Microstructure and Coercivity of Granular Nanocomposite FePt–Ag Multilayer Films

S. C. Chen[1], P. C. Kuo[2], A. C. Sun[2], C. Y. Chou[2], Y. H. Fang[2], and S. Y. Kuo[3]

[1]Department of Materials Engineering, Ming Chi University of Technology, Taipei 243, Taiwan, R.O.C., and Center for Nanostorage Research, National Taiwan University, Taipei 106, Taiwan, R.O.C.
[2]Institute of Materials Science and Engineering and Center for Nanostorage Research, National Taiwan University, Taipei 106, Taiwan, R.O.C.
[3]Department of Civil Engineering, De Lin Institute of Technology, Taipei 236, Taiwan, R.O.C.

The face-centered-tetragonal granular $L1_0$ FePt nanoparticles with large in-plane coercivity ($Hc_{//}$) of about 3923 Oe can be achieved by an Ag-capped layer of 5-nm thickness deposited onto the FePt magnetic layer, with thickness of 20 nm, after annealing at 400 °C for 30 min. The perpendicular coercivity ($Hc_{\perp}$) of the films is increased by increasing the annealing temperature (T_s), and the $Hc_{\perp}$ value is about equal to the $Hc_{//}$ value when the annealing temperature is increased to 600 °C. TEM-energy disperse spectrum analysis reveals that the Ag mainly distributed at the grain boundary of FePt and results in the isolation of the FePt grains as well as the increase of the grain boundary energy, which will enhance coercivity and change the preferred orientation of the FePt film.

***Index Terms*—Ag-capped layer, coercivity, FePt–Ag multilayer films, magnetron sputtering.**

I. Introduction

RECENTLY, FePt nanograins have attracted much attention for their potential applications for high-density magnetic recording media [1], because of high-magnetocrystalline anisotropy (Ku $\sim 7 \times 10^7$ erg/cm^3) of the orderd $L1_0$ phase. In general, the formation of the $L1_0$ FePt phase requires a heat treatment at a higher temperature around 600 °C [2], [3], which results in the grain growth.

The grain size must be reduced to below 10 nm for high-density magnetic recording media applications. Therefore, high-coercivity materials consisting of the ordered $L1_0$ FePt phase dispersed in nonmagnetic matrices to refine grain sizes of FePt have been reported in FePt–C [4], FePt–SiO_2 [5], and FePtCr–Si_3N_4 [6] thin films. Current studies have been focused on decreasing the energy barrier of phase transformation of FePt from an fcc-disordered phase to an fct-ordered phase. Several works on the lowering of ordering temperature and the enhanced coercivity of FePt films have been reported, such as doping a third element [7]–[9] or introducing an underlayer [10]–[13] and capped layer [14], [15] to the FePt films.

An Ag underlayer is found to be effective in reducing the FePt ordering temperature [10], [11]. The Ag is immiscible with either Fe or Pt. Instead, it tends to segregate at the grain boundary of FePt [15] and increases the grain boundary energy, which can change the preferred orientation of the FePt film [16]. However, there is still no direct evidence to confirm the exact position of the Ag in the FePt layer. In this paper, the TEM-energy disperse spectrum (EDS) is used to analyze the Ag distribution in the FePt layer. The microstructures, magnetic properties, and preferred orientation of the FePt layer of the granular nanocomposite MgO–FePt–Ag multilayer films are also investigated.

Digital Object Identifier 10.1109/TMAG.2005.854703

II. Experiment

The MgO underlayer of 5-nm thickness is deposited onto naturally oxidized Si(100) substrates by rf magnetron sputtering at an ambient temperature under an Ar pressure of 10 mtorr. The FePt magnetic layer with a thickness of 20 nm and an Ag-capped layer of 5-nm thickness are deposited subsequently by dc magnetron sputtering onto the MgO underlayer. The as-deposited films are annealed at 400 °C, 500 °C, and 600 °C for 30 min in vacuum higher than 5×10^{-7} torr. The composition of the FePt film determined by X-ray EDS is $Fe_{50.2}Pt_{49.8}$. The microstructures of the film are investigated by a Philips Tecnai F30 field emission gun (FEG) TEM and by an X-ray diffractometer (XRD) with Cu–K_α radiation. Compositions of the films are determined by EDS and the depth profiles of elements in the film are analyzed by Auger electron spectroscopy (AES). The magnetic properties of the films are measured using a vibrating sample magnetometer (VSM) at room temperature.

