Ion Induced Magnetization Reorientation in Co/Pt Multilayers for Patterned Media

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Abstract

Co/Pt multilayer films with perpendicular magnetic anisotropy and large out-of-plane coercivities of 3.9 - 8.5 kOe have been found to undergo a spin reorientation transition from out-of-plane to in-plane upon irradiation with 700 keV nitrogen ions. X-ray reflectivity experiments show that the multilayer structure gets progressively disrupted with increasing ion dose, providing direct evidence for local atomic displacements at the Co/Pt interfaces. This effectively destroys the magnetic interface anisotropy, which was varied by about a factor of two, between $K_S \cong 0.4$ erg/cm^2 and $K_S \cong 0.85$ erg/cm^2 for two particular films. The dose required to initiate spin-reorientation, $6 \cdot 10^{14}$ N$^+$/cm^2 and $1.5 \cdot 10^{15}$ N$^+$/cm^2, respectively, scales with K_S. It is roughly equal to the number of Co interface atoms per unit interface area contributing to K_S.

Introduction

Ion beam patterning of magnetic thin films through suitable stencil masks has been suggested recently as a prospective path towards patterned, ultrahigh-density magnetic recording media [1,2]. Co/Pt multilayers, in particular, are ideally suited for this application, since bombardment with a suitable dose of energetic ions may induce interface mixing [3], thereby triggering a spin-reorientation transition from easy axis out-of-plane to easy axis in-plane [1]. He$^+$ irradiation at 30 keV ion energy and 10^{16} ions/cm^2 dose was reported by Chappert et al. [1] to effectively reduce the anisotropy, coercivity and Curie temperature in Pt/Co/Pt sandwich and multilayer structures. The mechanism leading to the observed modification of magnetic properties is not well understood, but it is likely due to local atomic relaxation and alloy formation [1]. Here we provide direct evidence of interfacial mixing, based upon X-ray reflectivity and magneto-optic data.

Experimental

For the specific experiments discussed here, Co/Pt multilayers with representative structure [Si-substrate/40 nm SiN$_x$/20 nm Pt buffer/10%(0.3-0.4 nm Co/1 nm Pt)/2 nm Pt cap layer] were fabricated. The SiN$_x$ films were dc-magnetron sputtered and the SiN$_x$-coated substrates were heated to about 400°C before electron beam evaporation of the multilayers at growth temperatures $T_G \cong 210 - 230^o$C. Growth rates were ~0.05 nm/s for Co and Pt and the pressure during evaporation was in the low 10^{-7} Torr range. The resulting films were characterized using Rutherford backscattering spectrometry (RBS) and transmission electron microscopy (TEM with Philips CM200FEG) as well as X-ray diffraction (XRD) for compositional and structural analysis, respectively. Nitrogen ion bombardment of 8%8 mm^2 areas at 700 keV energy was performed using a rastered beam from a NEC 3UH Pelletron accelerator. Energy and ion species were selected to resemble the density of ionization conditions used in [1]. Magnetic measurements were performed using polar and transverse Kerr hysteresis measurements and torque magnetometry.

Results and Discussion

Fig. 1 shows a plan view TEM micrograph and an electron diffraction pattern of a 20nmPt/10%(0.3 nm Co/1 nm Pt)/1 nm multilayer grown at 250°C onto a SiN$_x$ membrane.
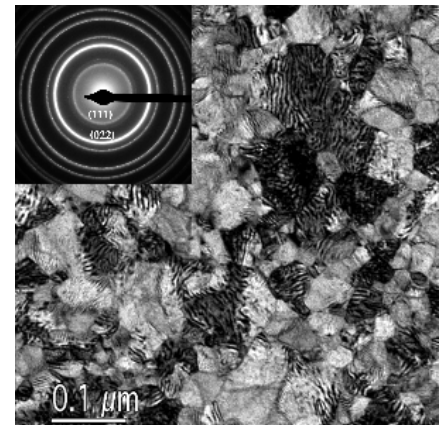

Fig. 1: Transmission electron micrograph and selected area diffraction pattern (SADP) of a representative Co/Pt multilayer (see text).

The film is (111) textured and shows evenly distributed grains varying in diameter between 30 – 80 nm. The polycrystalline structure, with [111] Pt and Co directions out of plane, is verified by selected area diffraction patterns (SADP) (see inset to Fig.1). The fine structure visible on some of the grains is attributed to Moiré fringes caused by small lattice mismatch

and rotation between Co and Pt planes. CoPt(111) rocking curve widths of ~9.3^o were found in XRD.

