Observation of irregular labyrinthine magnetic domains with dendritic edges in a Co-rich CoCu alloyed film

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We report on the observation of irregular labyrinthine magnetic domains with dendritic edges in a 1500 nm thick $Co_{96}Cu_4$ alloyed film grown via radio-frequency magnetron sputtering on a polyimide substrate. The film shows strong texture along the c-axis of the hexagonal closed packed system. As a result, its magnetic properties resemble those of single-crystalline hexagonal (001) Co films. Indeed, magnetization hysteresis loops recorded under fields up to 16 kOe via a home-made magneto-optic Kerr-effect magnetometer reveal strong tendency for perpendicular magnetic anisotropy. Hysteresis loops with the field parallel or perpendicular to the film plane present zero remanence. Magnetic force microscopy after perpendicular magnetization of the film shows the formation of regular bubble ferromagnetic domains in a metastable state with the local spins oriented up and down, perpendicular to the film plane. After perpendicular demagnetization, however, the labyrinthine ferromagnetic domains show irregularly distributed dendritic edges. This effect is attributed to Cu precipitates in a Co matrix, which act as pinning sites in the free motion of domain walls.

Keywords: Magnetic thin films, magnetic force microscopy, bubble, labyrinthine, dendritic domains, Cobalt, pinning sites

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Progress in magnetic domain imaging by Scanning Probe Microscopy techniques has allowed for high-resolution study of ferromagnetic domains formed on magnetic thin films and nanostructures and its correlation with structure and magnetic anisotropy.[1-6] Films with perpendicular magnetic anisotropy (PMA) are important for magnetic and magneto-optic recording applications, see e.g. Ref. [7]. When PMA is weak, the films show canted magnetization configurations and a mixture of parallel and perpendicular magnetic domains.[8,9] On the other hand, films with a significant PMA component show usually three types of ferromagnetic domains depending on the magnetic history of the sample: Bubble magnetic domains for field slightly smaller than the nucleation field or at remanence after perpendicular magnetization of the film, stripe magnetic domains after parallel magnetization or demagnetization of the sample and labyrinthine domains after perpendicular demagnetization of the samples.[5] In all such cases, the spins point up or down with respect to the film plane and the domain edges (boarders) are usually smooth.

In this communication, we report the formation of irregular labyrinthine magnetic domains with dendritic edges in a thick Co-rich CoCu film with very strong c-axis hexagonal closed packed (*hcp*) texture. Domain images showing some similarity to our results have been recorded on different materials and with different physical properties compared to our system; namely, either on thermomechanically processed (die-upset) melt-spun NdFeB bulk hard permanent magnets with very large coercivity (interaction domains),[10] or on magnetostrictive FeGa bulk alloys,[11,12] related to inhomogeneous stress. However, to our knowledge, such domain patterns have not been reported previously for magnetic thin films. Our film has a significant component of PMA, as magneto-optic Kerr effect (MOKE) hysteresis loop reveal, allowing for the formation of magnetic domains (bubbles, labyrinth or stripes) with spins oriented up or down with respect to the film plane. The particular and unusual decoration of the labyrinth domain edges is attributed to Cu precipitates in a Co matrix, which act as pinning sites in the free motion of domain walls.

A $Co_{96}Cu_4$ film, 1500 nm thick, was deposited on polyimide (Kapton) substrate by radio-frequency (r.f.) magnetron sputtering at about 400 K. The thickness was selected to be in the range of 1 micron, as we have shown recently that in such a case films with strong texture are favored.[13] The base pressure of the vacuum chamber was 5 x 10^{-8} mbar. The Ar pressure during deposition had been kept constant with the help of a fine valve at a pressure of about $6x10^{-3}$ mbar. The r.f. power had been kept constant at 50 W with the help of a feedback power supply and a matching circuit. This power resulted in a deposition rate of 0.4 nm/s. Deposition rate was monitored with the help of a precalibrated quartz balance. The calibration was performed as follows: In the inset of Fig. 1, we see the appearance of Kiessig fringes in the small-angle XRD spectrum of a thin CoCu film. These are present for homogeneous samples with small surface roughness; with the help of them, the film thickness may be accurately determined.[7] In order to form the alloy, a Cu foil (99.9 at.% purity) was sandwiched between a Co foil (99.99 at.% purity), having small holes randomly distributed on its surface, and the water-cooled magnetron head with the help of attractive magnetic forces. The composition of the sample has been checked by energy-dispersive X-ray analysis (EDAX) measurements carried out in a Scanning Electron Microscope (SEM) Zeiss Supra 35VP model. In our film, the Co concentration was found to be 96 at.%.

