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Magnetostriction and Microstructure of As-Deposited and Annealed Co Thin Films

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ABSTRACT

The magnetostriction of as-sputtered and annealed 400 nm thick Co films has been studied in longitudinal and transverse magnetic fields. The appreciable change of the magnetostriction behavior after annealing above 250 °C is correlated to grain growth and to the related change of the texture (from nearly randomly distributed hcp-Co crystallites to a c-axes texture perpendicular to the film plane). The magnetostriction behavior in the annealed samples cannot be explained by a domain magnetization within the film plane. It is assumed that a rotation of the spontaneous magnetization out of the film plane occurs due to the development of a perpendicular magnetic anisotropy.

INTRODUCTION

Cobalt thin films are of great interest as soft magnetic single layers as well as in magnetic multilayers, e.g., Cu/Co multilayers [1]. Due to the magnetoelastic coupling, the magnetostriction also affects the magnetic anisotropy. Deposition conditions and post-growth annealing may have a profound influence on the microstructure and therefore also on the magnetostrictive state.

The present contribution is focussed on the magnetostriction in sputtered 400 nm thick Co films in the as-deposited state and after annealing up to 450 °C. The results are correlated to findings of microstructural analyses concerning phase formation, texture, and grain morphology. The results are complemented with studies of: (i) the stress development as an indicator for microstructural processes as well as a factor in the magnetoelastic coupling, and (ii) the longitudinal magneto-optical Kerr effect (MOKE) for the characterization of the surface magnetization of the films.

EXPERIMENTAL DETAILS

The films were deposited by magnetron sputtering onto rotating oxidized 3-inch silicon wafers at room temperature. The deposition conditions were: base pressure 1×10^{-6} mbar, sputtering pressure 4×10^{-3} mbar Ar, and sputtering power 150 W for a 4-inch target. The film thickness was measured using a Dektak stylus profiler after wet-chemical etching of an edge into the film, and was determined to be 399 nm.

For the annealing procedure, the coated substrate was cut into slabs of 55 mm × 7 mm. The heat treatment was combined with in-situ stress measurements under high vacuum (1×10^{-5} mbar) using a sensitive laser-optical stress-measurement system described elsewhere [2]. The heating and cooling rate amounted to 4 K/min and the isothermal annealing was done at 150, 250, 350, and 450 °C for 2 h. The annealed slabs were cut into smaller samples for all other measurements.

The magnetostriction was determined by a laser-optical measurement of the substrate deflection in an applied magnetic field using a two-beam free-sample set-up described elsewhere [3]. The deflection δ of the substrate at the position of the laser beams (having a distance l of 25 mm) with respect to the sample center was determined in longitudinal (||) and transverse (⊥) magnetic fields H applied always within the film plane.

X-ray diffraction (XRD) investigations were performed using a Philips-XPert diffractometer with CuK_{α} radiation, Eulerian cradle, and thin-film equipment. Scans with symmetrical beam, grazing incidence, and pole-figure cuts were measured.

The transmission electron microscopy (TEM) studies employed cross-sectional specimens. The preparation was carried out by means of a focused ion beam (FIB) technique. For TEM observations, a PHILIPS CM20FEG was employed in imaging and diffraction mode. Grain-size distributions were determined from measurements in bright and dark field image modes.

RESULTS

The stress-temperature curves $\sigma(T)$ during the annealing (Fig. 1) reveal initial irreversible stress changes already above 150 °C and more distinct changes above 250 °C. Similar to CuNi thin films [4,5], the irreversible tensile stress contributions are expected to be the result of material densification due to grain-boundary relaxation and elimination (grain growth is proved by TEM, see below). No feature associated with a martensitic transformation from a hexagonal-closed-packed (hcp) to a face-centered-cubic (fcc) phase at about 400 °C, as it was reported in the literature for ≥500 nm thick Co films [6], was observed in our $\sigma(T)$ curves.

The magnetostrictive deflection during the second magnetization loop (Fig. 2) shows distinct changes between the as-deposited and the annealed state. For as-deposited samples (as well as for T_{max}=150 °C), a soft magnetic behavior with a hysteresis of about 10 kA/m is mainly observed and the deflections $\delta_{\parallel}$ and $\delta_{\perp}$ are of opposite sign. Full saturation of the deflection signal was reached only above 200 kA/m. For the samples with T_{max}=350 °C and 450 °C, the films show a hard magnetic behavior without remarkable hysteresis, the magnetostriction response being not nearly as saturated at 500 kA/m, and the deflections $\delta_{\parallel}$ and $\delta_{\perp}$ are relatively high and both negative. The curves for the sample with T_{max}=250 °C indicate a transition between the soft and hard magnetic case. The reason for these changes and the calculation of the magnetostriction value given at the right-hand axis of Fig. 2 will be discussed below. Experimental magnetostriction data, being in many points similar to our results, were also reported for 165 nm thick Co films [7].