III. Results and Discussion

Fig. 1 shows the in-plane coercivity and perpendicular coercivity of MgO 5 nm–FePt 20 nm–Ag 5 nm multilayer films as a function of the annealing temperature. It is found that a large $Hc_{//}$ value of about 3923 Oe can be obtained after annealing at 400 °C. Compared with the reports of Sellmyer *et al.* [17] and Kuo *et al.* [18], an $Hc_{//}$ value of only 1500–3000 Oe is obtained by annealing at the same temperature for pure FePt films. On the other hand, the $Hc_{\perp}$ value of the films is increased by increasing the annealing temperature, and the $Hc_{\perp}$ value is about equal to the $Hc_{//}$ value when the annealing temperature is increased to 600 °C. This indicates that the preferred orientation of the FePt films will be changed from (111) plane to random as the annealing temperature increases from 400 °C to 600 °C.

Fig. 2 shows the XRD patterns of the MgO 5 nm–FePt 20 nm–Ag 5 nm multilayer films after annealing at various temperatures. It is found that the preferred orientation of the

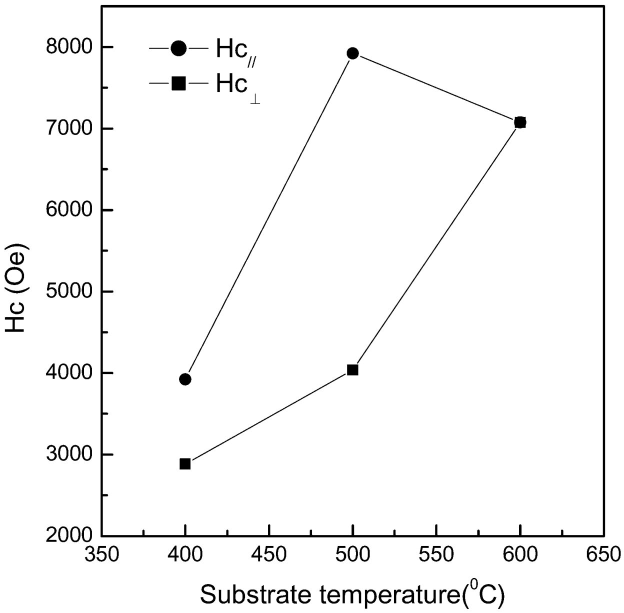


Fig. 1. In-plane coercivity and perpendicular coercivity of MgO 5 nm–FePt 20 nm–Ag 5 nm multilayer films as a function of annealing temperature.

