Microstructure and magnetic properties of CoPt-SiN$_x$/Ag thin films

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Different amounts of amorphous SiN$_x$ nonmagnetic material and magnetic $Co_{50}Pt_{50}$ alloy were cosputtered on the different thicknesses of Ag films at room temperature then annealed at different temperatures. When the thickness of Ag underlayer is 25 nm, the CoPt/Ag film has a minimum in-plane squareness ($S_{\parallel}$) which is about 0.35. The out-of-plane squareness ($S_{\perp}$), out-of-plane coercivity ($H_{c\perp}$), and saturated magnetization (M_s) values of the CoPt/Ag (25 nm) film are about 0.95, 15 kOe, and 420 emu/cm^3, respectively. From the microstructure analysis of CoPt-SiN$_x$/Ag (25 nm) films with different volume percent of SiN$_x$ content, it is found that the average grain size of CoPt decreases from about 80 to 9 nm when the volume percent of SiN$_x$ is increased from 0% to 50%. The $S_{\perp}$, $H_{c\perp}$, and M_s values of the $(CoPt)_{50}$-$(SiN_x)_{50}$/Ag films are about 0.5, 7.5 kOe, and 200 emu/cm^3, respectively. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2832342]

Both the CoPt and FePt alloy films have high magnetocrystalline anisotropy and high thermal stability as they apply to the ultrahigh density magnetic recording medium.[1] These two films possess a face-centered-cubic (fcc) phase which could be transferred to a face-centered-tetragonal (fct) phase by introducing the proper underlayer behind the FePt or CoPt films.[2–4] It has been reported that the strain energy caused by the misfit between the CoPt layer and Ag layer provides a driving force for the ordering of CoPt film as the Ag underlayer is introduced beneath the CoPt films.[5] According to Zhang *et al.*,[6] in order to decrease the recording noise, the grain size of the magnetic film has to be smaller than 10 nm. Moreover, the exchange coupling effect should be minimized in order to lower the transition noise. Several researchers controlled the grain size of the magnetic film by adding nanomagnetic materials [BN,[7] SiO_2,[8] Al_2O_3,[9] and C (Ref. 10)]. In our previous study,[11] we added the SiN$_x$ into the FePt films and examined the magnetic properties of $(FePt)_{1-y}$-$(SiN_x)_y$ films. In this work, we will discuss the addition of SiN$_x$ ceramic material to the CoPt films that have many good properties such as oxidation resistance, corrosion resistance, and wear resistance. Here, we introduced the different thicknesses of Ag underlayers beneath the CoPt films to achieve a transformation of CoPt films from fcc to fct structure, and added SiN$_x$ ceramic materials to the CoPt films in an attempt to reduce the grain sizes of CoPt films.

Different thicknesses of Ag underlayers were deposited on naturally oxidized Si (100) wafer and glass substrates at room temperature. Different volume percent of SiN$_x$ ceramic material and 20 nm $Co_{50}Pt_{50}$ films were cosputtered on the Ag films with different thicknesses at room temperature. Finally, the films CoPt-SiN$_x$/Ag were annealed at different temperatures for 30 min. The composition and thickness of the film were determined by an energy dispersive x-ray spectrometer and an atomic force microscope, respectively. Magnetic properties of the films were measured by using a vibrating sample magnetometer with a maximum applied field of 15 kOe and a superconducting quantum interference device. The film structure was examined by an x-ray diffractometer (XRD) and a field emission gun high resolution transmission electron microscope (FEG-TEM).

