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Short communication

Orientation and magnetic properties of FePt and CoPt films grown on MgO(1 1 0) single-crystal substrate by electron-beam coevaporation

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Abstract

We have studied the orientation and magnetic properties of FePt and CoPt films deposited by electron-beam co-evaporation on MgO(1 1 0) single-crystal substrates at substrate temperatures from 500 to 700 °C. We observed that long-range chemical ordering of the $L1_0$ structure occurred over the entire range of substrate temperatures in FePt films and at 600 °C and up in CoPt films. Growth of FePt and CoPt yielded epitaxial films with cube-on-cube orientation of the pseudo-cubic $L1_0$ lattice with respect to the cubic MgO. X-ray diffraction patterns and magnetization loops of the FePt and CoPt films revealed the existence of $L1_0$ domains with the tetragonal c axis inclined at 45° to the film plane, orientations (0 *h h*) and (*h* 0 *h*), as well as $L1_0$ domains with the tetragonal c axis in the plane of the film, orientation (*h h* 0). The FePt and CoPt films for which X-ray diffraction indicated tetragonal phase was present all exhibited hard magnetic properties with easy axis along the [0 0 1] substrate direction as well as large in-plane magnetocrystalline anisotropy.

Keywords: Epitaxial film; L10 phase; Magnetic film

1. Introduction

The growth of epitaxial FePt and CoPt films is of great interest since Fe_xPt_{1-x} and Co_xPt_{1-x} films (with $x \approx 0.5$) can form the $L1_0$ tetragonal, chemically ordered (CuAu I) phase, which has high saturation magnetization and large magnetocrystalline anisotropy [1,2]. The anisotropy fields are as high as 11.6 and 12.3 T for bulk FePt and CoPt, respectively. The structure of the $L1_0$ ordered phase is based on a face centered cubic (fcc) lattice with alternating pure Fe (or Co) and Pt planes stacked along one of the ⟨0 0 1⟩ directions. This produces a tetragonal distortion and *P*4/*mmm* symmetry along the ⟨0 0 1⟩ directions and the possibility of three orientations of the $L1_0$ ordered phase defined by the three axes along which the fcc lattice can order. In order to obtain the $L1_0$ phase ordered along a single direction, growth and/or annealing must occur in the presence of an external field that breaks the symmetry, e.g. an external magnetic field or mechanical constraint [3].

Epitaxial magnetic hard layers can form the basis of bilayer soft magnet/hard magnet exchange-coupled systems. Such systems can provide ideal one-dimensional models for studying the influence of different magnetic and physical properties, e.g., magnetic parameters and interface condition, on exchange coupling [4]. Combinatorial synthesis and measurement techniques can provide an efficient means to effect such studies; such techniques have recently been used to study the inter-phase exchange coupling in Fe/Sm–Co polycrystalline bilayers with gradient Fe thickness [5]. The magnetic properties of FePt and CoPt $L1_0$ films make them excellent choices for the epitaxial hard layer, development of which could facilitate research in exchange coupled nanocomposite magnets and ultrahigh density magnetic recording [6,7].

A large area of homogeneous and epitaxial hard magnetic film is required to serve as the substrate for deposition of the array of soft magnetic films in order to employ combinatorial fabrica-

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tion and analysis techniques. Epitaxial FePt and CoPt thin films have been fabricated by different thin film growing techniques. Farrow et al. grew epitaxial $L1_0$ FePt(1 1 0) on a MgO(1 1 0) substrate by means of molecular beam epitaxy (MBE); nearly complete chemical ordering was achieved for growth on a substrate at 500 °C, with an easy magnetization axis along the [0 0 1] substrate direction [8,9]. Epitaxial cube-on-cube growth of sputtered CoPt was also achieved by Abes et al. on a MgO(1 1 0) substrate at 627 °C, but the dominant (0 *h h*) and (*h* 0 *h*) orientations, rather than the desired (*h h* 0) orientation, placed an easy magnetization axis along the [1 $\bar{1}$ 0] substrate direction [3]. In this paper the use of electron-beam evaporation to grow epitaxial FePt and CoPt $L1_0$ films on MgO(1 1 0) single-crystal substrates is described. The films exhibited substantial anisotropy of the in-plane magnetic properties (magnetocrystalline anisotropy) with an easy magnetization axis along the [0 0 1] substrate direction.

