Nanostructure and magnetic properties of polycrystalline FePdPt/MgO thin films

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Atomic ordering, nanostructure and the magnetic properties of polycrystalline FePdPt thin films deposited with MgO underlayers have been investigated. The film thickness was fixed at 5 nm in all films, a thickness where perpendicular anisotropy is dominant. Films deposited at ambient temperature were postannealed at various temperatures to study the kinetics of atomic ordering. The Pd additions to FePt were effective in reducing the temperature needed for atomic ordering. The mean grain size determined by transmission electron microscopy is about 10 nm in annealed films. Within the limits of our observations, FePdPt films do not show phase separation into more than one phase of the $L1_0$ structure. The maximum coercivity (H_c) for annealed FePdPt films is about ~3000–4000 Oe. *In situ* ordered FePdPt films at 530 °C show no indication of a lower temperature for atomic ordering. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1453328]

$L1_0$ materials are of current interest for magnetic media application because of their high uniaxial magnetocrystalline anisotropy, K_u, and high saturation magnetization, M_s, which make it possible to decrease the magnetic layer thickness for better writing and high recording resolution.[1] There have been many reports on large anisotropy of $L1_0$ FePt and CoPt thin films.[2–4] We have found that a polycrystalline MgO underlayer is beneficial in the control of the *c*-axes orientations of $L1_0$ CoPt and FePt films.[5] FePd is known to order at lower temperatures than FePt.[6,7] In addition, it has similar M_s (1100 emu/cc) but much smaller K_u ($\sim 1.8\times 10^7$ erg/cc) and H_c than for FePt.[6,7] Therefore, Pd additions to FePt can provide lower ordering temperatures than in FePt, high M_s and also mechanisms for the easy tuning of H_c for adequate overwrite[3] in the recording process by controlling K_u. However, there have been few reports on this ternary alloy in the literature.

In this article, the structure and magnetic properties of polycrystalline FePdPt films on MgO underlayers are investigated. The comparison between the FePt and FePdPt will be performed based on the same underlayer structure. The composition of FePdPt was selected to be near the equiatomic composition in the pseudo-binary system, FePd–FePt (near Fe_2PdPt).

All specimens discussed here were fabricated by rf diode sputtering on oxidized silicon substrates. The sputtering pressure of the Ar gas was in the range of 10–50 mTorr. Rapid thermal annealing (RTA) as a postannealing procedure was applied in an Ar atmosphere. Magnetic properties were measured using alternating gradient force magnetometry, in fields up to 13–14 kOe. Structural analysis was performed using x-ray diffractometry (Cu $K\alpha$). The analysis of the microstructure was accomplished via transmission electron microscopy (TEM). The chemical compositions of the films were found to be close to $Fe_{53}Pd_{24}Pt_{23}$ and $Fe_{55}Pt_{45}$[5] as determined by energy dispersive x-ray analysis and x-ray fluorescence method.

To investigate the possibility of phase separation in the thin film, 20-nm-thick FePdPt films on oxidized Si substrates were synthesized at ambient temperature. The films subsequently were annealed by RTA for 30 min at 650 °C. Based on previous reports and recent results, the ternary $L1_0$ films may be susceptible to phase separation into two $L1_0$ phases.[8,9] In-plane XRD and selective area diffraction (SAD) patterns for RTA annealed FePdPt films are shown in Fig. 1. These show a clear peak separation [Fig. 1(a)] and SAD

