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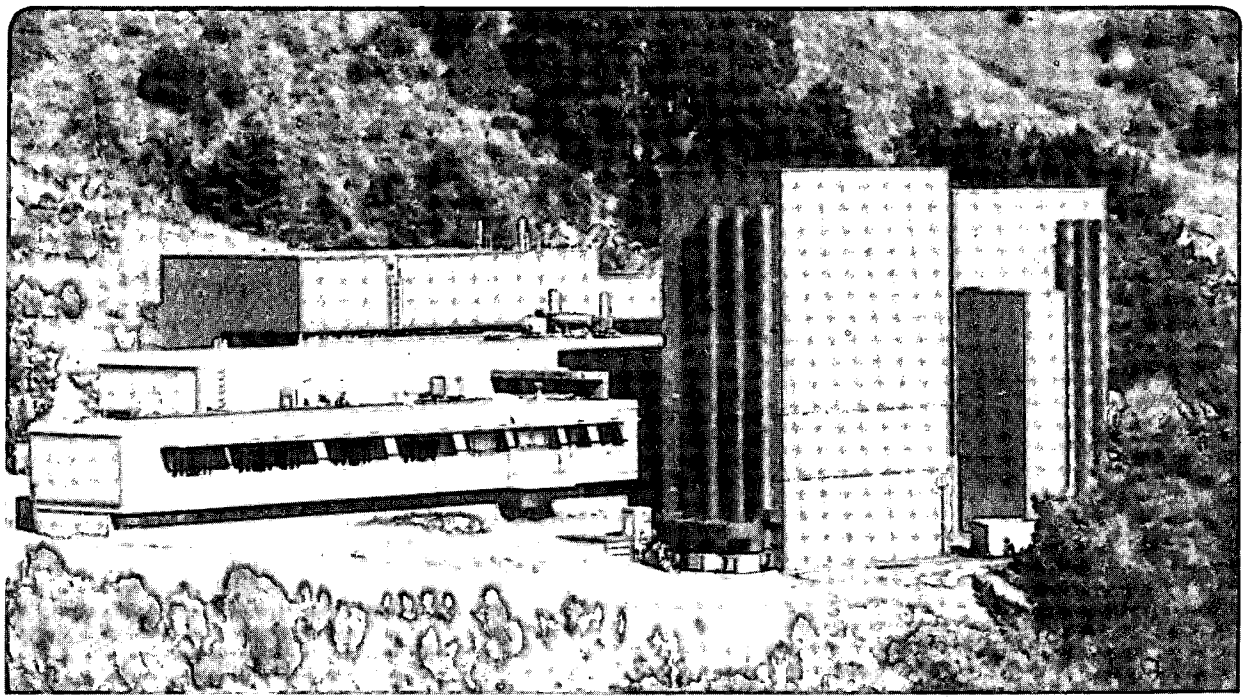
July 1994



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Micromagnetics and microstructure of epitaxially grown Co and Co-Cr films suitable for perpendicular magnetic recording

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Abstract --- Highly c-axis oriented, single crystal films of $Co_{1-x}Cr_x$ ($0 \leq x < 0.3$) have been grown epitaxially on mica substrates by e-beam evaporation. The orientation relationship is basically $(00.1)_{mica} \| (00.1)_{underlayer} \| (00.1)_{Co}$, and $[11.0]_{mica} \| [10.0]_{underlayer} \| [10.0]_{Co}$. Films grown on Ru underlayers have an average grain size of 50-80nm, negligibe fcc content and very narrow c-axis dispersions ($\Delta\theta \sim 0.7$-$1.5°$). For Co films ($x = 0$), the as-grown magnetization structure are mainly 180° domain walls with a uniform distribution of cross-ties for thinner samples ($\leq$ 300 Å), whilst thicker (> 400Å) ones show stripe domains. These images were analysed in detail to measure the wall widths and associated energy densities for as-grown, remanent and ac-magnetised samples. As expected, the magnetic properties of these films are composition dependent. However, for any Cr concentration, these films exhibit the largest saturation magnetisation when compared with either sputtered or evaporated samples. This enhancement can be attributed to a nanometer-scale segregation of Cr, which in these samples, could be particularly aided by the diffusion on the close-packed planes of the films with very narrow c-axis dispersions. Preliminary x-ray microanalysis and NMR data support this interpretation.

