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Magnetic properties and structure of Pd/Co and Pd/Fe multilayers

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Pd/Co and Pd/Fe multilayer films containing ultrathin Co and Fe layers were prepared by vapor deposition on substrates at room temperature. Their modulated structure, even for films containing 2-Å-thin Co and Fe layers, was proved by x-ray diffraction and transmission electron microscopy. Below a Co layer thickness of about 8 Å, the Pd/Co multilayers acquire an easy magnetic axis perpendicular to the film, which is mainly caused by magnetic interface anisotropy. This leads for multilayers containing Co monolayers to almost rectangular hysteresis loops, by which these films may be very suitable as a perpendicular magnetic recording medium. Pd/Fe multilayers also have a perpendicular interface anisotropy, but the shape anisotropy dominates. Per unit Co volume the Pd/Co multilayers have a higher saturation magnetization than pure Co, which is attributed to an induced ferromagnetism on Pd interfacial atoms.

Many years ago, Néel[1] predicted the existence of a magnetic surface anisotropy, caused by the reduced symmetry in the surroundings of a surface atom. In principle, such an anisotropy may also be present at the interface between a magnetic and a nonmagnetic metal. In a multilayer structure where there is an abundance of interfaces, it may then affect the magnetic anisotropy of the film as a whole. A large anisotropy found for compositionally modulated Cu-Ni thin films may have been caused by an easy plane interface anisotropy.[2] Recently, Carcia, Meinhaldt, and Suna[3] reported that sputtered Pd/Co multilayers with Co layers thinner than 8 Å had an easy magnetic axis normal to the film plane, as a result of a perpendicular interface anisotropy.

The present paper deals with structural and magnetic properties of Pd/Co multilayers prepared by vapor deposition on cold substrates. We found that in these films composition changes are very sharp, resulting in a large interface anisotropy. So far unreported multilayers containing Co monolayers then show almost rectangular hysteresis loops in perpendicular fields. The obtained results prompted us to investigate also some Pd/Fe multilayers with ultrathin Fe.

The multilayers were prepared in UHV by *e*-beam evaporation from two sources onto Si substrates at room temperature, with a base layer of 200-Å Pd. The vapor streams were interrupted alternatingly during predetermined times with mechanically driven shutters, while the deposition rate was kept constant at a fixed value in the range of 0.1–1 Å/s as monitored by a quartz resonator.

Table I summarizes constitutional details of the prepared Pd/Co films. Chemical analysis of a representative series of multilayers showed that within a few percent the intended amount of Co was present, while that of Pd was 10–20% higher. The multilayer structure was checked by x-ray diffractometry (XRD) using Cu$k\alpha$ radiation. Pure Pd films showed a pronounced [111] fiber texture. The multilayers gave near the $(111)_{\mathrm{Pd}}$ peak 2–3 superlattice reflections, which allowed the determination of the bilayer period **D**. As an example, Fig. 1 shows the XRD profile for a multilayer

TABLE I. Constitution and magnetic properties of Pd/Co multilayers; N = number of bilayers; t_{Co} = Co layer thickness; t_{Pd} = Pd layer thickness; D = bilayer period determined by XRD, I_s = saturation magnetization per unit Co volume; I_R = remanent magnetization; $\mu_0 H_c^{\perp}$ = coercivity.

N	t_{Co} (Å)	t_{Pd} (Å)	D (Å)	I_s (T)	$I_r^{\perp}/I_r^{\parallel}$	$I_r^{\perp}/I_s$	$\mu_0 H_c^{\perp}$ (T)
52	12.3	45	66	2.02	0.20	0.09	0.020
34	10.2	45	–	2.10	0.19	0.07	0.015
56	8.2	45	37	2.02	0.63	0.13	0.045
59	6.2	45	67	1.96	1.8	0.27	0.04
61	4.1	45	58	1.99	7.4	0.45	0.06
100	4	18	26.5	2.70	3.2	0.35	0.09
150	4	9	15.3	2.69	2.1	0.20	0.095
250	4	4.5	8.5	2.36	1.0	0.12	0.08
150	2	18	22.2	2.80	11	0.96	0.16
150	2	13.5	20.0	2.66	9	0.96	0.215
200	2	11.2	17.7	2.81	13	0.94	0.245
200	2	6.7	9.8	3.05	18	0.96	0.25
200	2	4.5	6.55	2.77	20	0.93	0.245

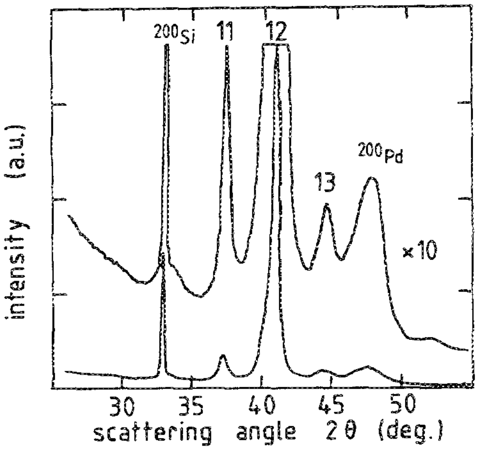

FIG. 1. XRD profile (Cukα) of a multilayer containing 4-Å Co layers and 18-Å Pd layers ($N = 100$). Indicated are the substrate (Si,Pd) and the multilayer reflections with order numbers L, defined by $2D \sin \theta = L\lambda$.

composed of 18-Å Pd and 4-Å Co layers. It is to be noted that a Co thickness of 2 Å is that of a monolayer. As it appears from Table I, D generally exceeds the programmed bilayer period, presumably owing to a higher Pd content.

