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Layer-dependent magnetic-moment distribution in an epitaxial double spin valve structure: Si(001)/Cu/FeNi/Cu/Co/Cu/FeNi/Cu

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Polarized neutron reflectivity was used to determine, layer selectively, the magnetic moments, spin orientations, and thicknesses of an epitaxial double spin valve structure of the form Si(001)/Cu/FeNi/Cu/Co/Cu/FeNi/Cu. Absolute values of the average magnetic moment per atom were determined within an error of $\sim\pm5\%$. At saturation, the layer-averaged magnetic moment per Co atom was found to be $1.71\pm0.08\mu_B$, while the top (bottom) FeNi layer moment was $1.08\pm0.06\mu_B$ ($0.95\pm0.08\mu_B$). For an applied field strength H_a smaller than the Co layer coercive field of $\sim$100 Oe, the Co and FeNi layer magnetization vectors are found to lie in plane but canted with respect to each other in contrast to the fully antiparallel state predicted by simple energy minimization. The observed canting is attributed to the presence of magnetic domains in the Co layer. [S0163-1829(99)10233-9]

Recently many efforts have been devoted to the investigation of spin valve (SV) structures which exhibit a giant magnetoresistance (GMR) effect, used in applications such as magnetic sensors and read head devices.[1–5] The GMR effect in a SV structure is associated with a change in the relative orientations of the magnetization of adjacent ferromagnetic layers when an external field is applied. In the spin valve model the GMR effect is expressed as $R_{\mathrm{GMR}}(\theta)\propto\cos\theta$ where θ is the angle between the layer magnetic moments.[2,6] Thus it is important to know the layer-dependent spin configuration and magnetization in each layer, in order to maximize the GMR effect. The most commonly investigated structures are either single SV of the form ferromagnetic (FM)(1)/nonmagnetic (NM)/ferromagnetic (FM)(2) or a double SV of the form FM(1)/NM/FM(2)/NM/FM(3). In the latter case it is important to control the orientations of the magnetization vector in all the ferromagnetic layers in order to maximize the GMR. Due to inequivalent interface properties in each of the layers and domain formation for example, complex spin structures can result. Thus it is important to be able to determine the magnetization vectors in each layer as a function of applied field.

Polarized neutron reflectivity (PNR) provides a means of directly probing the layer dependent magnetization vector profile in a multilayer system. It yields the absolute value of the magnetic moment per atom and layer thickness in a magnetic thin film with high accuracy.[1–5] Our previous work[5] successfully demonstrated that PNR can be used to determine such parameters as the layer-dependent magnetic-moment distribution, interface roughness amplitude as well as layer thicknesses, in single trilayer spin valve structures.

In this paper, we present the results of PNR measurements on a double spin valve system with a composite structure, Si(001)/Cu/FeNi/Cu/Co/Cu/FeNi/Cu, for the first time. In such a structure the GMR is maximized when both the FeNi layer magnetic moments are antiparallel to the Co magnetic moment. Thus it is important to determine the conditions under which it is possible to control the relative alignment of the FeNi and Co layers. For example, external fields of an appropriate strength can be used to rotate the soft (FeNi) layers while not altering the hard (Co) layer. We chose to study an epitaxial structure since a magnetocrystalline anisotropy in the Co layer can be used to control the switching fields and *M-H* characteristic.[5] The absolute values of the magnetization and the spin orientations are determined independently of each other with high accuracy. Each magnetic layer is found to have a near bulk magnetic moment, but the magnetic moment orientations in each layer depend on the direction and strength of the applied magnetic field.

