Atomic Ordering and Coercivity Mechanism in FePt and CoPt Polycrystalline Thin Films

Sangki Jeong, Yu-Nu Hsu, David E. Laughlin, and Michael E. McHenry

***Abstract*—40 nm thick CoPt and FePt films were prepared on oxidized Si substrates with 10 nm MgO underlayers. The maximum coercivity (H_c) for CoPt films was found to be ~10 kOe after annealing at 700 °C for ~20–30 minutes (min). Structural analysis showed a significant amount of FCC phase as well as the ordered $L1_0$ phase in these films. FePt films showed an abrupt increase of ordered volume fraction and H_c in the initial stage of annealing and predominance of the tetragonal $L1_0$ phase after 10 min. at 700 °C. The maximum H_c reached ~16 kOe after annealing at 700 °C for more than 20 minutes. Dark field (DF) images of the annealed CoPt films showed individual grains which exhibited a possibility of several variants or disordered phase with dimensions similar to the exchange correlation length, b_{cm}. The temperature dependence of H_c seems to indicate a weak pinning mechanism in the highly ordered FePt films. Magnetic force microscopy indicated a complex domain structure consisting of clusters with dimensions of several hundred nanometers.**

***Index Terms*—Atomic ordering, coercivity mechanism, CoPt and FePt, thin films.**

I. Introduction

FePt AND CoPt thin films have potential application for many magnetic devices such as high density recording media, magnetic bias films of magneto-resistive elements, and magnetic tips for magnetic force microscopy because of their high magnetic anisotropy, high H_c, and good corrosion resistance [1]–[3]. The source of high coercivity in these alloys has been of great interest [3]–[5]. It has been reported that the presence of finely dispersed mixture of the ordered and disordered phases is responsible for the high H_c in bulk alloys [3]–[5]. Other reports on thin films suggested that H_c values increase with the proportion of the ordered volume fraction until full ordering is achieved [3]. It is of great interest to study coercivity and reversal mechanism in nanocrystalline films since grains are smaller than the critical single domain size and the nanostructure within a grain may differ from that of bulk magnets [5]. It is interesting to simultaneously investigate the kinetics of atomic ordering, the nanostructure, magnetic properties, and their interrelationship to develop an understanding of the coercivity mechanism. It has been found that a polycrystalline MgO underlayer film is beneficial in the control of the c-axes orientations of $L1_0$, CoPt, and FePt films. Interestingly, the CoPt films with the thickness >20 nm exhibited predominance of in-plane c axes and consequently in-plane anisotropy [6]. Here the details of nanostructure and atomic ordering as well as a possible coercive mechanism of 40 nm thick FePt and CoPt films with an MgO underlayer are discussed based on structural and microstructural observations, magnetic measurements, and previous theoretical work. On the other hand, the very thin films (~5 nm) showed preferential growth of c axes in the perpendicular to the film surface. The characterization of 5 nm films with perpendicular anisotropy is presented in [7].

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II. Experimental Procedure

All specimens were fabricated by RF diode sputtering on oxidized Si substrates (growth temperature ~100 °C). An alloy target of CoPt, an MgO target, and a FePt composite target were used to synthesize the films. The chemical compositions of the films were found to be close to $Co_{46}Pt_{54}$ and $Fe_{55}Pt_{45}$ by an X-ray fluorescence method. Rapid thermal annealing was applied in an Ar atmosphere (heating rate: 100 °C/sec, cooling rate: ~100 °C/min.). Magnetic properties were characterized by Vibrating Sample Magnetometry (VSM) and SQUID Magnetometry, with maximum fields of 15–50 kOe. The structure and microstructure were examined by X-ray diffraction (XRD) (Cu–Kα) as well as transmission electron microscopy (TEM). The morphology and magnetic microstructure of the samples were examined by atomic force microscopy (AFM) and magnetic force microscopy (MFM).

III. Results and Discussion

We have prepared 40 nm thick polycrystalline FePt and CoPt films with an MgO underlayer (10 nm thick). Fig. 1 presents XRD spectra and TEM micrographs of CoPt and FePt films. Detailed discussions on conventional $\theta/2\theta$ scans of XRD patterns and electron diffraction patterns (EDP) can be found in [6]. In-plane $\theta/2\theta$ scans make it possible to generate similar intensity profiles to EDP which can distinguish more quantitatively the perpendicular and in-plane structural variants based on analysis of the (001) and (110) super-lattice reflections. The tetragonality, c/a shows ~0.98, ~0.97 in CoPt and FePt films, respectively.