The structural characterization was mainly performed with the help of x-ray diffraction (XRD) patterns recorded using a standard powder diffractometer (Berthold) with Ni- and Co-filtered CuKa_1 radiation (λ = 0.154059 nm). The magnetic anisotropy of the samples was studied by recording hysteresis loops with the applied field parallel and perpendicular to the film plane at room temperature via a home-made magneto-optic Kerr effect (MOKE) magnetometer.[14] The magnetic domain morphology in our films has been observed at room temperature by using the Magnetic Force Microscopy (MFM) mode of a Multimode Microscope with a Nanoscope IIIa controller and a 120 μm x 120 μm magnet-free scanner (Model AS-130VMF) developed by Digital Instruments. We used a Co/Cr coated Si probe tip whose magnetization is parallel to its axis. The images were recorded at magnetic remanence by using the amplitude detection mode.[1]

In Fig. 1, the high-angle XRD pattern of the 1500 nm $Co_{96}Cu_4$ film grown on Kapton is shown. The reference data for polycrystalline *hcp* and face centered cubic (*fcc*) Co powders have also been also introduced in Fig. 1.[15,16] The amount of *fcc* grains is obviously negligible as (i) the *fcc*(200) diffraction peak is very small and (ii) the largest diffraction peak is much closer to the *hcp*(002) than the *fcc*(111). The slight difference of $2\theta = 0.07^0$ in the (002) peak of our sample with respect to Co-reference may be attributed to the presence of some residual strain of about 0.15% due to growth conditions and the diferrence of the thermal expansion coefficient between the metallic film and the polymer substrate. Similar shifts in the (002) peak have been also observed in pure Co films grown under similar experimental conditions.[9] The presence of Cu should not have any effect due to the immiscibility between Co and Cu. Moreover, the fact that the *hcp*(002) peak is much larger than all the other *hcp* ones, unlike the ratios of the vertical lines which correspond to a reference random polycrystalline powder,[15] reveals that there is a strong texture with the c-axis perpendicular to the film plane. We have shown recently that ~ 1 μm thick CoPd alloyed films prepared under similar experimental conditions show perfect *fcc* {111} texture and this has a large influence on the magnetic properties.[13] Texture is influenced mainly by the total film thickness due to competition between residual strain, which may vary with film thickness and surface energy.[17-19] Application of the Scherrer formula to the *hcp*(002) peak taking the corrected full width at half-maximum (FWHM),[13] estimates a relatively large average grain size of about 85 nm.

In Fig. 2 we show hysteresis loops of the $Co_{96}Cu_4$ film recorded via MOKE magnetometry with the external magnetic field H applied parallel and perpendicular to the film plane, as indicated. MOKE is not an absolute magnetometry technique as, for example, Superconducting Quantum Interference Device (SQUID).[20] For this reason, we have calibrated the y axis in magnetization units taking the magnetization for bulk CoCu alloys of the same composition as ours from literature data.[21] The sample seems to be easier magnetized in the film plane, however, zero remanence is observed for both field directions. From the area between the anhysteretic hard- and the easy-magnetization axes, one can calculate the so-called effective magnetic anisotropy energy K_{eff} to be equal to 6 x 10^6 erg/cm^3. If one considers K_{eff} to be the sum of the shape anisotropy $K_d = 2\pi M^2$ and a first order uniaxial anisotropy K_{u1}, then K_{u1} = -5.3 x 10^6 erg/cm^3. (The sign '-' in our case reveals tendency for PMA. This tendency increases with the absolute value of the quantity $Q = K_{u1}/K_d$;[6] for our film Q = 0.47) The absolute value of K_{u1} is even by about 20% larger than the value of magnetocrystalline anisotropy of single crystalline *hcp* Co along the c axis. As discussed in Ref. [13], error bars at least as large as 10% in the determination of the anisotropy constants by the hysteresis loops may be expected for films with Q values like ours due to appearance of perpendicular magnetic domains. The excess of the uniaxial anisotropy with PMA tendency in our film compared to epitaxial Co should be attributed to the magnetoelastic anisotropy due to the presence of residual anisotropic strain in our film, for example due to the large difference between the thermal expansion coefficient between the metallic film and the polymer substrate.