Figure 1 shows the ratio of I_{001}/I_{111} as a function of Ag underlayer thickness with different annealing temperatures, where I_{001} and I_{111} are the related integrated intensities of the (001) and (111) peaks. It is found that the I_{001}/I_{111} ratio of CoPt/Ag film is increased from 0 to 13 as Ag thickness increases from 0 to 50 nm, then decreases rapidly with further increase of Ag thickness when the annealing temperature is 700 °C. The maximum I_{001}/I_{111} ratio occurs at about 100 nm Ag underlayer when the annealing temperatures are 600 and 500 °C. It has been found that the orientation of the easy axis of CoPt magnetic layer was strongly influenced by the Ag underlayer, and the Ag underlayer would also promote the formation of the CoPt (001) orientation; these resulted in enhancing the CoPt $L1_0$ ordering easily.[12,13] For the CoPt/Ag film annealed at 700 °C, the I_{001}/I_{111} ratio of the CoPt films decreases rapidly as the Ag underlayer thickness

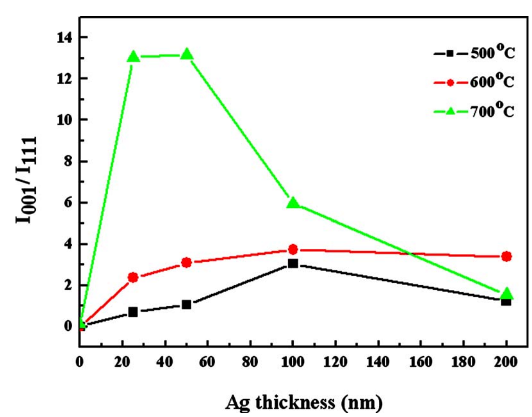

FIG. 1. (Color online) I_{001}/I_{111} ratio of the $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films as a function of Ag underlayer thickness annealed at different temperatures.

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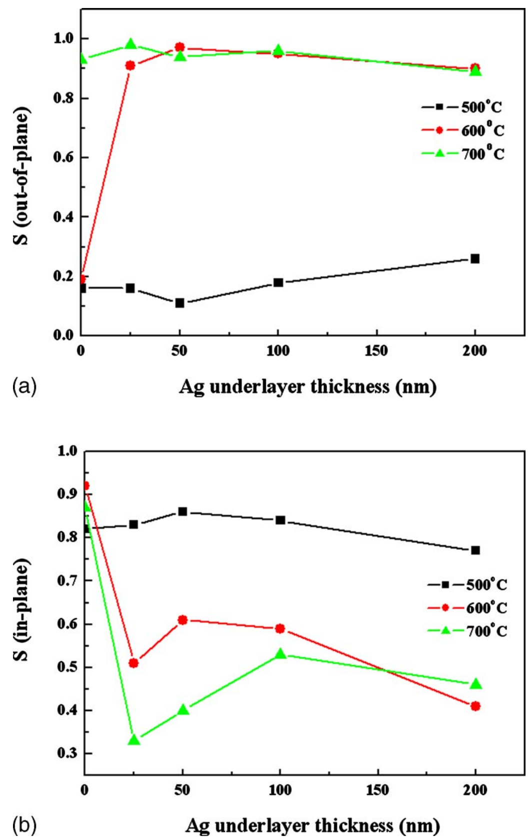

FIG. 2. (Color online) [(a) and (b)] Relationships among the $S_\perp$, $S_\parallel$ and thickness of Ag underlayers of CoPt/Ag films annealed at different temperatures.

is more than 50 nm. This is because the Ag (002) orientation will be formed and increased gradually as the Ag thickness is increased. However, the Ag (002) orientation will suppress the Ag (111) preferred orientation and also break up the epitaxy between Ag (111) and CoPt (001). Figures 2(a) and 2(b) are the relationships among squareness ($S_\perp$), squareness ($S_\parallel$), and the thickness of the Ag underlayer of CoPt/Ag films at different annealing temperatures. It is found that as the Ag underlayer thickness increases from 0 to 200 nm, the $S_\perp$ value of CoPt/Ag film is kept above 0.9 and the minimum $S_\parallel$ value is about 0.35, which occurs at 25 nm Ag, when the CoPt/Ag film is annealed at 700 °C. From the XRD analysis of the CoPt/Ag (25 nm) film annealed at 700 °C, it is observed that the fct (001) and fct (002) superlattice lines appeared, whereas the intensity of the (111) peak is suppressed. This means that the Ag underlayer is effective in promoting both the growth of the $L1_0$ phase and the chemical ordering of CoPt, as shown in Fig. 3. The $S_\perp$, $S_\parallel$, $H_{c\perp}$, and M_s values of the CoPt/Ag (25 nm) film annealed at 700 °C are about 0.95, 0.35, 15 kOe, and 420 emu/cm^3, respectively. Therefore, the 25 nm Ag film is chosen to be the underlayer due to its large perpendicular magnetic anisotropy.

