Structure and magnetic properties of $L1_0$ CoPt(Ag/MgO,MgO) thin films

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Disordered fcc (face-centered-cubic) [100] fiber-textured CoPt thin films with thicknesses of 20–40 nm were deposited on a nonmagnetic sputtered MgO seed layer with and without a Ag intermediate layer. These were subsequently annealed at 600–750 °C for 5–10 min using rapid thermal annealing (RTA). The structural variants were determined to coexist in the in-plane and normal to the plane directions after the RTA process. The MgO underlayer without the intermediate layer revealed strong in-plane anisotropy by magnetic hysteresis measurement. Selected area diffraction (SAD) by TEM and XRD measurements for these samples confirmed the preferential growth of in-plane variants of *c* axis compared to the perpendicular direction. High remanence and coercive squareness (S^*) were obtained due to strong in-plane texture and especially due to exchange coupling among the grains. Evidence for this was provided by ΔM curves. Initial magnetization curves for samples annealed above 600 °C, showed the possibility of assembled single domain or domain pinning mechanism but with negligible free wall motion inside of the grains. [S0021-8979(00)61008-3]

CoPt and FePt thin films have potential applications in extremely high density recording[1] where their large anisotropy energy density provides a barrier to thermally activated switching of the magnetization. Many results on materials with the $L1_0$ structure have been reported by several authors.[2,3] It is well known that strong (111) textured films are developed without any underlayer.[4] Larger S^*, coercivity and exchange-decoupled small grains would be required to achieve a sharp bit transition and low noise. In-plane anisotropy has been developed in conventional recording media using nonmagnetic underlayers. It is certain that an in-plane texture can provide the higher remanence and S^* compared with 3D (three-dimensional) random orientation or that with an out-of-plane component.[5] There are reports on (001) oriented *in situ* ordered films showing that perpendicular anisotropy can be achieved by deposition of $L1_0$ materials on MgO substrates.[2,3] However, there are few reports[6] on the development of in-plane anisotropy for longitudinal recording media using these $L1_0$ systems.

In this paper, we present the results of experiments aimed at controlling the easy axis orientation in $L1_0$ CoPt films. The in-plane oriented CoPt polycrystalline thin films have been produced through the use of appropriate underlayers.

All films were prepared by rf diode sputtering on one-inch silicon (100) coupons, with the pressure of Ar gas of 3–10 mTorr. An alloy target (nominal composition of 50 at% Pt) was used to produce the CoPt thin films. The substrate temperatures were maintained at 200–250 °C. The post annealing process was performed by rapid thermal annealing (RTA) in an Ar atmosphere for periods of 5–10 min. Magnetic properties were measured using vibrating sample magnetometry (VSM), alternating gradient force magnetometry (AGFM), and SQUID (superconducting quantum interface device) magnetometry in fields of 1.2–5 T. Structural and microstructural studies were made using x-ray diffractometry (Cu–$K\alpha$, 35 kV, 20 mA) and transmission electron microscopy (TEM). The chemical composition of the films was measured by x-ray fluorescence.

In this study we prepared $Co_{46}Pt_{54}$ films with thicknesses of 20–40 nm. To induce a [100] fiber texture in the disordered fcc CoPt based on lattice parameter considerations, 10–40 nm thick nonmagnetic underlayers of Ag–MgO or MgO were deposited prior to the deposition of the magnetic CoPt films. As shown in Fig. 1, an ordered CoPt (001) superlattice reflection is observed in annealed samples, indicative of the tetragonal $L1_0$ phase. The orientation of the (001) axes of the transformed $L1_0$ crystals is important in determining the usefulness of these materials as longitudinal media, as compared with perpendicular recording media.

The [100] fcc fiber texture in the precursor films gives rise to three possible structural variants in the transformed tetragonal materials. This leads to three possible orthogonal orientations of the *c* axes of the tetragonal phase, two in the plane and one normal to the plane. XRD (x-ray diffraction) measurements can be used to identify the transformed $L1_0$ phase in the films. We can identify the fractions of in-plane and perpendicular variants for the transformed ordered phase by comparing the peak intensity of (200) and (002) reflections of $L1_0$ structure. However, the c/a of 0.98 and (200) peak of remaining fcc phase make it difficult to distinguish these peaks if they have broadened enough to significantly overlap. For samples annealed below 700 °C, this is the case as illustrated in Fig. 1.