The CoPt films do not show clear peak splitting of FCC (200) into $L1_0$ peaks in XRD spectra even after annealing at 700 °C for 30 minutes. The intensity ratio of I [$L1_0$ (001), (110)] / I [FCC (200), $L1_0$ (200), (002)] is about ~0.3 based on in-plane $\theta/2\theta$ scans. The theoretical values of I_{001}/I_{002} (in-plane variants) and I_{110}/I_{200} (perpendicular variants) for complete

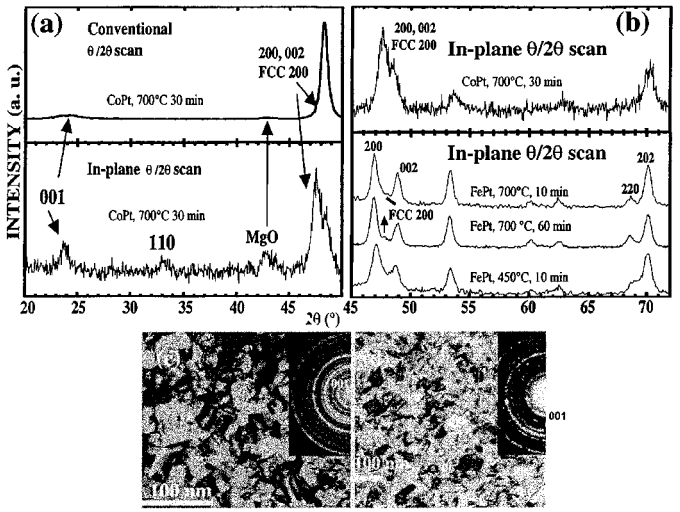

Fig. 1. XRD spectra of annealed (a) CoPt 40 nm/MgO 10 nm films (conventional $\theta/2\theta$ scan and in-plane $\theta/2\theta$ scan, in-plane $\theta/2\theta$ scan: grazing incident angle of 2°) and (b) CoPt and FePt 40 nm/MgO 10 nm films (in-plane $\theta/2\theta$ scans, grazing incident angle of 2°). SAD patterns and bright field (BF) images of (c) CoPt 40 nm/MgO 10 nm (annealed at 700 °C for 30 min.) and of (d) FePt 40 nm/MgO 10 nm (annealed at 700 °C for 30 min).

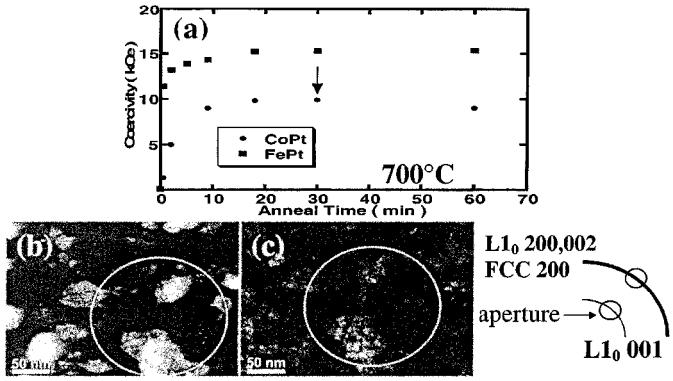

Fig. 2. (a) Anneal time versus H_c of the (CoPt, FePt) 40 nm/MgO 10 nm films at 700 °C. And the centered DF images of the CoPt film annealed at 700 °C for 30 minutes [designated by an arrow in (a)], using (b) {200} rings and (c) (001) ring.

