Orientational Dependence of the Interface Magnetic Anisotropy in Epitaxial Ni/Co/Ni Sandwiches

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Magneto-optical Kerr effect measurements have been used to investigate the orientational dependence of the magnetic anisotropy in ultrathin Ni/Co-wedge/Ni sandwiches deposited by molecular beam epitaxy on single-crystal Cu substrates. The results show a marked dependence of both the volume *and* interface anisotropy terms on the growth direction of the samples: [100], [110], and [111]. In addition, the (111) interface term was found to be significantly larger than existing literature values for Co/Ni multilayers. This effect is discussed in terms of the microstructure of the studied films.

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The investigation of perpendicular magnetic anisotropy (PMA), which by now has been well established in a variety of magnetic/nonmagnetic multilayers [1], has received a new impulse with the prediction and subsequent experimental confirmation of PMA in Co/Ni multilayers, where both constituents are ferromagnetic [2]. In understanding this PMA, two factors have been shown to play a crucial role [3], viz., the symmetry breaking by the interfaces, and the number of valence electrons which can place the Fermi energy in proximity to bands whose spin-orbit interactions favor perpendicular magnetization. Both of these factors are expected to be influenced by orientation. First, the extent of symmetry reduction, as determined by the number of broken bands at interfaces, is orientation sensitive, as shown in Néel's theory of surface anisotropy [4]. Second, since the lifting of twofold degeneracies by the spin-orbit coupling has been shown to play an important role in determining the PMA of [111]-oriented multilayers [3], a lower-symmetry interface—such as (110), which has no degeneracies—can be expected to exhibit a very different interface anisotropy. In this light, it is perhaps surprising that an orientation-*independent* interface contribution to the PMA has recently been reported for Co/Pd multilayers [5].

To investigate the orientational behavior of the anisotropy, we have prepared samples in the form of epitaxial Ni/Co/Ni sandwiches, deposited on Cu single-crystal substrates of crystallographic orientation [100], [110], and [111]. In this Letter, we verify an orientation-dependent PMA in these samples, which manifests itself in both the volume *and* interface contributions to the total anisotropy. Furthermore, we demonstrate that the interface anisotropy is larger for these epitaxially grown samples than for vapor-deposited Co/Ni multilayers [2,6]. Such a trend has recently been observed in (111) Pd/Co/Pd and Pt/Co/Pt epitaxially grown sandwiches [7], where the (respective) interface anisotropies were found to exceed all currently reported literature values for Co/Pd and Co/Pt multilayers (see [5,8], for example). This behavior will be discussed in terms of the microstructure of the various multilayered samples.

The complete thickness dependence of the anisotropy was determined from measurements on only three samples—one for each of the three crystallographic orientations—in which the Co layer was grown in the form of a wedge of uniformly varying thickness. The layers were deposited at room temperature in a multichamber molecular beam epitaxy (MBE) system (VG Semicon V80M), and the final sample composition was as follows: Cu(100)/Ni (8 Å)/Co-wedge (0–10 Å, 1.2 Å/mm)/Ni (17 Å)/Cu (7 Å)/Au (20 Å); Cu(110)/Ni (8 Å)/Co-wedge (0–13 Å, 1.2 Å/mm)/Ni (16 Å)/Cu (7 Å)/Au (25 Å); Cu(111)/Ni (10 Å)/Co-wedge (0–10 Å, 0.9 Å/mm)/Ni (10 Å)/Cu (7 Å)/Au (20 Å). The Co wedge shape was formed by slowly withdrawing an eclipsing shutter located between the single crystals and the Co source. The Co and Ni thicknesses were determined using a quartz crystal monitor calibrated from comparison with chemically analyzed reference samples, and the thicknesses thus determined were subsequently confirmed after deposition using combined *in situ* Auger electron spectroscopy and scanning electron microscopy. More precise details concerning the substrate preparation and growth technique can be found in [9].

