Origin of very large in-plane anisotropies in (110)-oriented Co/Pd and Co/Pt coherent superlattices

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Within the framework of a continuum free-energy calculation we show that the experimentally observed large uniaxial in-plane anisotropy in epitaxial single-crystal (110)-oriented Co/Pt and Co/Pd superlattices is caused by the magnetoelastic interaction as well as anisotropic lattice distortion, a process unique to coherently strained ultrathin layers. We find in agreement with existing experimental data that the easy axis is aligned in plane along the [001] axis, contrary to bulk cobalt, and that the absolute value of the anisotropy constant is dependent on the intermixing at the interfaces.

The recent availability of single-crystal magnetic-superlattice samples of (100), (110), and (111) orientation has focused interest to the origin of very large magnetic anisotropies found in these structures. It has been found that magnetocrystalline anisotropies and interface anisotropies, as well as anisotropies caused by the magnetoelastic interaction due to large elastic strain fields induced by the mismatch at the interfaces, contribute to the total anisotropy. In the case of Co(111)/Cu(111) superlattices, Lee *et al.* pointed out that the consideration of magnetocrystalline and magnetoelastic contributions alone yields a reasonably good estimate of the observed perpendicular anisotropy.[1]

However, the origin of the large in-plane uniaxial anisotropy for (110)-oriented systems like Co/Pd and Co/Pt systems with values as large as 5.3×10^7 erg/cm^3 for Co/Pt,[2,3] which is larger than the shape anisotropy, has not been addressed yet. We show that this anisotropy is dominantly magnetoelastic in nature caused by interface mismatch-induced strain fields and we calculate the value.

For the calculation, we assume that the superlattice structure is coherently strained by the lattice mismatch at the interfaces. The elastic strain fields are calculated by minimizing the elastic free energy of the system and by solving for the elastic boundary conditions at the interfaces. We note that this calculation yields limiting values for the strains which, however, might be partly released in a real system due to dislocation formation. For now we discard strain relaxation effects. We start with the free-energy density E of the system averaged over one bilayer period. The coordinate system is chosen such that the x_3 axis is perpendicular to the layers and the x_1 axis is along the in-plane (001) axis. Since the layers are homogeneously stressed along the x_1x_2 plane only the strains ϵ_1, ϵ_2, and ϵ_3 are nonzero. Let f_1 and f_2 be the fractional contributions of the Co layer thickness d_1, and the Pt or Pd layer thickness d_2, respectively, to the bilayer thickness, i.e., $f_i=d_i/(d_1+d_2)$. With $c_{ij,1}$ being the elastic constants of Co, $c_{ij,2}$ those of Pt or Pd, $\epsilon_{i,1}$ and $\epsilon_{i,2}$ the corresponding strains, b_{ij} the magnetoelastic tensor of Co, and α_i the magnetization direction cosines for axis x_i, the averaged free-energy density is

$$E=f_1\sum_{i,j}c_{ij,1}\epsilon_{i,1}\epsilon_{j,1}+f_2\sum_{i,j}c_{ij,2}\epsilon_{i,2}\epsilon_{j,2}+f_1\sum_{i,j}b_{ij}\alpha_i^2\epsilon_{j,1}\ . \tag{1}$$

The first two terms on the right-hand side are the elastic energies of the two layers of a bilayer period and the third term is the magnetoelastic energy in the magnetic layer. Here we use the 6×6 matrix notation for c_{ij} and b_{ij}. The tensor constants c_{ij} and b_{ij} are rotated from the crystallographic reference frame into the (110)-oriented layer frame. Since $\epsilon_i=0$ for $i>3$, the sums in Eq. (1) run over $i,j=1\ldots3$. At the interfaces the strains must accommodate the in-plane mismatch

$$a_1(1+\epsilon_{i,1})=a_2(1+\epsilon_{i,2})\ ,\quad i=1,2\ , \tag{2}$$

with a_i the lattice parameters of the two constituent materials. The calculation of the magnetic anisotropy is now performed in two steps. First the equilibrium conditions for Eq. (1) are solved neglecting the third term. Since the magnetostriction constants (see below) are several orders of magnitude smaller than the involved strains, we can neglect the magnetoelastic term in Eq. (1) for this step. We obtain a system of six linear equations for the six unknowns $\epsilon_{i,1}$ and $\epsilon_{i,2}$, $i=1\ldots3$, which we solve numerically.