We have grown an epitaxial double spin valve structure, with nominal composition: Si(001)/Cu(700 Å)/FeNi(60 Å)/Cu(60 Å)/Co(40 Å)/Cu(60 Å)/FeNi(60 Å)/Cu(50 Å). The sample was grown on an HF-passivated Si(001) surface at ambient temperature by molecular-beam epitaxy (MBE) under ultrahigh vacuum conditions (UHV) with a base pressure of $\sim3\times10^{-10}$ mbars. Prior to the deposition of the magnetic materials an epitaxial Cu(001) layer was deposited on Si(001) as a seed layer by using a Knudsen cell with a typical evaporation rate of $\sim$5 Å/min. Since the lattice mismatch for the Cu[100]/Si[110] is $\sim$6% good epitaxy can be obtained onto the HF-etched Si(001) substrate.[7] The 700-Å Cu thickness was chosen to be large enough to improve the epitaxy, and to give oscillations in the reflectivity curves in the low wave-vector range (0.015–0.045 Å^{-1}).[7] FeNi and Co layers were deposited on to the Cu(001) surface using electron-beam evaporation and the typical evaporation rate was ~1 Å/min. The pressure increased to $\sim 6\times 10^{-10}$ mbars while FeNi and Co were being deposited and to $\sim 3\times 10^{-9}$ mbars during Cu deposition. The deposition rates were calibrated using a quartz microbalance, which has an accuracy of ~±10%. Epitaxial growth was confirmed using *in situ* reflection high-energy electron diffraction (RHEED). The images obtained showed that the FeNi, Cu, and Co layers grew epitaxially in the (100) orientation, where the fcc Co{110} corresponds to the Si{100} direction.[7] The PNR measurements were made at the CRISP time-of-flight reflectometer at the Rutherford Appleton Laboratory.

Prior to the PNR measurements the magnetization loops were measured using *ex situ* magneto-optic Kerr effect (MOKE). From the angle dependent MOKE measurements the Co easy axes are found to be aligned parallel to the ⟨110⟩ crystalline directions indicating the presence of an in-plane fourfold anisotropy, as reported previously.[5,8] A typical *M-H* loop for the applied field along the fcc Co[110] direction is shown in Fig. 1(a). The loop shows that the FeNi layers reverse abruptly at low fields <10 Oe whereas the Co layer is saturated at the field of ~100 Oe. The plateau in the loop corresponds to a near antiferromagnetic (AF) alignment of the FeNi and Co moments.

Two distinct sets of PNR measurements were made, in which the magnetization directions of the three magnetic layers can be varied controllably via applied field. In the first set both the applied field strength and direction are varied as follows:

(1.1) The sample was first saturated by applying a ‘‘positive’’ field of +6 kOe along the fcc Co[110] easy axis. This field strength is strong enough to saturate all magnetic layers, and therefore, all magnetic moments are fixed in one direction denoted the FM state.

(1.2) A ‘‘negative’’ field of −6 kOe was first applied along the fcc Co [−1-10] direction and then reversed to the ‘‘positive’’ direction up to +50 Oe. For a field strength of 50 Oe, which is less than the Co switching field but greater than the FeNi coercivity, the Co vector is expected to be pinned in the ‘‘negative’’ direction due to the Co magnetic anisotropy ($2K_1/M > 200$ Oe), whereas the FeNi magnetic vectors follow the field and rotate by 180° to the ‘‘positive’’ direction. Hence an antiparallel orientation of the Co and FeNi layer magnetic moments is expected denoted the AF state.

First we fit the PNR data for the set (1.1) in which the magnitudes and orientations of the magnetic moments are well defined due to the saturation of the magnetizations in each layer. Figure (1b) shows spin dependent reflectivity (*R*) spectra, where *R* was determined as a function of perpendicular wave vector *q* both for spin-up and spin-down neutrons. One can see several well-pronounced reflectivity oscillations as a function of wave vector. It is more instructive to look at spin asymmetry curves as opposed to the reflectivity curves, which look very similar. Figure 1(c) shows the corresponding spin asymmetry curve, which is defined by $S=(R^+-R^-)/(R^++R^-)$. In fitting the data the nominal sample composition was assumed, in which a Gaussian interface roughness of variable amplitude is introduced.[2] Furthermore, in the fitting procedure the bulk scattering lengths and densities are assumed, whereas all the layer thicknesses, magnetic moments, and vertical interface roughnesses were freely varied. The fitting of the FM data reveal that all of the layers are aligned parallel within an error of ~1°, as expected in the case of the saturated ferromagnetic alignment. The absolute value of the magnetization vector of the Co layer was obtained to be $1.71\pm 0.08\mu_B$ ($1.70\mu_B$ for bulk fcc Co). The top FeNi layer moment was $1.08\pm 0.06\mu_B$ ($0.96\mu_B$ for bulk FeNi) whereas the bottom FeNi layer moment was $0.95\pm 0.08\mu_B$ (Table I).