TEM electron diffraction patterns can be used to distinguish between the perpendicular and in-plane scattering based on analysis of the (001) and (110) superlattice reflections which are well separated. The proportion of the easy (*c*) axes in the direction normal to the plane of the film, based on the relative intensities of the (200) and (002) x-ray reflec-

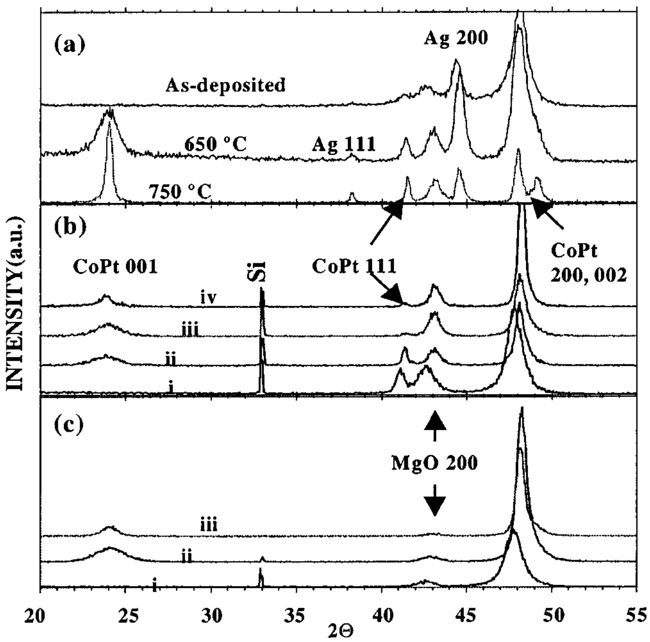

FIG. 1. XRD spectra for (a) CoPt 40 nm/Ag 40 nm/MgO 40 nm (8 min of RTA); (b) CoPt (20,40) nm/MgO 40 nm (8 min of RTA). From the bottom are (i) as-deposited 40 nm CoPt, (ii) annealed at 650 °C, (iii) 20 nm CoPt annealed at 650 °C, and (iv) 20 nm CoPt annealed at 750 °C; (c) CoPt 40 nm/MgO 10 nm (8 min of RTA), (i) as-deposited films, (ii) films annealed at 650 °C, and (iii) films annealed at 750 °C.

tions can be determined from the data in Fig. 1(a). However, the severe overlapping of these peaks and the possibility of the remaining fcc (200) peak make it difficult to determine the ratio between the perpendicular and in-plane variants for the samples annealed at temperature below 750 °C. The samples annealed at 750 °C. The samples annealed at 750 °C showed a clear splitting of (200) and (002) peaks and consequently the significant amount of perpendicular variants together with in-plane variants. The films without Ag showed no splitting of these peaks and much smaller intensity of the (001) peaks even after the higher temperature annealing as seen in Fig. 1(b). These samples revealed a small shoulder near the (002) peak. This is attributed to an incomplete ordering or dominant in-plane *c* axes.

As seen in Fig. 1(c), an improved in-plane texturing was obtained for CoPt films with 10 nm MgO underlayer (without a Ag intermediate layer). Figure 2 shows TEM results for these films annealed at 650 °C for 8 min using the RTA process. A strong (001) ring is observed in the SAD pattern.

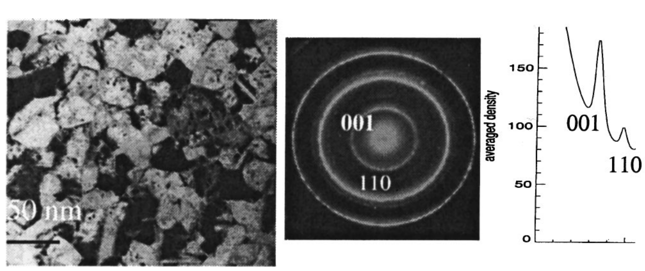

FIG. 2. Bright field image and SAD patterns of a 40 nm CoPt/10 nm MgO film after 8 min of RTA at 650 °C. Radial function of average peak intensity was mapped from SAD patterns by an IDL routine.