ordering are ∼1.9 and ∼0.78 in CoPt, respectively (based on structure, multiplicity, Lorentz-polarization, temperature, and absorption factors). Considering the fraction of perpendicular and in-plane structural variants [from (001) and (110) peaks [6], in-plane variants ∼>70–85%], $(I_{001}+I_{110})/I_{200,002}$ becomes ∼1.4–1.5. Therefore, the order parameter (S) seems to be <0.3. Here, the assumption is that the Lorentz-polarization factor in the conventional $\theta/2\theta$ scans can be applied to the in-plane $\theta/2\theta$ scans. SAD patterns in the same samples in the TEM seem to show $L1_0$ (400) and (004) as well as FCC (400) rings, although it is difficult to identify. FePt films showed clear peak splitting of FCC (200) reflection into $L1_0$ (200) and (002) as well as small intensity of FCC (200) peaks within 10 minutes at 700 °C. This makes it easier to do more quantitative analysis on ordering. FePt films annealed at 450 °C for 10 minutes have H_c of ∼10 kOe. In these films, in-plane XRD indicates a significant amount of ordered volume fraction, based on the peak splitting discussed above [Fig. 1(b)]. A small fraction of FCC phase seems to remain based on the ratio of peak intensity of FCC (200) and $L1_0$ (200) ($I_{FCC200}/I_{L10200} < 0.1$) after annealing for 60 minutes at 700 °C in FePt films. Therefore, it is believed that CoPt films show slower kinetics of ordering and have a significant amount of FCC volume fraction in films annealed at 700 °C for 30 minutes. The FePt films showed faster kinetics on atomic ordering and a small amount of FCC remains even after longer annealing. The reason for faster kinetics of ordering in FePt films is not understood completely. However, the FePt alloys do have a large driving force for ordering at 700 °C than do the CoPt alloys because FePt has a larger T_c (critical temperature) [3]. The grain size (10–50 nm, average grain ∼30 nm) of CoPt films is smaller than that (10–100 nm) of FePt films after the same annealing process [Fig. 1(c) and (d)].

Fig. 2 illustrates the relation between H_c and annealing time at 700 °C [along with DF TEM images of the films designated by an arrow in Fig. 2(a)]. These were taken using a (001) super-lattice and (200) FCC fundamental and $L1_0$ (200) and (002) reflections, respectively [the $L1_0$ (002), (200) and FCC (200) peaks are indistinguishable]. H_c reaches ∼10 kOe within ∼20 min. and did not change appreciably by 30 minutes in CoPt films (a mixture of FCC and $L1_0$ phase). H_c of FePt films increased abruptly within a few minutes and remained constant after ∼20 minutes. Annealing for longer times (>1 hour) in both films leads to the reduction in H_c. FePt films exhibit a maximum H_c at the time when the films become mostly ordered. BF TEM images indicate a grain size that is still smaller than the critical monodomain size [8]. Fig. 2 also shows that within a grain we observe intensity fluctuations attributed to strain, remaining FCC phase or other structural variants. DF images illuminating the same area [Fig. 2(b) and (c)] show different sized grains in CoPt films annealed at 700 °C for 30 minutes. This is attributed to structural variants or FCC phase within the inside of a grain. Although more detailed observations are required to determine the orientation of the other variants, some important aspects can be concluded in these films. According to the so-called spring exchange model of [9], the exchange correlation length (b_{cm}) is defined as $b_{cm} = \pi(A_s/2K_H)^{1/2}$, where A_s is the exchange energy of the soft magnetic phase and K_H is the magnetocrystalline anisotropy constant of the hard magnetic phase. To observe a sufficiently strong exchange coupling, the grain size of the soft magnetic phase should be smaller than $2b_{cm}$. The above TEM observation indicated that subgrains are still smaller than $2b_{cm}$ (∼10 nm), which is consistent with the single loop behavior in most of films although there is a significant fraction of FCC phase.

The coercive mechanism of bulk CoPt and FePt has been discussed by several authors [3]–[5]. In a polytwinned microstructure, high H_c can be attributed to the pinning of magnetic domains at twin or anti-phase boundaries (APBs) [5]. Bulk magnets usually show maximum H_c of <∼10 kOe in these materials [3]–[5]. Our films (FePt) show much higher H_c (∼16 kOe as a maximum value). BF TEM images show that APBs are hardly visible in our films. Structural twins are visible, but it is difficult to estimate their density within a grain. The fact that H_c reaches a maximum and remains constant for longer annealing, indicates