2. Experimental

The samples were prepared in an ultrahigh vacuum electron-beam evaporation system with base pressure of $\sim 5 \times 10^{-9}$ Torr. The chamber is equipped with two sources, each with movable crucibles, that allow controlled co-evaporation of two elements at a time. The evaporated elements were commercial products with purities higher than 99.99% for Fe and Co, and 99.9% for Pt. The MgO(1 1 0) substrates were attached to the heater plate using indium. With the substrates held at 700 °C, a 1 nm thick Pt seed layer was first deposited at a rate of 0.1 Å/s. The substrate temperatures were then adjusted to a temperature from 500 to 700 °C to grow the FePt or CoPt films. For the FePt films, the deposition rates of the elemental Fe and Pt sources were independently maintained at 0.4 and 0.5 Å/s, respectively, using feedback from two collimated sensors near the substrates; for the CoPt films the deposition rates were 0.3 Å/s for Co and 0.4 Å/s for Pt. The deposition rates were selected according to the elemental molar volumes to yield near equiatomic compositions in the films. Energy dispersive X-ray (EDX) analysis indicates the compositions of our films are around $Fe_{51}Pt_{49}$ and $Co_{48}Pt_{52}$, four samples have been analyzed for FePt and CoPt films, respectively. The composition error of our EDX analysis is within ±1 at.%. After depositing 30 nm thick films, the samples were cooled at a rate of 5 °C/min to below 50 °C prior to growth of a 10 nm thick protective capping layer of Au. X-ray diffraction (XRD) data were collected using a diffractometer with Cu Kα radiation. The magnetization measurements were carried out using a superconducting quantum interference device (SQUID) magnetometer at 300 K.

3. Results and discussion

Fig. 1 shows XRD patterns obtained in the θ–2θ (symmetric reflection) diffraction geometry from the FePt films grown at (a) 500 °C, (b) 600 °C and (c) 700 °C on MgO(1 1 0) substrates that were attached to the heater plate with indium. The XRD patterns reveal that various combinations of the three orientations (*h h* 0), (0 *h h*), (*h* 0 *h*) of the (psuedo) cube-on-cube oriented ordered tetragonal $L1_0$ phase coexisted in these films.

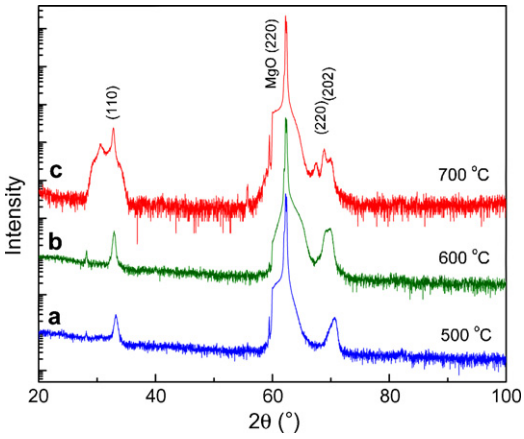

Fig. 1. X-ray diffraction patterns from FePt films grown on MgO(1 1 0) substrates at the indicated substrate temperatures. Substrates were bonded to the heater using indium (Cu Kα radiation).