The perpendicular and parallel lattice spacings were determined by measuring the energies of the primary Bragg low energy electron diffraction (LEED) reflections along the [00] rod, and by analysis of LEED patterns at constant electron energy, respectively. Both the Co and Ni displayed fcc surface nets which were nearly identical to that of the Cu substrate. Such coherent growth results in an in-plane expansion of the respective lattices by 2.0% (Co) and 2.5% (Ni), and introduces strains into the growing films. The in-plane lattice expansions were found to be accompanied by perpendicular lattice contractions, resulting in perpendicular Co-Co and Ni-Ni spacings of 1.71, 1.21, and 2.01 Å for the respective [100], [110], and [111] orientations—slightly less than the corresponding Cu-Cu lattice spacings of 1.81, 1.27, and 2.09 Å. Maintaining such a distortion requires a considerable amount of elastic energy. Above a certain critical film thickness, this energy becomes too great, and relaxation towards the bulk lattice parameters will occur via introduction of misfit dislocations. Co appears to be

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able to sustain such distortion up to considerable film thicknesses (>60 Å [9]); the larger lattice mismatch of Cu/Ni, however, induces relaxation at Ni thicknesses of ~ 25 Å, as determined from study of a separate Ni wedge grown on Cu(111). None of the three samples contained Ni layers of such thickness, and no clear indication of stress relaxation was observed in the LEED patterns, which remained sharp throughout the growth. The only exception was the (110) sample, where deposition of Co on the Ni underlayer produced a certain amount of streaking in the LEED patterns, particularly in the [100] directions, where the surface is most open.

The magnetic anisotropy was determined at various positions along the Co wedges (i.e., at different Co thicknesses) using a magneto-optical Kerr effect (MOKE) apparatus in which the probing laser beam had been focused to a spot of width ~ 0.1 mm. Anisotropies at thicknesses where the magnetization was *in plane* could be determined by forcing the sample towards saturation in a field applied along the film normal. On the other hand, *perpendicular* anisotropies were derived by forcing the magnetization away from the film normal using an external planar field, as described in detail in [7]. In what follows, we assume the phenomenological relationship

$$KD = K_V^{\mathrm{Co}} t_{\mathrm{Co}} + 2K_S^{*}, \tag{1}$$

where

$$2K_S^{*} = 2K_S^{\mathrm{Co/Ni}} + 2K_S^{\mathrm{Cu/Ni}} + K_V^{\mathrm{Ni}} t_{\mathrm{Ni}}. \tag{2}$$

Here, K is the measured total anisotropy per unit volume, D is the magnetic sandwich thickness (i.e., the sum of the respective Co and Ni total thicknesses, t_{Co} and t_{Ni}), K_S^{*} is the effective total interface anisotropy, K_V^{Co} and K_V^{Ni} are the respective partial volume anisotropies of Co and Ni, including shape anisotropy, and $K_S^{\mathrm{Cu/Ni}}$ and $K_S^{\mathrm{Co/Ni}}$ are the interface anisotropies for the Cu/Ni and Co/Ni interfaces, respectively. Positive K values correspond to perpendicular preferential orientations. Plotting KD against t_{Co}, as in Fig. 1, we can extract K_V^{Co} from the slope of the resulting straight line, and K_S^{*} from the vertical intercept at $t_{\mathrm{Co}}=0$. Subtracting the second and third terms of Eq. (2), we thus obtain the value of $K_S^{\mathrm{Co/Ni}}$; a single anisotropy measurement at a position before the start of the Co wedge (i.e., at $t_{\mathrm{Co}}=0$) is sufficient to determine these two terms.

From Fig. 1(c), it is seen that the [111]-oriented sample displays PMA for all Co thicknesses investigated. Application of the method discussed above yields $K_V^{\mathrm{Co}} = -0.8$ MJ/m^3, and $2K_S^{*} = 0.98$ mJ/m^2. Subtracting the volume contribution of the Ni and the Cu/Ni interface contribution (measured total value 0.14 mJ/m^2), we obtain $K_S^{\mathrm{Co/Ni}} = 0.42$ mJ/m^2, i.e., an interface contribution favoring PMA. The importance of the interface in enhancing the perpendicular anisotropy is emphasized in the region below $t_{\mathrm{Co}}=2$ Å. Here we have, on average, a Co thickness of less than one monolayer, so that the film will consist of isolated Co patches; between these patches, the Ni overlayer will grow directly onto the Ni underlayer, and no Co/Ni interface will form, so that the PMA is reduced. This explains the observed deviation from linear behavior below 2 Å [Fig. 1(c)].

