Magnetic Properties and the Order-disorder Phase Transformation of (Fe_{1-X}Co_X)Pt Magnetic Thin Films

K. H. Na, C. H. Park, J. G. Na, P. W. Jang¹, C. S. Kim² and S. R. Lee³

Thin Film Technology Center, Korea Institute of Science and Technology, Seoul 136-791, Korea

¹Department of Physics, Chongju University, Cheongju 360-764, Korea

²Department of Physics, Kookmin University, Seoul 136-702, Korea

³Department of Metallurgical Engineering, Korea University, Seoul 136-701, Korea

(Received 16 July 1999)

Magnetic properties and crystal structures of $(Fe_xCo_{1.x})Pt$ (X=0,0.2,0.4,0.5,0.6,0.8 and 1.0) ternary thin films were investigated. The order-disorder phase transformation of FePt thin films during annealing was also studied by x-ray diffraction and Mössbauer spectroscopy. The magnetic thin films were deposited on glass substrates using a dc sputtering method and were subsequently annealed at 400~700 °C in a high vacuum. The asdeposited films exhibited a high degree of the <111> preferred orientation and the preferred orientation was not destroyed even after the subsequent post annealing. The coercivity of the $(Fe_xCo_{1.x})Pt$ thin films annealed at 700 °C showed a minimum value at the equiatomic composition of the Fe and Co atoms. The ordered structure of the FePt alloy was thought to have formed from the disordered structure by an inhomogeneous process, which was confirmed by the asymmetric peak shapes and Mössbauer spectra.

1. Introduction

FePt and CoPt binary alloy films with a tetragonal $L1_0$ structure have received significant attention as magnetic and magneto-optic recording materials due to their high magnetocrystalline anisotropy [1, 2]. The magnetic anisotropy constants of FePt and CoPt alloys in ordered structure were reported as $7 \times 10^7 \, \mathrm{Jm^{-3}}$ and $5 \times 10^7 \, \mathrm{Jm^{-3}}$, respectively [3]. Because FePt and CoPt alloys have the tetragonal $L1_0$ structure, substitution of Co atoms for Fe atoms or vice versa may show interesting magnetic properties. To obtain high magnetic anisotropies of CoPt and FePt thin films with the tetragonal $L1_0$ ordered structure, the thin films with disordered structure are generally annealed in a high vacuum for phase transformation from disordered phase to ordered one [4]. Therefore, it is important to understand the phase transformation of FePt thin films during annealing.

In this study, the magnetic properties and the structure of $(Fe_xCo_{1-x})Pt$ ($x = 0 \sim 1$) films were investigated. Also the order-disorder transformation in FePt magnetic thin films during annealing was studied using x-ray diffraction (XRD) and Mössbauer spectroscopy.

2. Experimental Procedure

 $(Fe_xCo_{1-x})Pt$ (x = 0~1) magnetic thin films were sputtered on Corning 7059 glass substrates using a dc-magnetron-sputter system. For CoPt and the FePt binary films, com-

posite targets were used, for which 5 mm \times 10 mm Pt sheets were placed on the erosion tracks of pure Co or Fe targets, respectively. The (Fe_xCo_{1-x})Pt thin films were deposited from Co-Fe-Pt composite targets, for which additional 5 mm \times 10 mm Fe or Co sheets were placed on preset Co-Pt or Fe-Pt composite targets. For Mössbauer measurement, BN discs were used as substrates. Table 1 lists the sputtering conditions for (Fe_xCo_{1-x})Pt thin films. The as-sputtered films were annealed in a high vacuum of 7×10^{-6} Torr at $400 \sim 700$ °C for $10 \sim 60$ min.

The structure and composition of the thin films were analyzed using a Philps X'Pert diffractometer with a $CuK\alpha$ radiation source and an electron probe microanalyzer (EPMA). The magnetic properties were measured using a vibrating sample magnetometer (VSM) with a maximum field of 16 kOe. Mössbauer spectra were recorded using a conventional spectrometer of the electromechanical type

Table 1. Sputtering conditions for $(Fe_xCo_{1-x})Pt$ magnetic thin films

Background pressure	6×10^{-7} Torr		
Sputtering pressure	1×10^{-3} Torr		
Substrate temperature	room temperature		
Input power	100 W		
Film thickness	200 nm		
	20 μm		
	(for Mössbauer analysis)		

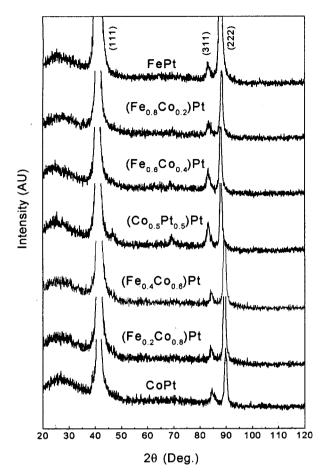


Fig. 1. X-ray diffraction patterns of as-deposited $(Fe_xCo_{1-x})Pt$ films.

with a ⁵⁷Co source in a rhodium matrix [5].

