Interface selective vector magnetometry of FeNi/Cu/Co trilayer spin-valve structures

J. A. C. Bland, C. Daboo, M. Patel, and T. Fujimoto
Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, United Kingdom

J. Penfold
Rutherford Laboratory, Chilton, Oxon., OX11 OQX, United Kingdom

(Received 25 November 1997)

Using polarized neutron reflectometry (PNR) together with superconducting quantum interference device magnetometry, the interface and interior magnetic moments have been determined for each of the ultrathin FeNi and Co layers within an epitaxial FeNi/Cu/Co trilayer structure, so demonstrating interface selectivity in layers of the same nominal chemical composition. The reduced moment found for the Co/Cu and FeNi/Cu interface regions are consistent with a model of enhanced electron spin-flip scattering at rough interfaces proposed to explain the temperature dependence of the giant magnetoresistance amplitude in FeNi/Cu/Co spin-valve structures. We further show that the layer-dependent vector moments can be determined by PNR with high precision. [S0163-1829(98)00117-9]

The spin configuration and magnetization at an interface between two layers within a magnetic multilayer structure determine such key properties as the magnetoresistance behavior, interlayer exchange coupling, and interface anisotropy.[1] Testing the interface spin structure of overlayers and multilayers predicted by theoretical models, for example, Ref. 2, requires experimental techniques capable of selectively probing buried single interfaces. Interest is now rapidly growing in the application of polarized radiation techniques such as x-ray magnetic circular dichroism (XMCD),[3] or second harmonic generation[4] to the study of interface magnetic structure. For example, the element specific selectivity of XMCD has been successfully used to determine the spin structures in Mn layers within Fe/Mn/Fe trilayers.[5] On the other hand, polarized neutron reflection (PNR) and diffraction techniques[6–10] can be used to probe the spin orientation within a magnetic multilayer structure with layer specific selectivity. Neutron scattering methods are the method of choice when a determination of the absolute value of the magnetization vector is required. In the case of multilayer structures, the interface structure can be obtained on a nm scale or better and PNR studies of magnetic ordering and interfacial structure in Gd/Fe multilayers,[11] Fe/Cr,[12] and Fe/Si (Ref. 13) multilayers have been recently reported. For simple overlayer structures, e.g., Gd/Fe (Ref. 14), the resolution with which the interface structure can be determined is reduced due to the limited wave vector range over which reflectivity data can be obtained. However, in the case of magnetic multilayers, in order to analyze the PNR data, it is usually necessary to assume a superlattice structure (i.e., *equivalent* repeat units within the structure). The presence of unknown domain structures further complicates the analysis of field dependent data in practice.

In the present work we have chosen to study FeNi/Cu/Co single trilayer spin valve structures using PNR in order to probe the interface spin configuration in a system without a periodic structure. We have selected a simple trilayer since the soft FeNi layer undergoes coherent spin rotation in an external field of appropriate strength[15] and so permits a test of the vector magnetometry capability of PNR. The total sample moment deduced from PNR is tested against superconducting quantum interference device (SQUID) magnetometry measurements. A further motivation in studying this system is provided by the results of recent studies of the temperature dependence of the giant magnetoresistance in FeNi/Cu/Co spin valve structures which suggest that the spin-flip electron scattering (which varies quadratically with temperature T at low temperature) is controlled by spin-wave excitations at the ferromagnetic/nonferromagnetic interfaces with a strength dependent on the interface structure.[16]

Structures of the form Si(100)/Cu(500 Å)/FeNi/Cu(60 Å)/Co/Cu(60 Å) were grown on HF-passivated p-Si(100) substrates in UHV at ambient temperature, with the pressure during growth maintained below 5×10^{-8} mbar. The Co and FeNi layer thicknesses estimated from quartz microbalance measurements were typically in the range 20–40 Å. Prior to deposition of the single trilayer, the substrate was heated to 200 °C for 30 min after which a thick Cu buffer layer ($\sim$500 Å) was deposited at room temperature. The Cu buffer layer is deposited to improve epitaxy[17] and is of a thickness chosen to give rise to oscillations in the neutron reflectivity at low wave vectors. *In situ* reflection high-energy electron diffraction (RHEED) images obtained during growth confirm the epitaxial growth of the FeNi, Cu, and Co overlayers in the (100) orientation.[17] Equivalent single trilayer polycrystalline structures grown directly onto Si substrates exhibit uniaxial anisotropies and giant magnetoresistance (GMR).[16] During growth the sample was rotated to suppress uniaxial anisotropies and the resulting epitaxial FeNi and Co layers both exhibit a fourfold anisotropy.[18] SQUID measurements reveal a significant reduction in the magnetization with increasing temperature in the range 1.5–300 K and thus the temperature range of the PNR measurements was extended to 1.5 K. The PNR measurements were made using the CRISP time of flight reflectometer at the ISIS facility at the Rutherford Appleton Laboratory. The spin-dependent specular reflectivity $R^{\pm}$ was determined as a function of perpendicular wave vector q for spin parallel (+) and