I. INTRODUCTION

Cobalt based alloy thin film media, widely used in longitudinal magnetic recording[1], display a range of microstructural features (grain size, shape, preferred orientation) depending on their growth conditions. These in turn, significantly affect their magnetic properties[2]. However, achieving higher densities in such recording is limited by the demagnetising field that approaches a maximum near the transition and hence the alternative perpendicular recording geometry is proposed[3,4]. In light of the above, further developments of this technology can be facilitated if the fundamental magnetic properties and the crystallography of growth, microstructure and magnetization of the individual grains constituting the thin film media is known. Hence, a basic effort incorporating the epitaxial growth of c-axis *oriented* single crystal Co and Co-alloy films on suitable underlayers, evaluation of their microstructure and measurements of their magnetic properties as well as their magnetization structure was carried out.

II. EPITAXIAL GROWTH

Thin films were grown by e-beam evaporation in a water cooled open hearth apparatus. The chamber was baked at 150°C for ~2hr and a vacuum of ~1×10^{-6} torr prior to deposition was achieved. The evaporation rates during growth were monitored by a crystal oscillator and varied from 2Å/s for Ru to 5-6 Å/s for Co/Ti. The accuracy of the crystal oscillator was subsequently verified by measuring the thickness of the films using a profilometer. The substrates, prepared by cleaving the single crystals in air outside the chamber just prior to the deposition process, were heated from the back side using a W heating coil and their temperature monitored using a thermocouple. A preliminary feasibility study of various substrates [MgO(100), NaCl(100), GaAs(100), mica (00.1)] and underlayers [Ti, Ru] indicated that *c-axis*

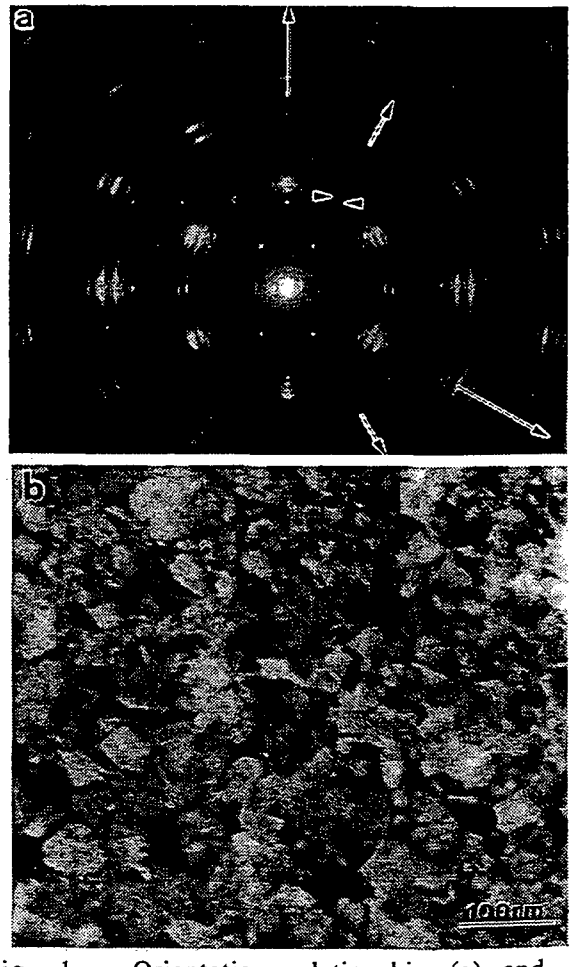

Fig. 1. Orientation relationship (a) and microstructure (b) of the single crystal Co films grown on mica with a Ru underlayer.

