

Magnetic properties and microstructure of low ordering temperature $L1_0$ FePt thin films

A. C. Sun,^{a)} P. C. Kuo, S. C. Chen, and C. Y. Chou

Institute of Materials Science and Engineering and Center for Nanostorage Research, National Taiwan University, Taipei 106, Taiwan

H. L. Huang and J. H. Hsu

Department of Physics and Center for Nanostorage Research, National Taiwan University, Taipei 106, Taiwan

(Presented on 8 January 2004)

Polycrystalline $\text{Fe}_{52}\text{Pt}_{48}$ alloy thin films were prepared by dc magnetron sputtering on preheated natural-oxidized silicon wafer substrates. The film thickness was varied from 10 to 100 nm. The as-deposited film was encapsulated in a quartz tube and postannealed in vacuum at various temperatures for 1 h, then furnace cooled. It is found that the ordering temperature from as-deposited soft magnetic fcc FePt phase to hard magnetic fct $L1_0$ FePt phase could be reduced to about 350 °C by preheating substrate and furnace cooling treatment. The magnetic properties measurements indicated that the in-plane coercivity of the films was increased rapidly as annealing temperature is increased from 300 to 400 °C, but it decreased when the annealing temperature is higher than 400 °C. X-ray diffraction analysis shown that the as-deposited FePt thin film was a disorder fcc FePt phase. The magnetic measurement indicated that the transformation of disorder fcc FePt to fct $L1_0$ FePt phase was started at about 350 °C, which is consistent with the analysis of x-ray diffraction patterns. From scanning electron microscopy observation and selected area energy disperse spectrum analysis, the distributions of Fe and Pt elements in the films became nonuniform when the annealing temperature was higher than 500 °C due to the formation of the Fe_3Pt phase. After annealing at 400 °C, the in plane coercivity of $\text{Fe}_{52}\text{Pt}_{48}$ thin film with film thickness of 100 nm is 10 kOe, M_s is 580 emu/cm³, and grain size is about 12 nm. © 2004 American Institute of Physics. [DOI: 10.1063/1.1689762]

It is well known that order $L1_0$ FePt phase thin films have high coercivity H_c , good corrosion resistance, and very high magnetocrystalline anisotropy energy ($K_u \sim 7 \times 10^7$ erg/cm³).¹⁻³ However, the as-deposited FePt film is a soft magnetically disordered face-centered-cubic phase. The high coercivity film will be obtained by high temperature annealing treatment or substrate heating to transform the fcc FePt phase into hard magnetically ordered face-centered-tetragonal $L1_0$ FePt phase. This ordering temperature is usually higher than 500 °C.^{2,4-7} The disadvantages will be encountered during device fabrication for this high temperature treatment. In order to overcome these problems, some methods have been developed to reduce the ordering temperature of FePt film, such as the addition of a third element,⁸⁻¹⁰ multilayering,^{11,12} ion irradiation,¹³ and introduction of the underlayer.¹⁴ Most of these processes are complicated or high cost. In this work, we deposited a single FePt layer on the preheated substrate (without cap layer or underlayer) then furnace cooled, where the ordering temperature could be reduced to 350 °C. The magnetic properties, film structure, uniformity of the film composition, and grain size of the film at various annealing temperatures and film thicknesses were also investigated.

Single layer polycrystalline $\text{Fe}_{52}\text{Pt}_{48}$ alloy thin film was

deposited on a nature-oxidized silicon wafer substrate by dc magnetron sputtering at room temperature. The substrate was rotated at 45 rpm in order to get uniform film thickness and composition. A mosaic target consisting of high purity iron disk (99.99%) overlaid with high purity platinum pieces (99.99%) was used. The base pressure in the sputtering chamber is lower than 5×10^{-7} Torr. Before sputtering FePt film, the substrate was preheated at 300 °C for 1 h in order to burn out the vapor, N_2 , and CO_2 adhered on the substrate that come from the air during expose in air, to clean the substrate surface and reduce the oxygen content in SiO_x , which is on the nature-oxidized silicon wafer substrate. The preheated substrate is cooled to room temperature in the sputtering chamber. The argon pressure during depositing FePt thin film was fixed at 10 mTorr. Thickness of the FePt film was varied from 10 to 100 nm. The as-deposited film was encapsulated in a quartz tube and postannealed in vacuum at various temperatures for 1 h, then furnace cooled. The vacuum condition for postannealing is 5×10^{-6} Torr.