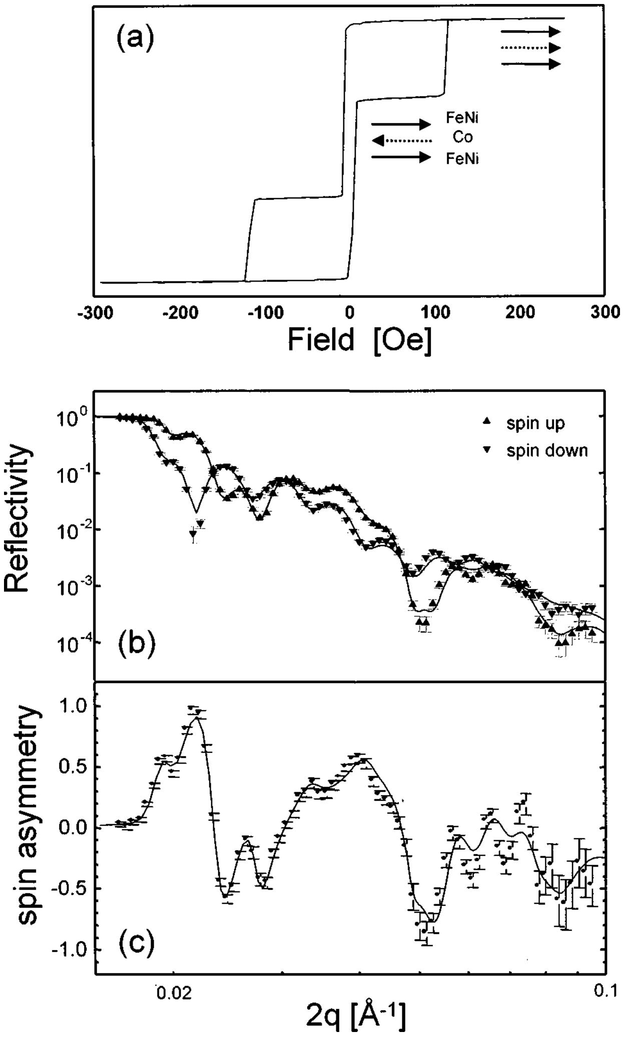

FIG. 1. (a) Hysteresis loop measured by MOKE with the applied field along the Co easy magnetic axis, [110] direction. The arrows represent the magnetization vectors in each magnetic layers in different PNR measurement configurations. The dotted (solid) arrows are for Co (FeNi). (b) Spin dependent reflectivity (*R*) determined as a function of perpendicular wave vector *q* both for spin-up and spin-down neutrons. (c) Spin asymmetry curve, which is defined by $(S=R^+-R^-)/(R^++R^-)$, where R^+ (R^-) is the reflectivity of the spin-up (spin-down) neutrons. For the PNR measurement the sample was saturated by applying a field of 6 kOe along the fcc Co [110] easy axis.

Figure 2 shows both the reflectivity and spin asymmetry curves for the corresponding AF data, i.e., the set (1.2). The moments obtained for this data set agree well with that in the FM state within error. However, the layer-averaged Co mag-

TABLE I. Summary of results fitted by PNR measurements. The date are obtained from the FM (1.1) and AF (1.2) states. Moments are given in Bohr magnetons (μ_B) while the values in the parentheses are the corresponding bulk values at room temperature.