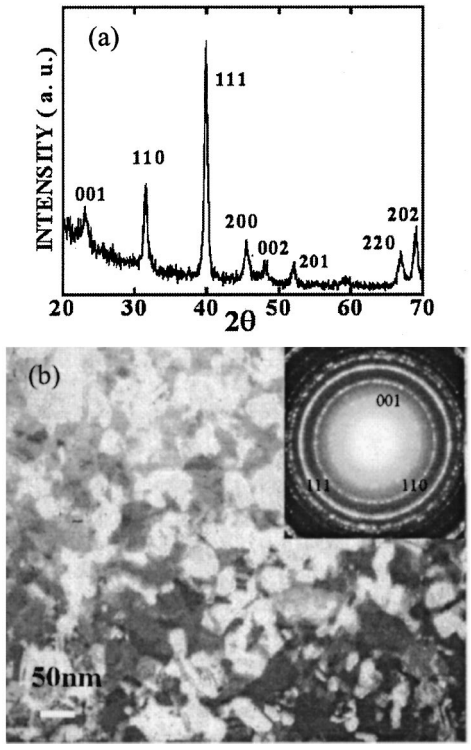

FIG. 1. (a) XRD patterns and (b) bright field image and SAD patterns for (a) FePdPt 20 nm/(on oxidized Si substrate) after RTA process for 30 min.

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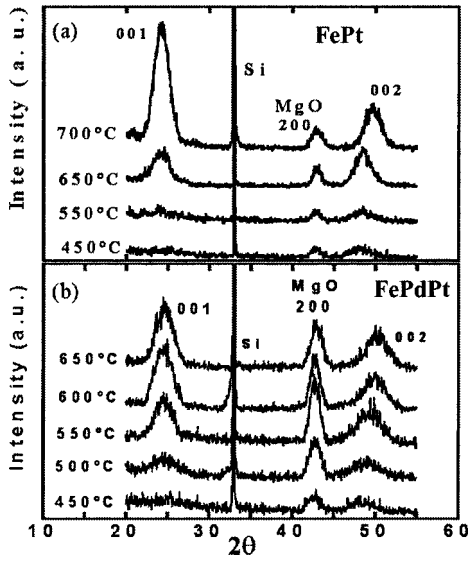

FIG. 2. XRD patterns after RTA process for 10 min at various temperatures (a) for FePt 5nm films/MgO 10 nm and (b) for FePdPt 5 nm / MgO 10 nm films.

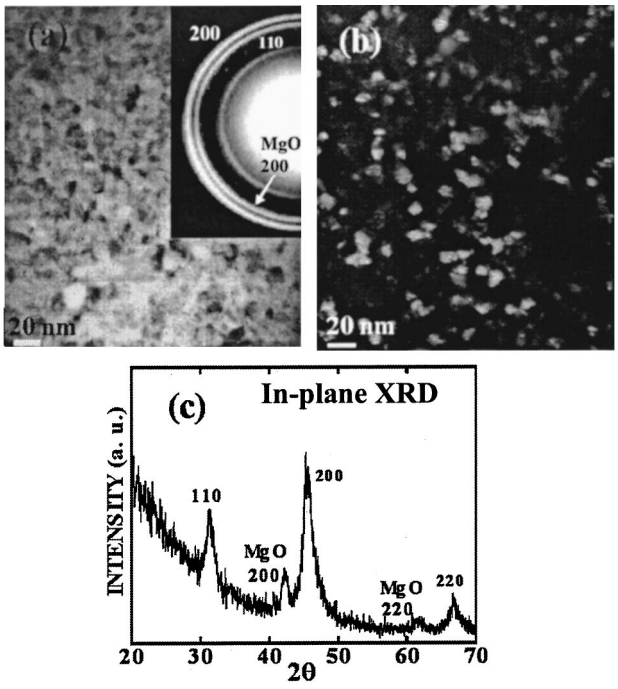

FIG. 3. (a) SAD patterns, bright field (BF), (b) dark field (DF) images and (c) in-plane XRD spectra ($\theta/2\theta$ scan with grazing incidence beam of 2°) for FePdPt 5 nm/MgO 10 nm films after RTA at 650 °C for 18 min.

patterns in TEM and also confirm the clear separation of (200) and (002) peaks [Fig. 1(b)]. This implies that these films consist primarily of a single $L1_0$ phase, without a significant amount of fcc phase. In the bright field image, there is no indication of phase separation of $L1_0$ phase to within the spatial resolution of our observations. This is in contrast to observations of fine fringes with the length scale of ~6 nm due to two $L1_0$ phase modulation in the bulk materials.[9] A detailed study on the phase separation based on the theoretical and experimental work in bulk FePdPt is presented elsewhere.[9] Long-time annealing might be used to induce the phase separation in thin films. However, in fine-grained films prepared with short annealing times it is not kinetically favorable to achieve equilibrium.

