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Thickness dependence of structure and magnetic properties of annealed [Fe/Pt]$_n$ multilayer films

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The $L1_0$ FePt phase material has potential applications for magnetic recording and permanent magnets due to its high magnetocrystalline anisotropy energy density. The heat treatment of [Fe/Pt]$_n$ multilayer films is one approach to form the $L1_0$ FePt phase at a lower processing temperature, which is highly desirable for its applications. This paper reports the influence of film total thickness (8–100 nm) on the structure and magnetic properties of annealed [Fe/Pt]$_n$ multilayer films. A novel technique based on hollow cone dark field transmission electron microscopy is used to determine the $L1_0$ phase fraction and grain size in the annealed films. It was found that the $L1_0$ phase fraction and grain size, ordering, and magnetic properties are strongly dependent on the total film thicknesses. An $L1_0$ FePt phase fraction close to 100% is achieved for a thick [Fe/Pt]$_n$ multilayer film (100 nm) annealed at 400 °C for 1 h, while a value of only 36% is achieved for a thin film (8 nm) annealed at the same condition. In this work the $L1_0$ FePt phase nucleation density is also presented. These results suggest that the $L1_0$ phase formation is nucleation limited, and that the subsequent growth of $L1_0$ phase grains also strongly influences the structure and magnetic properties of the annealed films. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3073842]

I. INTRODUCTION

The ordered $L1_0$ FePt phase has potential applications for high-density magnetic recording media and high-energy permanent magnets due to its large magnetocrystalline anisotropy energy density ($K_u \sim 7 \times 10^7$ ergs/cm^3), high Curie temperature (~ 480 °C), and large saturation magnetization ($\mu_0 M_s \sim 1.43$ T).[1] FePt films with the stoichiometric composition of 50 at. % Fe that are deposited at room temperature have a disordered fcc structure, which has a low anisotropy energy density and therefore lacks attractive hard magnetic properties. Postannealing at high temperature is necessary to induce the disorder (fcc) to order ($L1_0$) phase transformation. The heat treatment of [Fe/Pt]$_n$ multilayer thin films is a promising approach to form the $L1_0$ FePt phase at a reduced temperature, which is highly desirable for many of the applications.[2,3]

The study of the film thickness dependence of $L1_0$ FePt formation and magnetic properties is of interest, as different applications of this material require different film thicknesses. For example, films with a thickness less than 10 nm are desirable for magnetic recording medium. For magnetic microelectromechanical system applications, however, films thicker than several hundred nanometers or several micrometers are usually necessary. Furthermore, understanding of the $L1_0$ FePt formation in films with different thicknesses is also helpful to make clear the mechanism of the reactions in [Fe/Pt]$_n$ films upon annealing.

In cosputtered FePt alloy films, it has been reported that the structure and magnetic properties of annealed films are dependent on the film thickness.[4–8] For example, Berry and Barmak[4] modeled the time-temperature-transformation reaction diagram of fcc$\rightarrow L1_0$ phase transformation based on differential scanning calorimetry experiments, which, along with the experimentally measured $L1_0$ phase fraction of annealed FePt films (10 nm thick) by Ristau and co-workers,[9,10] demonstrated that the phase transformation is more difficult in thin FePt films (10 and 50 nm) than in thick films (1 μm). Other studies,[5–8] primarily based on magnetic properties, have reported an increase in coercivity with increasing thickness, implying a similar thickness dependence.

In this paper, the film thickness dependence of the structure and of the magnetic properties of annealed [Fe3.6 nm/Pt4.7 nm]$_n$ ($n=1, 3, 6, 9, 12$) multilayer films are reported. The $L1_0$ phase formation reaction in [Fe/Pt]$_n$ multilayers is generally believed to be different from that of FePt alloy films due to the existence of a long distance compositional gradient. In this paper, a method based on hollow cone dark field (HCDF) transmission electron microscopy (TEM) imaging was applied to examine the volume fraction and grain size of the $L1_0$ FePt phase in the annealed films. This technique, along with other measured results of the structure and magnetic properties, gives a clearer understanding on the influence of film thickness in [Fe/Pt]$_n$ multilayer films.

II. EXPERIMENTS

The samples sputtered for the investigation are listed in Table I. It can be noted that all films have similar composition and the same bilayer periodicity. A relatively large bilayer periodicity is chosen based on our previous studies of periodicities less than 50 nm which demonstrated that a

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TABLE I. A list of samples sputtered at room temperature for investigation.

