Perpendicular Magnetic Anisotropy of Co-Au Multilayers Induced by Interface Sharpening

F. J. A. den Broeder, D. Kuiper, A. P. van de Mosselaer, and W. Hoving
Philips Research Laboratories, 5600 JA Eindhoven, The Netherlands

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Co-Au multilayers with Co layer thicknesses between 22 and 5 Å, prepared by ion-beam sputtering, have an easy-plane magnetic anisotropy. Their saturation magnetization indicates the presence of diffuse interfaces, caused by ion-beam mixing. Annealing of the multilayers at 250–300 °C leads to a perpendicular anisotropy for Co thicknesses below about 14 Å. This effect is attributed to a strongly enhanced interface anisotropy, which is due to sharpening of the interfaces as revealed by x-ray diffraction.

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Magnetic thin films which are preferentially magnetized in a direction perpendicular to the film plane are said to have a "perpendicular" magnetic anisotropy. They are currently of great interest in relation to high-density magnetic recording. A popular example is CoCr thin films whose anisotropy has a magnetocrystalline origin.

In the search for other candidate recording media, multilayered thin films offer the possibility that, because of the abundance of interfaces, the anisotropy is dominated by the existence of interface anisotropy. This has recently been demonstrated by Carcia, Meinhaldt, and Suma[1] for sputtered multilayers and by Draaisma, den Broeder, and de Jonge[2] for vapor-deposited Co-Pd multilayers. In both cases the preferred magnetization direction became perpendicular to the film plane for a Co layer thickness below about 8 Å. The somewhat higher interface anisotropy for the vapor-deposited films was attributed to their sharper interfaces. As expected, diffusion treatments and ion bombardment gave a decrease of the anisotropy by Co-Pd interfacial mixing.[3]

In contrast, Co-Pd multilayers which we prepared by ion-beam sputtering (IBS) invariably had an easy-plane anisotropy. This may be explained by a drastic decrease of the interface anisotropy due to the occurrence of ion-beam mixed interfaces. Mixing of this kind had been demonstrated for Cu-Fe multilayers by Mössbauer spectroscopy.[4]

The present Letter is concerned with the magnetic anisotropy of Co-Au multilayers prepared by IBS. For these, too, we found an easy-plane anisotropy, while their saturation magnetization indicated Co-Au interfacial mixing. Since Au and Co are mutually insoluble, we then speculated whether a "back" diffusion of Au and Co, by a suitable thermal treatment, could lead to sharper interfaces. It will be shown later that, in spite of a marked susceptibility for the coarsening of the Co-Au multilayers, this subtle effect can indeed be induced while we maintain the periodic structure. It then gives rise to a greatly enhanced magnetic interface anisotropy which, below a certain Co layer thickness, leads to a preferred magnetization direction normal to the film plane.

The Co-Au multilayers were prepared by an IBS technique in which the modulation is achieved by our periodically deflecting a 1.5-kV Ar^+ beam to two separate targets.[5] The background pressure was $p \approx 10^{-8}$ Torr, while during sputtering the Ar pressure (containing less than 0.2 ppm O) was 2×10^{-5} Torr. The films were deposited at a rate of about 0.3 Å/s on to oxidized silicon strips ($4\times12\ \mathrm{mm}^2$) with an intervening 1000-Å Au layer. Their periodic structure was checked by x-ray diffractometry (XRD) with the scattering vector perpendicular to the film plane. Table I lists structural data of the films, deposited at room temperature. The thickness t_{Co} of the Co layers was derived from wet-chemical analysis, while that of the Au layers, t_{Au}, was obtained by subtraction of t_{Co} from the modulation period D determined by XRD.

The high-angle multilayer reflections were located around the 111 reflection of the Au base layer. This indicates a texture in which (111) fcc Au layers alternate with (00.2) hcp or (111) fcc Co layers. An fcc structure for the Co layers, however, is less likely on account of the magnetic anisotropy data given below. Also, recent electron-diffraction studies of Au-Co-Au sandwich films revealed an hcp Co structure.[6]

The magnetization of the films was measured with a

TABLE I. Structural data and saturation magnetization values M_s per unit Co volume of Co-Au multilayers before and after annealing at 250 °C or 300 °C; N = number of bilayers.