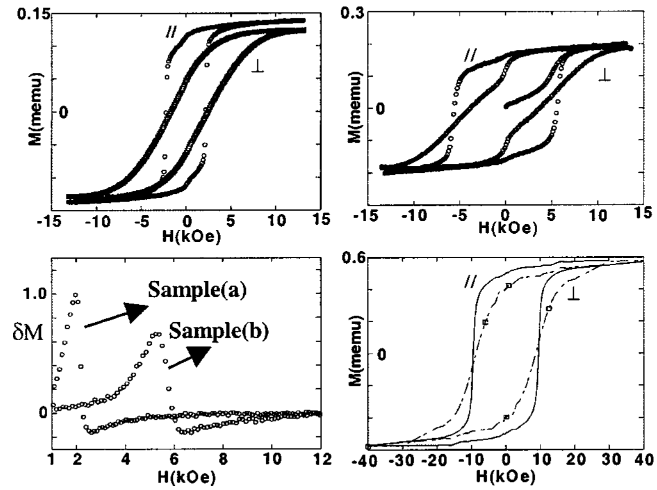

FIG. 3. Hysteresis loops for 40 nm CoPt/40 nm Ag/40 nm MgO films annealed at (a) 600 °C (8 min of RTA); (b) 650 °C (8 min of RTA), including initial magnetization after ac demagnetization in plane direction; (c) ΔM of samples (a) and (b); (d) 700 °C, 8 min of RTA.

The (110) reflection ring that comes from the perpendicular structural variant is very weak but nonvanishing. This means that a large proportion of the tetragonal *c* axes are in-plane and a small fraction of *c* axes along the direction perpendicular to the films. The local integrated intensity[7] of an (hkl) electron diffraction ring is proportional to $P_{hkl}|f_{hkl}|^2 d_{hkl}{}^2 V$, where P_{hkl}, f_{hkl}, d_{hkl}, and V denote the multiplicity factor of the (hkl) planes, the atomic scattering factor of the (hkl) planes, the *d*-spacing of the (hkl) planes and the volume of the diffracting crystals, respectively. For the case of equal volume fractions of the [001] and [100] oriented grains in films with the $L1_0$ structure, we obtained the following equation:

$$\left(\frac{I_{(001)}}{I_{(110)}}\right)=\left(\frac{P_{001}}{P_{110}}\right)\left(\frac{|f_{001}|}{|f_{110}|}\right)^2\left(\frac{d_{001}^2}{d_{110}^2}\right)\approx 1.11. \quad (1)$$

We have calculated the ratio of the average peak intensity of the (001) and (110) rings and found it to be greater than 7. This means that the volume fraction of the in-plane variants of the ordered phase is greater than 85%. In the case of CoPt–Ag–MgO films, the (110) rings showed much higher intensity for the samples annealed with the same conditions. From Fig. 2 we also see that the grain size of the CoPt films after an 8 min RTA at 650 °C is in the range of 10–50 nm.

Magnetic data are illustrated in Figs. 3 and 4. The in-plane and perpendicular coercivities were similar for the CoPt–Ag–MgO films except for the case of Fig. 3(b) which is not completely understood. This might be due to relatively different volume fractions of the perpendicular texture component depending on annealing temperature. As seen in Fig. 4(a), 40 nm CoPt/10 nm MgO films exhibited much larger coercivity for applied fields oriented with in-plane directions, consistent with the strong in-plane anisotropy. Figure 4(b) shows CoPt films with a thick MgO underlayer exhibits a more open hysteresis loop. This is attributed to the existence of a random distribution of *c* axes based on the (111) peaks in XRD and SAD patterns. Similar effects have been dis-