We have prepared a 5-nm-thick polycrystalline FePdPt with a 10 nm MgO underlayer and a SiO_2 capping layer. Figure 2 shows XRD data illustrating the evolution of the $L1_0$ phase starting from the fcc disordered structure in these FePdPt films. For comparison, 5 nm $Fe_{55}Pt_{45}$ films with an MgO underlayer were produced. The films were rapid thermally annealed (RTA) at various temperatures for a fixed time of 10 min. The calculated I_{001}/I_{002} (R) intensity ratio and the measured I_{001}/I_{002} ratio in the XRD patterns can be used to estimate the approximate order parameter in $L1_0$ structure, considering the product of the structure and Lorentz-polarization factors.[4,5] Pd and Pt atoms in $L1_0$ FePdPt are considered to be randomly distributed in the second lattice site of the $L1_0$ superstructure. The FePt/MgO films exhibit a fully ordered $L1_0$ phase based on I_{001}/I_{002} after RTA at 700 °C for 10 min [Fig. 2(a)].[5] FePdPt films were found to be close to the full ordering after RTA at 600 °C for 10 min [Fig. 2(b)]. There is no standard experimental data for fully ordered FePdPt available for comparison, therefore it is difficult to confirm the order parameter. However, longer annealing more than 10 min or higher temperature annealing than 600 °C in FePdPt films does not increase either I_{001}/I_{002} [Fig. 2(b)] or H_c, supporting the assertion that the ordering process has been completed at a nearly fully ordered phase within 10 min at 600 °C.

Figure 3 shows the SAD patterns, bright and dark field images in TEM for the samples annealed at 650 °C for 18 min. The mean grain size is about 10 nm from the dark field image by (200) reflection [Fig. 3(b)]. SAD patterns as well as the in-plane XRD with grazing incident beam of 2° confirmed the perpendicularly oriented (001 oriented) *c* axes.

Figure 4(a) shows the magnetic hysteresis of the FePdPt samples annealed at 650 °C for 18 min, showing H_c of

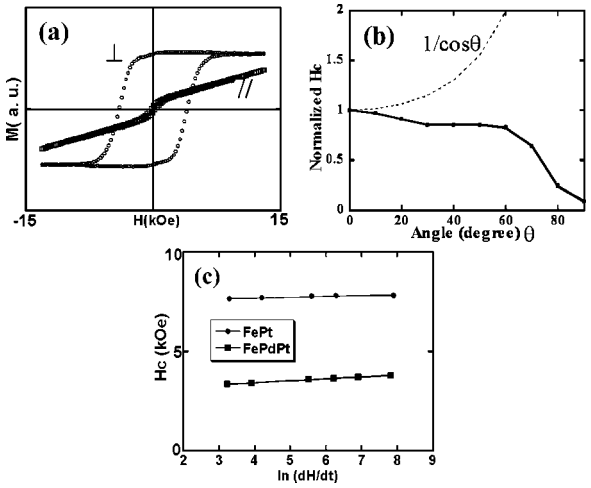

FIG. 4. (a) Hysteresis loops and (b) angular dependence of coercivity for FePdPt 5 nm/MgO 10 nm films after RTA at 650 °C for 18 min. Angle is defined between the magnetic field and perpendicular direction to film plane (c) Coercivity vs field sweep rate for the fully ordered FePt 5 nm/MgO 10 nm and FePdPt 5 nm/MgO 10 nm films, annealed (RTA) at 700 and 650 °C for 18 min, respectively.