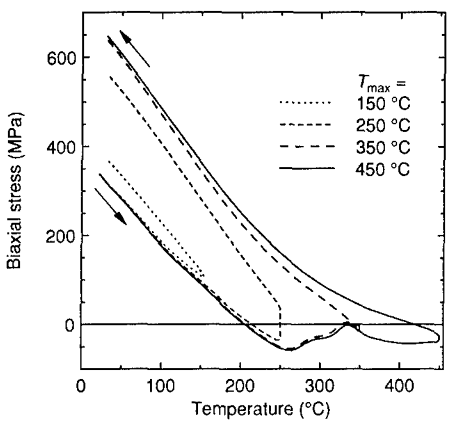

Figure 1. Stress development during annealing of Co thin films at various temperatures.

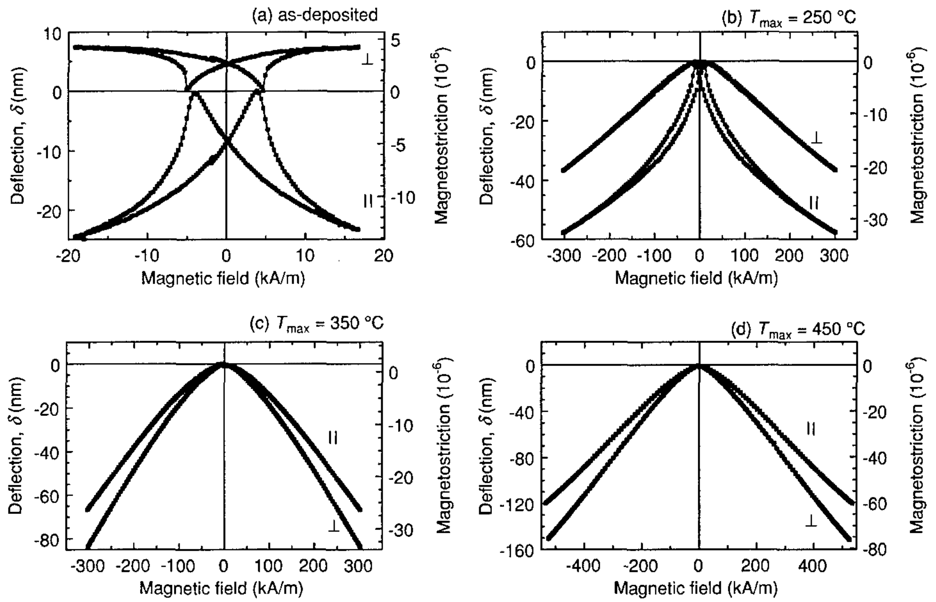

Figure 2. Magnetostrictive deflection and magnetostriction in longitudinal (||) and transverse (⊥) magnetic fields during the second magnetization loop. The behavior changes strongly during annealing at $T_{max} \geq 250$ °C.

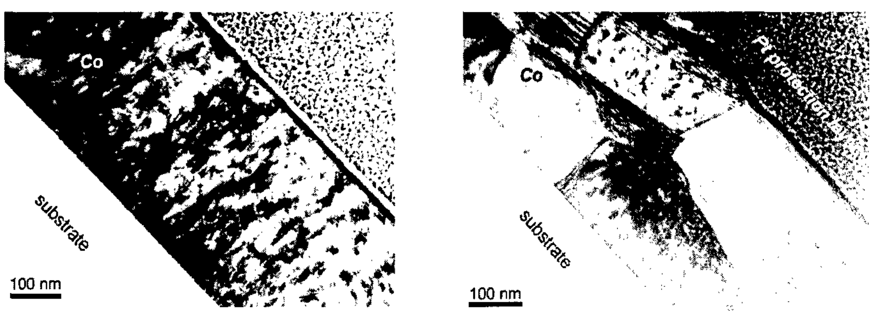

Figure 3. Cross-sectional TEM micrographs of Co thin films (FIB specimen preparation). Left: as-deposited state, right: after annealing at 450 °C. Grain growth is observed during annealing.

Two representative cross-section TEM images in Fig. 3 demonstrate the microstructural evolution during the annealing procedure. In the as-deposited state, small columnar grains with a mean diameter of 30 nm and a length of 80 nm dominate the morphology. These grains were found to grow at 250 °C starting at the film-substrate interface. For T_{max} = 350 °C and 450 °C, all original small grains are converted into large grains. The average lateral grain dimension amounts to 650 nm for T_{max} = 450 °C. Such a secondary recrystallization was also reported for CuNi(Mn) thin films [5].