A few samples were also studied by transmission electron microscopy (TEM). Planar sections showed a polycrystalline fcc structure with a grain size of 200–700 Å. Cross sections of the films revealed a columnar structure with [111] texture, with many [111] microtwins parallel to the multilayer plane. The modulated structure could be made visible in bright field by interference between the primary beam and the $L = 1$ superlattice reflection, or in dark field by interference between a strong high-order superlattice reflection and one of its satellites. As an example, Fig. 2 demonstrates the modulation in a multilayer containing 2-Å Co layers. This result provides strong evidence for very steep concentration changes in all the multilayers, possibly occurring across one or a few lattice planes.

FIG. 2. TEM micrograph of a cross section of a multilayer composed of 6.5-Å Pd and 2-Å Co layers. The picture is a dark-field image, in which the multilayer fringes at a distance of about 9.5 Å were made visible by interference between the $L = 4$ and $L = 5$ superlattice reflections.

The magnetic moment of the samples was measured at room temperature with a vibrating sample magnetometer, applying fields up to 1.7 T, both parallel and perpendicular to the film plane. The magnetic properties of the Pd/Co films are collected in Table I.

It appears that the saturation magnetization I_s, calculated per unit Co volume, is for all samples significantly higher than for pure Co ($I_s^0 = 1.76$ T). Since it is well known that in Pd-Co alloys Pd atoms are polarized by neighboring Co atoms,[4] the present deviation may be explained similarly. Then, assuming that the induced magnetization ΔI_s on the Pd atoms is confined to the Pd planes nearest to each Co layer at a distance $d \approx 2.15$ Å, one should have

$$I_s = I_s^0 + 2\Delta I_s d / t_{\mathrm{Co}} , \tag{1}$$

where t_{Co} is the thickness of an individual Co layer. We then find $\Delta I_s = 0.55 \pm 0.25$ T, which roughly agrees with a quoted value of $0.6\mu_B$ per Pd atom as the nearest neighbor of a Co atom in Pd-Co alloys.[4]

Figure 3 illustrates the hysteresis curves for a representative series of Pd/Co multilayers. It is seen that with decreasing t_{Co} the films become easier to magnetize in perpen-

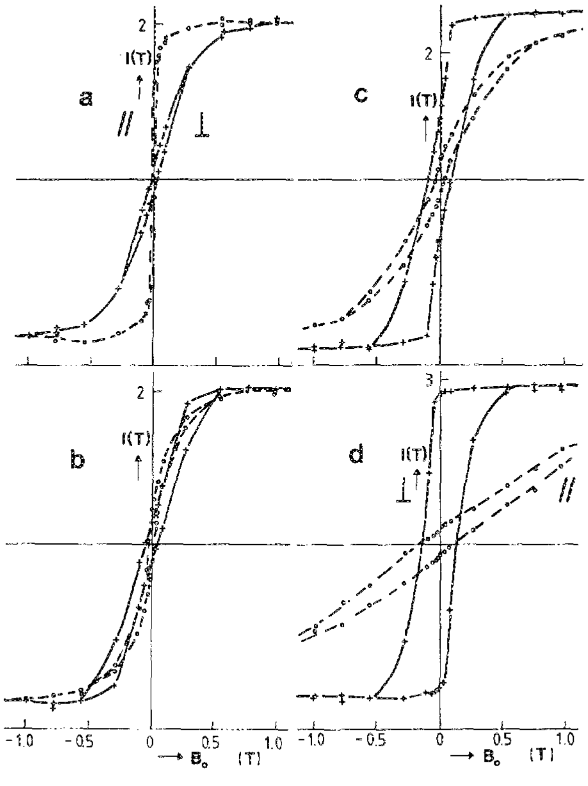

FIG. 3. Magnetic hysteresis loops of a series of Pd/Co multilayers, showing the effect of decreasing t_{Co}. They were measured in fields parallel (---) and perpendicular (—) to the film plane and are shown up to $B_0 = 1.1$ T. The vertical scale is the magnetization per unit Co volume. (a) $t_{\mathrm{Co}} = 12.3$ Å, $t_{\mathrm{Pd}} = 45$ Å; (b) $t_{\mathrm{Co}} = 8.2$ Å $t_{\mathrm{Pd}} = 45$ Å; (c) $t_{\mathrm{Co}} = 4$ Å, $t_{\mathrm{Pd}} = 18$ Å; (d) $t_{\mathrm{Co}} = 2$ Å, $t_{\mathrm{Pd}} = 18$ Å. To be noted is the almost rectangular shape of the perpendicular loop in case (d).