Nominal composition	Moment (bulk) [μ_B], FM	Moment (bulk) [μ_B], AF	Angle AF	Thickness [Å]
Cu (50 Å)				48.6±2.5
FeNr (60 Å)	1.08±0.06 (0.96)	1.18±0.08 (0.96)	0°	48.1±3.0
Cu (60 Å)				62.2±2.0
Co (40 Å)	1.71±0.08 (1.70)	1.60±0.09 (1.70)	35°±2.7°	33.7±2.5
Cu (60 Å)				48.7±2.5
FeNi (60 Å)	0.95±0.08 (0.96)	0.89±0.09 (0.96)	0°	56.2±3.0
Cu (700 Å)				599.7±10

netic moment ($1.6\pm0.3\mu_B$) is found not to remain in the negative [−1-10] direction, but is rotated by 35°+2.7° with respect to the [−1-10] direction. Thus we obtain a spin configuration where the FeNi layers have changed the magnetization direction to become parallel to the applied field, whereas the Co magnetization is rotated from the fully antiparallel direction and makes an angle of ~145° with respect to the FeNi magnetization (Fig. 2). We estimate a relative Co magnetization of 80% corresponding to the spin configuration of Fig. 2, where a ''net'' Co magnetic moment of $1.3\mu_B$ is calculated from the best-fit Co magnetic moment of $1.6\mu_B$ projected onto the [−1-10] direction using a canting angle of 35°. This net moment is larger than the relative moment (~40% of M_S) obtained by Kerr effect at the same field shown in Fig. 1(a). However the PNR net moment is larger than the corresponding net Kerr signal shown in Fig. 1(a) since the magneto-optical constant of Co is significantly larger than that of FeNi.

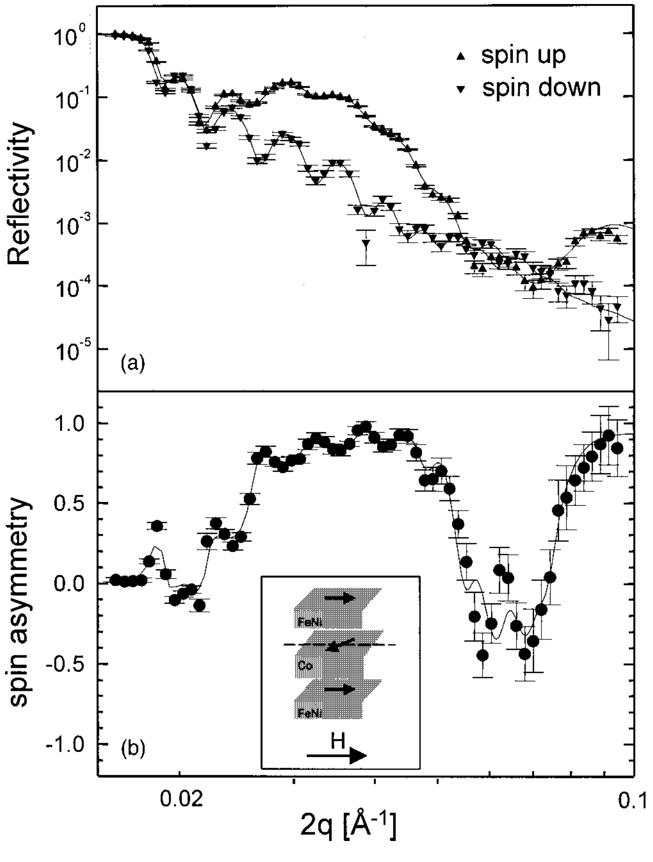

FIG. 2. (a) Spin dependent reflectivity (R) and (b) spin asymmetry as a function of perpendicular wave vector q both for spin-up and spin-down neutrons. A field of 6 kOe was first applied along the fcc Co [-1-10] direction and then reversed to the [110] direction up to 50 Oe. In the inset of (b) we show a schematic of the magnetization vectors in the antiferromagnetic state as described in the text.