Short ID	Film structure	Composition	Thickness (nm)
"e1"	$[\text{Fe3.6 nm/Pt4.7 nm}]_1$	$\text{Fe}_{49.0}\text{Pt}_{51.0}$	8
"e2"	$[\text{Fe3.6 nm/Pt4.7 nm}]_3$	$\text{Fe}_{49.0}\text{Pt}_{51.0}$	25
"e3"	$[\text{Fe3.6 nm/Pt4.7 nm}]_6$	$\text{Fe}_{49.0}\text{Pt}_{51.0}$	50
"e4"	$[\text{Fe3.6 nm/Pt4.7 nm}]_9$	$\text{Fe}_{49.5}\text{Pt}_{50.5}$	73
"e5"	$[\text{Fe3.6 nm/Pt4.7 nm}]_{12}$	$\text{Fe}_{49.5}\text{Pt}_{50.5}$	100

larger $L1_0$ phase fraction, a higher long-range order parameter, and a higher coercivity can be achieved for $[\text{Fe/Pt}]_n$ multilayer films with this larger periodicity, given equal annealing conditions.[11]

All samples were sputtered onto Si (100) substrates having a surface layer of 200 nm of thermally grown SiO_2. The deposition rate for Pt is 0.31 Å/s and that for Fe is 0.48 Å/s. The deposition rate of Fe and Pt was characterized with Rutherford backscattering spectroscopy. All depositions were performed in 4 mTorr of Ar+3% H_2 and controlled by an automated system. A deposition chamber pressure in the 10^{-8} Torr range was obtained prior to film deposition. The film samples were annealed in a tube furnace in 1 atm of flowing Ar+5% H_2 process gas. The plan view TEM samples were prepared using a back-etch technique, thinning initially with HF+HNO_3 solution and subsequently thinning with ion milling.[12] The microscopy was performed in a Tecnai F30 microscope with a field emission gun operating at 300 kV. The magnetic properties were measured with an alternating gradient force magnetometer (AGFM, Princeton Measurements Corp. MicroMag Model 2900) with a maximum field of 20 kOe.

III. RESULTS AND DISCUSSION

Figure 1 shows the HCDF TEM images of samples annealed at 400 °C for 1 h. The images were formed using the 001 and 110 superlattice reflections of $L1_0$ FePt phase, and the illuminated grains indicate the $L1_0$ FePt phase grains with 001 and 110 orientations. Clearly evident in the figure is the increase in $L1_0$ phase grain size with the increase in film thickness, which can be understood from the common scal-
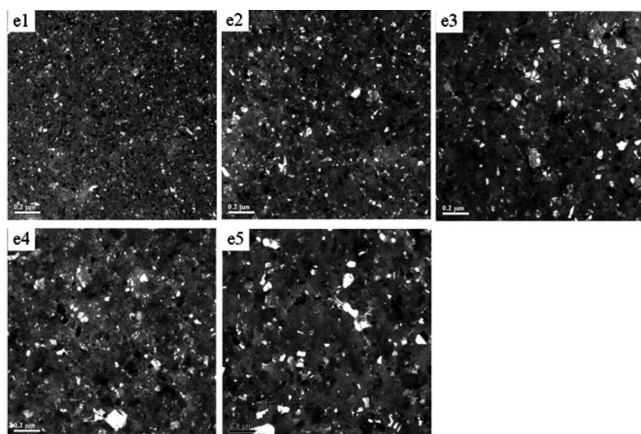

FIG. 1. The HCDF TEM images using 001 and 110 reflections of $L1_0$ FePt phase for samples e1, e2, e3, e4, and e5 annealed at 400 °C for 1 h.
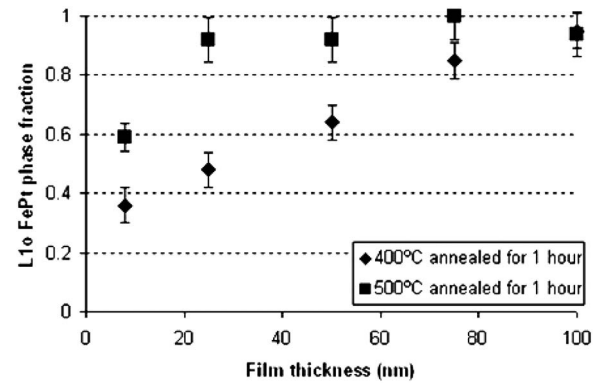

FIG. 2. The $L1_0$ FePt phase fraction of vs film thickness for samples e1, e2, e3, e4, and e5 annealed at 400 and 500 °C for 1 h. The volume fraction was examined using a technique based on HCDF TEM imaging. Briefly, the illuminated grain areas of HCDF TEM images of specific pair of reflections (such as 001 and 200) are used to extract the volume fraction (Ref. 14).

ing of grain size with the film thickness. Grain growth in thin films generally starts in both the film thickness direction and in-plane directions to form a columnar structure, wherein single grains extend from the top to the bottom surface of the film. When the in-plane grain size becomes nearly equal to film thickness, further grain growth is inhibited. Higher annealing temperatures are needed to provide adequate mobility for further in-plane grain growth.[13] For the $[\text{Fe/Pt}]_n$ multilayer films with different thicknesses, it is observed, as expected, that the grain growth of $L1_0$ FePt phase in the thicker films is stronger and results in larger grains than those in the thinner films.