Magnetic properties of the FePt thin films were measured by a vibrating sample magnetometer at room temperature with maximum applied field of 13 kOe. The structure of the films was determined by x-ray diffraction (XRD). The microstructure of the film was observed by field emission scanning electron microscope (FE-SEM). The grain size of the FePt film was obtained from the XRD diffraction peak by using the Scherrer formula.¹⁵ Composition and homogeneity of the FePt film were analyzed by an energy-dispersive spec-

^{a)}Author to whom correspondence should be addressed; electronic mail: d90527009@ntu.edu.tw

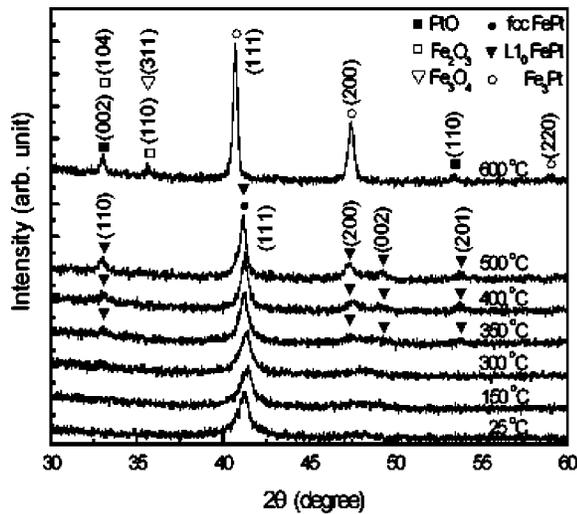


FIG. 1. X-ray diffraction patterns of the $\text{Fe}_{52}\text{Pt}_{48}$ films which annealed at various temperatures.

trum (EDS). Film thickness was measured by an atomic force microscope.

Figure 1 shows the XRD patterns of the $\text{Fe}_{52}\text{Pt}_{48}$ thin films which annealed at various temperatures. The film thickness is 100 nm. It reveals that the fcc FePt phase is dominated when $T_{\text{an}} \leq 300^\circ\text{C}$ and the $L1_0$ FePt phase appeared as $T_{\text{an}} = 350^\circ\text{C}$. The superlattice peaks (110), (111), (200), (002), and (201) of the $L1_0$ FePt phase are observed. This means that the order–disorder transformation temperature of the film starts at about 350°C . The superlattice peaks of the $L1_0$ FePt phase are more clearer as $T_{\text{an}} = 400^\circ\text{C}$. The superlattice peaks of the $L1_0$ FePt phase disappeared as T_{an} increased to 600°C and some Fe_3Pt , PtO, Fe_2O_3 , and Fe_3O_4 phases are appeared at this temperature. It is difficult to identify the peaks of PtO(002) and $\text{Fe}_2\text{O}_3(104)$, and $\text{Fe}_2\text{O}_3(110)/\text{Fe}_3\text{O}_4(311)$ peaks, because their d spacings are very close. Watanabe⁴ and Kuo *et al.*^{5,16} had used high temperature annealing ($T_{\text{an}} = 600^\circ\text{C}$) to get enough energy for the formation of the superlattice $L1_0$ FePt phase. Our experimental data show that annealing at 350°C for 1 h is enough to overcome the energy barrier for order–disorder transformation of the FePt film, if the substrate is preheated and the film is furnace cooled after being postannealed. When T_{an} is increased to 600°C , homogeneity of the FePt film will be changed and some Fe and Pt rich areas are formed in the film due to internal diffusion at high temperature. Owing to oxidation, the PtO phase is formed in the Pt rich area, and Fe_2O_3 , Fe_3O_4 , and Fe_3Pt are formed in Fe rich area.