To verify the observed canted spin configuration, we have simulated the spin dependent reflectivity assuming that the Co and FeNi moments are aligned fully antiparallel (that is, not canted) using the net Co magnetic moment of $1.3\mu_B$, calculated from the best-fit Co magnetic moment of $1.6\mu_B$ projected by 35° to the [-1-10] direction (Fig. 3). The result reveals that the reflectivity does not fit the data, indicating that the PNR measurement is highly sensitive to spin

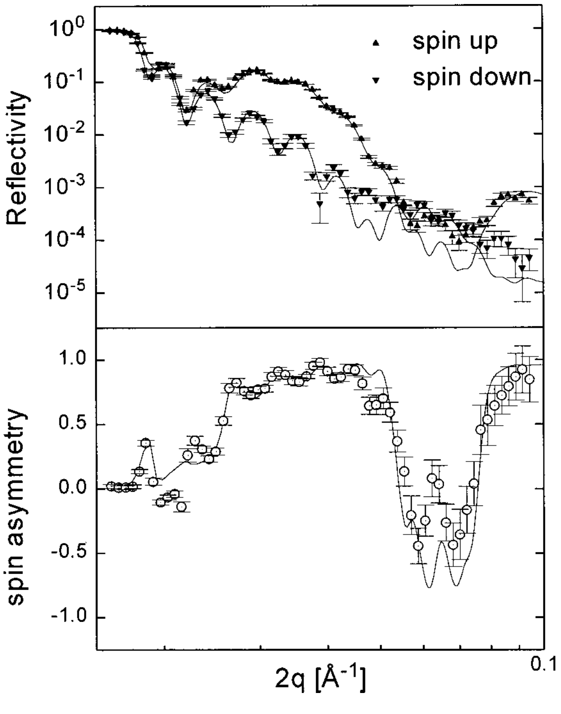

FIG. 3. Simulated spin dependent reflectivity (R) and spin asymmetry curve as a function of perpendicular wave vector q both for spin-up and spin-down neutrons. In the simulation the Co and FeNi moments are assumed to be aligned antiparallel with the net Co magnetic moment of $1.3\mu_B$. The data and field preparation condition are the same as that of Fig. 2.

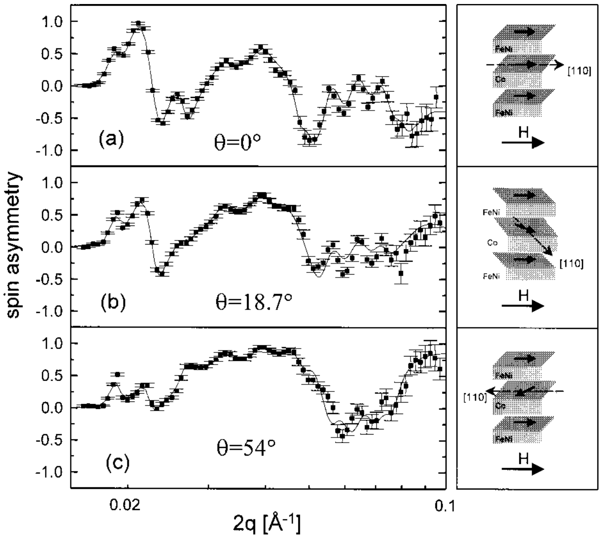

FIG. 4. Spin asymmetry curves for the second set of the PNR measurements. First the sample was magnetized by a field of 6 kOe along the [110] direction and then reduced to 50 Oe which is strong enough to switch the FeNi layer but not Co. The sample was then physically rotated in the applied field by ~90° (b) and ~180° (c). In the right-hand side of the spin asymmetry curves, the schematics of the measurement geometry are shown.

configuration, and we rule out the antiparallel alignment of the Co magnetization with respect to the FeNi.

The observed in-plane canting state can be interpreted as follows: the magnetic anisotropy of the Co layer is not strong enough to constrain the Co moments opposite to the field direction. However the field strength of +50 Oe is also not high enough to overcome the energy barrier due to the fourfold symmetry of the fcc Co layer for Co magnetization reversal. Therefore it cants towards the field direction, but an exact antiferromagnetic (AF) alignment of the FeNi and Co moments can not be realized at this field. The observed canting angle is compared with an analysis of the local minimum of the anisotropy energy $E(\theta)$, as will be discussed later.