Figure 2 shows the $L1_0$ FePt phase fraction versus film thickness of multilayer samples annealed at 400 and 500 °C for 1 h. The volume fraction was examined using a technique based on the HCDF TEM imaging. Briefly, the illuminated grain areas of HCDF TEM images of certain specific reflections (such as 001 and 200) are used to extract the volume fraction. The uncertainty from the film texture, image processing, and analysis has been considered, and details of this technique can be found elsewhere.[14] Figure 2 shows the strong thickness dependence of $L1_0$ phase fraction in the annealed films. A phase fraction close to 100% was achieved for sample "e5" annealed at 400 °C for 1 h, which indicates a nearly complete transformation to the $L1_0$ FePt phase in thicker $[\text{Fe/Pt}]_n$ films. On the other side, however, the $L1_0$ FePt phase fraction is only 36% in sample $[\text{Fe−Pt}]_1$, sample "e1," annealed at the same condition. Based on the measured $L1_0$ phase fraction and its average grain size, the $L1_0$ phase grain density (or nucleation density) can be estimated as 4.6×10^{17} cm^{-3} for sample e1 annealed at 400 °C, and 5.2×10^{17} cm^{-3} for the sample annealed at 500 °C.

The increase in $L1_0$ FePt volume fraction with film grain size (or thickness) indicates that the $L1_0$ FePt phase formation is nucleation controlled. Specifically, the $L1_0$ nucleation events are limited to sites that are significantly more widely spaced than the bilayer periodicity. In the course of the reaction, the limited number of $L1_0$ FePt nuclei formed give a small fraction of product phase and it is the subsequent growth of these $L1_0$ FePt grains, at the expense of fcc FePt
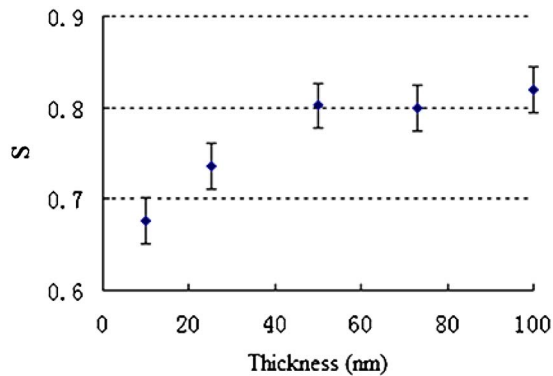
FIG. 3. (Color online) The long-range order parameter vs film thickness of samples e1, e2, e3, e4, and e5 annealed at 400 °C for 1 h. The S of samples e2, e3, e4, and e5 were derived from the integrated intensity of 001 and 002 XRD peaks, while the ordering of sample e1 was determined indirectly using the c/a ratio of TEM SADP.

FIG. 4. (Color online) The M-H curves for samples e1, e2, e3, e4, and e5 annealed at 400 °C for 1 h.

phase, that increases the $L1_0$ volume fraction. Obviously, the stronger grain growth behavior in thicker films is helpful to accomplish the phase transformation.

A decrease in the volume fraction transformed with decreasing sample thickness is consistent with relatively simple models of the phase transformations by nucleation and growth.[4,15] However, for quantitative use, these models require a uniform growth rate for the transformed phase, which we do not observe due to the tendency for grain growth to stagnate when the grain size becomes comparable to film thickness.

The long-range order parameter (S) of samples annealed at 400 °C for 1 h is shown in Fig. 3, as derived from the integrated intensity of 001 and 002 x-ray diffraction (XRD) peaks.[16] The S for sample "e1" was determined indirectly from the (c/a) ratio[17] from the $L1_0$ superlattice peaks of TEM select area diffraction pattern (SADP), due to the difficulty to derive a high quality XRD pattern for an 8 mm thickness. The (c/a) ratio for fully ordered sample ($S=1$) was taken as 0.964. The XRD instrument was calibrated using the standard Si powders provided by NIST. A slight increase in S along with film thickness was observed, which indicates that the grain growth of $L1_0$ FePt phase grains is also helpful to increase its degree of order.