The peaks have been labeled using lattice parameters for the bulk tetragonal FePt ($a = 3.8525$ Å, $c = 3.7133$ Å, *P4/mmm*, see JCPDS 43-1359) [10]. The superstructure (1 1 0) associated with the $L1_0$ ordering, the fundamental (2 2 0) Bragg peaks and the two-fold rotational symmetry of {1 0 0} planes about the [1 1 0] axis (the ϕ scan is not shown here) indicate epitaxial, pseudo cube-on-cube growth of the $L1_0$ ordered FePt film. The presence of the (2 0 2) peak indicates (0 *h h*) and (*h* 0 *h*) orientations of the $L1_0$ phase. The (2 0 2) peak overlaps the lower angle (2 2 0) peak when these orientations are present with the (*h h* 0) oriented $L1_0$ phase, the latter unambiguously indicated by the (1 1 0) peak. The apparent "shift" of this combined peak to lower angle as the growth temperature increases indicates an increasing fraction of (2 2 0) oriented film as the substrate temperature is increased; the area (height) of the (2 0 2) peak, degenerate with the (0 2 2) peak, would be twice that of the (2 2 0) peak for equivalent fractions of the three orientations. The other peaks are from impurities in MgO substrates, which include the peaks at 28.14°, 55.73°, 59.45° and those or small flats shown on the low-angle side of (1 1 0) peak. The overlap between (1 1 0) and the substrate impurity peaks and that between (2 2 0) and (2 0 2) peaks prevent us from accurate calculation of chemical ordering parameter of the ordered $L1_0$ phase. The use of clamps to anchor one substrate resulted in the growth of some grains with (0 0 *l*) orientation, possibly as a result of inhomogeneous substrate temperature and/or stress state. Results from that specimen are not included.

Magnetization loops obtained from the same specimens, measured at room-temperature for magnetic field applied along the in-plane [0 0 1] and [1 $\bar{1}$ 0] substrate directions, are shown in Fig. 2. The [1 $\bar{1}$ 0] substrate direction is magnetically hardest for all three deposition temperatures, with incomplete saturation of the magnetization M along this direction even for the largest applied field. The difference between the fields required to achieve saturation magnetization (M_s) in the two orthogonal directions, the (in-plane) anisotropy field, is estimated to be 6.5 T. The kinks on the loops (near applied field $H = 0$) are consistent with incomplete magnetic coupling between the different orientations of the tetragonal $L1_0$ phase in the film [3]. The appreciable field required to reach magnetization $M = 0$ (coer-

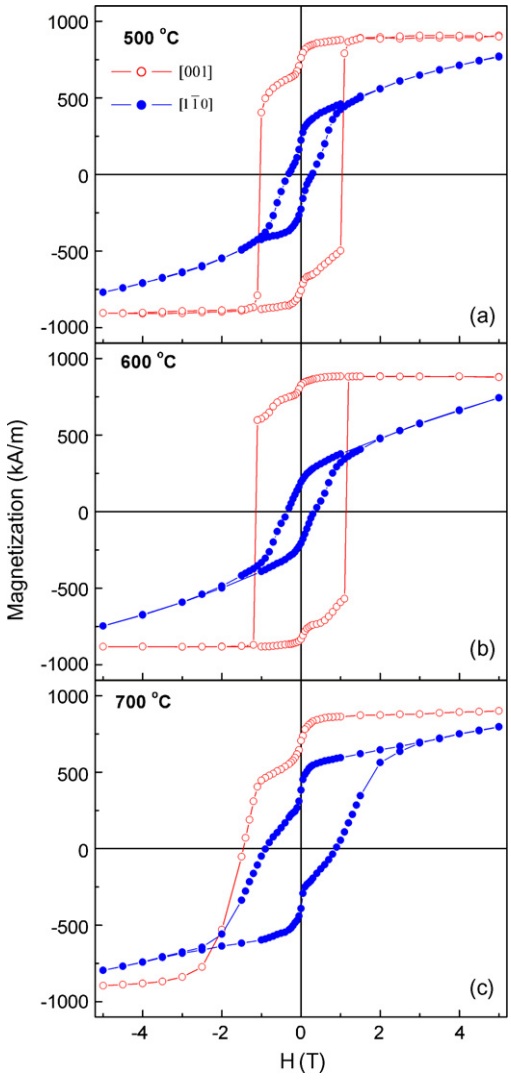

Fig. 2. Room-temperature magnetization loops and/or unidirectional scans for FePt films grown at the indicated temperatures. Magnetic field applied in the film plane along the [0 0 1] and [1 $\bar{1}$ 0] directions of the MgO substrate. Same specimens as in Fig. 1.