In Fig. 3(a), we present the topography of our film. One may see the presence of small crystallites with a mean diameter of about 85 nm, as aforementioned. In Fig. 3(b), a MFM image of our film is presented after perpendicular magnetization. This process includes magnetization to the perpendicular saturation field and sudden removal of this field.[5]

Magnetic domains are formed at the nucleation point (which in our case is practically equal to the saturation field), and have the configuration of magnetic bubbles. As the field is removed fast, the system does not have the time to reach equilibrium at remanence. Therefore, the bubble configuration is clearly maintained. This is one of the few times that many clearly circular bubbles are observed as usually they are elongated and merge tending to be transformed into labyrinths.[5] Even more spectacular is the image of Fig. 3(c) after perpendicular demagnetization. This means that after perpendicular saturation the film is demagnetized by periodic decrease of the field amplitude.[5] One may observe at remanence the appearance of irregular labyrinthine domains with dendritic edges. In the present work, the deposition process and film thickness are similar to the ones of fully textured $Co_{58}Pd_{42}$ alloyed thin film.[13] Moreover, the magnetic Q value of our $Co_{96}Cu_4$ film is almost equal to the one of a fully textured $Co_{58}Pd_{42}$ film. The latter film showed, however, regular labyrinthine magnetic domains after perpendicular demagnetization without any decoration of the edges. A typical image of this kind is presented in Fig. 3(d). The only difference between the two films is that, according to the binary alloy phase diagrams and verified experimentally, Co and Pd form solid solutions and therefore the CoPd film is structurally and magnetically homogeneous,[13] while Co and Cu do not show any miscibility.[22] Therefore, presence of Cu precipitates in a Co matrix is expected. The Cu nanograins should act as obstacles (pinning sites) in the free motion of domain walls during the slow pseudo-static demagnetization process, and the labyrinthine domains present irregular dendritic edges.

In conclusion, we have investigated the magnetic properties of a $Co_{96}Cu_4$ film with a thickness of 1500 nm, deposited on polyimide by r.f. magnetron sputtering. The film presented a *hcp* structure with very strong c-axis texture. As a result, it showed enhanced perpendicular magnetic anisotropy as compared to epitaxial single-crystalline Co films.[5] Bubble magnetic domains were observed at remanence after perpendicular magnetization of the film; some of them retained their original circular shape as they did not merge with others. Finally, we showed the unusual formation of irregular labyrinthine magnetic domains with dendritic edges after perpendicular demagnetization of the film. This effect was attributed to structural and local magnetic inhomogeneity of the film due to separation of Co and Cu phases.

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Figure Captions

Figure 1 (coloured on-line)

High-angle XRD pattern of the 1500 nm $Co_{96}Cu_4$ film grown on Kapton. The reference data for random polycrystalline *hcp* and *fcc* Co powders have been also introduced by vertical lines ended in squares or circles, respectively. The inset shows Kiessig fringes for a thin sample. With the help of them, we determine the film thickness and calibrate our quartz balance.

Figure 2 (coloured on-line)

Hysteresis loops of the $Co_{96}Cu_4$ film recorded via MOKE magnetometry with the external magnetic field H applied parallel and perpendicular to the film plane, as indicated. The Y-axis has been calibrated with the help of Ref. [21].

Figure 3 (coloured on-line **please use double column width**)

Topography (a) and magnetic domain configurations after (b) perpendicular magnetization and (c) perpendicular demagnetization of the $Co_{96}Cu_4$ film. In (b) we may observe bubble magnetic domains and in (c) irregular labyrinthine ones. In (d) a MFM image of regular labyrinthine domains for a CoPd film is presented for comparison. Light (dark) color corresponds to up (down) oriented domains with respect to the film plane. The images size is 10x10 μm^2.