Figure 2 shows the relation between the in-plane coercivity ($H_{c\parallel}$) and the annealing temperature (T_{an}) of various FePt films. Thickness of the FePt film was varied from 10 to 100 nm. We can see that the film has very low $H_{c\parallel}$ (<100 Oe) as $T_{\text{an}} \leq 300^\circ\text{C}$. The $H_{c\parallel}$ value increases rapidly as T_{an} is increased from 300 to 400°C . Then it decreases abruptly as $T_{\text{an}} > 400^\circ\text{C}$. The maximum $H_{c\parallel}$ value of the film occurred at $T_{\text{an}} \sim 400^\circ\text{C}$ and the film thickness is 100 nm; it is about 10 kOe. The variation of $H_{c\parallel}$ value is small as the film thickness is larger than 100 nm. When $T_{\text{an}} \leq 300^\circ\text{C}$, low $H_{c\parallel}$

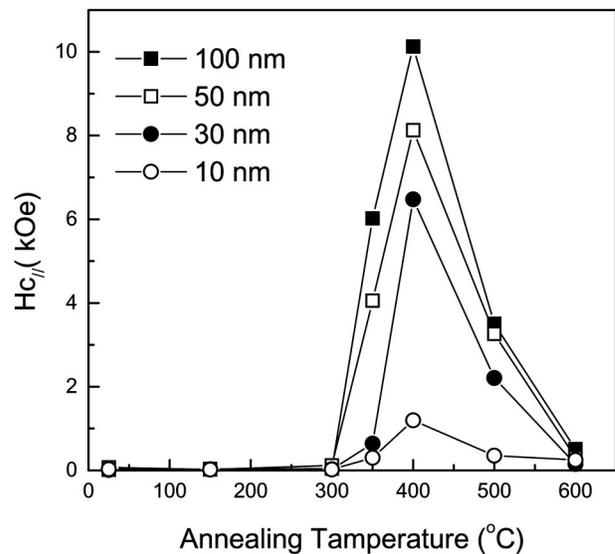


FIG. 2. The relations between the in-plane coercivity ($H_{c\parallel}$) and the annealing temperature of the FePt films with different thicknesses.

(<100 Oe) of the film is due to the film being in the soft magnetically fcc phase. As $T_{\text{an}} > 300^\circ\text{C}$, the rapid increase of $H_{c\parallel}$ is due to the rapid increase of the amount of the fct $L1_0$ FePt phase¹¹ (see Fig. 1). As previously discussed, the formation of the $L1_0$ FePt phase is usually at 600°C ,^{2,4–7} but the $H_{c\parallel}$ value is lower than 4 kOe when $T_{\text{an}} > 500^\circ\text{C}$, as shown in Fig. 2. This is due to the film being oxidized and some chemical reactions occur in the FePt film when $T_{\text{an}} > 500^\circ\text{C}$ (see Fig. 1).

Figure 3 shows the relations between the saturation magnetization (M_s) and the annealing temperature of various FePt films. The film thickness is varied from 10 to 100 nm. When $T_{\text{an}} \leq 300^\circ\text{C}$, the M_s value of the film is kept at about the same as that of as-deposited film. This means that the film was not oxidized and film structure was still a soft magnetic fcc phase as $T_{\text{an}} \leq 300^\circ\text{C}$. When T_{an} increases from

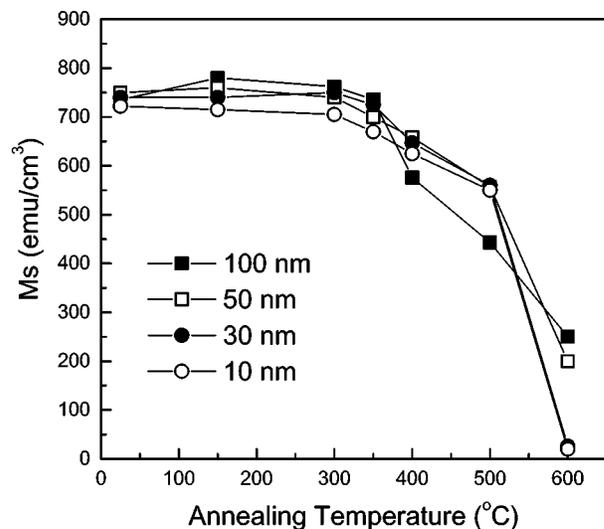


FIG. 3. The relations between the saturation magnetization (M_s) and the annealing temperature of the FePt films with different thicknesses.