Sample	N	t_{Au} (Å)	t_{Co} (Å)	As dep. M_s (T)	$T_a = 250\,^\circ\mathrm{C}$ M_s (T)	$T_a = 300\,^\circ\mathrm{C}$ M_s (T)
a	25	54.1	22.4	1.59	1.71	
b	25	57.0	18.2	1.55		1.68
c	50	27.7	10.9	1.59	1.71	
d	50	28.6	9.5	1.53		1.76
e	80	26.8	8.7	1.51	1.72	
f	80	27.5	8.0	1.51		1.71
g	100	26.3	5.6	1.11	1.34	
h	100	26.3	5.2	0.95		1.59

vibrating-sample magnetometer in fields up to 1300 kA/m, both parallel and perpendicular to the film plane. The values of the saturation magnetization M_s, taken per unit volume Co, are given in Table I. It is seen that M_s becomes increasingly lower than the value for pure Co ($M_s^0 = 1.76$ T) as the Co layers become thinner. This loss of magnetization is interpreted by the presence of Co-Au mixed interfaces, in which the Co is not ferromagnetic. From the M_s data at various t_{Co}, the amount of this mixing is estimated to be equivalent to the loss of about 2-Å Co per period. All the multilayers showed a preference for magnetization in the film plane. A discussion of their anisotropy as a function of t_{Co} is postponed to later in this Letter.

Au and Co are practically insoluble in each other in the solid state. In view of a possible relation between the absence of perpendicular anisotropy and the interfacial diffuseness, we wondered whether the interfaces could be made sharper by a thermal treatment which causes a backward diffusion of Co and Au from the mixed regions to the pure metal layers. This would be expected to occur when the width of the mixed regions is larger than the width of an Au-Co interface at thermal equilibrium, which might even be atomically sharp.

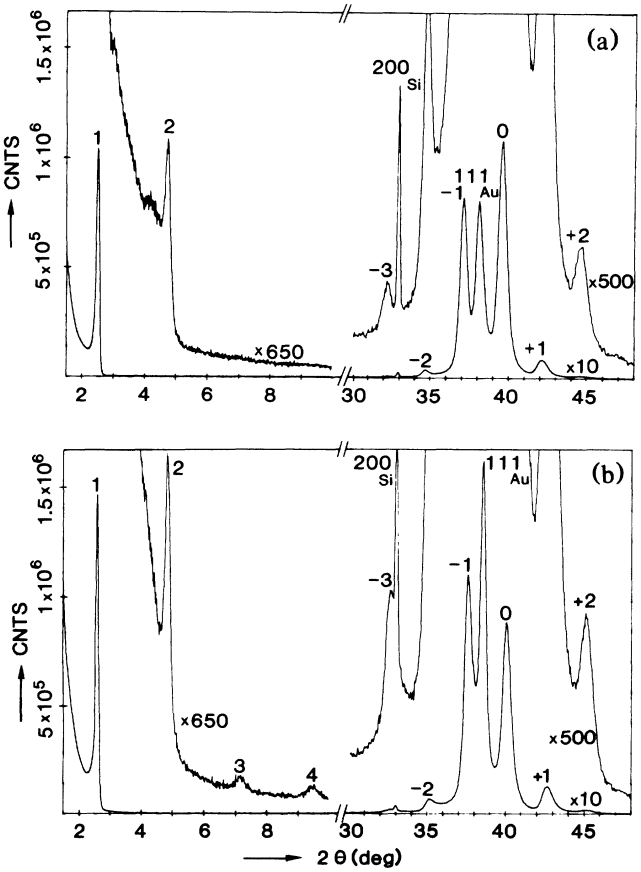


FIG. 1. XRD patterns (Cu $K\alpha$) of multilayer d (a) before and (b) after annealing for $\frac{1}{2}$ H at 300 °C.

However, it was anticipated also that, because of the positive Au-Co interfacial energy,[7] the periodic structure might disappear by the agglomeration of the layers to form a coarse two-phase structure. But since interface sharpening involves a much shorter diffusion distance than structural coarsening, one may expect it to be established much sooner.

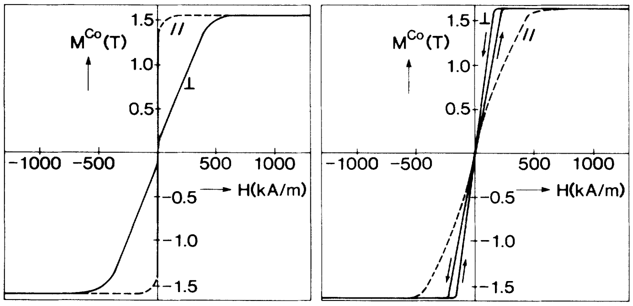
The multilayers listed in Table I were annealed at 250–400 °C in a reducing H_2/N_2 atmosphere. XRD then showed for all films annealed for 1 h at 400 °C only intense Au(111) and Co(0.02) reflections, indicating that the periodic structure was destroyed. The preferred magnetization direction remained in the film plane, while M_s increased up to the value for pure Co.