civity) and appreciable magnetization measured at zero field (remanence) along the hard axis [1 $\bar{1}$ 0] direction are attributed to the incomplete ($h\,h\,0$) growth and/or presence of ($0\,h\,h$) and ($h\,0\,h$) orientations of the ordered $L1_0$ phase; low coercivity and remanence are expected along the [1 $\bar{1}$ 0] hard axis of the ordered $L1_0$ phase [2], which lies along the [1 $\bar{1}$ 0] substrate direction for ($h\,h\,0$) oriented film but not for ($h\,0\,h$) or ($0\,h\,h$) orientations. Such behavior is observed for the highly textured ($h\,h\,0$) oriented FePt films of Ref. [3]. Because of the fourfold rotational symmetry of the $L1_0$ structure about the ordering direction, similar behavior is also expected along the [1 1 0] axis of the ordered $L1_0$ phase, which is normal to the MgO substrate for ($h\,h\,0$) oriented film but not for the ($h\,0\,h$) or ($0\,h\,h$) orientations. This point will be returned to later.

The FePt films exhibit a saturation magnetization M_s of approximately 850–900 kA/m along the easy [0 0 1] substrate direction, which is lower than the 1080 kA/m value obtained for highly ($h\,h\,0$) oriented $Fe_{58}Pt_{42}$ films grown at 500 °C by MBE [9]. The reason is our films have ~7 at.% lower Fe concentration. Consistent with the XRD results in Fig. 1, as well as the results of Ref. [9], the significant anisotropy and coercivity indicate that chemical ordering of the FePt $L1_0$ phase takes place for substrate temperature of 500 °C (and above). The film grown at 500 °C exhibits coercivity along the hard [1 $\bar{1}$ 0] substrate direction of 0.318 T, similar to that along the hard [1 $\bar{1}$ 0] axes for highly ($h\,h\,0$) ordered FePt films [9]; the ≈1 T coercivity along the easy [0 0 1] substrate direction is significantly greater than the 0.275 T value reported in the same study. The anisotropy field is estimated to be ≈7.5 T. The deep kinks on the magnetization loops are ascribed to poor magnetic coupling of the ($0\,h\,h$), ($h\,0\,h$) and ($h\,h\,0$) oriented domains. Growth at 600 °C (Fig. 2b) yields a smaller kink and squarer magnetization loop for the FePt film that is consistent with a larger fraction of the desired [1 1 0] orientation [9]; such a trend is also consistent with the increased intensity of the (1 1 0) XRD peak in Fig. 1b and the "shift" of the combined 220/202 peak toward lower angles, i.e., the increased height of the (2 2 0) peak. However, further increase of the substrate temperature to 700 °C (Fig. 2c) results in significantly more isotropic magnetic behavior with reduced squareness of the magnetization loops (unidirectional [0 0 1] scan) and apparent increase of the remanence and coercivity along the hard [1 $\bar{1}$ 0] substrate direction to values substantially closer to those along the easy [0 0 1] substrate direction. The deep kink presumably indicates reduced coupling between the different orientations and/or phases present [3].

The CoPt films were grown on MgO(1 1 0) at substrate temperatures of 500, 600, 650 and 700 °C. The XRD patterns and magnetization loops are displayed in Figs. 3 and 4, respectively. The magnetization loops were taken with the external field along the MgO substrate in-plane [0 0 1] and [1 $\bar{1}$ 0] and plane-normal [1 1 0] directions. Comparison of Figs. 1 and 3 makes clear that CoPt requires a higher substrate temperature than FePt to achieve the chemical ordering that underlies the $L1_0$ phase, consistent with literature results [3,8]. Specifically, the CoPt film grown at 500 °C (Fig. 3a) exhibits no (1 1 0) XRD peak of the $L1_0$ phase and only a single, broad peak is observed