Figure 3 shows the x-ray diffraction patterns of the $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films annealed at 700 °C for 30 min. It is found that the peak intensity of CoPt (001) and CoPt (002) peaks are very high for the CoPt/Ag film ($y=0$). This indicates that the CoPt/Ag film without adding the SiN_x possesses good perpendicular magnetic anisotropy. As the SiN_x

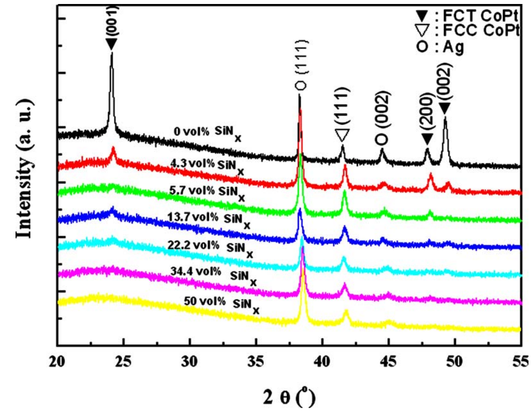

FIG. 3. (Color online) X-ray diffraction patterns of the $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films annealed at 700 °C for 30 min.

content of $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films increases, the intensities of CoPt (001) and CoPt (002) superlattice lines decrease rapidly. This implies that the SiN_x will break up the epitaxy between CoPt (001) and Ag (111), and lower the perpendicular magnetic anisotropy of the film.

Figures 4(a)–4(d) show the FEG-TEM images of CoPt/Ag, $(CoPt)_{95.7}$-$(SiN_x)_{4.3}$/Ag, $(CoPt)_{77.8}$-$(SiN_x)_{22.2}$/Ag, and $(CoPt)_{50}$-$(SiN_x)_{50}$/Ag films, respectively. The bright regions and the dark regions are SiN_x matrix and CoPt grains, respectively. The average grain sizes of Figs. 4(a)–4(d) are about 80, 50, 30, and 8 nm, respectively. For the $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films with low SiN_x contents, it contains a nonuniform distribution of CoPt particles connected to each other, as shown in Figs. 4(b) and 4(c). As the SiN_x content achieves 50 vol %, the CoPt particle sizes are decreased drastically to about 9 nm, as shown in Fig. 4(d). In this film, the CoPt grains are isolated by the SiN_x matrix and are more uniform than that of lower SiN_x contents $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films. To observe the effect of SiN_x volume percent on the coercivity (H_c) of $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films annealed at 700 °C, the changes of H_c values with SiN_x contents are shown in Fig. 5. As the