However, on annealing for $\frac{1}{2}$ h at 250 °C and 300 °C, the samples e to h showed only partial coarsening (appearance of a faint Co(00.2) reflection, and weakening of the multilayer reflections), while the periodicity of the films a to d seemed to be fully preserved. For the latter samples, the intensities of the multilayer reflections showed interesting changes compared to those before annealing. This is illustrated in Fig. 1 for sample d. At low angles the first- and second-order reflections become stronger, while the third- and fourth-order multilayer reflections appear after annealing. This strongly indicates that the composition profile has become more rectangular. At high diffraction angles the reversal in intensity of the two strongest multilayer reflections (0 and −1) is remarkable.

In Table II the peak intensities of the high-angle reflections are compared with the intensities calculated kinematically for a multilayer with perfectly sharp interfaces of about the same composition (5 and 11 close-packed planes of Co and Au, respectively, per period), with bulk values for the Co and Au spacings.[8] It is seen that the intensities of most of the reflections approach those of the ideal multilayer rather well on annealing. In particular, the reversal in intensity of the reflections 0

TABLE II. Values of $d = \lambda/2\sin\theta$ and relative intensities of multilayer reflections for sample d before and after annealing at 300 °C, compared with calculated values for a corresponding model multilayer; L is defined by $2D\sin\theta = L\lambda$.

	As dep.		$T_a = 300°$ C		Calculated		
Order	d (Å)	I (%)	d (Å)	I (%)	L	d (Å)	I (%)
−3	2.78	0.3	2.74	1.0	13	2.78	3.3
−2	2.58	2.1	2.55	3.6	14	2.58	0.3
−1	2.42	75.9	2.39	100	15	2.41	100
0	2.28	100	2.25	79.7	16	2.26	66.5
+1	2.14	5.8	2.12	9.5	17	2.12	10.0
+2	2.03	0.5	2.01	0.9	18	2.00	1.3
Au_{111}	2.36		2.34			2.35	



(a) (b)

FIG. 2. Magnetization curves measured in fields parallel and perpendicular to the film plane for multilayer c (a) before and (b) after $\frac{1}{2}$ h annealing at 250 °C.

and -1 is in accordance with the calculation. It also appears from Table II that the d values corresponding to the superlattice reflections and d_{111} of the Au base layer have decreased. With the assumption of a Poisson contraction, this is interpreted by a relaxation of a compressive stress in the sputtered film. The most important conclusion, however, is that by annealing at 250 °C or 300 °C the Au-Co interfaces become sharper by an effective negative or "uphill" diffusion, while the periodic structure is maintained.

Apart from an increase of M_s, as seen from the data in Table I, this phenomenon is accompanied by a dramatic change of the anisotropy. While the field needed to saturate the multilayers a and b normal to their plane decreased from 1000 to about 400 kA/m, the multilayers with thinner Co layers acquired a perpendicular anisotropy.

Figure 2 shows for sample c the magnetization curves before and after annealing for $\frac{1}{2}$ h at 250 °C. In the as-deposited state, Fig. 2(a), the multilayer is more easily magnetized in the film plane, but upon annealing, Fig. 2(b), the film normal has become the easy direction. An interesting detail of the perpendicular curve in the latter case is that it only shows hysteresis in relatively high fields. This is an effect which is typical for thin films with perpendicular anisotropy when difficult nucleation of reverse domains from the saturated state is followed by easy domain-wall motion dictated by the changing equilibrium between magnetostatic energy, field energy, and domain-wall energy.[9]

Figure 3 demonstrates clearly for sample d, by the complete reversal of the torque curve after annealing for $\frac{1}{2}$ h at 300 °C, the change of the easy magnetization direction from parallel toward normal to the film plane.

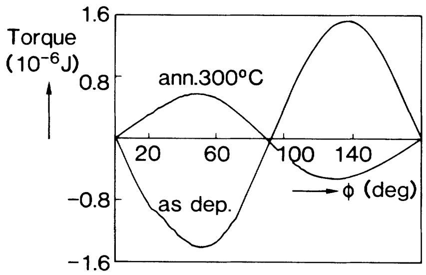


FIG. 3. Magnetic torque curves for sample d before and after $\frac{1}{2}$ h annealing at 300 °C, measured in a field of 1450 kA/m; ϕ is the angle between field direction and film normal.

The anisotropy energy K_u per unit volume Co was determined from the area between the parallel and perpendicular magnetization curves. For a few films K_u was also obtained from torque curves, with K_u then defined as the torque integrated from $\phi=0$ to $\phi=\pi/2$. The values obtained from both methods agree very well. Figure 4 shows plots of $K_u t_{\mathrm{Co}}$ as a function of t_{Co} for the films before and after annealing. While K_u is always negative for the as-deposited samples, the annealed films show a crossover from planar towards perpendicular anisotropy. This transition seems to occur at larger t_{Co} with higher annealing temperature. The perpendicular anisotropy occurring below $t_{\mathrm{Co}} \approx 14$ Å is in accordance with ferromagnetic resonance studies by Chappart *et al.*[10] of vapor-deposited Au-Co-Au sandwich films.