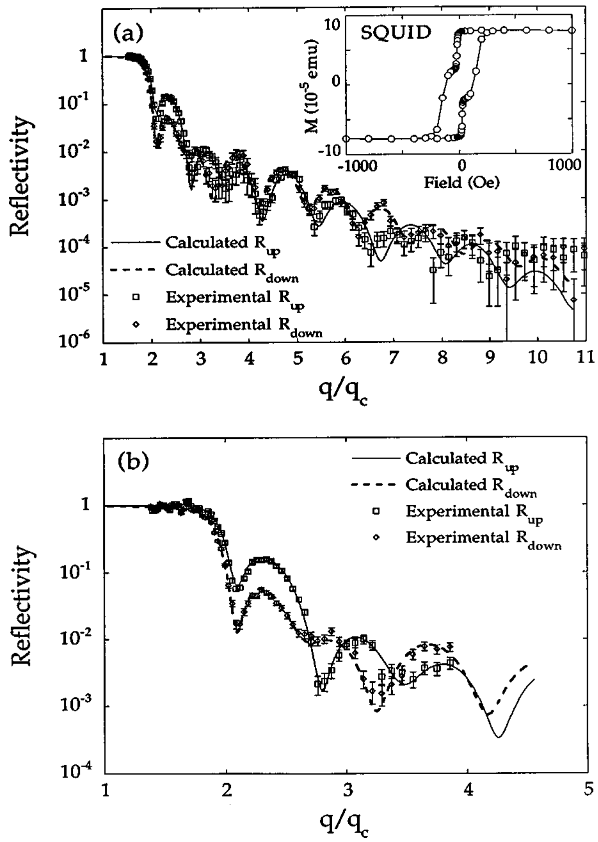

FIG. 1. (a) The spin-dependent reflectivity spectra plotted as a function of wave vector q for the epitaxial trilayer structure held at 100 K with the layer magnetizations aligned parallel by an applied field of 3 kG. The lines correspond to fits using the interface model (see text). The inset shows the magnetic hysteresis loop for the Si(001)/Cu/FeNi/Cu/Co/Cu epitaxial spin-valve structure obtained by SQUID magnetometry. (b) The spin-dependent reflectivity spectra plotted as a function of wave vector q obtained at low wave vectors for an epitaxial trilayer held at 1.5 K with the layer magnetizations aligned parallel by an applied field of 3 kG. The lines correspond to fits using the interface model.

antiparallel (−) to the applied field parallel to the [100] direction (easy axis for the FeNi layer) as a function of temperature with the sample magnetically saturated in plane and at room temperature as a function of field orientation. Immediately following the PNR measurements, magnetic hysteresis loops as a function of temperature were obtained from SQUID magnetometry with the field applied along the easy axis direction. The room temperature loops show saturation fields for the Co layers of ~200 Oe and reveal abrupt reversal of the FeNi layers at low fields [inset Fig. 1(a)]. The plateaux which occur following the reversal of the FeNi layer correspond to a near antiferromagnetic (AF) alignment of the FeNi and Co moments and thus can be used to estimate the magnetic moment of the FeNi and Co layers. The magnetic anisotropy of the Co layers is strong enough to constrain the Co moments at low fields.

Figure 1(a) shows spin-dependent reflectivity spectra in the low wave vector range obtained for an epitaxial trilayer held at 100 K with the layer magnetization aligned parallel by an applied field of 3 kG. Seven well pronounced reflectivity oscillations as a function of wave vector are seen within the wave vector range accessed. In fitting the data, two models of the structure were used: one in which the nominal sample structure is assumed and a second model in which additional intermixed FeNi-Cu and Co-Cu layers of variable composition and thickness are introduced at the interfaces. In both models, a Gaussian interface roughness of adjustable amplitude is introduced[7] and the layer thicknesses are determined from the reflectivity data available throughout the temperature range studied whereas the magnetic moments are fitted separately for each temperature. In the nominal structure model, the bulk scattering lengths and densities are assumed for each nonmagnetic layer, while all layer thicknesses, the scattering densities for the magnetic layers, and the magnetic moments were freely varied. The result, while reproducing well the features of the reflectivity data in the low wave vector range[19] is not consistent with the total moment measured by SQUID magnetometry and therefore can be excluded (see Table I). In the interface model, the interior regions in the ferromagnetic layers are assumed to have the full bulk moment of FeNi and Co and bulk scattering densities. In the interface regions the densities, moments, and thicknesses are adjusted (Table I). The resulting best fit using the interface model is shown as solid (spin-up) and dashed (spin-down) lines in Fig. 1(a) and the corresponding parameters are given in Table II. Layer thicknesses are determined in this way to a precision of 2–3 Å. It can be seen that the resulting fit closely reproduces all features of the data very well.