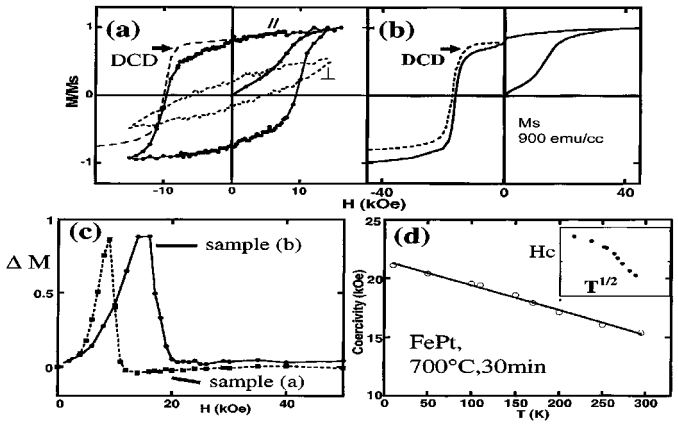

Fig. 3. (a) Hysteretic responses of FePt 40 nm/MgO 10 nm films annealed at 450 °C for 10 minutes, Dotted lines (designated by an arrow) represent remanent DC Demagnetiztion (DCD) curves. //: in-plane, ⊥: perpendicular hysteresis. (b) Initial, demagnetization, and DCD curve of FePt 40 nm/MgO 10 nm films annealed at 700 °C for 30 min.(c) ΔM curves of sample (a) and (b). (d) Temperature dependence of H_c of sample (b).

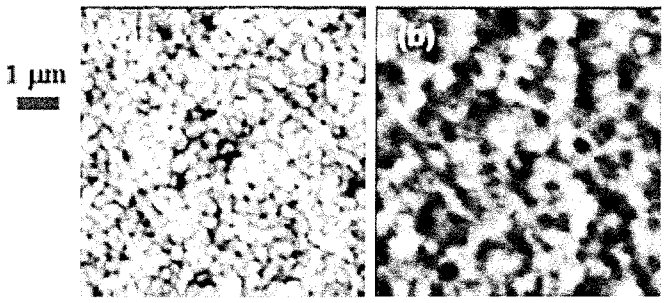

Fig. 4. (a) AFM and (b) MFM images of FePt 40 nm/MgO 10 nm films. Annealed at 700 °C for 30 min. [sample (b) in Fig. 3], Size: 5 μm × 5 μm.

that grain growth and increasing $L1_0$ volume fraction are not essential to obtaining the highest H_c. Apparently, above a certain $L1_0$ phase volume fraction, H_c reaches maximum value and does not increase for longer times of annealing.

Fig. 3 shows the magnetic properties of FePt films. The remanent demagnetization curve in Fig. 3(a) shows a reversible behavior typical of a spring-exchange nanocomposite [9]. However, highly ordered films with large grains exhibit constricted hysteresis loops due to a small fraction of FCC phase. This decreases the amount of recoil in the remanent demagnetization though the ordered grains remain strongly coupled [Fig. 3(b)]. The reason for the decreased coupling between ordered and disordered phase is not clear and further detailed characterization is required. Therefore, irreversible switching behavior exhibits sharp transitions in both cases and consequently collective switching due to strong coupling between grains [Fig. 3(c)]. Since these films show strong exchange coupling [based on ΔM curves Fig. 3(c)], the Stoner–Wohlfarth model can not adequately explain the magnetic response. The initial magnetization is the pinning type behavior in CoPt films [6] and close to a pinning type in highly ordered FePt films. The temperature (T) dependence of highly ordered FePt samples demonstrates the possibility of weak pinning mechanism based on the linear relationship (the best fit) between the H_c and T in Fig. 3(d) (H_c versus $T^{1/2}$ is plotted in Fig. 3(d) as well but no linear relationship exists) [7], [10]. However, lack of data on the temperature dependence of K_1 and M_s makes it difficult to do further analysis. FCC regions can be nucleation sites and domain walls can be pinned in this soft phase or at planar defect boundaries. H_c can be determined by the density of pinning sites and pinning field [5], [11]. On the basis of domain pinning theory [11], the maximum H_c occurs when the domain wall thickness (w) is equal to the size of defect (d) in the influence of the pinning field. The thickness of domain wall in these materials is much larger than the size of planar defects such as structural twins, APB's and grain boundaries. Based on Reference [5] and [11] (Krönmuller type analysis), the estimated H_c would be $<\sim 0.1\ H_K$ in the case of d/w $\sim$0.1. The theory based on APBs pinning predicts H_c $\sim$1–10 kOe in FePt [5]. Therefore, the coercive mechanism of FePt films would be somewhat different from conventional pinning theory which can be applied to CoPt films. MFM and simultaneous AFM of the highly ordered FePt films indicated a complex domain structure consisting of clusters with dimensions of several hundreds nanometers, which seems to consist of interaction domains aligned along the boundaries of the polycrystalline aggregates.

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