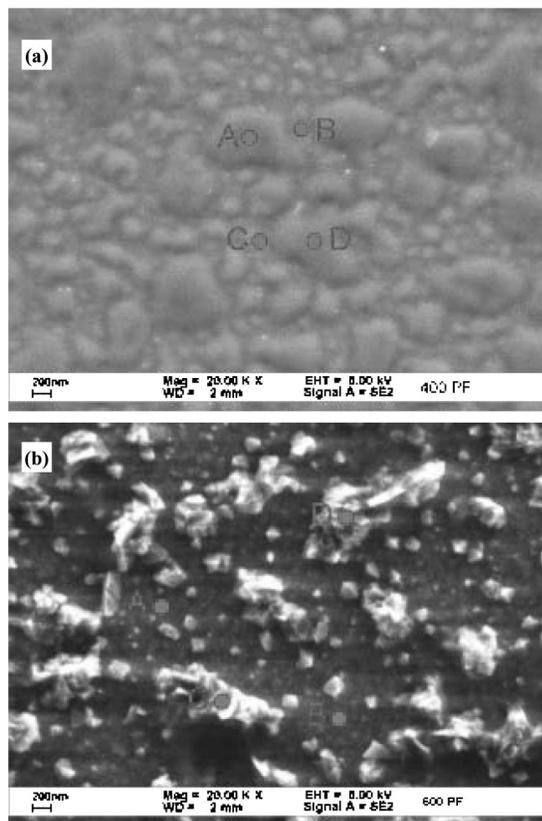


FIG. 4. The FE-SEM micrographs and selected area of EDS composition analysis of the FePt films which annealed at (a) 400 °C and (b) 600 °C.

300 to 500 °C, the M_s value decreases slowly. This is due to the disordered FePt phase being transformed gradually into an ordered $L1_0$ FePt phase, because the M_s value of the ordered $L1_0$ FePt phase is lower than that of the disordered FePt phase.¹⁷ When $T_{an} > 500$ °C, the M_s value of the film decreases abruptly. This is due to the oxidation of FePt film and produces some Fe_2O_3 , Fe_3O_4 , PtO, and Fe_3Pt phases, as shown in Fig. 1.

Figure 4 shows the FE-SEM micrographs and selected area of EDS composition analysis of the FePt film with annealing temperature of (a) 400 °C and (b) 600 °C. The film thickness is 100 nm. The mole ratios of Fe to Pt at different areas measured by EDS are shown in Table I. We can see that the mole ratios of Fe to Pt at different areas are all around 50:50 after annealing at 400 °C. But the mole ratios of Fe to Pt are changed at different areas when T_{an} is increased to 600 °C. This indicates that the internal diffusion occurs in the film at higher annealing temperature.

TABLE I. The mole ratios of Fe to Pt in the areas A, B, C, and D of Figs. 4(a) and 4(b).

	A (mole ratio) Fe:Pt	B (mole ratio) Fe:Pt	C (mole ratio) Fe:Pt	D (mole ratio) Fe:Pt
400 °C	51:49	52:48	50:50	52:48
600 °C	39:61	35:65	79:21	67:33

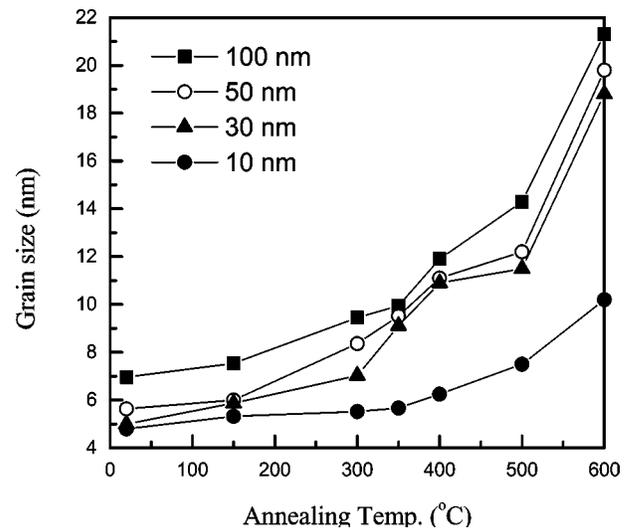


FIG. 5. Average grain sizes of the FePt films with different thicknesses as a function of annealing temperature.