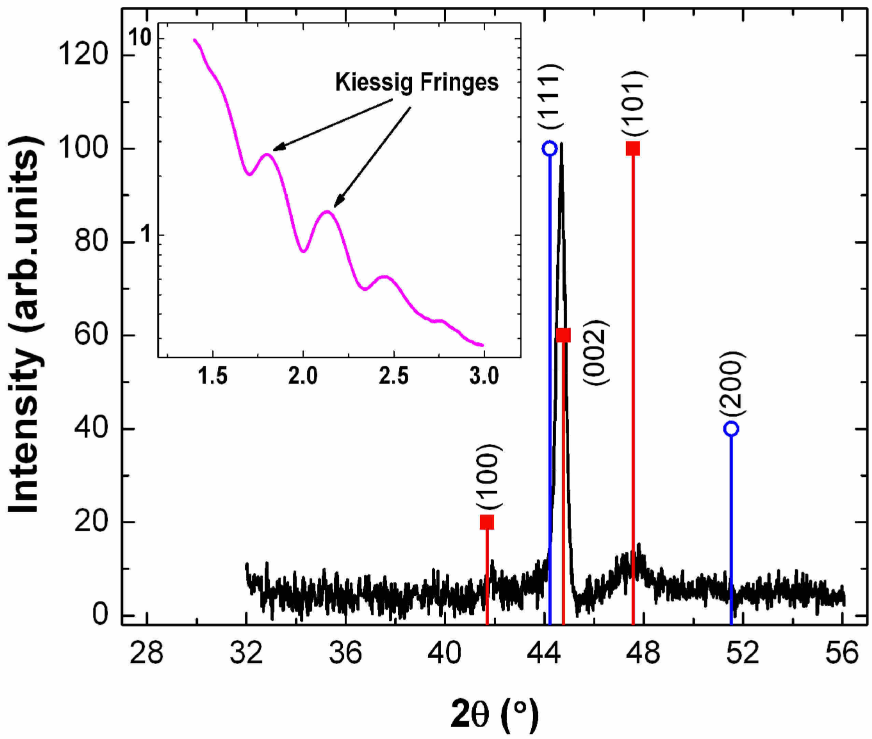

Figure 1, Pappas et al.