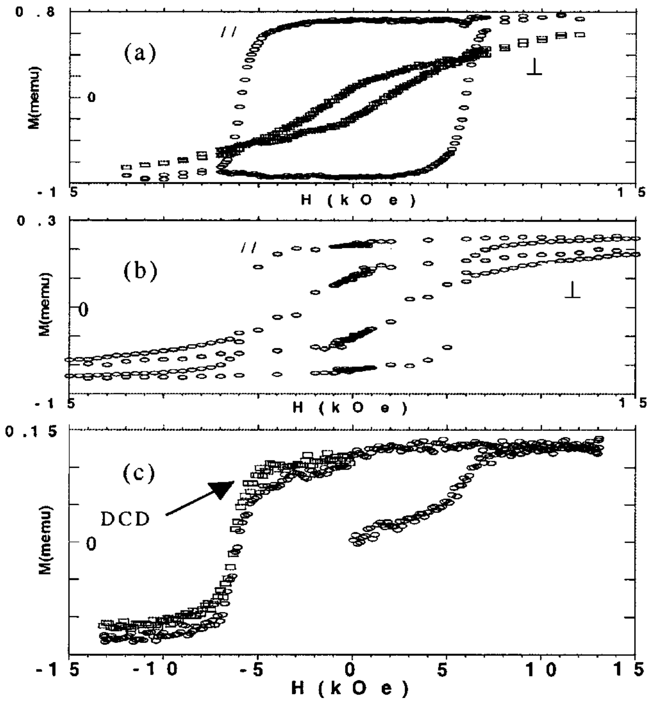

FIG. 4. Hysteresis loops for (a) 40 nm CoPt/10 nm MgO; (b) 40 nm CoPt/40 nm MgO after 8 min of RTA at 650 °C; (c) initial and demagnetization curve after ac demagnetization for 20 nm CoPt/40 nm MgO films, in-plane direction, 8 min of RTA at 650 °C.

cussed in a previous study of MgO films by Li-Lien Lee.[8] The ΔM curve of Fig. 3 reveals strong exchange coupling between the grains that contributes to the large switching volume and consequently the large S and S^* for samples annealed above 600 °C. Figure 3(c) shows the peak value of ΔM curve decreases with an increase of the annealing temperature. Based on these results, it is possible that the underlayer is influencing the cubic to tetragonal phase transformation. The elucidation of the detailed transformation mechanism is the subject of future work.

From the observation of grain size, we believe that the grains are single domain (>0.17 μm for the fcc phase). Therefore, the hysteretic response may be dominated by rotation mechanisms including the strong possibility of incoherent rotation due to intergranular interactions and the smaller exchange length of the ordered phase. However, the possibility of three variants within individual grains makes it necessary to consider a subgrain structure which makes analysis of the reversal mechanism more complicated. Based on the initial magnetization curve[9] of Figs. 3(b) and 4(c), the reversal process for the samples annealed above 600 °C can be divided into two regimes. At low fields, the susceptibility is relatively small, indicating free domain wall motion does not occur. In higher fields, close to the coercive field, the irreversible susceptibility increases strongly. This behavior shows that either the films consist of an assembly of single domain crystallites with no domain walls, or that domain wall pinning occurs at the grain boundaries. However, there would still be a possibility that several mechanisms co-exist depending on the grain size and the details of the complicated nanostructure within the grains.

It has been reported that field annealing of FePd showed preferential alignment of c axes with the applied field directions.[10] This can be attributed to forced magnetostriction ($\lambda_{100}<0$).[10] In the case of CoPt, a previous report indicates that the sign of λ_{100} is positive.[11] However, it was reported that directional ordering occurred and produced an induced anisotropy in the field direction.[12] Another report says that the field only induced more ordering.[13] We have annealed the CoPt–Ag–MgO (40 nm each) films in 5–7 kOe applied field using a VSM oven with an inert Ar atmosphere. The temperature was slowly increased to ~500 °C and held for 1 h to promote nucleation of the ordered phase. The temperature was then increased and held at 600 °C for 1 h ($T>T_c$). The in-plane coercivity was observed to be ~4 kOe. The coercivity in the perpendicular direction was observed to be ~2 kOe. However, when we annealed these samples using the same thermal cycle without a field, the results were almost the same as those of the field annealed samples within experimental error. In comparison with RTA annealing protocals, the above thermal processing routes did lead to larger in-plane coercivities for the same samples.

In summary, a sputtered polycrystalline MgO underlayer has shown promise in promoting enhanced in-plane magnetic anisotropy in CoPt thin films. SAD and XRD patterns confirmed the preferential in-plane orientations of c axes.

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