PRODUCTION OF C-AXIS ORIENTED PTFE THIN FILMS WITH PERPENDICULAR MAGNETIC ANISOTROPY

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We report a technique for producing the ordered PtFe intermetallic compounds with perpendicular magnetic anisotropy and a preferred c-axis orientation perpendicular to the film plane. These tetragonal alloys possess large magnetocrystalline anisotropy and magnetizations, suggesting them as candidates for magneto-optic and perpendicular magnetic recording.

KEYWORDS: PtFe, EPITAXY, MAGNETO-OPTIC, MAGNETIC, THIN FILM

Thin-film Pt/Fe and Pt/Co superlattice structures have recently received much attention owing, in part, to their potential application in magneto-optic recording[1]. These artificial superlattice structures are generally based upon layer stacking along the [111] Pt direction. Polycrystalline $Pt_{1-x}Fe_x$ and $Pt_{1-x}Co_x$ alloys, some possessing perpendicular anisotropy, have also been fabricated and suggested for magneto-optic recording[2,3]. Here, we report perpendicular magnetic anisotropy in films composed of the natural superlattice PtFe, with Pt and Fe layers stacked along the [001] direction, produced by annealing epitaxial Pt and Fe artificial superlattices stacked along the [001] Pt and Fe directions. Annealing of these epitaxial artificial superlattices produced the tetragonal PtFe intermetallic phase with the crystallographic c axis preferentially oriented out of the film plane.

Epitaxial Pt/Fe multilayers were grown by sputtering onto 200 Å thick epitaxial Pt(001) oriented substrate layers. These substrate layers were deposited onto polished single crystals of MgO(001) using a seeding technique described elsewhere[4]. The equiatomic multilayers consisted of bilayers with Fe thicknesses from 1.4Å to 23Å and Pt thickness from 2Å to 30Å, deposited at 100°C. These depositions yielded epitaxial multilayer structures with the Pt[001] and Fe[001] axes oriented out of the plane of the sample. X-ray diffraction detected no other phases present in the as deposited samples. Asymmetric diffraction scans showed directly the presence of BCC Fe and FCC Pt in the multilayer for Fe thicknesses of 23Å or larger, with Pt[100]/Fe[110], i.e. a 45° in-plane rotation between the Pt and Fe unit cell axes. After magnetic and x-ray measurements, the samples were annealed in a 5×10^{-7} Torr vacuum at a temperature

Figure 1) Symmetric X-ray diffraction from various bilayer period epitaxial Pt/Fe multilayers before annealing. Composition modulation is evident down to a bilayer period of 6.5Å, corresponding to 2 Fe monolayers and 2 Pt monolayers in each bilayer.

of 475°C for 14 hours, forming the tetragonal FePt phase.

Symmetric X-ray scans were performed on a Philips 3100 X-ray diffractometer equipped with a graphite monochromator. Figure 1 shows high angle symmetric X-ray scans from 450Å thick multilayers of various modulation wavelengths. All of the scans include peaks at 43° due to the MgO

single crystal substrate and at 46° from the epitaxial Pt seed layer. Other features in the scans are due to the epitaxial multilayer. For bilayer periods of $53\text{\AA}$ and $26\text{\AA}$, high angle superlattice lines around the average position of the out-of-plane lattice spacing for Pt and Fe are observed. Below this thickness, a peak near 48° appears, corresponding to the disordered PtFe alloy. Peaks corresponding to the composition modulation also appear.

Figure 2 shows symmetric X-ray scans of a $(23\text{\AA}\text{ Fe}/29\text{\AA}\text{ Pt})_8$ multilayer before annealing (top scan) and after annealing (bottom scan) using an X-ray wavelength $\lambda=1.542\text{\AA}$ (Cu $K\alpha$). The top scan has been shifted upward by 1000x to provide clarity in the figure. The as-grown sample shows high-angle artificial superlattice diffraction peaks, indicating crystalline order between the Fe and Pt

Figure 2) Symmetric scan of a $(23\text{\AA}\text{ Fe}/29\text{\AA}\text{ Pt})_8$ multilayer before and after annealing. Diffraction peaks from the MgO single crystal substrate and epitaxial Pt seed layer, as well as high angle artificial superlattice peaks, are present before annealing. After annealing, the high angle peaks disappear, while peaks corresponding to the natural superlattice PtFe appear. The apparent shoulder on the low side of the MgO substrate peaks is due to the presence of a Ni absorption filter; weak peaks from the MgO(002) from Cu $K\beta$ appear at 38° . A very weak peak in the pre-anneal scan at 26.2° is of unknown origin.

lattices comprising the superlattice. The positions of these superlattice peaks depend on the modulation wavelength of the structure. No ordered PtFe peaks were observed in the symmetric X-ray scan prior to annealing.

After annealing, diffraction peaks appear at $2\theta = 24.0^\circ$ and 49.0° , corresponding to interplanar spacings of $3.71\text{\AA}$ and $1.85\text{\AA}$, respectively. We associate these peaks with the (001) and (002) reflections of c-axis oriented, tetragonal PtFe, which has the AuCu(I) structure with c-axis and a-axis lattice parameters of $3.71\text{\AA}$ and $3.85\text{\AA}$, respectively[5]. Another effect of the anneal is that the high angle artificial superlattice lines disappear. We attribute this to the removal of the Fe/Pt composition modulation in the structure. The disappearance of the composition modulation is also manifest in low angle scans. Before the anneal, we observe clear low angle peaks out to the 3rd order in the modulation wavelength, at 4.7° , while after the anneal, only a very weak first order composition modulation peak occurs.