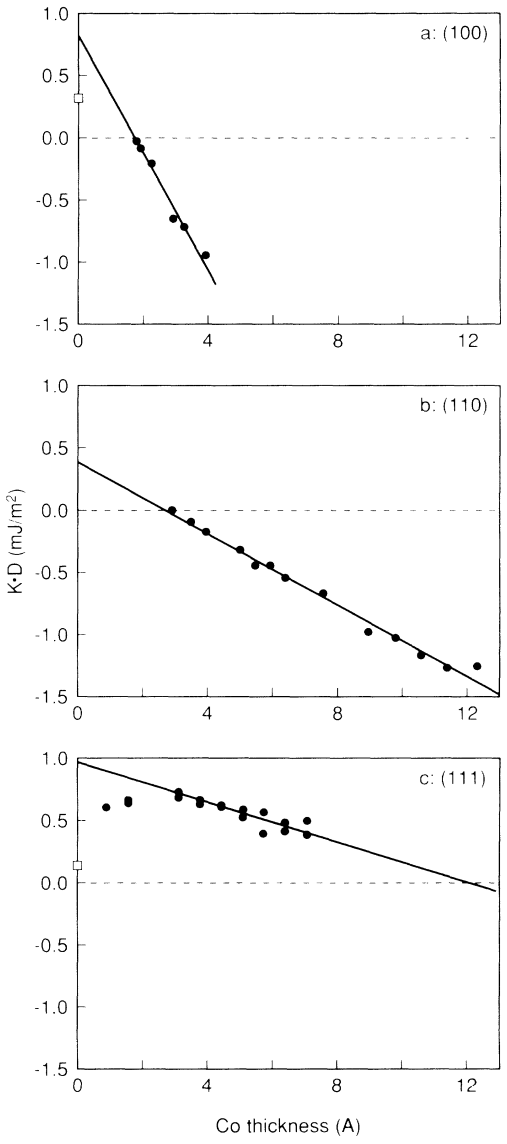

FIG. 1. The product of total anisotropy K and total magnetic layer thickness D, plotted as a function of Co thickness for the [100], [110], and [111] orientations [(a), (b), and (c), respectively]. Each single data point at $t_{\mathrm{Co}}=0$ (square symbol) yields the sum of the second and third terms of Eq. (2).

Similar analysis of Fig. 1(a) for the (100) sample yields $K_V^{\mathrm{Co}} = -4.5$ MJ/m^3 and $2K_S^{*} = 0.78$ mJ/m^2. The reference measurement before the start of the Co wedge produced a value of 0.32 mJ/m^2 for the sum of the second and third terms in Eq. (2), thus yielding $K_S^{\mathrm{Co/Ni}} = 0.23$ mJ/m^2. Note that only a narrow range of Co

thicknesses is plotted in the figure. This is due to the exceptionally high negative value of K_V^{Co}, which precludes perpendicular saturation of thick Co layers with the available perpendicular fields ($\sim$1 T), thus preventing anisotropy determination at Co thicknesses above 4 Å.

In the case of the (110) system, Fig. 1(b) reveals a slope $K_V^{Co} = -1.5$ MJ/m^3 and a vertical intercept $2K_S^* = 0.38$ mJ/m^2. Reference measurements before the start of the wedge were fruitless, since it was found that the anisotropy at zero Co thickness was always positive, but that its value depended on the orientation of the sample axes with respect to the applied planar field. This ambiguity did not arise in the measurements conducted above 3 Å Co, since these employed a perpendicular field configuration. We can thus conclude at most that $K_S^{Co/Ni} < 0.19$ mJ/m^2. Nevertheless, this is enough to show that the three samples demonstrate different volume *and* interface anisotropy contributions, as summarized in Table I.

The origin of the differing Co volume anisotropy contributions is most probably attributable to a stress-related magnetoelastic anisotropy (in combination with the constant shape anisotropy $K_{shape} = -1.27$ MJ/m^3). As discussed in [5,6,8,10], an additional anisotropy K_σ will arise as a result of the strained coherent growth of the Co and Ni. For the [100] and [111] orientations, this takes the form $K_\sigma = -\frac{3}{2} E\eta\lambda$, where E is Young's modulus for the strained metal, η the lattice misfit, and λ the orientation-dependent magnetostriction constant. As approximate values for fcc Co, we take $E \approx 2\times10^{11}$ N/m^2 [11], $\lambda_{100} \approx +130\times10^{-6}$ [10], and $\lambda_{111} \approx -50\times10^{-6}$ [12]. With $\eta = 2\%$, we thus obtain $K_\sigma^{100} \approx -0.8$ MJ/m^3 and $K_\sigma^{111} \approx +0.3$ MJ/m^3. Taking $K_V = K_{shape} + K_\sigma$, we therefore find $K_V^{100} \approx -2.07$ MJ/m^3 and $K_V^{111} \approx -0.97$ MJ/m^3. The K_V^{111} value is in reasonable quantitative agreement with the experimental value given above, considering the approximations in E and λ_{111} for fcc Co. The value of K_V^{100} is indeed more negative than K_{shape}, but falls short of its experimental counterpart by a factor of 2, suggesting perhaps that bulk values for E and, in particular, λ do not satisfactorily apply to films in the 1–2-monolayer range. In any case, the calculations verify the presence of significant magnetoelastic strain due to the coherent growth. A similar analysis for the (110) system is far more complicated [13], and is not attempted here.