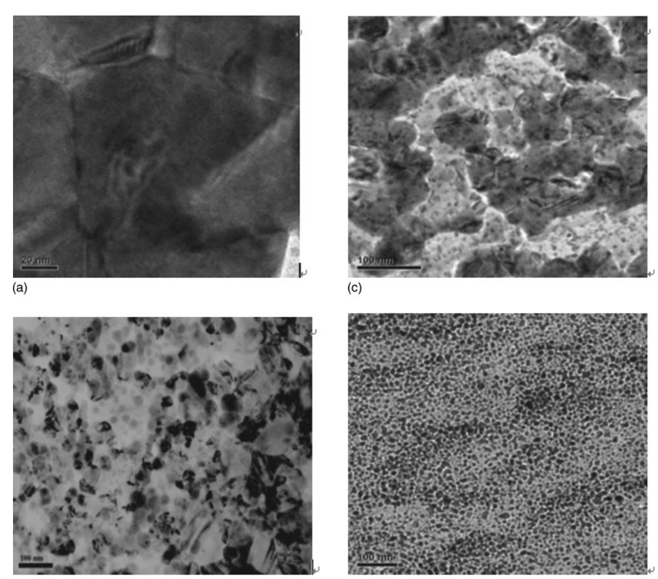

FIG. 4. FEG-TEM images of (a) CoPt/Ag, (b) $(CoPt)_{95.7}$-$(SiN_x)_{4.3}$/Ag, (c) $(CoPt)_{77.8}$-$(SiN_x)_{22.2}$/Ag, and (d) $(CoPt)_{50}$-$(SiN_x)_{50}$/Ag films.

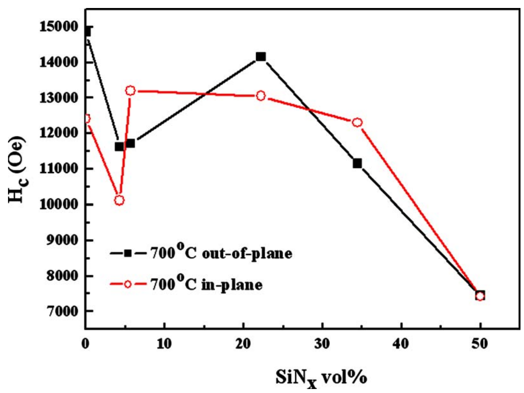

FIG. 5. (Color online) Effect of SiN_x volume percent on the H_c of $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films annealed at 700 °C.

SiN_x content is 4.3 vol % [Fig. 4(b)], the CoPt particles are isolated partially and the particle sizes are not uniform. It is suggested that a few CoPt particles below the single particle domain diameter (D_s) cause a decrease of $H_{c\perp}$ and $H_{c\parallel}$ of the $(CoPt)_{95.7}$-$(SiN_x)_{4.3}$/Ag film. When the SiN_x content is between 4.3 and 22.2 vol %, the particle size distribution of CoPt becomes more uniform. In these films, the SiN_x matrix will impede the reverse of the spin moments of CoPt. Therefore, the high $H_{c\perp}$ and $H_{c\parallel}$ values of these films are due to pinning site effect of SiN_x matrix. When the SiN_x content is higher than 22.2 vol %, the $H_{c\perp}$ and $H_{c\parallel}$ of $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films decrease rapidly when the SiN_x content is increased. The FEG-TEM image of Fig. 4(d) showed that the particle size distribution of CoPt is uniform and some particles are smaller than 8 nm. By calculating the D_p (minimal stable particle diameter) with anisotropy energy density ($K_u=3.6\times10^6$ erg/cm^3) and anisotropy field (H_k =30 kOe) of CoPt,[14] we obtained a D_p of about 8.8 nm. When the particle size is smaller than 8.8 nm, the thermal agitation effect will cause the $H_{c\perp}$ and $H_{c\parallel}$ of $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films to decrease drastically.

A further important research is to reduce the ordering temperature of CoPt/Ag film and $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films. To obtain the ordered CoPt phase at low temperatures, some researchers focused on the addition of Ag into the CoPt film to reduce the activation energy and promote the ordering process.[15,16] Moreover, in order to lower the ordering temperature of FePt films, some researchers increased the atomic diffusivity, thereby enhancing the kinetics of ordering of FePtCu films.[17,18] These studies suggest that lowering the ordering temperature of fcc to $L1_0$ FePt phase transformation can be obtained by adding Cu nanoparticles. Based on the above references, experiments involving the addition of Cu into the $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films are on-going in our laboratory. We expect that the (001) texture of $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films can be maintained and the ordering temperature can be lowered. On the other hand, the addition of Ag onto $(CoPt)_{1-y}$-$(SiN_x)_y$/Ag films will be investigated in the near future.