TABLE II. Constitution and magnetic properties of Pd/Fe multilayers; t_{Fe} = Fe layer thickness; t_{Pd} = Pd layer thickness; D = bilayer period determined by XRD; I_s = saturation magnetization per unit Fe volume; $\mu_0 H_s^{\perp}$ = perpendicular field which gives saturation.

N	t_{Fe} (Å)	t_{Pd} (Å)	D (Å)	I_s (T)	$\mu_0 H_s^{\perp}$ (T)
125	6	18	26.5	2.70	1.1
136	4	18	24.4	2.74	0.85
150	2	18	22.2	2.52	0.46

dicular fields and increasingly more difficult in parallel fields. This leads to a crossover from easy plane to easy axis anisotropy between $t_{\mathrm{Co}} = 8$ and 6 Å, as is demonstrated by the remanence ratio $M_R^{\perp}/M_R^{\parallel}$ becoming larger than one (see Table I). This agrees with the results reported for sputtered Pd/Co multilayers which were prepared down to $t_{\mathrm{Co}} = 4.7$ Å.[3] In our case the increasing perpendicular anisotropy extends even to multilayers with $t_{\mathrm{Co}} = 2$ Å. For the latter films the hysteresis curves become almost rectangular with remanences approaching the saturation magnetization [see Fig. 3(d)] They also exhibit high coercivities up to about 0.25 T (2.5 kOe). Inspection of the data in Table I shows that the dramatic increase in perpendicular remanence and coercivity when going from $t_{\mathrm{Co}} = 4$ Å to $t_{\mathrm{Co}} = 2$ Å occurs for all multilayers, irrespective of t_{Pd}.

Sputtered Pd/Co multilayers with an easy perpendicular axis have been proposed before as candidate media for perpendicular recording.[3] In such an application the high remanences of the vapor-deposited Pd/Co multilayers with $t_{\mathrm{Co}} \approx 2$ Å will be beneficial in obtaining a high signal-to-noise ratio.

In a separate paper[5] it is shown that the uniaxial anisotropy constant K_u per unit Co volume, as determined from the parallel and perpendicular magnetization curves of the present Pd/Co multilayers, satisfies the relation

$$K_u = K_v + 2K_s/t_{\mathrm{Co}}, \tag{2}$$

with $K_v = -0.72 \times 10^6$ J/m^3 and $K_s = 0.26 \times 10^{-3}$ J/m^2. Here K_v is a volume contribution per unit Co volume, interpreted as the sum of magnetostatic and strain or crystal anisotropy, while K_s is the Co/Pd interface anisotropy energy. It appears that below $t_{\mathrm{Co}} = 7.2$ Å where $K_u > 0$, the interface term is the major source of the intrinsic anisotropy. The value for K_s is larger than that given $(0.16 \times 10^{-3}$ J/m$^2)$ for sputtered Pd/Co multilayers[3] which is probably due to steeper composition gradients in the vapor-deposited layers.

The above results prompted us to investigate a few, similarly prepared, Pd/Fe multilayers containing ultrathin Fe layers. Table II lists their constitution and their relevant magnetic properties. XRD demonstrated the modulated structure.

Electron diffraction showed the films containing a 2-Å Fe layer to be fcc. TEM results for the films with thicker Fe layers were not yet obtained. It is of great interest to determine whether the Fe atomic planes have an 110_{bcc} (α-Fe) or an 111_{fcc} (γ-Fe) symmetry, in view of the controversy on whether γ-Fe is antiferromagnetic,[6] or ferromagnetic with a higher magnetic moment than α-Fe.[7,8] Thus the higher I_s of the present Pd/Fe multilayer compared to pure α-Fe($I_s = 2.15$ T) could either be due to polarization of Pd, or due to γ-Fe ferromagnetism. This question is subject of further studies.

The Pd/Fe multilayers appeared to be magnetized more easily in their plane than along their film normal. But Table II also shows that the perpendicular field needed to achieve saturation ($\mu_0 H_s$) is lower than I_s, while this field sharply decreases with lower t_{Fe}. This points to a perpendicular interface anisotropy, which apparently even for the thinnest possible Fe layers does not overcome the high magnetostatic energy associated with a perpendicular magnetization.[5]

In conclusion, our structural studies of vapor-deposited Pd/Co and Pd/Fe multilayers indicate that these polycrystalline films have very sharp interfaces which may extend over only one or a few lattice planes. The saturation magnetization of the Pd/Co multilayers per unit volume Co exceeds that of pure Co, presumably by induced ferromagnetism on Pd interfacial atoms. They acquire a perpendicular easy axis below a Co layer thickness of about 8 Å, mainly through the effect of interface anisotropy. For multilayers containing monoatomic Co layers this results in high perpendicular remanences and coercivities. In Pd/Fe multilayers a perpendicular interface anisotropy is also present, but even for the thinnest Fe layers it does not overcome shape anisotropy.

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