Table 1: Selected properties of two Co/Pt multilayers grown on SiN$_x$ coated Si(001) substrates with nominally identical structures (10x(0.3nmCo/1.0nmPt)) grown at 230°C (sample I) and 210°C (sample II). The anisotropy constants are per Co volume. The large difference in K_u, K_S and H_C is attributed to structural differences and possibly slightly different t_{Co}.

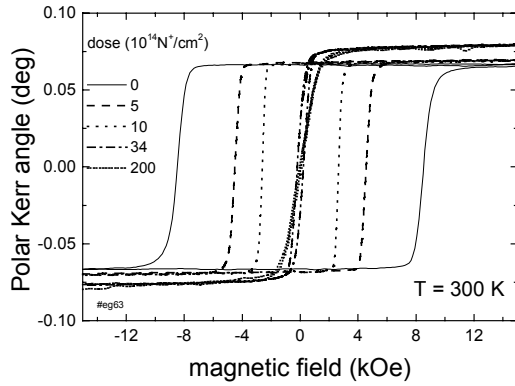| sample	H$_C^0$ (Oe)	K$_u^0$ (erg/cm^3)	K$_S^0$ (erg/cm^2)
I	8500	6.7·10^7	0.84-0.86
II	3950	3.7·10^7	0.39-0.41

Fig. 2: Polar Kerr hysteresis loops of a Co/Pt multilayer subjected to N$^+$ ion irradiation at various doses.

Two similar samples, I and II, as listed in table 1 and grown on SiN$_x$ coated Si(001) substrates, are discussed in the following. Fig. 2 shows perpendicular hysteresis loops of sample I, before and after irradiation with 700 keV N$^+$ ions at various doses. The initial coercivity is H_C^0=8500 Oe, similar to that in [4] and [5] and ten times larger than in [1]. This is attributed to the large intrinsic anisotropy and the granularity of the present films (see Fig. 1). As in [1], strong sensitivity of H_C to dose is observed. Doses as low as 10^{14} N$^+$/cm^2 reduce H_C, without, however, affecting the rectangular loop shape. Substantial decrease in the perpendicular remanence is observed for doses above ~10^{15} N$^+$/cm^2. It is interesting to note that this corresponds roughly to the Co interface atomic density of ~1.5·10^{15} atoms per cm^2 in these films. Note also, that the polar Kerr angle increases with dose. This is different from the results of Chappert et al. [1], where they reported a transition to paramagnetic behavior accompanied by a reduction of the Curie temperature, T_C, as well as the room temperature Kerr effect upon He-irradiation of Pt/Co/Pt sandwich structures. The present films have Curie temperatures, T_C>300°C, and do not show evidence of the onset of paramagnetism at room temperature. The enhancement of the Kerr rotation by about 15%, as evident in Fig. 2, is characteristic of these and a variety of other films.

Co/Pt multilayers deposited on different substrates. For example, Si/SiN$_x$, SiO$_2$ or Al$_2$O$_3$, all showed similar Kerr enhancements of the order of 10-15%, independent of ion induced reductions of the reflectivity. The effect is therefore intrinsic. It may be interpreted as a direct consequence of interface mixing of Co and Pt atoms, resulting in local "alloying". Such resulting CoPt alloys are well known for their enhanced magneto-optic effects because of stronger Pt-5d electronic contributions [4].
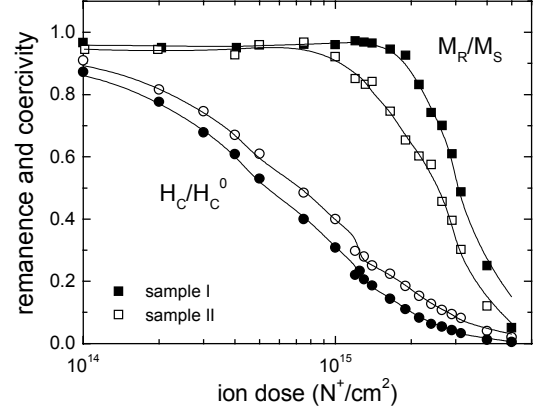

Fig.3: Perpendicular remanence (M$_r$/M$_S$) and normalized coercivity (H$_C$/H$_C^0$) as function of N$^+$ ion dose for two films with vastly different initial coercivity and anisotropy.