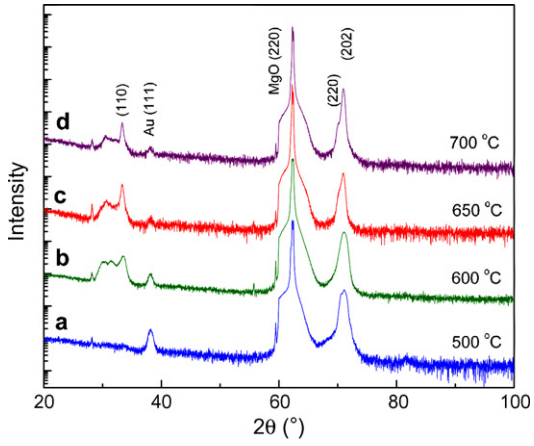

Fig. 3. X-ray diffraction patterns of CoPt films grown on MgO(1 1 0) substrates at the indicated substrate temperatures. All substrates bonded to the heater using indium (Cu Kα radiation).

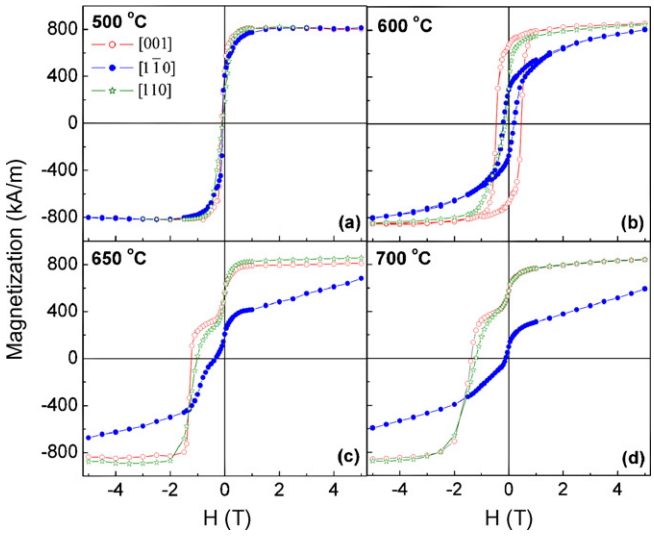

Fig. 4. Room-temperature magnetization loops and/or unidirectional scans for CoPt films grown at the indicated temperatures. Magnetic field applied in the film plane along the [0 0 1] and [1 $\bar{1}$ 0] directions and the plane normal [1 1 0] directions of the MgO substrate. Same specimens as in Fig. 3.

where the (2 2 0)/(2 0 2) peaks overlap; the corresponding magnetization loop in (Fig. 4a) also displays behavior consistent with the soft disordered (isotropic) fcc phase. Evidently, the substrate temperature of 500 °C does not increase the kinetics sufficiently for ordering of the Co and Pt atoms to occur. In contrast, substrate temperature of 600 °C (Fig. 3b) yielded a small (1 1 0) peak indicative of CoPt $L1_0$ chemical ordering along the MgO [0 0 1] in-plane direction, although the broad (1 1 0) and combined (2 2 0)/(2 0 2) peaks still suggest small grains and/or substantial disorder. The corresponding magnetization loops in Fig. 4b indicate increased coercivity and anisotropy that are consistent with partial formation of the ordered phase. The absence of obvious kinks in the magnetization loops presumably indicates the domains of the different orientations/phases are small enough for effective exchange coupling. The saturation magnetization M_s is 859 kA/m, and coercivities are 0.45, 0.206 and 0.145 T for the MgO in-plane [0 0 1] direction (easy axis) and [1 $\bar{1}$ 0] directions (hard axis) and the [1 1 0] plane normal, respectively. The in-plane anisotropy field is estimated to be 6.5 T. As with the FePt, the significant coercivity along the in-plane [1 $\bar{1}$ 0] direction is believed to arise from the presence of the (0 h h) and (h 0 h) orientations of the $L1_0$ phase; as noted earlier, the same argument should apply to the axis normal to the substrate. Visibly narrower (1 1 0) and (2 0 2) diffraction peaks at the higher substrate temperatures of 650 and 700 °C indicate increased size of and/or ordering within the variously ordered regions. This evidently degrades the exchange coupling, resulting in the deep kinks on the corresponding magnetization loops in Fig. 4c and d. Increased substrate temperature of 650 °C did not affect the saturation magnetization M_s significantly, however the coercivities increased to 1.23, 0.328 and 1.02 T for the in-plane easy [0 0 1] and hard [1 $\bar{1}$ 0] directions and substrate normal [1 1 0] direction, respectively. The in-plane anisotropy field also increased with deposition temperature; estimated values are 7.0 T for growth at 650 °C and 9.5 T for growth at 700 °C.