~3500 Oe. The samples annealed below 600 °C showed H_c to be smaller than 1000 Oe. The fully ordered samples annealed at >600 °C for 10–18 min show a H_c of ~3500 Oe. Longer annealing does not help increase the H_c. The M_s is estimated to be ~900–1000 emu/cc, consistent with bulk values. Strong perpendicular anisotropy in the fully ordered FePdPt films was obtained and an approximately perpendicular anisotropy field ($H_{K_\perp}$) is found by extrapolation to be about 20 kOe. The measured anisotropy of 20 kOe is much smaller than that observed for fully ordered FePt thin films (~40 kOe) previously reported in the literature.[10] From a consideration of the demagnetization energy ($K_u = K_\perp + 2\pi M_s^2$),the K_u is estimated to be close to $\sim 1.4\times 10^7$erg/cc. Therefore, the intrinsic anisotropy field (H_{Ku}) is predicted to be ~30 kOe. The angular dependence of H_c shows a large deviation from that predicted for a conventional domain wall pinning mode (where a $1/\cos\theta$ dependence is predicted), as shown in Fig. 4(b). Here it is seen that H_c in FePdPt films decreases as the angle (θ) between the applied field and easy axis increases. Further, a slight increase in H_c was detected as the angle θ increased to ~70° in FePt films.[5] This implies that grains in FePdPt films reverse more independently during the magnetization reversal process[11,12] than in FePt films.[5]

The switching volume (V^*) of the films can be evaluated by the measurement of the dependence of the H_c on the sweep rate of the applied field.[13] Since the presence of the demagnetizing field causes the internal field to change following the variation of the magnetization during the measurement, the measurement of the variation of the magnetization with time in the presence of a steady negative field (time-decay method) to determine the H_c cannot be applied to the films with perpendicular anisotropy.[14] Here we have measured the coercivities with the various sweep rates (25–2700 Oe/s), to determine the V^* for the fully ordered FePt and FePdPt films. As seen in Fig. 4(c), the H_c displays a linear relationship with the logarithm of the sweep rate. From the slope of this plot, we obtain the switching volumes. The estimated V^* for the fully ordered FePdPt films is $\sim 0.3\times 10^{-18}$ cm^3 which is close to the mean grain volume. A V^* of $\sim 10^{-18}$ cm^3 was determined for the fully ordered FePt films from the field rate dependence of the H_c. This method is thought to give more reliable results than those obtained by time decay method.[10] This V^* for the fully ordered FePt films is larger than the physical volumes of most of the grains in FePt films (average grain ~10 nm).[10] The measured H_c increases (4%–20%) with increasing sweep rate over the range obtainable with our apparatus in FePt and FePdPt films [Fig. 4(c)]. This indicates that thermal activation aids the reversal process in those thin films. The inference from this observation is that FePt films are more thermally stable than the FePdPt films due to their higher K_u as well as larger V^*.

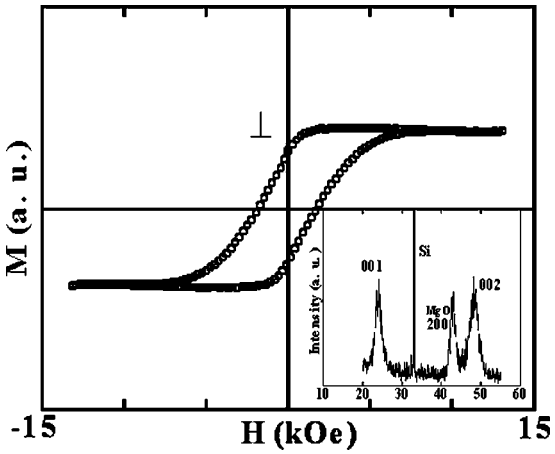

FIG. 5. Hysteresis loop (perpendicular to films) and XRD pattern (the inset figure) for *in situ* ordered 5 nm FePdPt films with an MgO underlayer (10 nm) on Si substrate at 530 °C.

We have also deposited 5 nm FePdPt films at the elevated temperature of ~530 °C with a MgO underlayer. As seen in Fig. 5, H_c is found to be ~1800 Oe. The inset figure in Fig. 5 shows the XRD patterns, indicating that there is no indication of lowering the ordering temperature by *in situ* deposition as compared to the postannealing (Fig. 2). FePt has a significant reduction in ordering temperature by *in situ* deposition.[4]

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