In summary, a large perpendicular magnetic anisotropy of a CoPt/Ag film can be obtained by adding 25 nm Ag underlayer to the CoPt (20 nm) film. The $S_\perp$, $S_\parallel$, $H_{c\perp}$, and M_s values of the CoPt (20 nm)/Ag (25 nm) film annealed at 700 °C are about 0.95, 0.35, 15 kOe, and 420 emu/cm^3, respectively. On the other hand, adding amorphous SiN_x nonmagnetic material to the CoPt film can isolate CoPt particles and reduce the particle size of CoPt film efficiently, and a uniform particle size distribution granular structure $(CoPt)_{50}$-$(SiN_x)_{50}$/Ag film with an average particle size of about 9 nm was obtained.

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[1]S. C. Chen, P. C. Kuo, S. T. Kuo, A. C. Sun, S. C. Lie, and C. Y. Chou, Mater. Sci. Eng., B **98**, 244 (2003).
[2]E. Manios, V. Karanasos, D. Niarchos, and I. Panagiotopoulos, J. Magn. Magn. Mater. **272–276**, 2169 (2004).
[3]K. R. Coffey, M. A. Parker, and J. K. Howard, IEEE Trans. Magn. **31**, 2737 (1995).
[4]M. Yu, Y. Liu, A. Moser, D. Weller, and D. J. Sellmyer, Appl. Phys. Lett. **75**, 3992 (1999).
[5]X. H. Xu, T. Jin, H. S. Wu, F. Wang, X. L. Li, and F. X. Jiang, Thin Solid Films **515**, 5471 (2007).
[6]Z. Zhang, K. Kang, and T. Suzuki, IEEE Trans. Magn. **40**, 2455 (2004).
[7]M. Daniil, P. A. Farber, H. Okumura, G. C. Hadjipanayis, and D. Weller, J. Magn. Magn. Mater. **246**, 297 (2002).
[8]C. P. Luo, S. H. Liou, and D. J. Sellmyer, J. Appl. Phys. **87**, 6941 (2000).
[9]J. Bai, Z. Yang, F. Wei, M. Matsumoto, and A. Morisako, J. Magn. Magn. Mater. **257**, 132 (2003).
[10]T. Maeda, T. Kai, A. Kikitsu, T. Nagase, and J. Akiyama, Appl. Phys. Lett. **80**, 2147 (2002).
[11]C.-M. Kuo and P. C. Kuo, J. Appl. Phys. **87**, 419 (2000).
[12]X.-H. Xu, Z.-G. Yang, and H.-S. Wu, J. Magn. Magn. Mater. **295**, 106 (2005).
[13]H. Wang, S. X. Xue, F. J. Yang, H. B. Wang, X. Cao, J. A. Wang, Y. Gao, Z. B. Huang, C. P. Yang, W. Y. Cheung, S. P. Wong, Q. Li, and Z. Li, Thin Solid Films **505**, 77 (2006).
[14]D. Weller, A. Moser, L. Folks, M. E. Best, W. Lee, M. F. Toney, M. Scgwickert, J.-U. Thiele, and M. F. Doerner, IEEE Trans. Magn. **36**, 10 (2000).
[15]C. Chen, O. Kitakami, S. Okamoto, and Y. Shimada, Appl. Phys. Lett. **76**, 3218 (2000).
[16]J. M. Hove, *Interfaces in Materials* (Wiley, New York, 1997), p. 56.
[17]Y. K. Takahashi, M. Ohnuma, and K. Hono, J. Magn. Magn. Mater. **246**, 259 (2002).
[18]X. Sun, S. Kang, J. W. Harrell, D. E. Nikles, Z. R. Dai, J. Li, and Z. L. Wang, J. Appl. Phys. **93**, 7337 (2003).