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Domain structures and temperature-dependent spin reorientation transitions in *c*-axis oriented Co–Cr thin films

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Highly *c*-axis oriented $Co_{95}Cr_5$ films with perpendicular anisotropy were grown epitaxially on Si (111), using an Ag seed layer, by physical vapor deposition. Films were characterized by x-ray diffraction, transmission electron microscopy (TEM), selected area electron diffraction, and Lorentz microscopy in a TEM. The following epitaxial relationship was confirmed: $(111)_{\mathrm{Si}} \| (111)_{\mathrm{Ag}} \| (0001)_{\mathrm{CoCr}}; [220]_{\mathrm{Si}} \| [220]_{\mathrm{Ag}} \| [1100]_{\mathrm{CoCr}}$. Magnetic domain structures of these films were observed as a function of thickness; *t*, in the range, $200\,Å < t < 700\,Å$ using a wedge-shaped sample, and temperature-dependent measurements were carried out by *in situ* resistance heating. Thickness was measured locally by electron energy loss spectroscopy. At room temperature, below a critical thickness, $t_c \approx 300\,Å$, the magnetization was found to be effectively in-plane of the film, and above t_c a regular, stripe-like domain pattern with a significant, alternating in sign, perpendicular component was observed. The spin reorientation transitions of the stripe domains to the in-plane magnetization were studied dynamically by observing the domains as a function of temperature by *in situ* heating up to 350 °C. The critical transition thickness, t_c, which is a function of K_u and magnetostatic energy, was found to increase with increasing temperature. The stripe-domain period, *L* observed at room temperature was found to increase gradually with thickness; $L = 90$ nm at $t = 300\,Å$, and $L = 110$ nm at $t = 700\,Å$. *© 2000 American Institute of Physics.* [S0021-8979(00)46408-X]

INTRODUCTION

Currently the Co-based alloy systems are the most popular media material for magnetic recording. Continued increase in longitudinal recording densities implies a drastic scaling down of the track pitch and bit-cell length, requiring films with smaller and smaller grain size. This approach, however, will collapse when the superparamagnetic and/or thermal decay limit is reached. As an alternative, based on the analysis of the demagnetization mechanisms, Iwasaki and Takemura[1] suggested the possibility of perpendicular magnetic recording (PMR), and computer simulation,[2] predict that an areal density of more than 300 Gbit/in.^{2} can be achieved for the PMR mode. However, the development of such ultrahigh density magnetic recording media is complex because the intrinsic magnetic properties depend on composition, segregation, and temperature, while the extrinsic magnetic properties are affected by the nature of the film (i.e., microstructure, thickness, grain size, shape, distribution, and isolation). Perpendicular media utilizing Co-based hexagonal-close-packed (hcp) alloys requires that the crystallographic *c* axis be oriented normal to the media and the magnetocrystaline anisotropy be larger than the shape or demagnetizing energy.[3] The growth conditions of highly *c*-axis oriented $Co_{95}Cr_5$ films on Si(111) substrates are discussed. When sufficient Cr is added to Co it lowers the saturation magnetization, M_s, and allows out of plane magnetization.[4] For intermediate Cr content a stripe domain with the magnetization effectively in-plane but with an out-of-plane component alternating in sign from one stripe to the next is expected. In this article we present details of these stripes and their behavior as a function of temperature and thickness.

EXPERIMENT

Single crystal *c*-axis oriented Co–Cr films were grown on Si(111) substrates by evaporation. Initially, Si(111) wafers were submersed in a $HF:H_2O(1:10)$ solution for 3–4 min to remove the native SiO_2 and to obtain a hydrogen-terminated surface. Upon etching wafers were immediately loaded into the vacuum chamber and heated to 300 °C. For optimum epitaxial growth, a 500 Å Ag underlayer was evaporated.[5] The Co–Cr layer was deposited at a substrate temperature of 300 °C by evaporation from the $Co_{95}Cr_5$ source. The vacuum achieved during evaporation was $\approx 10^{-6}$ Torr. The film structure (θ–2θ diffraction scans), *c*-axis dispersion (rocking curves, i.e., θ scans) were characterized by x-ray diffraction, while the microstructure (grain size, orientation relationship between substrate/underlayer/film) of the film was evaluated by transmission electron microscopy (TEM) and selected area diffraction (SAD) of plan-view samples. The composition of the film was verified by quantitative energy dispersive x-ray spectroscopy (EDS) in a TEM. The magnetic hysteresis loops with the external field ($\leqslant 1.4$ T) applied both parallel and perpendicular to the thin film sample were measured by vibrating sample magnetometry (VSM).