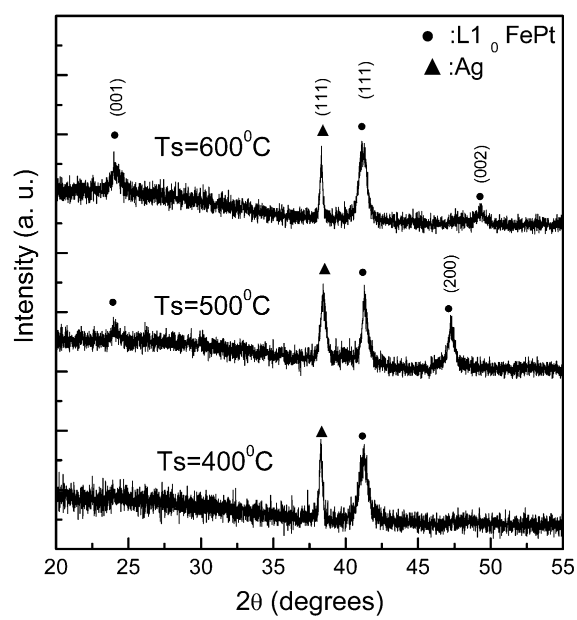
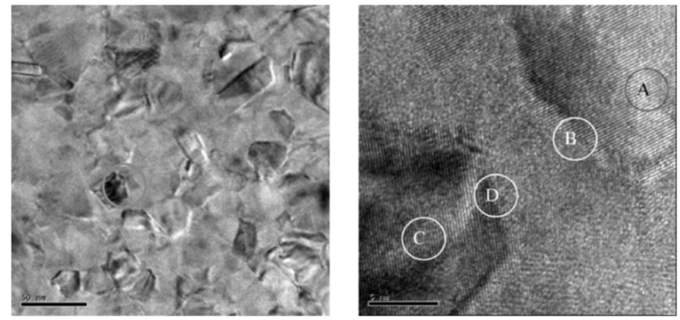


Fig. 2. XRD patterns of the various annealed MgO 5 nm–FePt 20 nm–Ag 5 nm multilayer films. Annealing temperatures are 400 °C, 500 °C, and 600 °C.

FePt films is (111) after annealing at 400 °C and 500 °C. As the annealing temperature increases to 600 °C, the (001) and (002) peaks of the FePt films are enhanced. However, the intensity of the (111) peak of the FePt films almost keeps constant. This means that the preferred orientation of the FePt film will change from the (111) plane to random when the annealing temperature is increased from 400 °C to 600 °C, which is consistent with the measurement of the coercivities in Fig. 1.

Fig. 3(a) is a TEM bright field image for the MgO 5 nm–FePt 20 nm–Ag 5 nm multilayer films which annealed at 600 °C for 30 min. The TEM image of the film shows a granular structure, and its grain size is about 24 nm. Fig. 3(b) shows the high-resolution TEM lattice image of the enclosed area of Fig. 3(a). In order to understand the exact position of Ag atoms in the FePt



(a) (b)

Fig. 3. (a) TEM bright field image of the MgO 5 nm–FePt 20 nm–Ag 5 nm multilayer films which annealed at 600 °C for 30 min. (b) High-resolution TEM lattice image of the enclosed area of Fig. 3(a).

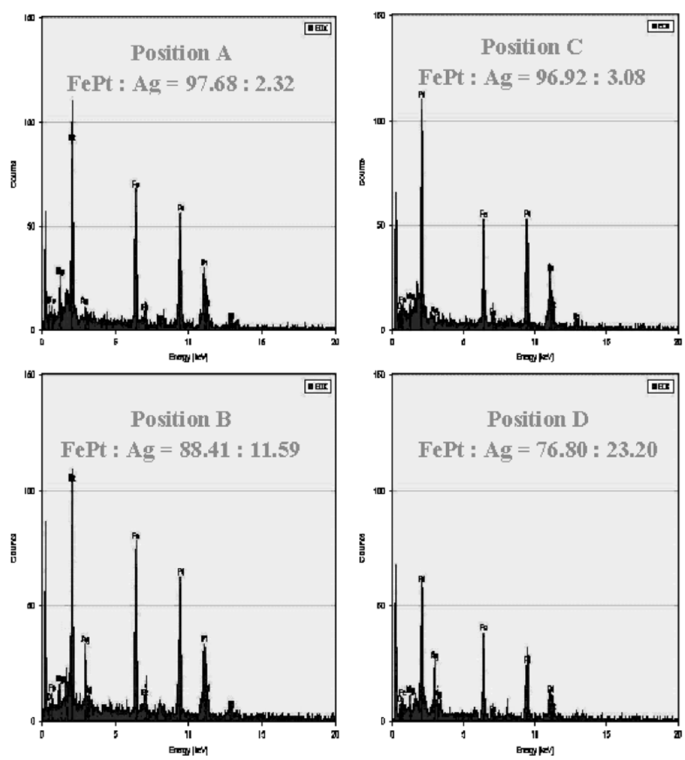


Fig. 4. Ag contents of the enclosed areas of A (inner grain), B (grain boundary), C (inner grain), and D (grain boundary), illustrated in the image of Fig. 3(b).