Fig. 3 shows room temperature perpendicular coercivity and remanence data as a function of N$^+$ ion dose in the range 10^{14}-5·10^{15} N$^+$/cm^2 for sample I and II. Both films show strong uniaxial perpendicular anisotropy, deduced from torque magnetometry using a 45° method (see table 1). Such large anisotropies are not uncommon for Co/Pt films [5, 6] and are attributed to an interface contribution, K_S, arising from the orbital overlap of Co-3d and Pt-5d wavefunctions. After irradiation with >3-5·10^{15} N$^+$ions/cm^2, K_S is quenched and the

$$K_u = K_V + 2\pi M_S^2 + \frac{2 \cdot K_S}{t_{Co}} \qquad (1)$$

total measured anisotropy energy has dropped to K=5·10^5 erg/cm^3. Commonly, one writes

with K$_V$ a total volume anisotropy contribution, $2\pi M_S^2$ the demagnetization energy and t_{Co} the Co layer thickness. We estimate K_S^I=0.84-0.86 erg/cm^2 and K_S^{II} =0.39-0.41 erg/cm^2, for samples I and II, respectively. The spread in K_S results from assumptions about K_V, which is not well known for 1.5 monolayer thick Co films. Measurements on sputtered and MBE grown (111) textured Co/Pt multilayers indicate K_V =1.2 – 2.6·10^6erg/cm^3 [6]. The demagnetization term is estimated to be $2\pi M_S^2 = 1.6 \cdot 10^7 erg/cm^3$ assuming a Pt polarization enhanced $M_S \cong$1600 emu/cm^3 per Co volume [6].

The normalized coercivities decrease gradually with dose, almost identical to the trend observed in K_u (some data indicated in Fig. 3). We therefore conclude that the loss in coercivity is mainly interface anisotropy driven. The transition from out-of-plane to in-plane easy axis orientation occurs over a dose range of $\Delta \cong 3\text{-}4\cdot 10^{15}$ N$^+$/cm^2 in both cases, which reflects the statistical nature of the bombardment. The onset of the transition, on the other hand, is sample dependent and correlates with the measured surface anisotropy K_S. It is determined by the balance of (positive) out-of-plane anisotropy energy and (negative) demagnetization energy contributions in eq.(1).

We estimate the atomic density of Co atoms per interface area to be $N_i^{Co} \cong 1.5\cdot 10^{15}$ cm^{-2}. A fraction of about 85% (sample I) and 40% (sample II) of these Co interface atoms, i.e. $N_I^{Co} \cong 1.3\cdot 10^{15}$ cm^{-2} and $N_{II}^{Co} \cong 6\cdot 10^{14}$ cm^{-2}, respectively, should contribute to K_S. This assumes a maximal, intrinsic interface anisotropy value of $K_S \cong 1$ erg/cm^2 (100%) (see e.g. results found in highly perfected MBE Co/Pt films [6]). Interestingly, these are exactly the doses needed to trigger spin re-orientation in the present samples.
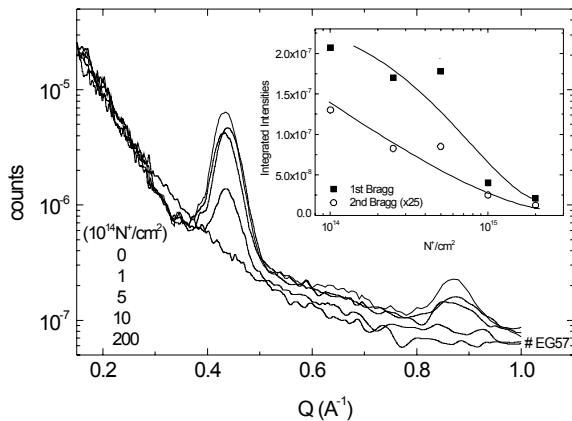

Fig.4: Diffuse or off specular X-ray reflectivity spectra and integrated Bragg peak intensities for different ion doses.

Fig. 4 shows diffuse or off specular X-ray reflectivity [7] data of a 20 nm Pt/10%(0.3nm Co/ 1 nm Pt) multilayer for different ion doses. The diffuse reflectivity is the scattered intensity obtained just off the specular reflection condition [7], and the presence of the 'Bragg' peaks in these data show that the multilayer roughness is conformal over long lateral length scales. The trend in coercivity is closely related to that in the integrated intensity of the first and second Bragg peak in the diffuse reflectivity indicated in the inset to Fig. 4. This drop in intensity shows directly that there is a significant interfacial intermixing on short lateral length scales with increasing ion dose and this destroys the perpendicular anisotropy, due to the disruption of abrupt Co/Pt interfaces. At the highest dose of $2\cdot 10^{16}$ N$^+$/cm^2 in this experiments, the multilayer is completely converted into a Co$_{\sim 25}$Pt$_{\sim 75}$ alloy.