The magnetic domain structures of these films, as a function of thickness (*t*) and temperature, were investigated by Lorentz microscopy in a Philips CM200FEG TEM using

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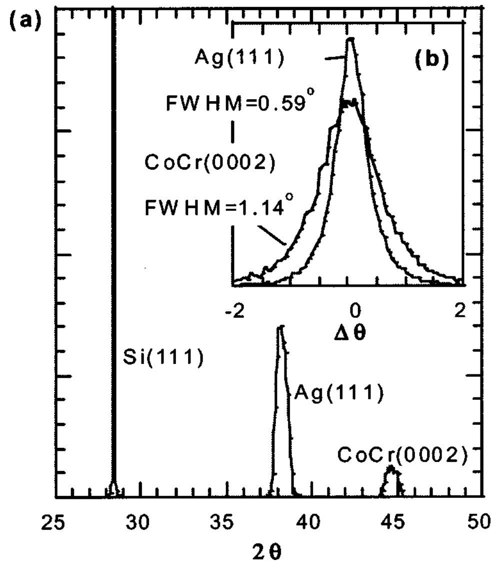

FIG. 1. (a) $\theta-2\theta$ x-ray diffraction spectra of a $Co_{95}Cr_5$ 100 nm/Ag 500 nm/HF–Si(111) film (b) rocking curves (θ scans) for the Ag(111) and $Co_{95}Cr_5$ (0002) peaks.

Fresnel, Foucault, and differential phase contrast (DPC) imaging methods.[6,7] The influence of thickness, in the range $200\,Å<t<700\,Å$, on the magnetic domain pattern was studied using a wedge-shaped sample prepared by low angle ion milling. Moreover, the temperature-dependent measurements were carried out by *in situ* resistance heating. Local thickness of the sample [Fig. 4(a)] was obtained from measurements of inelastic mean-free path lengths in transmission electron energy-loss spectroscopy (EELS).[8]

RESULTS AND DISCUSSION

A representative θ–2θ x-ray diffraction scan [Fig. 1(a)] shows only Si(111), Ag(111), and Co–Cr(0002) peaks, indicating an epitaxial growth and closed pack hexagonal structure of CoCr layer, with the *c*-axis oriented normal to the film plane. The corresponding rocking curves, θ scans, Fig. 1(b) for the Ag(111) and Co–Cr (0002) peaks show a very narrow full width at half maximum (FWHM) widths of 0.59° and 1.14°, implying a narrow dispersion of the (111) and (0002) orientation through the grains of the Ag and Co–Cr layers, respectively. An average grain size of 30–50 nm was observed and the indexing of the SAD pattern of the plan-view samples confirmed the following epitaxial relationship: $(111)_{Si}\|(111)_{Ag}\|(0001)_{CoCr};[\bar{2}20]_{Si}\|[\bar{2}20]_{Ag}\|[\bar{1}100]_{CoCr}$. Moreover, the Ag and Co–Cr spots exhibit arcs comparable to that of the Si substrate confirming that the rotation of the subgrains about the $[111]_{Ag}$ and $[0001]_{CoCr}$ axes is small ($\leqslant 3°$). The EDS analysis confirmed the composition of the film to be Co/Cr$=95/5$, which was the composition of the evaporation source used. Typical hysteresis loops (VSM) with the external field applied parallel ($H_{c\|}\approx 455$ Oe, $M_r\sim 0.15M_s$), and perpendicular ($H_{c\perp}\approx 230$ Oe) to the thin-film sample are shown in Fig. 2. From these curves it can be concluded that the preferred orientation of the magnetization is in the plane of the film. In addition, the wasp-like shape of the perpendicular hysteresis loop shows that the sample is not uniformly magnetized, but it is divided into magnetic domains with up and down magnetization components.[9] The analysis of the Fresnel images of the wedge shape sample with the thickness varying from 200 to 700 Å, revealed two types of domain configuration, above and below critical transition thickness $t_c\approx 300\,Å$. Below t_c the magnetization is in-plane, and film supports large domains with size varying between 0.6 and 1μm, with relatively straight domain walls. Above t_c a regular stripe like pattern was observed, with stripe period increasing gradually with thickness, from $\sim$ 90 nm at $t=300\,Å$ to $\sim$110 nm at $t=700\,Å$. In this region the magnetization is effectively in-plane with a small, out-of-plane component, and the stripe-like pattern is due to up and down periodic modulation of the out-of-plane magnetization component, as illustrated schematically in Fig. 3. The stripe patterns observed in our films are similar to the patterns observed by Kooy and Enz[10] in thin layers of garnet material. Using their model for the total energy of the film with stripe domains, one can estimate the critical transition thickness t_c at which this configuration will be energetically favorable over the in-plane magnetization. The model assumes straight domains with the preferential direction of magnetization normal to the surface of the plate, and a large anisotropy as compared with the demagnetization field, what implies no closure domains. Moreover, the magnetization is allowed to depart from the c axis (the μ effect), which was observed in our film. The energy terms, which are included in the model are: the magnetocrystaline energy E_{MC}; the wall energy E_γ; and the magnetostatic or demagnetizing energy E_{MS}. When the magnetization lies in the plane of the film the only contribution to the total energy of the layer is the $E_{T(1)}=E_{MC}$

