Reduction of Grain Size and Ordering Temperature in $L1_0$ FePt Thin Films

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Single-layer polycrystalline $Fe_{52}Pt_{48}$ alloy thin films were deposited on preheated natural-oxidized (100) silicon wafer b y conventional sputtering method at room temperature. The as-deposited films are soft fcc FePt phase. After suitable temperature annealing and furnace cooling, the as-deposited films are transformed from disordered soft fcc FePt phase into ordered fct $L1_0$ FePt phase. The ordering temperature of $L1_0$ FePt phase could be reduced to about 350 °C by preheating substrate to 300 °C followed by furnace cooling treatment. The grain size of FePt was found to decrease as the ordering temperature of $L1_0$ FePt phase was reduced. After annealing at 350 °C for 1 h, the in-plane coercivity $(H_{C//})$ of the 100-nm $Fe_{52}Pt_{48}$ alloy thin film is about 6 kOe, and the average grain size is about 6 nm.

***Index Terms*—Grain size, order–disorder transformations, sputtering.**

I. Introduction

$L1_0$ FePt alloy thin film has very high magnetocrystalline anisotropy energy ($K_u \cong 7 \times 10^7$ erg/cm^3), high coercivity (H_c), high saturation magnetization (M_s), good corrosion resistance, and large energy products $(BH)_{\max}$ [1]–[3]. Its minimal stable grain size reported is about 3.0 nm [1], which is thought to be a promising material for magnetic recording media with ultrahigh recording density. However, a high temperature treatment is necessary to obtain the ordered $L1_0$ FePt phase with high K_u by using a conventional sputtering process [2], [4]. Grain growth and interlayer diffusion will occur at such high annealing temperature ($T_{\rm an}$) [5]. Hence, an approach to reduce the ordering temperature is required. So far, some approaches have been proposed to reduce the ordering temperature of the FePt film [6]–[10]. But they are complicated and expensive for mass production processes. Until now, it has been hard to reduce the ordering temperature of the $L1_0$ FePt phase effectively by using conventional sputtering method. In this investigation, we propose a more effective procedure for reducing the ordering temperature of the $L1_0$ FePt thin film. The FePt thin film was deposited on pretreated natural-oxidized (100) silicon wafer substrate at room temperature. After suitable heat treatment, the $L1_0$ FePt ordering temperature could be reduced to about 350 °C and the coercivity of the film was about 6 kOe. The average grain size was about 6 nm.

II. Experiment

Single-layer polycrystalline FePt alloy thin film was deposited on natural-oxidized (100) silicon wafer substrate by conventional magnetron sputtering with a dc power supply at room temperature. The as-deposited FePt thin film was encapsulated in a quartz tube and postannealed in vacuum (about 5×10^{-6} Torr) at various temperatures for 1 h, then furnace cooled. The deposition rate of the FePt film was about 0.3 Å/s and the film thickness was varied from 10–200 nm. Before sputtering FePt thin film, the substrate was preheated to 300 °C for 1 h in order to burn out the vapor, N_2, O_2, and CO_2 adhered on the substrate, to clean the substrate surface to reduce the oxygen content in SiO_x layer which is on the substrate. The preheated substrate was then cooled to room temperature in the sputtering chamber.

Magnetic properties of the films were measured by vibrating sample magnetometer (VSM) at room temperature with maximum applied field of 13 kOe. The phases and microstructures of the films were characterized by X-ray diffraction (XRD) with Cu-Kα radiation and transmission electron microscopy (TEM) bright field image, respectively. Composition of the films was determined by energy dispersive X-ray diffractometer (EDS). Thickness of the films was measured by atomic force microscope (AFM).

III. Results and Discussion

Fig. 1 shows the XRD patterns of 50-nm-thick FePt thin films annealed at various temperatures, followed by furnace cooling. Only one (111) peak is observed when $\mathrm{T_{an}} \leq 300$ °C. There is no evidence that ordered structure exist in the film when $\mathrm{T_{an}} \leq 300$ °C. After annealing at $T_{\rm an} \geq 350$ °C, the $L1_0$ FePt peaks (001), (110), (200), (002), and (201) appeared. This indicates that the disorderd fcc FePt phase will be transformed into the ordered $L1_0$ FePt phase at about 350 °C. Watanabe [11] and Kuo *et al.* [4] had used high temperature annealing ($T_{\rm an} = 600$ °C) to get enough energy for the formation of the $L1_0$ FePt phase. But in our investigation, annealing at 350 °C for 1 h is enough

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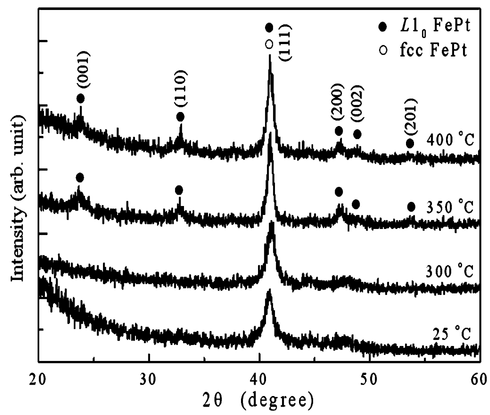

Fig. 1. X-ray diffraction patterns of 50-nm-thick FePt thin films annealed at various temperatures.