The present data indicate that a constant number of 3-4 impinging N$^+$ ions per surface atomic area is needed to rearrange one Co-Pt interface pair and remove their contribution to K_S. Essentially, all Co-Pt atomic pairs need to be displaced (randomized) to effectively remove the perpendicular anisotropy. This was found to hold for the number of bilayers varying between 2 and 20. Each bilayer evidently responds to the incident ion flux in identical fashion, as anticipated, since the energy and fluence of 700 keV N$^+$ ions are negligibly degraded on the present thickness scale.

The mechanism leading to atom rearrangements is not obvious. A ballistic recoil mechanism, predicted by simple TRIM [8] simulations, would be far too sparse to explain the present results. It is reasonable to assume that thermodynamically preferred local cluster configurations of Co$_{1-x}$Pt$_x$ atoms, as suggested by Chappert et al. [1], are assisted to form without a large ballistic energy transfer. Such local relaxation of the interface order could possibly be stimulated by energy transferred from the ion via ionization and lattice phonons. To test this concept, a detailed model will be required, taking account of the threshold energies for displacement of specific Co and Pt atoms in the interface configuration.

References

1 C. Chappert, H. Bernas, J. Ferré, V. Kottler, J. -P. Jamet, Y. Chen, E. Cambril, T. Devolder, F. Rousseaux, V. Mathet, H. Launois, "Planar Patterned Magnetic Media Obtained By Ion Irradiation", Science 280, 1919 (1998);J. Ferré, C. Chappert, H. Bernas, J.-P. Jamet, P. Meyer, O. Kaitasov, S. Lemerle, V. Mathet, F. Rousseaux, H. Launois "Irradiation Induced Effects on Magnetic Properties of Pt/Co/Pt Ultrathin Films", J. Magn. Magn. Mater. 198-199, 191 (1999); T. Devolder, C. Chappert, Y. Chen, E. Cambril, H. Bernas, J.-P. Jamet, J. Ferré, "Sub 50 nm planar magnetic nanostructures fabricated by ion irradiation", Appl. Phys. Lett. 74, 3383 (1999).

2 B. Terris, L. Folks, D. Weller, J.E.E. Baglin, A.J. Kellock, H. Rothuizen, P. Vettiger, "Ion Beam Patterning of Magnetic Films Using Stencil Masks", Appl. Phys. Lett. 75, 403 (1999).

3 P.F. Carcia, S.I. Shah, W.B. Zeper, "Effect of energetic bombardment on the magnetic coercivity of sputtered Pt/Co thin-film multilayers", Appl. Phys. Lett. 56, 2345 (1990).

4 D. Weller, H. Brändle, C. Chappert, "Relationship between Kerr effect and perpendicular magnetic anisotropy in Co1-xPtx and Co$_{1-x}$Pd$_x$ alloys", J. Magn. Magn. Mater. 121, 461 (1993).

5 S. Shiomi, T. Nishimura, T. Kobayashi, M. Masuda, "Effect of Substrate Heating on Magnetic Properties of Co/Pt Multilayers on Pt Buffer Layers", Advanced Materials'93 II/B: Information Storage Materials ed. M. Abe et al., Trans. Mag. Res. Soc. Jpn., Vol 15B, 1007 (1993).

6 D. Weller, R.F.C. Farrow, R.F. Marks, G.R. Harp, H. Notarys, G. Gorman, "Interface and volume anisotropy of MBE-grown Co/Pt(111), (110) and (001) and sputtered Co/Pt multilayers", Mat. Res. Soc. Symp. Proc. vol 313, 791 (1993).

7 V. Holy, U Pietsch, T Baumbach, "High-Resolution X-ray Scattering from Thin Films and Multilayers", Springer, Berlin, 1999.

8 TRIM (TRansport of Ions in Matter) Monte Carlo code simulating interaction of ions in matter: J.P. Biersack and L. Haggmark, Nuclear Instruments and Methods in Physics Research 174, 257 (1980). The TRIM software for PCs is available from J.F. Ziegler, jfz@us.ibm.com.
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