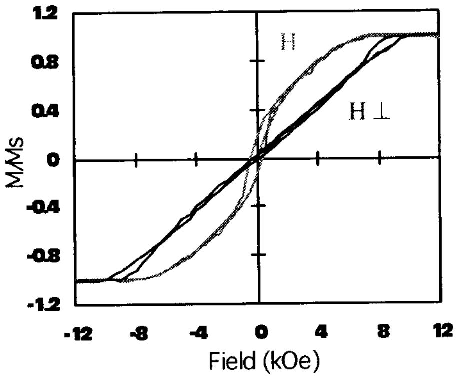

FIG. 2. Magnetic hysteresis loops for both in-plane and perpendicular field directions, measured with VSM.

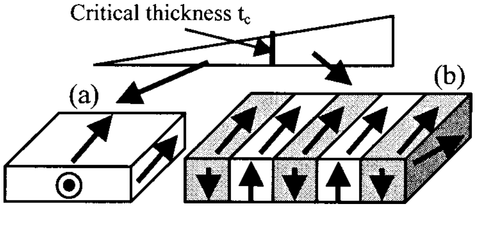

FIG. 3. Schematic diagram showing the domain patterns observed as a function of specimen thickness. (a) Below t_c large domains with magnetization in-plane. (b) Above t_c stripe domains with magnetization effectively in-plane with a small up or down out-of-plane component.

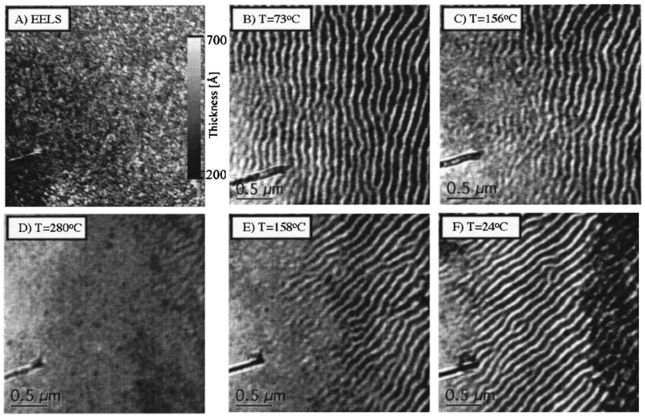

FIG. 4. Temperature dependent domain images observed by TEM Lorentz microscopy. (a) EELS thickness map. (b)–(d) Fresnel images, heating: $T=73$ °C, 156 °C, 273 °C, (e) and (f) cooling: 158 °C, 24 °C.