oriented single crystal Co films could be grown on mica substrates. For the latter substrate, growth conditions were successfully optimized (underlayer ~ 500°C, 300 Å; film ~ 200 °C, 100-600Å) and the quality of the films routinely monitored by x-ray diffraction[5]. Pole figure x-ray measurements confirmed the absence of fcc Co for the Ru underlayer films. The orientation relationship (figure 1a) is basically: $(00.1)_{mica} \parallel (00.1)_{Ru} \parallel (00.1)_{Co}$, & $[11.0]_{mica} \parallel [10.0]_{Ru} \parallel [10.0]_{Co}$, i.e. the basal planes of all three crystals are parallel, the hexagonal grids of Ru and Co are in registry but are rotated with respect to that of mica by 30°. However, a small fraction of the grains also exhibit an alternative orientation relationship given by: $(00.1)_{mica} \parallel (00.1)_{Ru} \parallel (00.1)_{Co}$ & $[10.0]_{mica} \parallel [10.0]_{Ru} \parallel [10.0]_{Co}$, i.e. the basal planes as well as all the principal crystallographic axes are aligned. Moreover, the Ru and Co diffraction spots exhibit arcs due to the presence of small subgrains rotated about the (00.1) axis with respect to each other by about 20°. For Ru underlayer, an average grain size of 500-800Å, with a small fraction of smaller grains (~150Å) is observed (fig. 1b). In summary, appropriate conditions for the growth of c-axis oriented Co films with single crystal like SAD patterns, narrow c-axis dispersion as observed in x-ray diffraction rocking scans (FWHM ~ 1.6°) and negligible fcc content were established.

III. MAGNETIC PROPERTIES

Typical hysterisis loops (VSM) with the external field applied parallel (Hc ~ 50 Oe, Ms ~ 1450 emu/cc) and perpendicular (Hc ~ 600 Oe, Ku ~ 3 x 10^6 erg/cm^3) to the thin film sample is shown in fig. 2. Even though the single crystal film is grown in the (00.1) orientation, the shape anisotropy or demagnetization energy ($2\pi M_S^2$ ~1.32 x 10^7 erg/cm^3) exceeds the perpendicular anisotropy constant and renders the effective anisotropy in-plane. Moreover, the small hysterisis observed when the field is applied perpendicular cannot be explained by the presence of Co_{fcc} (not observed in pole figure measurements) or in terms of a broad c-axis dispersion

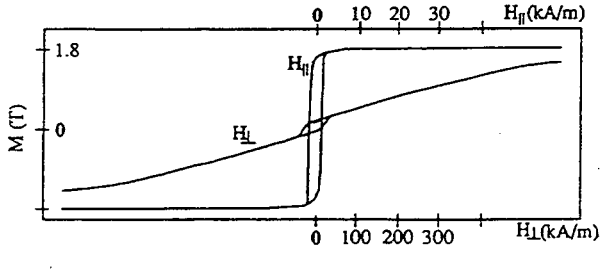

Fig. 2. Hysteresis loops of a 300Å thick cobalt film with the field applied parallel and perpendicular to the film plane.

$(Co(00.2)_{FWHM}$ ~ 1.7 °). However, these features in the hysterisis loops are consistent with a simple non-interacting particle model invoking the shape anisotropy contribution of the small sized grains [6]. For thinner samples, the validity of this model has been verified by annealing.

IV. MAGNETIZATION STRUCTURE

Following the epitaxial growth and characterisation of these single crystal, c-axis oriented, hcp cobalt thin films, we investigated their micromagnetic domain properties by Lorentz Electron Microscopy (LEM). Both the Fresnel and differential phase contrast (DPC) imaging modes of LEM were employed[7]. The latter technique allows a wide range of quantitative analysis to be carried out directly from the observed maps of in-plane induction. The domain structures as a function of the cobalt film thickness were studied in the as-grown, ac-demagnetised