3. Results and Discussion

Figure 1 shows XRD patterns of the as-deposited (Fe_xCo_{1-x}) Pt thin films. It can be seen from Fig. 1 that the as-deposited (Fe_xCo_{1-x}) Pt thin films have a disordered face-centered cubic structure because peaks related with the $L1_0$ -ordered phase could not be detected. A strong <111> fiber texture can be clearly seen in Fig. 1, which agrees with the facts that a fiber texture with a closely packed plane parallel to the film plane develops when the deposition is done at a low temperature, while a low-index plane texture develops at a high temperature.

As-deposited (Fe_xCo_{1-x})Pt thin films showed low coercivities of less than 100 Oe, which implies that the thin films have disordered crystal structure [4]. However, annealed films with the ordered *L*1₀-type structure exhibited high coercivities. Figures 2 and 3 show changes in the coercivities of (Fe_xCo_{1-x})Pt thin films annealed at 550 and 700 °C for 10 to 60 min, respectively. When the films were annealed for 10 min at 550 °C, the CoPt binary films showed a low coercivity of 100 Oe while the FePt binary films exhibited a high coercivity of 8,000 Oe. After annealing for 30 min, the coercivities of CoPt and FePt thin films increased little.

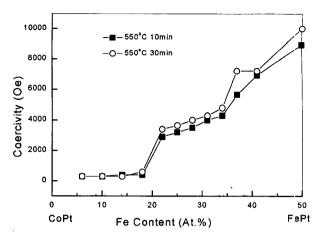


Fig. 2. Variation of coercivity of (Fe_xCo_{1-x})Pt films with Fe content when annealed at 500 °C for 10~60 min.

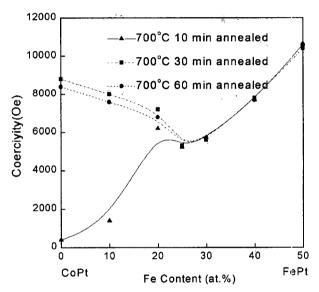


Fig. 3. Variation of coercivity of $(Fe_xCo_{1-x})Pt$ films with Fe content when annealed at 700 °C for $10\sim60$ min.

When the films were annealed for 10 min at 700 °C, the CoPt binary films showed a low coercivity of 100 Oe, while the FePt binary films exhibited a very high coercivity of 10,000 Oe. After annealing for 30 and 60 min, the coercivities of CoPt increased dramatically up to 9,000 Oe, while that of FePt thin films increased little. These results are attributed to the different order-disorder transition temperature of FePt and CoPt alloys. The transition temperature of the CoPt and FePt alloys is 825 and 1300 °C, respectively [5]. For the thin films annealed at 700 °C for 60 min, the coercivity of the thin films showed V-shape with increasing Fe content in (Fe_xCo_{1-x})Pt thin films and a minimum coercivity of 5,500 Oe at the equiatomic composition of the Fe and the Co atoms. According to the result, the coercivity of (Fe_xCo_{1-x})Pt thin films can be controlled by adjusting composition.

To understand the order-disorder transformation of (Fe_xCo_{1-x})Pt thin films during annealing, XRD and Mössbauer experiments were conducted. Figure 4 shows XRD

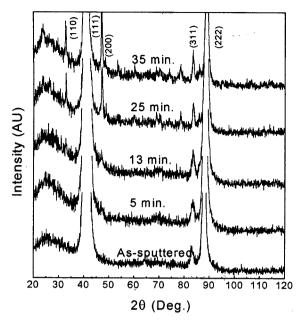


Fig. 4. XRD patterns of FePt thin films annealed at 400 °C for 0, 5, 13, 25 and 35 min.

patterns of the FePt thin films annealed at 400 °C for 0, 5, 13, 25 and 35 min, respectively. As can be seen from Fig. 4, there were no peaks related to superlattices such as (001) and (002) in the as-deposited FePt thin films, but (001) and (002) peaks can be seen clearly in the thin films annealed for 25 and 35 min, respectively. These results indicate that the H_c increase of the FePt thin films during annealing is mainly attributed to the phase transformation from disordered phase to ordered one.

For the order-disorder transformation in FePt thin films, two types of ordering processes can be available: homogeneous and inhomogeneous one. Taking into account that the diffraction angle of the disordered FePt (222) peak is lower than that of the ordered one, the symmetric (222) peak of the as-sputtered thin film represents the thin film has a disordered structure [6, 7]. If the thin films become ordered by the homogeneous ordering process, in which ordered phase is formed gradually in a disordered grain by nucleation and growth, gradual shift of symmetric (222) peak to high angle can be observed. On the other hand, if the thin films become ordered by the inhomogeneous ordering process, in which ordered phase is formed abruptly like the martensitic transformation in a disordered grain, the symmetric (222) peak of disordered phase becomes gradually broadened into band and the (222) peak of the ordered phase emerged from the band. From Fig. 5, one can see that the symmetric (222) peak of the disordered phase becomes asymmetric with a long tail to high angle in the early stage of the transformation and then move to high angle with a long tail to low angle after 25 min annealing. These results imply that the FePt thin films become ordered during annealing by an inhomogeneous ordering process.