layer, the TEM-EDS is used to analyze the Ag contents of the enclosed areas of A (inner grain), B(grain boundary), C(inner grain), and D(grain boundary), which are illustrated in the lattice image of Fig. 3(b). As shown in Fig. 4, the Ag contents of the enclosed areas of A, B, C, and D are determined to be 2.32, 11.59, 3.08, and 23.20 at.%, respectively. The TEM-EDS datas reveals that the Ag atoms in the FePt magnetic layer mainly distribute at the grain boundary and result in the isolation of the FePt grains as well as the increase of the grain boundary energy. This will reduce grain size, enhance coercivity, and change the preferred orientation of the FePt film. Therefore, as shown in Fig. 1, the $L1_0$ FePt nanoparticles with a large in-plane coercivity of about 3923 Oe can be obtained by annealing at a low temperature of 400 °C for the MgO 5 nm–FePt 20 nm–Ag 5 nm multilayer films. Furthermore, the perpendicular coercivity of the films increases with increasing the annealing temperature, and the $Hc_{\perp}$

value is almost equal to the $Hc_{//}$ value as the annealing temperature is increased to 600 °C. This indicates the preferred orientation of the FePt films will change from the (111) plane to random as the annealing temperature increases from 400 °C to 600 °C, which is confirmed by XRD measurement, as shown in Fig. 2. This result is also ascribed to the Ag atoms distributed at the grain boundary of FePt to increase the grain boundary energy; hence, it changes the preferred orientation of the FePt films. Similarly, Yuan *et al.* [14] reported that an Au capped layer on FePt film can largely enhance coercivity, which is also due to the diffusion of Au into the grain boundary of the FePt layer.

IV. Conclusion

The TEM-EDS analysis results reveal that the Ag mainly distributed at the grain boundary of FePt, which will hinder the grain growth, enhance coercivity, and change the preferred orientation of FePt film. The granular $L1_0$ FePt nanoparticles with a large in-plane coercivity of about 3923 Oe can be obtained from MgO 5 nm–FePt 20 nm–Ag 5 nm multilayer films after annealing at a low temperature of 400 °C for 30 min.

Acknowledgment

This work was supported in part by the National Science Council under Grant NSC 93-2216-E-002-023 and by the Ministry of Economic Affairs, Taiwan, R.O.C., under Grant 93-EC-17-A-08-S1-0006.

References

[1] D. Weller, A. Moser, L. Folks, M. E. Best, W. Lee, M. F. Toney, M. Schwickert, J.-U. Thiele, and M. F. Doerner, "High Ku materials approach to 100 Gbits/in^2," *IEEE Trans. Magn.*, vol. 36, no. 1, pp. 10–15, Jan. 2000.

[2] T. Suzuki and K. Ouchi, "Sputter-deposited (Fe-Pt)-MgO composite films for perpendicular recording media," *IEEE Trans. Magn.*, vol. 37, no. 2, pp. 1283–1285, Mar. 2001.

[3] P. C. Kuo, S. C. Chen, Y. D. Yao, A. C. Sun, and C. C. Chiang, "Microstructure and magnetic properties of nanocomposite FePtCr-SiN thin films," *J. Appl. Phys.*, vol. 91, pp. 8638–8640, 2002.

[4] J. A. Christodoulides, Y. Huang, Y. Zhang, G. C. Hadjipanayis, I. Panagiotopoulos, and D. Niarchos, "CoPt and FePt thin films for high density recording media," *J. Appl. Phys.*, vol. 87, pp. 6938–6940, 2000.