TABLE I. Summary of Co/Ni anisotropy data pertaining to the three investigated orientations. $t_{Co}^{\perp}$ is the (extrapolated) Co thickness at which the transition from perpendicular to in-plane magnetization occurs; the other symbols are elucidated in the main text.

Orientation	t_{Ni} (Å)	$t_{Co}^{\perp}$ (Å)	K_V^{Co} (MJ/m^3)	$K_S^{Co/Ni}$ (mJ/m^2)
[100]	25	2	−4.5	+0.23
[110]	24	3	−1.5	$<$+0.19
[111]	20	12	−0.8	+0.42

Note that the measured value of $K_S^{Co/Ni}$ for the (111) sample (0.42 mJ/m^2) significantly exceeds the values cited in [6] for evaporated 120-Å (111)-textured Co/Ni multilayers grown on coated glass (0.31 mJ/m^2 maximum). This trend has also been observed recently in Co/Pd and Co/Pt, where the values of K_S measured for MBE-grown ultrathin (111) Pd/Co/Pd and Pt/Co/Pt sandwiches deposited on single-crystal substrates (0.92 and 1.15 mJ/m^2 [7], respectively) significantly exceed corresponding values for MBE-grown (111)-oriented 600-Å Co/Pd multilayers and 400-Å Co/Pt multilayers grown on coated GaAs (0.63 [5] and 0.82 mJ/m^2 [14], respectively). A possible explanation of this discrepancy draws on the microstructure of the deposited layers. It is well established that columnar structures frequently form via *geometrical shadowing* during growth of thick layers, resulting in rounded grains separated by relatively deep valleys [5,15,16]. For example, the (111) multilayers grown in [5] exhibited grains of diameter 500 Å, and valleys of width and depth 50 Å. This granularity reduces the effective area of the interfaces, so that the measured K_S is also diminished. In contrast, ultrathin MBE-grown sandwiches such as those studied here and in [7] are unlikely to exhibit grain formation, since the analysis in [16] suggests that columnar growth only occurs above a certain thickness (typically $>$ 100 Å), whose exact value depends on growth conditions. As a result, a majority of the surface area will contribute to the interface anisotropy, yielding an "intrinsic" value of K_S. Since the (100), (110), and (111) Co/Pd multilayers studied in [5] all displayed different ratios of grain area to valley area, and yet yielded identical *measured* interface anisotropies, one must conclude that the intrinsic values of $K_S^{Co/Pd}$ for the three orientations are indeed different, as we would expect.

The large measured values of K_S for the (111) Ni/Co/Ni and Pd/Co/Pd [7] sandwiches are in close agreement with the first-principles calculated values in [2,17]. For (111) Co/Ni, a hypothetical Co_1Ni_2 cell is predicted to display an anisotropy $KD = 0.65 \pm 0.1$ mJ/m^2 [2], whereas application of the K_S value measured here to the same Co_1Ni_2 cell yields $KD = 0.53$ mJ/m^2, after compensating for shape anisotropy. The agreement is equally good for (111) Co/Pd: The value $KD = 1.85 \pm 0.25$ mJ/m^2 predicted in [17] is closely matched by the value $KD = 1.6$ mJ/m^2 corresponding to the measured K_S term in [7], after compensation for shape anisotropy. The quality of this agreement reinforces the assertion that the interface terms measured in thin epitaxial sandwiches are close to the intrinsic values.

In conclusion, we have used MOKE to investigate the orientational dependence of the magnetic anisotropy in MBE-grown ultrathin Ni/Co/Ni sandwiches deposited on single-crystal Cu substrates. The results show a marked dependence of both the volume *and* interface anisotropy

terms on the growth direction of the samples, and have yielded the highest (111) interface anisotropy contribution observed for Co/Ni to date. Relating these data to similar results for Co/Pd and Co/Pt, it is concluded that granulation effects arising from columnar growth in thick multilayers are likely to artificially lower the interface anisotropies in these systems with respect to the intrinsic values measured for epitaxial ultrathin sandwiches.

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