As a general result we obtain that in a superlattice structure the elastically softer material accommodates the larger fraction of interface strains. In particular, in Co/Pt superlattices the Co layers contain the larger part of strains which in turn (see below) increases the corresponding magnetic anisotropy contributions.

Figure 1 displays the results obtained for Co/Pt (solid

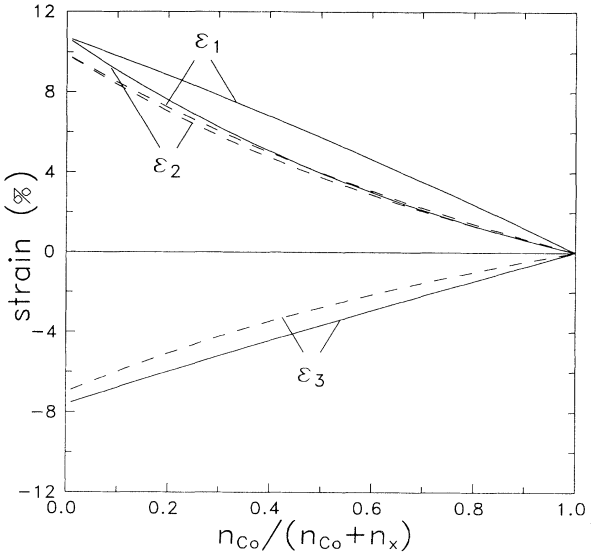

FIG. 1. Strain components ϵ_i as a function of the number of Co atomic layers, n_{Co}, relative to the total number of atomic layers in each bilayer, $n_{\mathrm{Co}}+n_x$, where x = Pt,Pd. The solid (dashed) lines correspond to the results calculated for (110)-oriented Co/Pt (Co/Pd) superlattices.

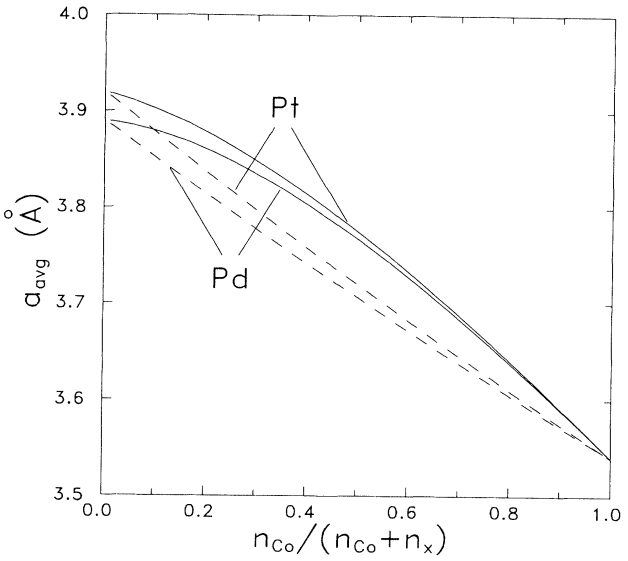

FIG. 2. Calculated average perpendicular lattice constants for (110)-oriented Co/Pt and Co/Pd superlattices (solid lines) as a function of the relative Co layer thickness. For comparison, the lattice constants scaled with the relative Co layer thickness, as appropriate for intermixed structures, are shown as dashed lines (Vegard's law).

lines) and Co/Pt (dashed lines) superlattices. The strain components ϵ_i are plotted as a function of the relative Co layer thickness, $n_{\mathrm{Co}}/(n_{\mathrm{Co}}+n_x)$ with $n_i=d_i/a_i$ the number of atomic layers per layer with index i=Co,x and x=Pt,Pd. With decreasing Co layer thickness, the strains within the Co layers increase and the Co lattice spacing perpendicular to the layers decreases. This effect is more pronounced for the Co/Pt system than for the Co/Pd system, since the elastic properties of the constituent materials are more different in the Co/Pt system. In Fig. 2 the calculated average perpendicular lattice constant $a_{\mathrm{avg}}=(\epsilon_{3,1}d_1+\epsilon_{3,2}d_2)/(n_1+n_2)$, is shown as solid lines, as can be measured using x-ray diffraction.

