaxial anisotrpy field and constant of $Fe_{3-x}Mn_xO_4$ thin films as a function of composition x.

$$M_s(T)=M_s(0)\ [1-T^{\frac{3}{2}}-CT^{\frac{5}{2}}] \qquad (6)$$

The quantity B is related to the spin wave stiffness constant, D with

$$B=\frac{\zeta(\frac{3}{2})\cdot\mu_B}{M_s(0)}\ (\frac{k_B}{4\pi D})^{\frac{3}{2}} \qquad (7)$$

$$D=(2.612)^{2/3}(\frac{g\ \mu_B}{M_s(0)B})^{2/3}(\frac{k_B}{4\pi}) \qquad (8)\ \cdot$$

$\zeta(3/2)$ is Riemann-Zeta functions. k_B is Boltzmann cosntant. The effective magnetizations at 0 K, $M_{eff}(0)$ = 487 emu/cm³ (x=0.000), 438 emu/cm³ (x=0.006) respectevly, were obtained by extrapolating from 80 K to 0 K via simple spin wave approximation. The spin wave stiffness constants of Fe_3O_4 and $Fe_{2.994}Mn_{0.006}O_4$ are 55.63 meVÅ, 46.05 meVÅ respectively. Physical parameters obtained from our experiments are listed in Table 1.

Table 1. Effective magnetization $4\pi M_{eff}$, spectrscopic splitting factor g, uniaxial anisotrpy field and constant H_A, K_u, Bloch coefficient B, saturation magnetization at 0 K, $M(0)$ and spin wave stiffness constant D of the $Fe_{3-x}Mn_xO_4$ thin films.

composition(x)	0.000	0.006	0.010	0.015	0.023
$4\pi M_{eff}$(Oe)	5823	5947	5941	5957	5935
g	2.12	2.15	2.15	2.16	2.16
HA(Oe)	-106	500	430	892	726
K_u (× 105erg/cm³)	-0.24	1.28	1.09	2.43	1.92
B(× 10^{-4}K $^{-3/2}$)	1.443	2.161			
M(0) (emu/cm³)	486.7	437.8			
D (meVÅ²)	55.7	46.1			

Ⅳ. Conclusion

The g values increases from 2.12 to 2.16 with increasing Mn content except for x=0.010. In the case of x=0.000, the value of g agrees well with that of a single crystal published results[15]. The resonance magnetic field as a function of applied the angle of H_{app} with respect to the normal axis to the film plane shows good coincidence with theoretcal estimation. The uniaxial anisotropy fields are very large(from -106 Oe to 892 Oe). The uniaxial anisotropy constant of $Fe_{3-x}Mn_xO_4$(x=0.000) is larger about ten or twenty times than the types of polycrystallline bulk(-2.1 × 10⁵ erg/cm³)[16] and single crystal(-1.1 × 10⁵ erg/cm³)[17]. The values of others(x=0.006~0.025) change positive sign.

The tendency of temperature dependence of effective magnetization is similar to that reported in the literature of I.Abu-Aljarayesh[18,19]. The spin wave stiffness constant of Fe_3O_4 is larger than $Fe_{2.994}Mn_{0.006}O_4$. In conclusion, we confirm that the temperature dependence of the magnetization of $Fe_{3-x}Mn_xO_4$ thin films agrees with the prediction of spin wave excitation theory, and that ferrite plating method is proper to ferrite-film formation.

Acknowledgement : This work was supported by the basic research program, The Ministry of Education.

References

[1] M. Matsuoka, M. Naoe and Y. Hoshi, J. Appl. Phys, **57**, 4040 (1985).

[2] I. Zaquine, H. Benazizi and J.C. Mage, J.Appl. Phys, **64**, 15 (1988).

[3] M. Abe, T. Itoh, Y. Tamaura, Y. Gotoh, and M. Gomi, IEEE Trans. **Magn.-23**, 3736 (1997).

[4] M. Abe and Y. Tamaura, J. Appl. Phys, **55**, 2614 (1984).

[5] M. Abe, Y, Tanno and M. Tamaura, J. Appl. Phys., **57**, 3795 (1985).

[6] Jeong Sik Lee, T. Itoh and M. Abe, JKPS, **28**, 375 (1995).

[7] Jeong Sik Lee, T. Itoh and M. Abe, J. Kor. Magn. Soc, **8**, 145 (1996).

[8] T. Itoh, M. Abe and Y. Tamaura Jpn. J. Appl. Phys, **27**, 839 (1988).

[9] M. Abe and Y. Tamaura, Proc. ICF-4, 1984, Sanfrancisco, Advances in Ceramics, **15**, 639 (1985).

[10] K. Ishikawa, M. Oishi, T. Saitoh, M. Abe and Y. Tamaura, Digests of Intermag '87, EB-07.

[11] M. J. Park, K. H. Kim, H. J. Lee, Y. H. Kim, J. Kor. Magn. Soc, **6**, 298 (1996).

[12] E. Schlomann and J. R. Zeender, J. Appl. Phys, **29**, (1958).

[13] M. Rubinstein, B.N. Das, N.C. Koon, D.B. Chrisey and J. Horowitz, Phys. Rev, B **501**, 184 (1994).

[14] V. Seshu Bai, S.M. Bhagat, R. Krishnan, M.Seddat, J. Magn. Magn. Mater, **147**, 97 (1995).

[15] S. E. Harrison, C. J. Kriessman, and S. R. Pollack, Phys. Rev, B, **110**, 4 (1958).

[16] J. B. Birks, Proc. Phys. Soc. (London) B63, 65 (1950).

[17] L. R. Bickford, Phys. Rev. **78**, 449 (1950).

[18] I. Abu-Aljarayesh, A. Al-Rawi a nd H. Abu-Safia, J. Magn. Magn. Mater, **119**, 87 (1993).

[19] I. Abu-Aljarayesh, A. Al-Bayrakdar and S.H. Mahmood, J. Magn. Magn. Mater, **123**, 267 (1993).