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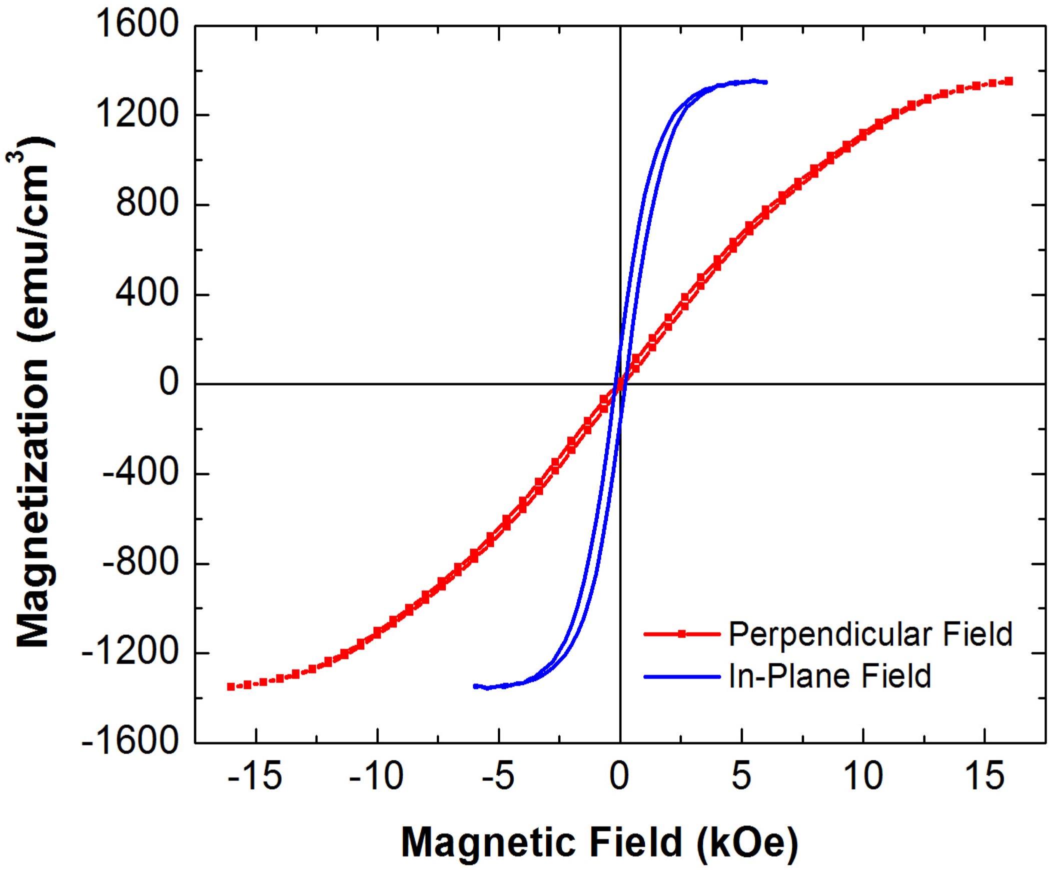
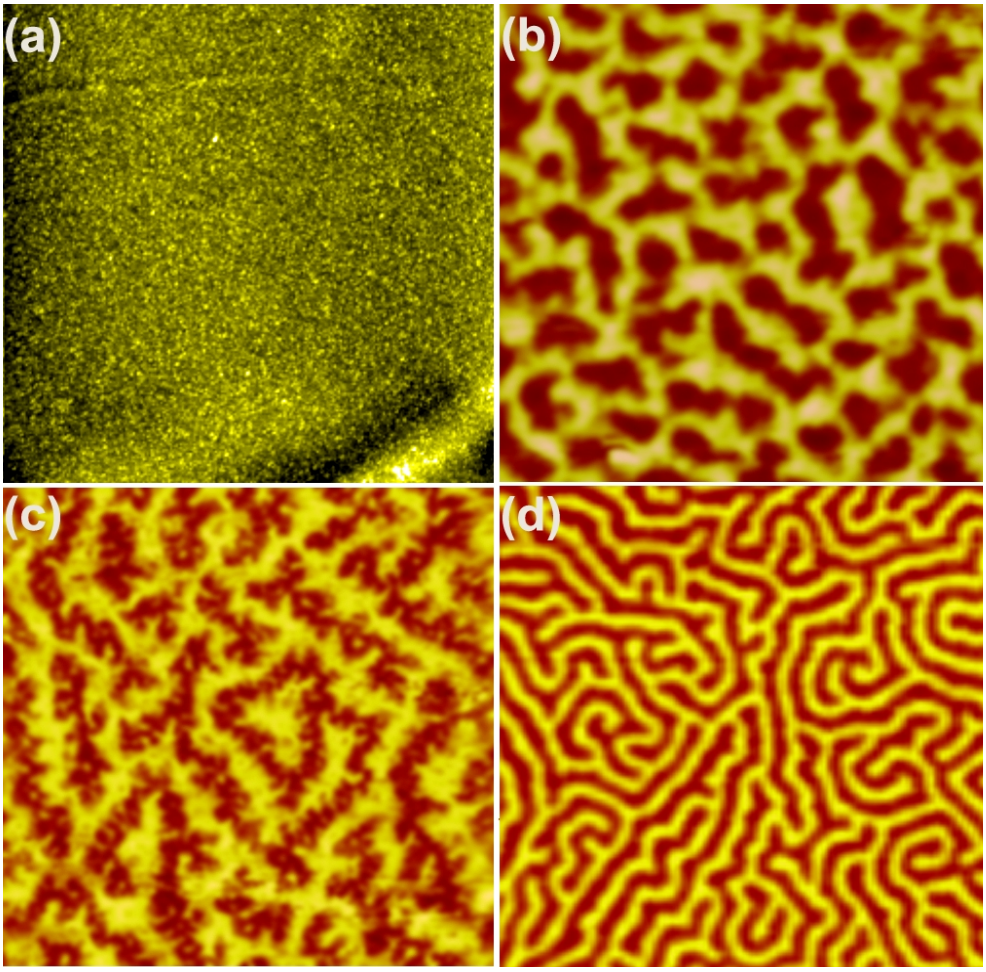


Figure 2, Pappas et al.

Observation of irregular labyrinthine magnetic domains with dendritic edges in a Co-rich CoCu alloyed film



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