The XRD measurements of all samples show patterns (Fig. 4) representative of hcp Co. In the as-deposited state, broad reflections occur indicating small grains with (100), (001), and (101) planes parallel to the surface (lattice parameters: a= 0.2507 nm and c= 0.4067 nm). The reflection intensity relations correspond nearly to a random orientation distribution, however, with a weak preference of the hexagonal c-axis perpendicular to the film plane. Furthermore, texture measurements showed an additional weak (101) texture. During annealing, the texture strongly changes. After annealing at and above 350 °C, only a c-axis texture is found in the XRD scans. Further measurements indicate an additional small fraction of face-centered-cubic crystallites, which show a (111) texture after annealing.

Two examples of MOKE results are given in Fig. 5. An isotropic magnetic behavior within the film plane was confirmed by measurements in different azimuthal directions. A close correspondence of the magnetostrictive deflection curves ("layer-bulk" effect) to the MOKE curves (surface related signal down to about 10 nm) is ascertained. Note, that the hysteresis loop of the annealed sample reflects a hard-axis behavior for the in-plane magnetization.

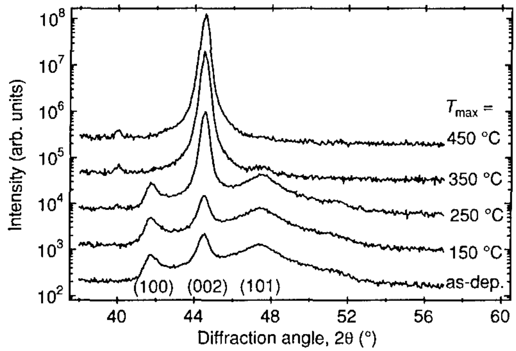

Figure 4. Reflections of hcp-Co as measured by XRD for the as-deposited and annealed Co films (annealing temperature T_{max}, as indicated). The increasing abundance of c-axis texture component is indicated by the intensity changes of the reflections.

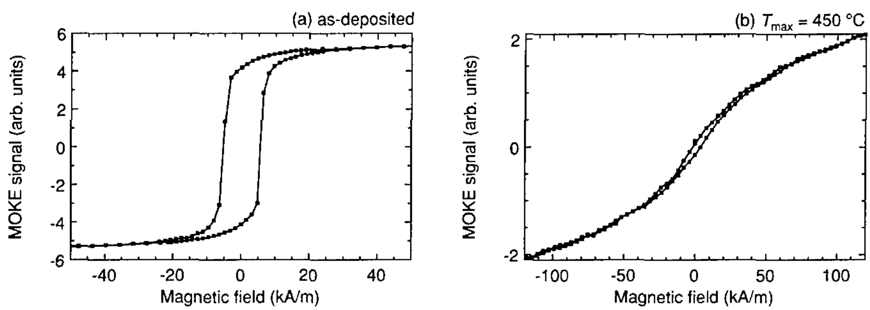

Figure 5. MOKE signals of the as-deposited and an annealed (450 °C) Co film demonstrate the surface magnetization behavior. Main features correspond to the deflection changes in figure 2.

DISCUSSION AND CONCLUSIONS

The microstructural studies have shown that the as-deposited film is polycrystalline, weakly textured, whereas the annealed state with T_{max}>250 °C is c-axis textured. Because the annealed state is more definite than the as-deposited one, only the annealed state is considered in the following. To discuss the magnetostriction behavior, (i) the influence of the texture on the magnetostriction and (ii) the correlation between magnetostriction and substrate deflection will be considered, and (iii) the experimental results will be compared to model calculations.

In the first approximation for hexagonal materials, the following equation describes the saturation magnetostriction in a single crystal with the material parameters λ_A, λ_B, λ_C, and λ_D:

$$\begin{aligned}\lambda_{\alpha\beta} &= \lambda_A\left[(\alpha_1\beta_1+\alpha_2\beta_2)^2-(\alpha_1\beta_1+\alpha_2\beta_2)\alpha_3\beta_3\right]+\lambda_B\left[(1-\alpha_3^2)(1-\beta_3^2)-(\alpha_1\beta_1+\alpha_2\beta_2)^2\right]\\ &+\lambda_C\left[(1-\alpha_3^2)\beta_3^2-(\alpha_1\beta_1+\alpha_2\beta_2)\alpha_3\beta_3\right]+4\lambda_D(\alpha_1\beta_1+\alpha_2\beta_2)\alpha_3\beta_3\end{aligned} \tag{1}$$

where the measuring direction has the direction cosines α_1, α_2, and α_3 and the magnetic field, β_1, β_2, and β_3 [8]. The direction cosines refer to a cubic reference system with x and z along the hexagonal [100] and [001] direction, respectively. The magnetostriction reference is related to the ideally demagnetized state (IDS) with a regular three-dimensional domain distribution along the easy directions.