Figure 5 shows the average grain size of various FePt films as a function of annealing temperature. Grain sizes of the film are calculated from the Scherrer formula¹⁵ by using the x-ray diffraction peaks of disordered FePt (111) and ordered $L1_0$ FePt (111). The grain size of the film is increased with increased annealing temperature. After being annealed at 400 °C, the average grain size of the 100 nm thick film is about 12 nm. The $H_{c\parallel}$ and M_s of this film are about 10 kOe and 580 emu/cm³, respectively, as shown in Figs. 2 and 3.

The authors are grateful to Dr. H. W. Chang for VSM measurement. This work was supported by the National Science Council and Ministry of Economic Affairs of Taiwan, R.O.C., through Grant Nos. NSC 92-2216-E-002-020 and 92-EC-17-A-08-S1-0006, respectively.

- ¹M. H. Hong, K. Hono, and M. Watanabe, *J. Appl. Phys.* **84**, 4403 (1998).
- ²T. Suzuki, N. Honda, and K. Ouchi, *J. Appl. Phys.* **85**, 4301 (1999).
- ³J. A. Christodoulides, Y. Zhang, G. C. Hadjipanayis, and C. Fountzoulas, *IEEE Trans. Magn.* **36**, 2333 (2000).
- ⁴K. Watanabe, *Mater. Trans. JIM* **32**, 292 (1991).
- ⁵C. M. Kuo, P. C. Kuo, and H. C. Wu, *J. Appl. Phys.* **85**, 2264 (1999).
- ⁶K. R. Coffey, M. A. Parker, and K. Hoear, *IEEE Trans. Magn.* **31**, 2737 (1995).
- ⁷M. R. Visokay and R. Sinclair, *Appl. Phys. Lett.* **66**, 1692 (1995).
- ⁸T. Maeda, T. Kai, A. Kikitsu, T. Nagase, and J.-I. Akiyama, *Appl. Phys. Lett.* **80**, 2147 (2002).
- ⁹T. Maeda, A. Kikitsu, T. Kai, T. Nagase, H. Aikawa, and J. Akiyama, *IEEE Trans. Magn.* **38**, 2796 (2002).
- ¹⁰Y. K. Takahashi, M. Ohnuma, and K. Hono, *J. Magn. Magn. Mater.* **246**, 259 (2001).
- ¹¹C. P. Luo and D. J. Sellmyer, *IEEE Trans. Magn.* **31**, 2764 (1995).
- ¹²Y. Endo, N. Kikucki, O. Kitakami, and Y. Shimada, *J. Appl. Phys.* **89**, 7065 (2001).
- ¹³D. Ravelosona, C. Chappert, V. Mathet, and H. Bernas, *J. Appl. Phys.* **87**, 5771 (2000).
- ¹⁴Y. N. Hsu, S. Jeong, D. E. Laughlin, and D. N. Lambeth, *J. Appl. Phys.* **89**, 7068 (2001).
- ¹⁵B. D. Cullity, *Elements of X-ray Diffraction* (Addison Wesley, Reading, MA, 1978), p. 102.
- ¹⁶C. M. Kuo, P. C. Kuo, H. C. Wu, Y. D. Yao, and C. H. Lin, *J. Appl. Phys.* **85**, 4886 (1999).
- ¹⁷T. Katayama, T. Sugimoto, Y. Suzuki, M. Hashimoto, P. de Haan, and J. C. Lodder, *J. Magn. Magn. Mater.* **104–107**, 1002 (1992).