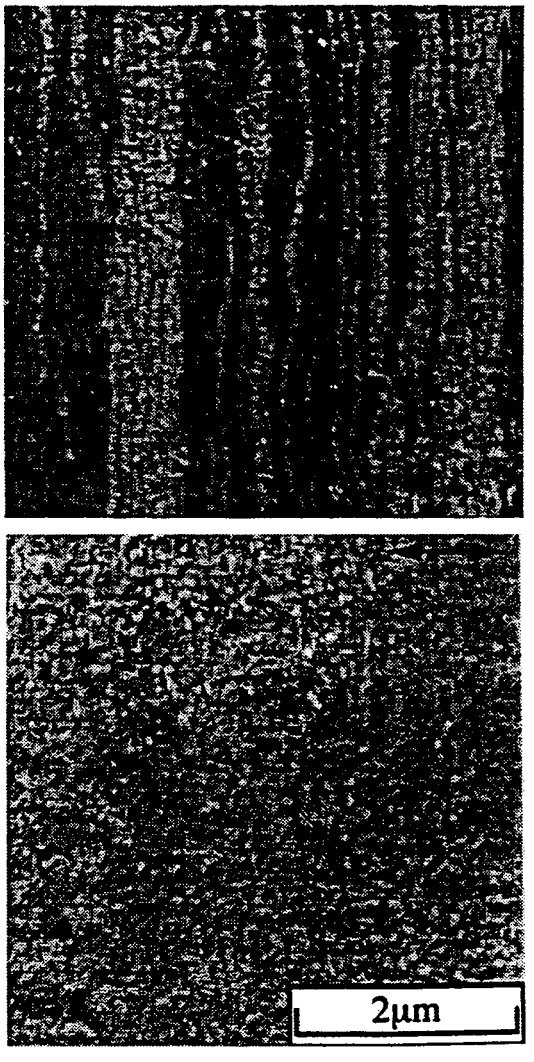

Fig. 3. Differential phase contrast images of stripes and ripple structures in a 40nm thick film of cobalt in the remanent state.

and remanent states. For thinner films (t ≤ 300 Å), large domains with regular magnetisation ripple and cross-tie wall structures were observed in the as-grown state. The period of the cross-ties was observed to be ~600 nm and did not change with ac-demagnetisation. The width of the domain walls (well away from the cross-ties) was measured to be ~46nm. In thicker films [t ≥400Å], the in-plane hysteresis loop is no longer square and exhibits considerably curved nucleation points with long tails to saturation. No cross-ties were seen, since Bloch type walls are expected for this thickness of Co, and instead, stripe domains were observed (fig. 3). This type of dual domain pattern is qualitatively the same as that first observed in permalloy films [8] and the stripe pattern can be explained by the direction of magnetisation deviating (alternatively up and down) from the easy direction (in-plane). For larger thickness, the magnetocrystalline anisotropy is more dominant than the shape anisotropy and allows a component of magnetisation to be out of plane. The period of the stripes was measured to be ~110 nm from these DPC images. From line scans of the intensities of the images, the angle θ, that the magnetisation deviates from the in-plane direction as a stripe pattern is traversed was estimated [8] to be ~44°. Finally, using the Kooy and Enz model [9] and based on the observed stripe domain periods (140 and 122 nm], the specific domain wall energy was calculated to be 33.6 and 16.9 mJ/m^3 for 400Å and 600Å thick samples, respectively. Details of these measurements are given elsewhere [7].

V. EPITAXIALLY GROWN Co-Cr FILMS

Extending this work to alloy films, highly c-axis oriented, single crystal films of $Co_{1-x}Cr_x$ were also grown epitaxially on mica substrates. The epitaxial films exhibit peak widths, $\Delta\Theta_{(00.2)}$~ 1.0 -1.5° in x-ray diffraction (fig. 4) and show exactly the same orientation relationship as the Co films grown earlier, i.e. the hexagonal grids of Ru and Co-Cr are in registry but are rotated with respect to mica by 30° [9]. As expected, these films show perpendicular magnetization and their magnetic properties are composition dependent. For example, the saturation magnetization decreases linearly with increasing Cr content in the film (fig. 5). However, at any given Cr concentration, it is clear that M_S is a minimum for the powder/alloy samples and a maximum for our epitaxially grown films. The values of Ms for the films sputtered on glass or evaporated on polymide substrates are intermediate between these two extremes.