If the demagnetized state corresponds to the IDS, one obtains the longitudinal and transverse magnetostriction, $\lambda_\parallel$ and $\lambda_\perp$, for a c-axis-textured film from Eq. (1) [9]. It is

$$\lambda^{\mathrm{IDS}}_{\parallel,\mathrm{text}} = \lambda_A \qquad\qquad \lambda^{\mathrm{IDS}}_{\perp,\mathrm{text}} = \lambda_B \tag{2}$$

In thin films, however, one has an initial magnetic state (IMS), which departs from the IDS. Therefore, one has an initial change of the dimension of the sample. Considering a film with magnetic moments under a fixed angle Θ to the plane's normal, Schelp et al. [9] obtained for the magnetostriction of the IMS (in relation to the IDS)

$$\lambda^{\mathrm{IDS}}_{\mathrm{IMS}} = \frac{1}{2}(\lambda_A+\lambda_B)\sin^2\Theta \tag{3}$$

The extreme cases are Θ=90° and Θ=0° for in-plane and perpendicular spontaneous magnetization. The magnetostriction responses of the thin films as measured from the saturated state in relation to the state without magnetic field (H=0) are given by

$$\lambda_\parallel = \lambda^{\mathrm{IDS}}_\parallel - \lambda^{\mathrm{IDS}}_{\mathrm{IMS}} \qquad\qquad \lambda_\perp = \lambda^{\mathrm{IDS}}_\perp - \lambda^{\mathrm{IDS}}_{\mathrm{IMS}} \tag{4}$$

In thicker magnetic films, one has mostly an in-plane magnetization. But for films with strong magnetic anisotropy, the magnetization may also rotate out-of plane.

In the general case, the relation between magnetostriction and substrate deflection is complicated and no practicable equations exist, because of the tensor character of both elastic properties and magnetostriction [10]. The most complicated case with still manageable efforts is the case of isotropic mechanical properties of film and substrate and anisotropic magnetostrictive strains in two perpendicular directions within the film plane [11]. In this case one obtains a linear equation set between the deflections $\delta_\parallel$ and $\delta_\perp$ and the magnetostrictive strains $\lambda_\parallel$ and $\lambda_\perp$. For the simplified case of equal Poisson's ratios of film and substrate, the equation set decouples, and one finds:

$$\lambda_{\parallel,\perp} = \frac{4}{3}\frac{E_s}{E_f}\frac{t_s^2}{t_f}\frac{1}{l^2}\delta_{\parallel,\perp} \tag{5}$$

where E and t are Young's modulus and thickness of film (f) and substrate (s), respectively. Using Eq. (5), $E_f = 210$ GPa for (polycrystalline) Co, $E_s = 148$ GPa as an average in the Si (100) plane, and t_s=380 μm, the magnetostrictive strain values given in the right-hand axes of Fig. 2 were calculated.

For a comparison between theory and experiment one can use Eqs. (2) – (4) and the material parameters of single-crystalline hcp Co, i.e., $\lambda_A = -50\times10^{-6}$, $\lambda_B = -107\times10^{-6}$, $\lambda_C = 126\times10^{-6}$, and $\lambda_D = -105\times10^{-6}$ [12]. The saturation magnetostriction values of a c-axis-textured film in relation to the IMS are: $\lambda_{\parallel} = -52\times10^{-6}$, $\lambda_{\perp} = 52\times10^{-6}$ and $\lambda_{\parallel} = -50\times10^{-6}$, $\lambda_{\perp} = -107\times10^{-6}$ for in-plane and perpendicular initial magnetization, respectively. Comparing to the experimental results of Fig. 2, one has to take into account the incomplete saturation of the c-axis-textured films. Already within a qualitative (sign-wise) comparison, one may speculate that the magnetization in the IMS is not confined to the film plane but may have a strong out-of-plane component at least.

A c-axis-textured Co film should be expected to develop a perpendicular crystalline anisotropy. This may result in a rotation of the magnetization in the H=0 state into the direction perpendicular to the film plane, e.g., via formation of stripe domains [13] or with a perpendicular magnetization [9]. For further insight into the spontaneous magnetization behavior, measurements of the domain structure by means of Kerr microscopy and of the magnetization behavior in large magnetic fields applied within and perpendicular to the film plane are now under investigation.

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