The changing properties are undoubtedly linked to the ordering and changing microstructure detailed in the XRD scans. Increasing substrate temperature clearly promotes improved long-range chemical ordering that manifests as narrower XRD peaks, as was previously described for FePt [9]. However, unlike the XRD results for FePt (Fig. 1), the XRD results for CoPt films in Fig. 3 do not suggest substantial increase of the fraction of (h h 0) oriented grains as the temperature increases farther beyond the temperature where ordering is evident. In this the results are consistent with the absence of a preference for (h h 0) orientation as was previously observed for growth of CoPt on (1 1 0) MgO surfaces [3]. However, the results are like those for the FePt films in that the hard axis for the CoPt films of this study lies along the [1 $\bar{1}$ 0] substrate direction rather than along the [0 0 1] substrate direction observed previously for CoPt by Abes et al. [3] Uniquely, the magnetic behavior obtained along the out-of-plane [1 1 0] direction is similar to that obtained along the easy [0 0 1] substrate direction. This contrasts with the similarity between the out-of-plane [1 1 0] and the hard [1 $\bar{1}$ 0] substrate direction for the highly textured FePt films [9], as expected for the fourfold symmetry about the (0 0 1) tetragonal $L1_0$ axis noted earlier. It also contrasts with the different magnetic properties for all three directions and easy axis along the [1 $\bar{1}$ 0] substrate direction previously published for poorly (1 1 0) ordered $L1_0$ CoPt [3].

Taken together, the electron-beam evaporated FePt and CoPt films yielded $L1_0$ ordering achieved for all conditions studied with the exception of CoPt at 500 °C. The films exhibited temperature-dependent density of (h h 0) oriented grains, the FePt exhibiting a temperature-dependent fraction as well. The easy axis for all films remained along the [0 0 1] MgO substrate direction and a large in-plane magnetocrystalline anisotropy was achieved for the ordered films.

4. Conclusions

Epitaxial growth of FePt and CoPt $L1_0$ films on MgO(1 1 0) substrate using electron-beam co-evaporation is detailed. X-ray diffraction indicates that chemical ordering of the $L1_0$ phase is achieved over the entire range of substrate temperatures 500–700 °C for FePt films and for substrate temperatures 600–700 °C for CoPt films. Psuedo cube-on-cube epitaxial growth of the $L1_0$ tetragonal phase was obtained in all cases. The epitaxial growth occurred in three orientations that differed only in the ordering direction of the Fe(Co) and Pt elemental planes. All the films with ordered $L1_0$ phase exhibited an easy magnetization axis along the in-plane [0 0 1] substrate direction, and hard axis along the [1 $\bar{1}$ 0] substrate direction. A large in-plane anisotropy field varying over the range of 6.5–9.5 T was estimated for in-plane magnetization. The existence of (0 h h) and (h 0 h) in addition to (h h 0) orientations of the ordered $L1_0$ phase contributes to the kinks in the magnetization curves.

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[10] The correct crystallographic description of the $L1_0$ structure is: *P*4/*mmm*; $a' = a/\sqrt{2}$; $c' = c$; two Wyckoff sites: Fe 0 0 0; Pt 1/2 1/2 1/2.