In the second set of PNR measurements we probe the vector orientation of the magnetic moments. The magnetization directions in each magnetic layer are varied by rotating the sample physically whereas the applied field direction is fixed. The soft FeNi layers undergo coherent spin rotation following the external field direction. The measurement sequence is as follows:

(2.1) The sample was first saturated by applying a field of +6 kOe along the [110] direction then reduced to +50 Oe. It is denoted as the ''0°'' state.

(2.2) The sample is then physically rotated by ~90°. The FeNi layer magnetization is, as a consequence, rotated with respect to the Co magnetization. This is denoted as the ''90°'' state.

(2.3) Subsequently, the sample is again physically rotated by a further 90°. This is denoted as the ''180°'' state.

The spin asymmetry data in Fig. 4 shows dramatic variations according to the rotation of the sample with respect to the applied field. Fitting the magnetic moments and spin orientations both of the Co and NiFe layers, the measurement (2.1) [Fig. (4a)] reveals perfect parallel alignment of the layers as shown in the case of the FM state. The measurement set (2.2) [Fig. (4b)] reveals that the layer-averaged Co magnetic moment makes an angle of $\theta = 18.7° \pm 3.4°$ with respect to the original [110] direction, whereas the FeNi vectors follow the external field direction. Finally the measurement set (2.3) [Fig. (4c)] shows that the Co moment makes an angle of $\theta = 54° \pm 3°$ to the [110] direction, i.e., the FeNi and Co moments make an angle of ~126° instead of an antiparallel alignment. From this result it is concluded that the FeNi layers have followed the applied field of ~50 Oe to become aligned parallel to the field, whereas the magnetic anisotropy of the Co layer is again not strong enough to constrain the Co magnetization against this field strength. Therefore the Co layer becomes canted with respect to the applied field direction upon rotating the sample.

The canting of the Co magnetization with respect to the applied field direction described above is compared with the results of a local minimization of the anisotropy energy. Under the assumption of a single domain and fourfold symmetry for the fcc Co(001) layer, the anisotropy energy of the Co magnetization can be expressed as:[9]

$$E = 1/4\, K_1 \sin^2 2\phi - MH \cos(\phi - \alpha), \tag{1}$$

where K_1 describes the cubic anisotropy constant in the Co layer which is magnetocrystalline in origin. The value of K_1 is assumed to be -2.0×10^{-6} erg/cm^3.[10] The second term is the Zeeman term due to the applied magnetic field, respectively. ϕ is the angle between the magnetization and the [100] direction, α is the angle of the applied field direction with respect to the [100] direction. Using the above equation, the energy $E(\phi)$ at the applied field strength of 50 Oe is minimized to determine the Co magnetization orientations. This rough analytical method reveals a local minimum of E at ~6° with respect to the [110] direction in the (2.2) configuration, whereas the canting angle determined by PNR was ~19°. With the field along the [-1-10] direction favoring the ''180°'' state in the (2.3) configuration, energy minima at 94° and 181° were calculated, whereas the Co magnetization orientation is experimentally determined to be 54° at an applied field of 50 Oe. From this discrepancy we conclude that the magnetization vector cannot be predicted from simple energy minimization suggesting that the energetics of magnetic domains need to be considered.

Evidence for domain formation is given by the results of angle dependent magnetoresistance (MR) measurements, where the sample resistance has been measured as a function of the angle θ between the initially magnetized sample direction and the applied magnetic field (Fig. 5). In these measurements the sample is first saturated by applying a magnet field of 6 kOe along the fcc Co [110] direction and then the magnet field strength is reduced to 50 Oe, as in the PNR measurement set (2.1). By rotating the sample at this reduced field strength, the FeNi layer magnetization should follow the applied field, whereas the Co magnetization is naively expected to be held in the [110] direction due to the magnetocrystalline anisotropy. Hence an antiparallel orientation of the Co and FeNi layer magnetic moments is expected from energy minimization [Eq. (1)] to occur at ~180° giving rise to a single maximum in the resistance as denoted by the dotted curve in Fig. 5. The dotted curve corresponds to the function $\sin^2(\theta/2)$ for illustration only. Detailed fitting of