Figure 1(b) shows spin-dependent reflectivity spectra in the low wave vector range obtained for an epitaxial trilayer held at 1.5 K with an applied field of 3 kG. Three well pronounced reflectivity oscillations as a function of wave vector are seen within the wave vector range accessed. The best fit (solid lines) again assumes the presence of interfacial layers as given in Table II and the resulting value for the total moment of the sample is found to be in good agreement within errors with the result of the SQUID measurements (Table I). High quality fits are also obtained for the 300 K

TABLE I. Moments fitted by PNR (interfacial model and nominal layer models) compared to moments fitted by SQUID magnetometry. The sample area used for the SQUID measurements is 0.165 cm^2, leading to the following conversions: for Co, 1.376×10^{-6} emu/μ_B Å; for FeNi, 1.373×10^{-6} emu/μ_B Å

Layer	Moment ($\times10^{-5}$ emu) at RT		Moment ($\times10^{-5}$ emu) at 100 K			Moment ($\times10^{-5}$ emu) at 1.5 K	
	PNR (Interfacial)	SQUID	PNR (Interfacial)	PNR (Nominal)	SQUID	PNR (Interfacial)	SQUID
Co	5.09±0.84	5.03	5.39±1.02	4.46±0.49	5.40	5.56±0.88	5.51
FeNi	2.87±0.48	2.86	3.16±0.79	3.17±0.42	3.13	3.23±0.83	3.21

TABLE II. Thicknesses and moments (in Bohr magnetons) used to fit the PNR data using an interfacial layer model. The errors shown for the thicknesses are those determined for the room temperature fit and are typical of the values found for the other fits.

Layer	Thickness (Å)	PNR Moment (μ_B)		
		RT	100 K	1.5 K
Cu	62.65±2.24			
Cu-Co	10.25±1.13	1.44±0.21	1.52±0.13	1.57±0.26
Co	4.44±0.94	1.60±0.41	1.67±0.83	1.73
Co-Cu	10.60±1.14	1.42±0.22	1.53±0.20	1.58±0.31
Cu	64.45±2.05			
Cu-FeNi	11.14±2.14	0.82±0.15	0.91	0.93
FeNi	5.06±1.93	0.86±0.06	0.94	0.96
FeNi-Cu	8.88±1.72	0.83±0.13	0.91±0.22	0.93
Cu	451.13±2.86			

data using the interface model (see Table II) which again agree with the SQUID magnetometry measurements. As above, it is again possible to exclude a model in which the interface regions are not included. The average moments of the FeNi and Co based layers (i.e., effective moments averaged over both the interior and interface regions) are found to decrease by 11.1 and 7.4% (Ref. 19) in the temperature range 1.5–300 K, in excellent agreement with the results from SQUID magnetometry of 10.5 and 8.9%, respectively.

The interface regions are found to extend over a thickness of 9–11 Å and to have significantly lower effective magnetizations than the corresponding values expected for the pure FeNi and Co layers. Such a reduction is consistent with chemical intermixing but while no significant moment is expected for the Cu atoms, the values we obtain for the effective moments of the mixed composition regions are slightly larger than might be expected from the effect of simple dilution by the Cu atoms. However, the existence of reduced coordination effects associated with atomic scale roughness can increase the local moment on the magnetic atoms as has been observed in epitaxial Fe/Ag(001) layers, see, for example, Ref. 20. The existence of such a diffuse interface supports the interpretation of measurements of the temperature dependence of the giant magnetoresistance amplitude for FeNi/Cu/Co spin-valve structures which suggests the presence of intermixed interface regions.[16] For relatively rough interfaces, as in the case of FeNi/Cu/Co spin-valve structures, the interface exchange coupling is expected to be weakened in comparison with the ideal interface and the spin-fluctuation intensity and electron spin-flip scattering accordingly increased.