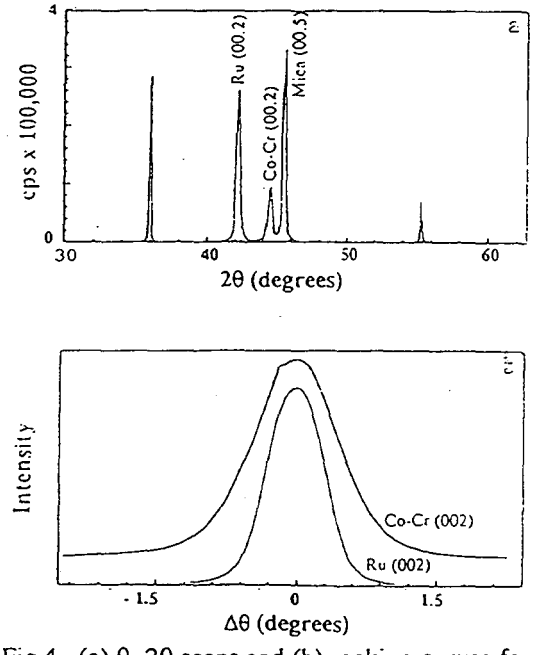

Fig.4. (a) θ–2θ scans and (b) rocking curves for the Ru and Co-Cr (00.2) peaks for a representative sample of mica/Ru(500°C, 300Å)/Co-Cr

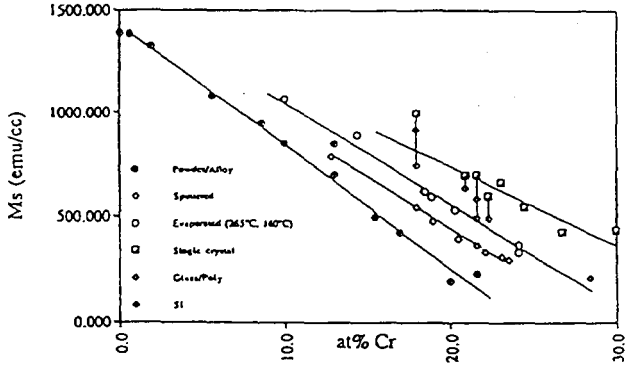

Fig. 5 Variation of Ms with Cr content. Data for powders/alloys ($\Delta\Theta_{50}$-random) and sputtered/evaporated samples ($\Delta\Theta_{50}$ ~ 3-8°) from the literature is compared with our single crystal films ($\Delta\Theta_{50}$ ~ 0.7 - 1.4°). Our data for films on glass/silicon substrates at the same time as on mica are linked by vertical lines.

From the structural point of view, the c-axis orientation of the powder/alloy is random; the sputtered/evaporated samples are reported to exhibit a reasonably good c-axis dispersion ($\Delta\Theta_{50}$ ~ 3 - 8°) whilst our epitaxially films grow well oriented with narrow Co-$Cr_{(00.2)}$ peaks ($\Delta\Theta_{50}$ ~ 1 - 1.5°). From these observations, it can be concluded that the saturation magnetization of these films is directly correlated with the dispersion of the c-axis about the film normal, culminating in a maximum for these epitaxial films. Fundamentally speaking, even though M_s is strictly dependent on the number of atomic moments per unit volume, we suspect that aided by the enhanced mobility in the basal planes, a narrow c-axis dispersion facilitates the segregation of Cr on the nanometer scale.

To further investigate this relationship between $\Delta\theta_{50}$ and M_S, four samples of different compositions (i.e. *x = 22.31, 21.65, 21.3, 18.0*) were grown (at the same time)

on both mica and glass (or silicon) substrates. In each of these four cases there was an appreciable drop in M_S compared to the epitaxially grown single crystal films on mica (fig. 5). Structural measurements (θ–2θ x-ray scans and Co-$Cr_{(002)}$ rocking curves) for a set of three representative samples (x = 21, 3) grown under identical conditions on mica, silicon and glass substrates are shown in figure 6. The film grown on mica is highly c-axis oriented with $\Delta\theta_{50}$ (Co-$Cr_{00.2}$) ~ 1.06°and exhibits the largest M_S. On the other hand, the films grown on glass and silicon show weak Co-$Cr_{00.2}$ peaks; in addition, the latter has an extra peak at 2θ=32.95° (d=2.716Å) that cannot be attributed to either silicon, ruthenium or cobalt and may possibly be due to alloying or chemical reaction at the Ru/Si interface during the high temperature deposition of the underlayer. Finally the rocking curves, $\Delta\theta_{50}$ (Co-$Cr_{00.2}$) for the silicon/glass substrates, show broad peaks, i.e. large c-axis dispersion.