Figure 4 shows the M-H loops of samples annealed at 400 °C for 1 h. An increase in coercivity with film thickness is apparent, although it should be noted that the range of magnetic field available may be insufficient to ensure saturation of the samples. The increased coercivity is not surprising, as it is expected that a larger fraction of ordered phase and a greater extent of chemical order will provide an increased magnetic anisotropy and coercivity in polycrystalline thin films.

IV. CONCLUSIONS

This paper reports our investigation on the influence of film total thickness (8–100 nm) on the structure and mag-
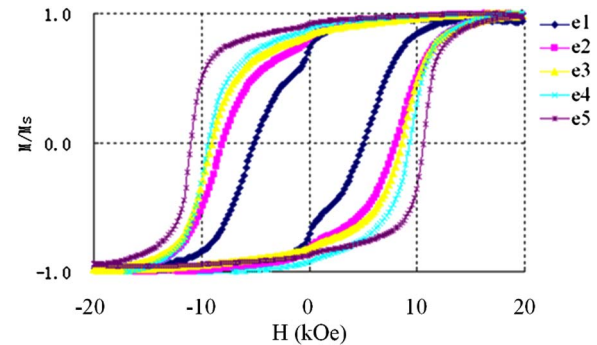
netic properties of annealed $[Fe3.6 \text{ nm}/Pt4.7 \text{ nm}]_n$ multilayer films. A novel technique based on HCDF TEM is used to determine the $L1_0$ phase fraction and grain size in the annealed films. It was found that the $L1_0$ phase fraction and grain size, ordering, and magnetic properties are strongly dependent on the total film thickness, i.e., the number of bilayer periods. An $L1_0$ FePt phase fraction close to 100% is achieved for a thick $[Fe3.6 \text{ nm}/Pt4.7 \text{ nm}]_n$ multilayer film (100 nm) annealed at 400 °C for 1 h, while a value of only 36% is achieved for a thin film (8 nm) annealed at the same condition. In this work the $L1_0$ FePt phase nucleation density is also presented. These results suggest that the $L1_0$ phase formation is nucleation limited, and that the subsequent grain growth strongly influences the structure and magnetic properties of the annealed films.

[1] R. Skomski, J. Phys.: Condens. Matter **15**, R841 (2003).
[2] Y. Endo, N. Kikuchi, O. Kitakami, and Y. Shimada, J. Appl. Phys. **89**, 7065 (2001).
[3] Y. Endo, K. Oikawa, T. Miyazaki, O. Kitakami, and Y. Shimada, J. Appl. Phys. **94**, 7222 (2003).
[4] D. C. Berry and K. Barmak, J. Appl. Phys. **101**, 014905 (2007).
[5] B. C. Lim, J. S. Chen, and J. P. Wang, J. Magn. Magn. Mater. **271**, 431 (2004).
[6] F. Michael, J. Appl. Phys. **93**, 15 (2003).
[7] P. T. L. Minh, N. P. Thuy, and N. T. N. Chan, J. Magn. Magn. Mater. **277**, 187 (2004).
[8] S. K. Chen, F. T. Yuana, S. N. Hsiaoa, H. C. Changa, C. Y. Lioua, and W. C. Chang, J. Magn. Magn. Mater. **282**, 198 (2004).
[9] K. Barmak, R. A. Ristau, K. R. Coffey, M. A. Parker, and J. K. Howard, J. Appl. Phys. **79**, 5330 (1996).
[10] R. A. Ristau, K. Barmak, L. H. Lewis, K. R. Coffey, and J. K. Howard, J. Appl. Phys. **86**, 4527 (1999).
[11] B. Yao and K. R. Coffey, J. Appl. Phys. **103**, 07E107 (2008).
[12] B. Yao and K. R. Coffey, J. Electron Microsc. **57**, 47 (2008).
[13] C. V. Thompson, Annu. Rev. Mater. Sci. **20**, 245 (1990).
[14] B. Yao and K. R. Coffey, "On the phase quantification of annealed $[Fe/Pt]_n$ multilayer films," J. Appl. Phys. **105**, 033901 (2009).
[15] R. W. Balluffi, S. M. Allen, and W. C. Carter, *Kinetics of Materials* (Wiley, Hoboken, NJ, 2005), pp. 537–538.
[16] K. Barmak, J. Kim, L. H. Lewis, K. R. Coffey, M. F. Toney, A. J. Kellock, and J.-U. Thiele, J. Appl. Phys. **98**, 033904 (2005).
[17] W. Roberts, Acta Metall. **2**, 597 (1954).