[5] C. P. Luo, S. H. Liou, and D. J. Sellmyer, "FePt:SiO_2 granular thin film for high density magnetic recording," *J. Appl. Phys.*, vol. 87, pp. 6941–6943, 2000.

[6] S. C. Chen, P. C. Kuo, A. C. Sun, C. T. Lie, and C. C. Chiang, "Effects of Cr and SiN contents on the microstructure and magnetic grain interactions of nanocomposite FePtCr-SiN thin films," *IEEE Trans. Magn.*, vol. 39, no. 2, pp. 584–589, Jan. 2003.

[7] T. Maeda, T. Kai, A. Kikitsu, T. Nagase, and J.-i. Akiyama, "Reduction of ordering temperature of an FePt-ordered alloy by addition of Cu," *Appl. Phys. Lett.*, vol. 80, pp. 2147–2149, 2002.

[8] S. S. Kang, D. E. Nikles, and J. W. Harrell, "Synthesis, chemical ordering, and magnetic properties of self-assembled FePt-Ag nanoparticles," *J. Appl. Phys.*, vol. 93, pp. 7178–7180, 2003.

[9] H. Y. Wang, W. H. Mao, X. K. Ma, H. Y. Zhang, Y. B. Chen, Y. J. He, and E. Y. Jiang, "Improvement in hard magnetic properties of FePt films by N addition," *J. Appl. Phys.*, vol. 95, pp. 2564–2568, 2004.

[10] Y. N. Hsu, S. Jeong, D. E. Laughlin, and D. N. Lambeth, "Effects of Ag underlayers on the microstructure and magnetic properties of epitaxial FePt thin films," *J. Appl. Phys.*, vol. 89, pp. 7068–7070, 2001.

[11] X.-H. Xu, H.-S. Wu, F. Wang, and X.-L. Li, "Structure and magnetic properties of FePt and FePt/Ag thin films deposited by magnetron sputtering," *Thin Solid Films*, vol. 472, pp. 222–226, 2005.

[12] C.-H. Lai, C.-H. Yang, C. C. Chiang, T. Balaji, and T. K. Tseng, "Dynamic stress-induced low-temperature ordering of FePt," *Appl. Phys. Lett.*, vol. 85, pp. 4430–4432, 2004.

[13] S. C. Chen, P. C. Kuo, S. T. Kuo, A. C. Sun, C. Y. Chou, and Y. H. Fang, "Improvement in hard magnetic properties of FePt films by introduction of Ti underlayer," *IEEE Trans. Magn.*, vol. 41, no. 2, pp. 915–917, Feb. 2005.

[14] F. T. Yuan, S. K. Chen, W. C. Chang, and L. Horng, "Effect of Au cap layer on the magnetic properties and the microstructure for FePt thin films," *Appl. Phys. Lett.*, vol. 85, pp. 3163–3165, 2004.

[15] Z. L. Zhao, J. Ding, K. Inaba, J. S. Chen, and J. P. Wang, "Promotion of $L1_0$ ordered phase transformation by the Ag top layer on FePt thin films," *Appl. Phys. Lett.*, vol. 83, pp. 2196–2198, 2003.

[16] Y. Z. Zhou, J. S. Chen, G. M. Chow, and J. P. Wang, "Structure and magnetic properties of in-plane oriented FePt-Ag nanocomposites," *J. Appl. Phys.*, vol. 93, pp. 7577–7579, 2003.

[17] M. L. Yan, N. Powers, and D. J. Sellmyer, "Highly oriented nonepitaxially grown $L1_0$ FePt films," *J. Appl. Phys.*, vol. 93, pp. 8292–8294, 2003.

[18] C.-M. Kuo, P. C. Kuo, and H.-C. Wu, "Microstructure and magnetic properties of $Fe_{100-x}Pt_x$ alloy films," *J. Appl. Phys.*, vol. 85, pp. 2264–2269, 1999.

Manuscript received February 7, 2005; revised April 1, 2005.