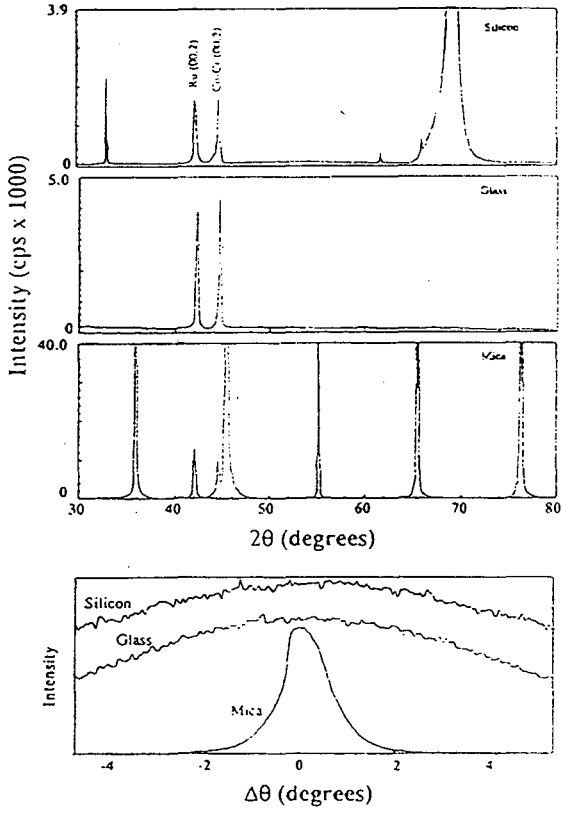

Fig 6. θ–2θ scans and rocking curves (θ scans) of the Co-Cr (00.2) peak for samples grown under identical conditions on mica, glass, and silicon substrates.

It may be argued that the enhanced M_S may also arise from errors in either the sample thickness measurements or changes in composition with depth. It may also be argued that the vacuum level and the small deposition rates used makes the oxidation of the Ru underlayer and Co-Cr film possible. This could then account for the M_s variation. However, neither was oxygen detected in sputtered Auger measurements nor were any additional oxide peaks observed in XRD experiments. Moreover, the decrease in M_S observed in samples *grown simultaneously* on glass/silicon substrates with respect to mica suggests that it is reasonable to conclude that M_S is strongly correlated with the degree of c-axis dispersion ($\Delta\Theta_{50}$) of the films. Even though other parameters such as sample thickness and growth temperature should be taken into consideration when comparing our results with the literature, the relationship between the dependence of M_S on the degree of c-axis dispersion (fig. 5) is convincing and the exact mechanism responsible for this enhancement needs to be investigated.

The most plausible explanation is the segregation of Cr on the nanometer scale. Preliminary x-ray microanalysis using a 1.5nm electron probe (broadened to a resolution of ~3nm for a 30nm thick Co-Cr film) and counting times (50-100 seconds) sufficient to generate statistically significant data (~5000 counts within a preset 90 eV window for the Co peak) gave inconclusive results on Cr segregation. Two grains were analyzed. The first grain indicated a uniform distribution of Cr but with an average value ~3-4% lower than the bulk concentration obtained by the ICPS measurements. The second grain analyzed contains two subgrain boundaries and the measurement of the Cr concentration profile seems to suggest the segregation of Cr to these boundaries. Even though more grains need to be analyzed at this high spatial resolution to make a definitive interpretation, it is clear that a variation in the distribution of Cr is present and may be responsible for the enhancement of M_s. Recent nuclear magnetic resonance measurements [10] support this interpretation of a microscopic segregation of Cr, perhaps on a scale smaller than the resolution of the above microanalysis.

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