$=K_u t$, and when the film assumes an out-of-plane magnetization with the stripe pattern the $E_{T(2)}=E_{\mathrm{MS}}+E_\gamma=1.7M_s^2D+\gamma t/D$, where K_u is the uniaxial anisotropy energy, t is the thickness of the CoCr film, M_s is the saturation magnetization of the material, D is the stripe domain period, $\gamma=(4AK_u)^{1/2}$ is wall surface energy, and A is the exchange constant. Solving for the optimal domain period yields the minimum total energy $E_{T(2)\min}=2M_s(1.7\gamma t)^{1/2}$. Comparing the energy terms for the two configurations the transition thickness can be approximated by $t_c=13.6M_s^2(A)^{1/2}/(K_u)^{3/2}$. For the 5% Cr film composition, M_s is approximately constant in the (0–400 °C) temperature range;[11] however, K_u decreases strongly with increasing temperature. Hence, the critical thickness $t_c=1/f(1/T)=f(T)$ is expected to increase with increasing temperature. This spin reorientation transition of the stripe domains with decreasing value of the anisotropy constant K_u was studied dynamically by observing the domains as a function of temperature by *in situ* heating up to 350 °C. As predicted, the critical transition thickness, t_c, was found to increase with increasing temperature, as summarized in Figs. 4(b)–4(f), which show the same area imaged at different temperatures. The thickness of this area [Fig. 4(a)] increases from ~300 Å in the lower left corner to −650 Å in the upper right corner of the image. At 73 °C [Fig. 4(b)] stripe domains are observed for the entire image region (300–700 Å). Upon heating, the stripe pattern gradually shifts to larger thickness, i.e., at 156 °C [Fig. 4(c)] only half of the area ($t>450$ Å) has stripe domains and finally at 280 °C [Fig. 4(d)] no stripe pattern is observed over the entire thickness range. These figures clearly illustrate that upon heating the stripe domain contrast shifts to larger thickness. The plot of the t_c as a function of temperature is shown in Fig. 5. Upon cooling the stripe pattern was found to return, Figs. 4(e) and 4(f), and the t_c was found to be similar for both heating and cooling directions. The stripe period was measured to be the same after cooling, no hysteresis was observed. However, the direction of the stripes was changed, as can be see by comparing Figs. 4(b) and 4(f). The spin reorientation transition, observed as a disappearance of the stripe pattern upon heating, is associated with the loss of the out-of-plane component rendering the magnetization strictly in plane. This novel *in situ* experiment has made it possible to track this spin reorientation in a spatially varying fashion. The temperature measurements of the anisotropy constant will be carried out in the future in order to further model this system.

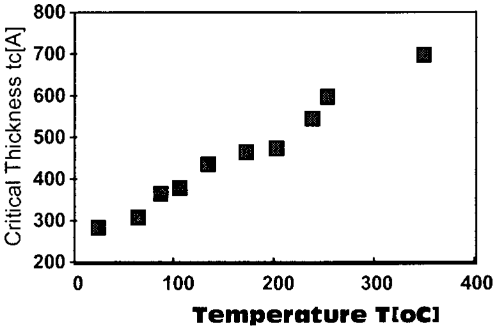

FIG. 5. Critical transition thickness $t_c=13.6M_s^2(A)^{1/2}/(K_u)^{3/2}$, as a function of temperature measured during *in situ* TEM experiment.

[1] S. I. Iwasaki and Takemura, IEEE Trans. Magn. **MAG-11**, 1173 (1975).
[2] Y. Nakamura, J. Magn. Soc. Jpn. **15**, 497 (1991).
[3] K. M. Krishnan, Y. Honda, Y. Hirayama, and M. Futamoto, Appl. Phys. Lett. **64**, 1499 (1994).
[4] R. D. Fisher, V. S. Au-Yeung, and B. B. Sabo, IEEE Trans. Magn. **MAG-20**, 806 (1984).
[5] W. Yang, D. N. Lambeth, L. Tang, and D. E. Laughlin, J. Appl. Phys. **81**, 4370 (1997).
[6] J. N. Chapman, J. Phys. D **17**, 623 (1984).
[7] A. C. Daykin and A. K. Petford-Long, Ultramicroscopy **58**, 365 (1995).
[8] R. F. Egerton, *Electron Energy-Loss Spectroscopy in the Electron Microscope* (Plenum, New York, 1996).
[9] V. Gehanno, A. Marty, B. Gilles, and Y. Samson, Phys. Rev. B **55**, 12552 (1997).
[10] C. Kooy and U. Enz, Philips Res. Rep. **15**, 7 (1960).
[11] I. M. Song, S. Ishio, M. Ishizuka, T. Tsunoda, and M. Takahashi, J. Magn. Magn. Mater. **119**, 261 (1993).