A very important aspect of PNR is its sensitivity to the vector orientation of the magnetic moments of the layers within the sample.[9] In the case of noncollinear structures the $+$ and $-$ reflectivities are both dependent on the in-plane components of the magnetization vector as described by a reflectivity matrix.[6] However, until now this capability has not been tested to our knowledge in trilayer samples in which the magnetic orientation can be accurately controlled. Figure 2(a) shows the spin asymmetry defined by $S=(R^+ - R^-)/(R^+ + R^-)$ at room temperature measured by first saturating the sample along an easy axis with an applied field of

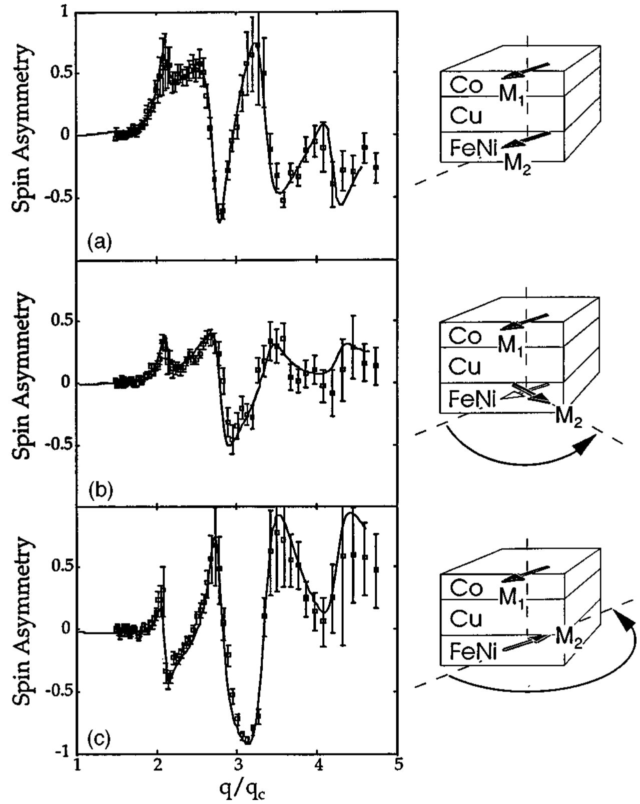

FIG. 2. The spin asymmetry spectra as a function of the relative layer magnetization orientation at room temperature for (a) parallel alignment and after the sample is rotated with respect to the applied field by approximately (b) 90° and (c) 180° causing the FeNi layer to rotate as shown in the schematic (right insets). The lines correspond to fits described in the text.

3 kOe and then reducing the field to 50 Oe, which is enough to switch the FeNi layer but not the Co layer magnetization. Strong variations in the spin asymmetry as a function of wave vector are seen. A best fit is shown as a solid line obtained with all the magnetic layers aligned parallel to the applied field. It can be seen that all the features of the data are well reproduced by the model. The sample was then physically rotated in the applied field by $\sim$90° [Fig. 2(b)] and $\sim$180° [Fig. 2(c)]. The spin asymmetry data display dramatic variations according to the direction of the sample with respect to the applied field, with pronounced oscillatory structure across the wave vector range. In fitting the data we first assume that the FeNi layer has coherently rotated to angles of 90° and 180°, respectively, and fit the moments of the layers; then we vary the angle of the FeNi layer ϕ keeping the moments fixed. For the 180° data the fitted angle is very close (within 1°) to 180° confirming that coherent rotation occurs. The inequivalence of the spin orientations in both orientations is clearly seen in the fit parameters. In each orientation, very good agreement between all the features of the data and the fitted curves is obtained indicating that to a good approximation the FeNi layer magnetization indeed rotates as a single domain to become aligned parallel to the applied field and that to a good approximation the orientation of the Co layer is unchanged, thus exhibiting ideal spin-valve behavior. For the 90° state the best fit parameters yield an error of 8°–14° indicating that in this configuration the spin

asymmetry is less sensitive to the spin-separation angle. We emphasize that in addition to the vector orientation of the layers, PNR also yields the absolute value of the total moment of each layer. This is important in determining the extent to which the layers are uniformly magnetized.

In conclusion we have used polarized neutron reflectometry measurements to demonstrate interface selective magnetometry in an epitaxial FeNi/Cu/Co trilayer spin-valve structure. Interface and interior magnetic moments in each of the FeNi and Co layers are obtained which agree within errors with the results of SQUID magnetometry measurements of the total sample moment. This is the first time to our knowledge that such a detailed comparison between SQUID magnetometry and PNR has been successfully performed. The results are consistent with the presence of chemical intermixing inferred from the strong temperature variation of the GMR amplitude in FeNi/Cu/Co spin-valve structures. We show that the layer-dependent vector moments can be determined quantitatively with high precision from measurements of the spin asymmetry as a function of wave vector. The capability of PNR in quantitatively probing the interface spin structure of layers with a common magnetic element is likely to provide an important complementary probe to XMCD techniques and to play an important part in unraveling the spin structure of magnetic interfaces in the future.