As possible causes for the increase of K_u (which, e.g., for sample c is about 5×10^5 J/m^3) induced by annealing, one may consider (i) relief of compressive stresses which were caused by recoil implantation of Ar; (ii) relief of coherency stresses; (iii) increase of perpendicular interface anisotropy.

By measurement of the curvature of some samples the magnitude of the compressive stresses was found to be about 0.2 GPa. With the negative magnetostriction of hcp Co ($\lambda_{\mathrm{Co}} \approx -50\times10^{-6}$), stress relaxation would yield a change of the magnetoelastic anisotropy energy

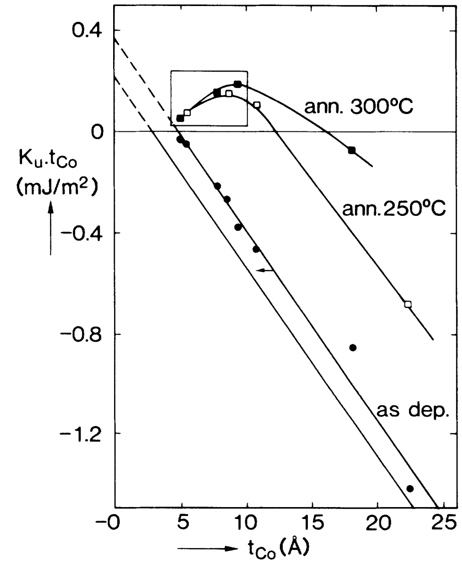


FIG. 4. Plot of $K_u t_{\mathrm{Co}}$ vs t_{Co} for multilayers before and after annealing at 250 °C and 300 °C; see text.

in the right direction, but its magnitude $\Delta K = 3/2\lambda\sigma = 1.5\times10^4$ J/m^3 is far too small.

When the Co layers are coherent with the Au layers, they are under a tensile stress due to the smaller size of Co atoms. With a negative λ_{Co} this would give a perpendicular magnetoelastic anisotropy. A relief of this stress by annealing would then *decrease* this anisotropy. Apart from this argument, it is improbable that for the present layer thicknesses the Co-Au interfaces are coherent, since the atomic size difference between Co and Au is as large as 15%. The interfaces are more likely to be incoherent, consisting of arrays of misfit dislocations. The slope of K_v for the unannealed films (Fig. 4) (see below) also points to the absence of coherency stresses.

Rejecting changes in magnetoelastic anisotropy as causes for the increased perpendicular anisotropy, we are left to conclude that the effect is due to an enhancement of the interface anisotropy by interface sharpening, as observed by XRD.

In Fig. 4, the data for the as-deposited films yield a straight line which obeys the relation $K_u t_{Co} = K_v t_{Co} + 2 \times K_s$, in which K_v is a volume contribution to the anisotropy and K_s is the interface anisotropy energy. Taking into account the above-mentioned effective Co thickness loss due to interfacial alloying, this line may be corrected by a 2-Å displacement to lower t_{Co}. We then obtain $K_v = -0.75$ mJ/m^3 and $K_s = 0.1$ mJ/m^2. It is to be noted that the value of K_v is almost equal to the sum of the demagnetization energy (-1.23 MJ/m^3) and the crystalline anisotropy (0.51 MJ/m^3) for hcp Co. Although the Co layers might be in a state of stress as discussed above, a strain anisotropy contribution to K_v is evidently small.

The data in Fig. 4 for the annealed multilayers do not show a linear behavior. With decreasing t_{Co}, $K_u t$ passes through a maximum but becomes less positive below $t_{Co} \approx 10$ Å. This effect is attributed to a decrease of the total Co-Au interface by partial coarsening, which becomes a more and more competitive reaction for thinner Co layers. If we exclude therefore the data contained in the box in Fig. 4, K_s can only be estimated to be about 0.45 mJ/m^2 for the multilayers annealed at 250 °C. The large increase of K_s caused by interface sharpening confirms a recently proposed pair-interaction model for the interface anisotropy which predicts that the magnitude of K_s is very sensitive to the precise distribution of magnetic and nonmagnetic atoms at the interface.[11]

In conclusion, we have shown that the diffuse interfaces of IBS Co-Au multilayers can be made sharper by an appropriate heat treatment while maintaining the periodic structure. This enhances the magnetic interface anisotropy, by which the easy magnetization direction becomes perpendicular to the film plane for Co layers thinner than about 14 Å. This is the first case in which increasing the sharpness of the interface by a thermal treatment has been shown to produce increased perpendicular anisotropy.

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