Figure 3 shows a detail of the diffracted intensity from $44\text{--}50^\circ$ 2θ after annealing. We attribute the three diffraction peaks, corresponding to interplanar spacings of $1.96\text{\AA}$, $1.94\text{\AA}$, and $1.85\text{\AA}$, to 1) Pt(002), 2) PtFe(200), and 3) PtFe(002), respectively. The ratio of the peak areas PtFe(002):PtFe(200) is 3.2:1. The Pt(002) peak is due to a small amount of the initial Pt underlayer remaining after the anneal. Some fraction of the center peak intensity may also be due to oriented, atomically disordered, PtFe, or to the ordered phases of $\text{Pt}_{1-x}\text{Fe}_x$. Ordering would cause the appearance of an (001) peak at 23.0° , such a peak is apparent as a small shoulder on the low angle side of the PtFe(100) peak. For the present discussion these other orientations and phases represent undesired features in the structure. Below, we assume that all of the intensity in the peak at 47° to be due to PtFe(200), establishing an upper bound for the amount of this phase present epitaxially. Rocking curves about the sample normal show that the rocking peak widths for the peaks at 47° and 49° are nearly indistinguishable, 1.9° for PtFe(002) and 1.8° for PtFe(200). This indicates that the difference in diffracted intensity from the two phases is due to the preferential orientation of the PtFe(001) axis out of the plane, since the structure factors for these two reflections are nearly the same. The relative intensities from the symmetric and rocking curve scans indicate that approximately 3 times as much of the tetragonal material is oriented with c axis out of the plane of the film as in the plane of the film.

Asymmetric diffraction scans showed these peaks to also be oriented in the plane of the sample,

with the PtFe[100]//MgO[100], with an in-plane PtFe rocking width of 2.2° FWHM. Longer, more detailed symmetric scans of the annealed sample showed no other diffraction peaks, indicating that

Figure 3) Diffracted intensities from the annealed sample (linear scale) in the range $44\text{--}51^\circ$.

randomly oriented PtFe or other crystalline phases were not present at the detection limit of our diffractometer. Cross sectional transmission electron microscopy work, however, has shown the existence of other unoriented and oriented phases, particularly in the region of the Pt underlayer, in addition to the oriented tetragonal PtFe[6].

The structural changes that occurred upon annealing were accompanied by changes in the multilayer magnetic properties. Figure 4 illustrates the in-plane and out-of-plane magnetic hysteresis loops before annealing, measured on a vibrating sample magnetometer (VSM). As can be seen in the figure, the sample was much more easily magnetized in the plane of the sample than out of the plane, indicating a preferred easy magnetic axis in the film plane. Almost no hysteresis occurs for applied field orientations either in or out of the film plane.

Figure 5 shows the hysteresis loops measured by VSM after the anneal. There is a dramatic change in the magnetic hysteresis loops, as can be seen by comparison to Figure 4. The films become much more easily magnetized out of the plane of the sample, and more difficult to magnetize in the plane. In addition, the magnetization becomes hysteretic, with a coercive field of 1.0 kOe out of the plane of the sample. As can be seen in the figure, the 15 kOe magnetic field is insufficient to saturate the magnetic moment in either direction.

Figure 4) Magnetic hysteresis of $(30 \text{ \AA Pt}/23 \text{ \AA Fe})_8$ multilayer before annealing.

Figure 5) Magnetic hysteresis of $(30 \text{ \AA Pt}/23 \text{ \AA Fe})_8$ multilayer after annealing at 475°C for 14 hours.

As is well known, the microstructure and physical properties of ordered PtFe and PtCo intermetallics is sensitive to processing conditions[7]. For instance, strain annealing of bulk PtCo crystals can result in uniaxial structural and magnetic properties, while coercivity varies strongly with annealing times and temperatures.

Strain annealing is generally not an available processing technique for thin films, which precludes the use of many uniaxial materials in thin film applications. The coercive field in our films also depends on processing. The composition and amount of epitaxial material formed during the anneal is also sensitive to processing.

Magnetic torque measurements were carried out on the annealed sample for which the hysteresis loop is shown in Figure 5. For all fields investigated, up to 22.6 kOe, hysteretic torque curves were obtained, i. e. it was not possible to completely saturate the sample. Therefore, our measured anisotropy at 22.6 kOe represents a lower bound on the anisotropy constant K_u [8]. From these torque measurements, we find a value $K_u \geq 8 \times 10^6 \text{ ergs/cm}^3$, where K_u is the effective anisotropy constant $K_u = K_{mc} - 2\pi M_s^2$, where K_{mc} is the magneto-crystalline anisotropy and M_s is the saturation magnetization, with the easy axis perpendicular to the sample surface.

In conclusion, we have shown that preferentially oriented epitaxial layers of FePt, with the CuAu(I) structure, can be prepared from initially epitaxial Fe(001) and Pt(001) multilayers. These natural superlattices possess magnetic anisotropy out of the film plane, with several potential applications. This technique can be used to prepare other epitaxial structures, for instance tetragonal PtCo(001).

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