We are grateful to the Toshiba Corporation and to the EPSRC for supporting this work, and we thank the Rutherford Appleton Laboratory for making reflectometry facilities available.

[1] For a review of these topics see, for example, *Ultrathin Magnetic Structures*, edited by J. A. C. Bland and B. Heinrich (Springer-Verlag, Berlin, 1994), Vols. I and II.

[2] A. Vega, C. Demangeat, H. Dreyssé, and A. Chouairi, Phys. Rev. B **51**, 11 546 (1995).

[3] V. Chakarian, Y. U. Idzerda, G. Meigs, E. E. Chaban, J. H. Park, and C. T. Chen, Appl. Phys. Lett. **66**, 3368 (1995); C. T. Chen, Y. U. Idzerda, H. J. Lin, G. Meigs, A. Chaiken, G. A. Prinz, and G. H. Ho, Phys. Rev. B **48**, 642 (1993).

[4] G. Spierings, V. Koutsos, H. A. Wierenga, M. W. J. Prins, D. Abraham, and Th. Rasing, J. Magn. Magn. Mater. **121**, 109 (1993).

[5] V. Chakarian, Y. U. Idzerda, H. J. Lin, C. Gutierrez, G. A. Prinz, G. Meigs, and C. T. Chen, Phys. Rev. B **53**, 11 313 (1996).

[6] G. P. Felcher, R. O. Hilleke, R. K. Crawford, J. Haumann, R. Kleb, and G. Ostrowski, Rev. Sci. Instrum. **58**, 609 (1987); S. J. Blundell and J. A. C. Bland, Phys. Rev. B **46**, 3391 (1992).

[7] J. A. C. Bland, in *Ultrathin Magnetic Structures* (Ref. 1), pp. 305–343.

[8] R. Rodmacq, K. Dumesnil, Ph. Mangin, and M. Hennion, Phys. Rev. B **48**, 3556 (1993).

[9] A. Schreyer, J. F. Ankner, T. Zeidler, H. Zabel, C. F. Majkrzak, M. Schafer, and P. Grunberg, Europhys. Lett. **32**, 595 (1995).

[10] A. Schreyer, K. Bröhl, J. F. Ankner, C. F. Majkrzak, Th. Zeidler, P. Bödeker, N. Metoki, and H. Zabel, Phys. Rev. B **47**, 15 334 (1993).

[11] W. Hahn, M. Loewenhaupt, Y. Y. Huang, G. P. Felcher, and S. S. P. Parkin, Phys. Rev. B **52**, 16 041 (1995).

[12] S. Adenwalla, G. P. Felcher, E. E. Fullerton, and S. D. Bader, Phys. Rev. B **53**, 2474 (1996).

[13] J. F. Ankner, C. F. Majkrzak, and H. Homma, J. Appl. Phys. **73**, 6436 (1993).

[14] O. F. K. McGrath, N. Ryzhanova, C. Lacroix, D. Givord, C. Fermon, C. Miramond, G. Saux, S. Young, and A. Vedyayev, Phys. Rev. B **54**, 6088 (1996).

[15] M. Patel, T. Fujimoto, E. Gu, C. Daboo, and J. A. C. Bland, J. Appl. Phys. **75**, 6528 (1994).

[16] T. Fujimoto, M. Patel, E. Gu, C. Daboo, and J. A. C. Bland, Phys. Rev. B **51**, 6719 (1995).

[17] R. Naik, M. Ahmad, G. L. Dunifer, C. Kota, A. Poli, K. Fang, U. Rao, and J. S. Payson, J. Magn. Magn. Mater. **121**, 60 (1993).

[18] A. Ercole, T. Fujimoto, M. Patel, C. Daboo, R. J. Hicken, and J. A. C. Bland, J. Magn. Magn. Mater. **156**, 121 (1996).

[19] M. Patel, T. Fujimoto, A. Ercole, C. Daboo, and J. A. C. Bland, J. Magn. Magn. Mater. **156**, 53 (1996).

[20] J. A. C. Bland, C. Daboo, B. Heinrich, Z. Celinski, and R. D. Bateson, Phys. Rev. B **51**, 258 (1995).