Fig. 6 shows the Mössbauer spectra of the FePt thin films

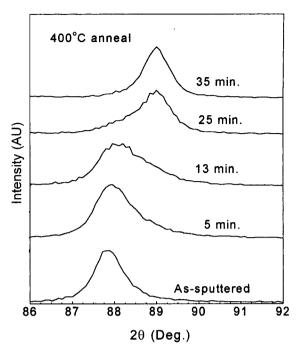


Fig. 5. Detailed XRD patterns near (222) peak of FePt thin films annealed at $400 \, ^{\circ}\text{C}$ for 0, 5, 13, 25 and 35 min.

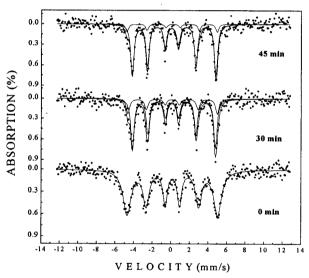


Fig. 6. Mössbauer spectra of FePt thin films annealed at 400 °C for a) 0 min., b) 30 min, and c) 45 min.

annealed at 400 °C for a) 0, b) 30, and c) 45 min, respectively and Table 2 gives the least-squares-fitting data deduced from the Mössbauer spectra shown in Fig. 6. It can be seen from Fig. 6 and Table 2 that for the thin film (a), one kind of sextets, which seems to be similar to the spectrum of cubic phase, is detected, while for the thin films (b) and (c), two kinds of sextets are detected. From Table 2, it can be seen that the Mössbauer parameters of the first subspectrum are nearly the same as that of the as-deposited thin film, while the second one is quite different from the first one. As the area ratio of the second subspectrum increases with increasing annealing time, the second sub-

Table 2. Mössbauer data of thin films deduced from spectra shown in Fig. 6

Sample	Assigned subspectra	H_{hf} (MAm ⁻¹)	ΔE_Q (mm s ⁻¹)	IS (mm s ⁻¹)	Area (%)
(a)	Α	24.1	0.14	0.00	100
(b)	A	23.9	0.02	0.12	25
	В	22.1	0.11	0.18	75
(c)	Α	24.0	-0.05	0.13	11
	В	22.3	0.14	0.16	89
		(± 0.8)	(± 0.01)	(± 0.01)	(± 0.01)

spectrum in the thin films (b) and (c) may correspond to the ordered FePt. The values of the Mössbauer parameters, H_{hf} and IS, deduced from the second subspectrum in Table 2 agree with those of Goto *et al.* [8].

4. Conclusion

(Fe_xCo_{1-x})Pt ternary thin films were deposited on glass substrates using a dc-sputtering method at room temperature and were subsequently annealed at a elevated temperature in a high vacuum.

- (1) The as-deposited thin films have disordered face-centered cubic structure with a high degree of the <111> preferred orientation and superlattice peaks related with the $L1_0$ -ordered phase could not be detected.
- (2) The ordered structure of the FePt alloy was thought to be formed from the disordered structure by an inhomoge-

- neous process, and this was confirmed by the asymmetric peak shapes and Mössbauer spectra.
- (3) The order-disorder phase transformation occurred in FePt thin films more rapidly than in CoPt thin films, which was attributed to different order-disorder transition temperatures of FePt and CoPt alloys.
- (4) For the thin films annealed at 700 $^{\circ}$ C for 60 min, the coercivity of the thin films showed V-shape with increasing Fe content in (Fe_xCo_{1-x})Pt thin films and a minimum coercivity of 5,500 Oe at the equiatomic composition of the Fe and Co atoms.

References

- [1] M. Watanabe and M. Homma, J. Appl. Phys., **35**, L1264 (1996).
- [2] B. M. Laison, M. R. Visokay, E. E. Mariero, R. Sinclare and B. M. Clemens, J. Appl. Phys., 74, 1922 (1993).
- [3] B. M. Laison and B. M. Clemens, Appl. Phys. Lett., **63**. 1438 (1993).
- [4] S. W. Yung, Y. H. Chang, T. J. Lin and M. P. Hung, J. Magn. Magn. Mater., 116, 411(1992)
- [5] T. B. Massalski, Binary alloy phase diagrams, Am. Soc. Metal, Ohio(1986), pp. 791.
- [6] Powder Diffraction File, Card No. 29-718.
- [7] Powder Diffraction File, Card No. 43-1359.
- [8] T. Goto, H. Utsugi and K. Watanabe, Hyperfine Inter., **54**, 539 (1990).