Our approach certainly is only valid if no other mechanisms exist which distribute the strains more equally between the two materials. Such mechanisms are interdiffusion and interface roughness. If such mechanisms are acting, the average perpendicular lattice constant would more or less linearly scale with the fraction f_1 of the Co layer thickness to the bilayer thickness (Vegard's law). For comparison we also calculated a_{avg} assuming the latter, shown in Fig. 2 as dashed lines. We would like to mention that for Co/Pt superlattices, x-ray data exist,[4,5] which corroborate our new approach, and which are not consistent with the assumption of interdiffusion.

The anisotropy contribution is obtained from the third term in Eq. (1) by a comparison to a general anisotropy energy expression of the form

$$E_{\mathrm{aniso}}=-K_s\alpha_3^2-K_p\alpha_1^2 , \tag{3}$$

with K_p (K_s) the strain-induced uniaxial in-plane (out-of-plane) anisotropy constant, and $\alpha_1^2+\alpha_2^2+\alpha_3^2=1$. The signs in Eq. (3) are chosen following the usual convention that a positive sign for each anisotropy constant denotes its easy axis. The symmetry axis for the in-plane anisotropy is the [001] axis. By performing the appropriate tensor rotations for b_{ij} and identifying the appropriate terms, we find

$$K_p=(b_{11}-b_{12})(-\epsilon_1+\epsilon_2/2+\epsilon_3/2)+b_{44}(\epsilon_2-\epsilon_3) , \tag{4a}$$

$$K_s=2b_{44}(\epsilon_2-\epsilon_3) . \tag{4b}$$

For clarity, we have dropped the index 1 in the strains referring to the Co layers. Of interest is the so-called effective perpendicular anisotropy, K_{eff}, which is a measure of the difference in free-anisotropy energy between the direction of magnetization lying in plane and out of plane. The sample is perpendicularly magnetized for K_{eff} larger than zero. For $K_p>0$ the in-plane anisotropy contributes to K_{eff} as well, since the layer normal lies in the hard plane for this anisotropy. Thus

$$K_{\mathrm{eff}}=K_s-2\pi M_s^2 \quad \text{for } K_p<0 , \tag{5a}$$

$$K_{\mathrm{eff}}=K_s-K_p-2\pi M_s^2 \quad \text{for } K_p>0 , \tag{5b}$$

with $4\pi M_s$ the saturation magnetization.

In order to quantitatively compare the calculations to experimental data, we need to know the magnetoelastic coefficients $(b_{11}-b_{12})$ and b_{44}. Because these or derived constants for pure fcc Co do not exist in the literature, we have used values obtained for Co-rich, fcc Pd-Co alloys.[6] The magnitudes of the tabulated constants, $\lambda_{100}=-2/3(b_{11}-b_{12})/(c_{11}-c_{12})=130\times10^{-6}$ and

$\lambda_{111}=-1/3\ b_{44}/c_{44}=-65\times10^{-6}$, are found to be independent of the Co concentration above 60% and hence should provide a reasonable estimate for pure fcc Co. We tested these values by calculating the magnetoelastic anisotropy constant for (111)-oriented layers, $K_s^{(111)}$ as well as for (0001)-oriented layers of hcp structure, $K_s^{(0001)}$:

$$K_s^{(111)}=-6\lambda_{111}(c_{11}-c_{12})(4/3\epsilon_1-\epsilon_3)\ \text{[fcc Co]} \tag{6a}$$