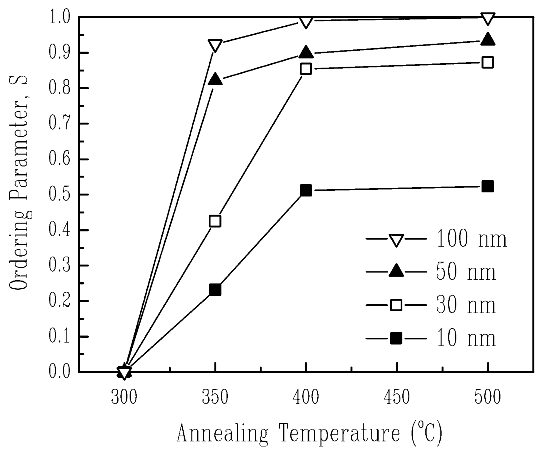

Fig. 2. Relationship between the ordering parameter (S) and annealing temperature ($T_{\rm an}$) of FePt thin films.

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.

Maeda *et al.* [6] added Cu to FePt film to increase the defects and strain energy in the film. Thus, the energy barrier of fcc FePt phase transformed into $L1_0$ FePt phase can be overcome and the transformation temperature could be lowered. On the other hand, Lai *et al.* [9] used ion irradiation to create excess vacancies into the FePt thin films. The vacancy is a point defect which can significantly enhance the diffusion and promote the formation of the ordered phase, thus, lower the transformation temperature. Here, we used the conventional sputtering system to fabricate FePt thin film. The deposition rate of FePt thin film is 0.3 Å/s. This is a high rate deposition process and easy to introduce vacancies into the FePt thin film. Because the vacancy is a kind of defect and a good diffusion path, some extra energy was introduced into the as-deposited film during deposition and reduced the order–disorder energy barrier of FePt film. Thus, $L1_0$ FePt phase could be formed after annealing at 350 °C and furnace cooling. In this study, the key points to reduce the ordering temperature to 350 °C are: 1) preheating the substrate to 300 °C for 1 h to clean the substrate surface; 2) introducing vacancies into as-deposited FePt film by high sputtering rate deposition; and 3) furnace cooling after postannealing.

Fig. 2 shows the relationship between $T_{\rm an}$ and the ordering parameter (S) of various FePt films. For the partially ordered phase, S can be calculated from the axial ratio (c/a) which is determined from the X-ray diffraction pattern. The S value is given by [8]

$$S^2 = \frac{1-(c/a)}{1-(c/a)_{sf}}$$

where $(c/a)_{sf} = 0.961$ is the axial ratio for the fully ordered phase (S_f), and (c/a) is the axial ratio of the annealed film lattice. Fig. 2 indicates that S was increased with $T_{\rm an}$ and FePt film thickness. This means that the amount of $L1_0$ FePt phase was increased with increasing $T_{\rm an}$ and film thickness. In Fig. 2, S is about zero at $T_{\rm an} = 300$ °C and increases rapidly as $T_{\rm an}$ increases to 300 °C–400 °C. The variation of S is small as $T_{\rm an} \geq 400$ °C. This means that the disordered fcc FePt phase starts to transform into $L1_0$ FePt phase at about 350 °C and transformation is completed at about 400 °C.

The magnetic hysteresis loops show that all as-deposited FePt films are soft magnetic phases. After annealing at 350 °C, the hard magnetic $L1_0$ FePt phase can be formed in the films as shown in Fig. 1. Fig. 3 shows the relation between in-plane coercivity ($H_{C//}$) and thickness of the FePt thin film at various annealing temperatures. When $T_{\rm an}$ is higher than 350 °C, ($H_{C//}$) increases with increasing film thickness. Tsoukatos *et al.* [12] had investigated the effects of film thickness on the magnetic hysteresis of CoPt films with film thickness lower than 200 nm. They found the coercivity of the films increasing with film thickness. Our experiment shows similar behavior because the amount of $L1_0$ FePt phase was increased with the FePt film thickness as the $T_{\rm an} \geq 350$ °C (see Fig. 2). On the other hand, ($H_{C//}$) is lower than 100 Oe as $T_{\rm an} \leq 300$ °C and increases rapidly when $T_{\rm an} \geq 350$ °C, as shown in Fig. 3. The higher ($H_{C//}$) value (about 10 kOe) occurred at $T_{\rm an} \sim 400$ °C with 100 nm film thickness. When the film thickness is thicker than 100 nm, the variation of ($H_{C//}$) value is small. As $T_{\rm an} \leq 300$ °C, low ($H_{C//}$) (<100 Oe) of the film is due to that the film is soft magnetic fcc phase. As $T_{\rm an} \geq 350$ °C, the rapid increase of ($H_{C//}$) is owing to the rapid increase of the amount of fct $L1_0$ FePt phase [7]. As previously discussed, the formation temperature of $L1_0$ FePt phase is usually higher than 500 °C [2], [4],