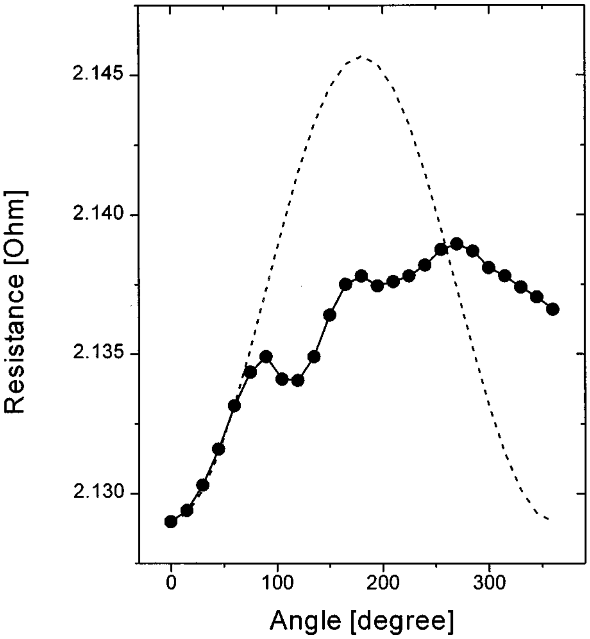

FIG. 5. Angular dependence of the magnetoresistance measured in the spin valve structure. The resistance has been measured as a function of the angle θ between the initially magnetized fcc Co [110] and the applied magnetic field direction. The dotted curve corresponds to the function $\sin^2(\theta/2)$ for illustration only.

the MR curve requires that both the anisotropic magnetoresistance (AMR) and GMR contributions are included and is beyond the scope of this paper. The MR data shows that the resistance increases when the magnetization of the FeNi films rotate from the parallel alignment with the Co film, but the resistance strongly depends on the angle between the applied field and the [110] direction. Most importantly the maximum resistance is found at 270°, indicating that the Co magnetic moment is held at some angle with respect to the applied field. Furthermore, it reveals that there is a periodically varying contribution to the resistance which results from the cubic anisotropy in the fcc Co(001) layer. Since this behavior is not predicted by the energy minimization calculation, domain formation in the Co layers may be important. While it is well known that domain formation sensitively influences the magnetoresistance, only a small fraction of the Co layer is in a multidomain state as confirmed by the fact that the layer-averaged Co magnetic moment in the AF state ($1.6\mu_B$) is only slightly lower than that of FM state ($1.7\mu_B$) and that the resistance value measured at 360° is slightly (~0.4%) larger than that at 0°. At 360° the averaged Co and FeNi layer magnetic moments do not align parallel due to the domains present in the Co layer, and the Co magnetization is therefore canted with respect to the initial field direction, as confirmed by the PNR data.

In conclusion, layer selective vector magnetometry using PNR has been successfully used to determine the magnetic moments and spin orientations for each of the ultrathin FeNi and Co layers in an epitaxial Si(001)/Cu/FeNi/Cu/Co/Cu/FeNi/Cu double spin valve structure. The absolute values of the magnetic moments in each layer were obtained within an error of $\sim\pm5\%$, where both of the FeNi layers and Co layer agree well with the bulk moments. The spin orientations of the ferromagnetic layers were also determined with layer specific selectivity. The spin orientation of the Co layer is found to be in plane canted with respect to the FeNi layers after reversing or rotating the external field with a moderate field strength of 50 Oe. Hence an exact antiferromagnetic (AF) alignment of the FeNi and Co moments predicted by simple energy minimization could not be realized. The observed canting (non-AF) alignment of adjacent magnetic layers is not ideal, since for a spin valve structure a well-defined AF configuration which maximizes the GMR is desirable for device applications. These results emphasize the possible role of domain formation in controlling the low-field magnetic configuration of spin valve structures.

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