$$K_s^{(0001)}=(\lambda_A+\lambda_B)(c_{11}+c_{12}-2c_{44}^2/c_{33})\epsilon_1\ \text{[hcp Co]} \tag{6b}$$

with λ_A and λ_B the usually defined magnetostriction coefficients[7] of bulk hcp Co.[8] With the elastic constants of fcc cobalt,[9] $c_{11}=242$ GPa, $c_{12}=160$ GPa, and $c_{44}=128$ GPa, as well as those for hcp cobalt,[9] we find $K_s^{(111)}=\epsilon_1$ (9.48×10^8 erg/cm^3) using Eq. (6a) and $K_s^{(0001)}=\epsilon_1$ (6.75×10^8 erg/cm^3) using Eq. (6b). The obtained values are in reasonably good agreement with each other, in particular taking into account that λ_{100} and λ_{111} refer to the fcc structure and λ_A and λ_B refer to the hcp structure. Since the volume anisotropy constant is known to depend very sensitively on the structure, it is even rather surprising that the strain-induced anisotropy constants calculated with both methods agree to within 30%. However, we note that the values of $b_{11}-b_{12}$ and b_{44} still need to be determined more accurately, in order to apply our approach to a more quantitative comparison with experimental data.

Figure 3 shows the obtained values of K_p and K_s as a function of the relative Co layer thickness. The positive sign of K_s denotes that the layer normal is an easy axis for the out-of-plane anisotropy, and the positive sign of K_p denotes that the [001] axis is an easy axis. The finding of an easy [001] axis for Co is unique to ultrathin, coherently strained Co(110) layers and is caused by the anisotropic magnetoelastic interaction. The anisotropic lattice distortion revealed by $\epsilon_1\neq\epsilon_2$ also contributes to the effect (cf. Fig. 1).

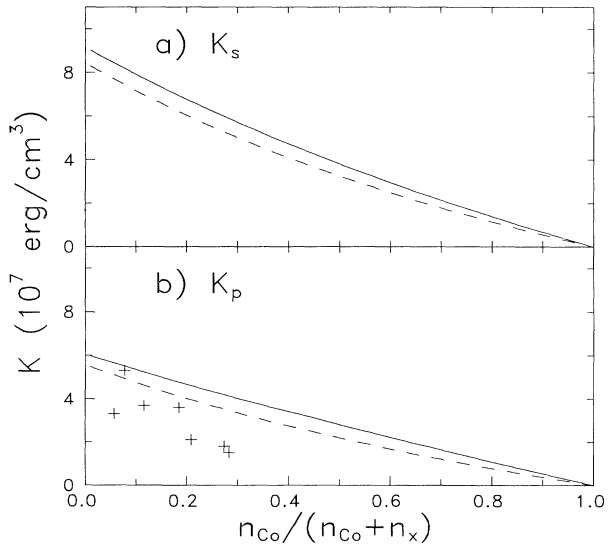

FIG. 3. Calculated in-plane (K_p) and out-of-plane (K_s) anisotropy constants as a function of the relative Co layer thickness for (110)-oriented Co/Pt (solid lines) and Co/Pd (dashed lines) superlattices. Experimental data for K_p taken from Refs. 2 and 3 are shown by crosses.

The obtained values for K_s, in particular for small Co layer thicknesses, are up to seven times larger than the shape anisotropy, $2\pi M_s^2$, and those for K_p up to 5.5 times. Due to the larger strain fields in the Co/Pt system as compared to the Co/Pd system, the anisotropy values are slightly larger in this system.