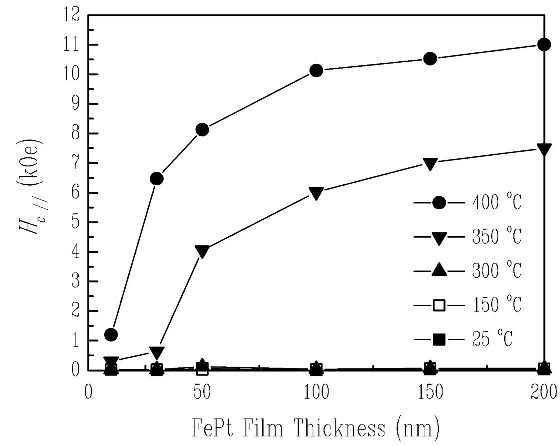

Fig. 3. Relation between in-plane coercivity ($H_{C//}$) of the FePt thin films and FePt thickness at various annealing temperatures.

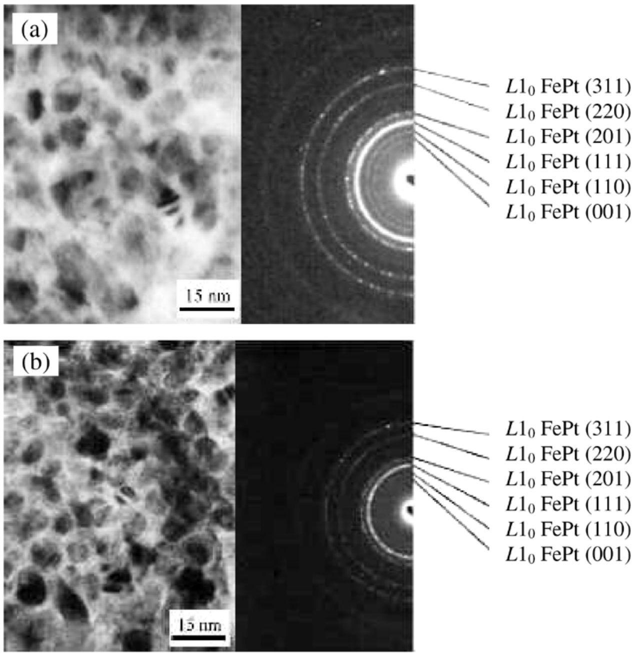

Fig. 4. TEM bright field images and electron diffraction patterns of the 100-nm films annealed at (a) 400 °C and (b) 350 °C.

[11], [12]. But in this work, the higher $(H_{C//})$ value can be obtained at $T_{\rm an} \sim 350$ °C.

Fig. 4(a) and (b) shows the TEM bright field images and electron diffraction patterns of the 100-nm films which were annealed at 400 °C and 350 °C, respectively. Both the film structures are ordered $L1_0$ FePt phase. The average grain sizes of FePt are about 9 and 6 nm in Fig. 4(a) and (b), respectively. It is well known that the grain size will decrease with reducing the annealing temperature. In Fig. 4, the grain size annealed at 350 °C is smaller than that annealed at 400 °C. The magnetic properties are also well maintained at 350 °C (see Fig. 3). Therefore, it is potentially a good candidate for future higher density magnetic recording media application.

IV. Conclusion

Single-layer polycrystalline $Fe_{52}Pt_{48}$ alloy thin films deposited on preheated natural-oxidized (100) silicon wafer by conventional sputtering method were investigation. The disordered soft fcc FePt phase started to transform into ordered fct $L1_0$ FePt phase at 350 °C. The grain size of FePt was also decreased as the ordering temperature of $L1_0$ FePt phase was reduced. After annealing at 350 °C for 1 h, the in-plane coercivity $(H_{C//})$ of the 100 nm $Fe_{52}Pt_{48}$ alloy thin film is about 6 kOe, and the average grain size is about 6 nm.

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