Experimental data are available for a few Co/Pt and Co/Pd samples. Lin *et al.*[10] find for a Co/Pt superlattice with $d_{Co}=3.7$ Å and $d_{Pt}=16.8$ Å an anisotropy constant of $K_p=3.6\times10^7$ erg/cm^3 which compares rather well to our calculated value of 4.8×10^7 erg/cm^3. Their value of $K_{eff}=0.2\times10^7$ erg/cm^3 is smaller than our value of 1.1×10^7 erg/cm^3 assuming for the shape anisotropy contribution the bulk value of the Co magnetization of $4\pi Ms=17.9$ kG.[11] Farrow *et al.* measured for a number of (110)-oriented Co/Pt superlattices the change in free energy, rotating the direction of magnetization from the layer normal into the easy and hard in-plane directions, respectively.[2,3] An analysis of their data yields with $d_{Pt}\approx25$ Å and d_{Co} in the range of 1.1 to 9.9 Å, values for K_p in the range of 1.5×10^7 to 5.3×10^7 erg/cm^3. These results are in good agreement with our calculations, which are in the range of 4.2×10^7 to 5.6×10^7 erg/cm^3. The experimental and calculated data are listed in Table I. For comparison, the experimental data of K_p are also shown as crosses in Fig. 3. For K_{eff}, a comparison between theory and experiment cannot unambiguously be made since an additional contribution from interface anisotropies cannot be separated in the analysis.

For Co/Pd (110)-oriented superlattices, a value of $K_p=0.35\times10^7$ erg/cm^3 has been found by Brillouin light scattering,[12] which compares to a calculated value of 1.89×10^7 erg/cm^3.

A possible strong contribution from magnetocrystalline anisotropies to K_p can be ruled out. This contribution would favor the [111] axis as the easy axis. A detailed discussion of magnetoelastic anisotropy contributions to oriented superlattice structures is presented elsewhere.[13]

Although our model predicts the correct sign and magnitude of the in-plane and out-of-plane anisotropies, a more quantitative comparison is still lacking. This is partly caused by the lack of reliable magnetoelastic constants for fcc Co. Also needed is a better understanding of the influence of interface interdiffusion and roughness on the anisotropy constants. Formations of dislocations and grain boundaries release the interface strains which in turn would decrease the related anisotropy contributions. Thus we regard the calculated anisotropy constants presented in this analysis as upper limits for the particular system.

Any interface anisotropy contributions were neglected in the comparison. We, however, note that the dependence of K_p and K_s on d_{Co} would suggest a large inter-

TABLE I. Measured (exp.) and calculated (calc.) in-plane (K_p) and effective out-of-plane (K_{eff}) anisotropy constants of (110)-oriented Co/Pt superlattices. The experimental data were obtained by analyzing data on K_{eff} of Refs. 2 and 3.

d_{Co} (Å)	d_{Pt} (Å)	K_p(exp.) (10^7 erg/cm^3)	K_p(calc.) (10^7 erg/cm^3)	K_{eff}(exp.) (10^7 erg/cm^3)	K_{eff}(calc.) (10^7 erg/cm^3)
1.1	20.1	3.3	5.6	0.36	1.6
1.6	21.1	5.3	5.5	1.7	1.5
2.6	22	3.7	5.2	1.4	1.3
3.7	18	3.6	4.9	0.1	1.1
6.2	26	2.1	4.7	−1.1	0.9
8.2	24.1	1.8	4.3	−0.8	0.5
9.9	27.7	1.5	4.2	−0.9	0.5

face anisotropy contribution in a commonly used separation of measured anisotropy values into a thickness-independent term and a term proportional to $1/d_{\text{Co}}$. The available data at present do not allow for the separation between thickness-dependent magnetoelastic contributions and pure interface contributions.

In summary, we have calculated the elastic strain fields as well as the in-plane and out-of-plane anisotropy constants for (110)-oriented Co/Pt and Co/Pd superlattice structures. We find a very large in-plane anisotropy making the in-plane (001) axis the easy axis of the layers in agreement with experimental data. We would like to conclude by commenting on the influence of interdiffusion on the strain fields and therefore on the anisotropy values. For very small Co layer thicknesses comparable to the interdiffusion length, the strains are more or less equally distributed between both materials according to the relative Co layer thickness f_1. If the layers are thicker, our approach applies and modified strain fields need to be considered. The model is thus valid in a thickness regime larger than the interdiffusion thickness and smaller than the critical thickness needed for dislocation formation. Applications of the model of the calculation of the effective elastic constants of the superlattice stack are in progress.

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