Giant magnetoresistance and oscillatory exchange coupling in disordered Co/Cu multilayers

1999 J. Phys.: Condens. Matter 11 81

(http://iopscience.iop.org/0953-8984/11/1/007)

J. Phys.: Condens. Matter **11** (1999) 81–88. Printed in the UK PII: S0953-8984(99)94381-3

Giant magnetoresistance and oscillatory exchange coupling in disordered Co/Cu multilayers

C H Marrows†, Nathan Wiser†§, B J Hickey†, T P A Hase‡ and B K Tanner‡

† The E C Stoner Laboratory, Department of Physics, University of Leeds, Leeds LS2 9JT, UK

‡ Department of Physics, University of Durham, Durham DH1 3LE, UK

Received 18 May 1998, in final form 2 October 1998

Abstract. We measured the exchange coupling strength and magnetoresistance for multilayers of Co/Cu grown by sputtering. The samples have (111) texture with a grain size of about 140 Å. Their mosaic spread, determined by high-angle x-ray rocking curves in triple-axis geometry, is as large as 16°, suggesting the description *disordered*. The magnetoresistance oscillates as a function of the Cu thickness and reaches 70% at room temperature with near zero remanence at the first antiferromagnetic peak. These results indicate that a sufficient condition for oscillatory exchange coupling with a period of ∼ 10 Å is a well defined separation between the magnetic layers. We also measured the exchange coupling strength as a function of the magnetic layer thickness and found no oscillations. The measured saturating magnetic field is accurately described by the 'orange-peel' coupling effect.

1. Introduction

The great interest in magnetic multilayers originated with the discovery of a giant magnetoresistance (GMR) arising from the exchange coupling between neighbouring magnetic layers. One of the most striking features of the interlayer exchange coupling is that it oscillates in magnitude as a function of the thickness of the non-magnetic layers [1]. Originally it was proposed that the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction could account for the oscillations apart from the fact that the experimentally measured period was considerably larger than $\lambda_F/2$. The so-called *aliasing* effect [2]—the discrete nature of the spacer thickness means that the RKKY oscillations can only be sampled at certain points—was proposed to account for the observed longer periods. Subsequently, it was shown [3] that the period of these oscillations can be derived from spanning vectors of the spacer Fermi surface, with multiple periods existing in certain crystal orientations. For the best crystalline samples grown by molecular beam epitaxy, the predicted multiple periods have been observed [4]. For sputtered samples, it is assumed that the observed periods have the same origin but are less well defined due to their polycrystalline structure. Nevertheless, the existence of oscillations in polycrystalline samples relies on some degree of texture [4, 5].

As with most experimental results, an important feature of the models of exchange coupling is that they refer to *ordered* crystalline spacers. Recently, attention has been turning to the role of *disorder* in exchange coupling. Experiments have shown that oscillations exist in the coupling through spacers composed of alloys and of amorphous metals [6]. Disorder is difficult to treat theoretically. The effect on the coupling of substitutionally disordered alloys has been

§ Permanent address: Department of Physics, Bar-Ilan University, Ramat-Gan, Israel.

0953-8984/99/010081+08$19.50

dealt with by Bruno *et al* [7] with the disorder being represented by different potentials sited on a crystal lattice as in the Anderson model. The coupling through amorphous silicon has also received some attention [8] but the calculation was performed for an ordered insulator. The calculation did explain the temperature dependence of the coupling, but the predicted oscillations were not observed.

Experimentally, the investigation of the coupling and the GMR in samples with amorphous spacers is impeded by the high resistivity of the material. In extreme cases, the magnetic layers can shunt the current away from the spacer layers. What is required is a topologically disordered material that does not have a very high resistivity. In this context, it is relevant to recall the results of Parkin [9], who found the same coupling period of about 10 Å for many transition-metal spacers. Though these films were not disordered, they represent a wide variety of crystal structures with various orientations and rather different Fermi surfaces. Nevertheless, the period was found to be almost the same in each case, independent of the particular crystalline order. On the other hand, the importance of grain size in multilayers was first pointed out by Bobo *et al* [10]. They found the grain size (estimated from x-ray scattering) in their Fe-buffered samples was approaching 20 Å, and the samples still displayed a reasonable GMR and oscillations. These results reveal a poorly understood phenomenon: the coupling through nanocrystalline materials, amorphous spacers and samples with different crystallinity always exhibits the same period of about 10 Å. In other words, the oscillatory exchange coupling seems to be *independent* of crystallinity.

Other previous work of importance is that of Egelhoff and Kief [11] who have emphasized the importance of texture for understanding the oscillations exhibited by MBE-grown multilayers on a Cu(111) substrate. Moreover, Miranda [12] has shown that MBE-grown samples may fail to exhibit oscillations because of island growth of the magnetic Co layer. Thus, our present studies on sputtered Co/Cu samples complement these previous MBE studies regarding the role of texture in the measurement of oscillations.

In this paper, we report measurements of the coupling strength for *disordered* Co/Cu sputtered samples. These samples, produced without buffers or caps, have reasonably low resistivities, a GMR that is as high as any published for Co/Cu [13], extremely smooth interfaces and they exhibit an oscillatory coupling with a period of 10 Å as a function of the Cu thickness.

In addition to oscillations in the magnitude of the exchange coupling as a function of the thickness of the *non-magnetic* spacer layers, such oscillations were also predicted [14, 15] as a function of the thickness of the *magnetic* layers. This latter prediction has been examined experimentally. For the Co/Cu system, Qiu *et al* [16] did not find any such oscillations, whereas Bloemen *et al* [17] reported oscillations with a period of 6–7 Å of Co. A similar situation exists for the Fe/Cr system. Okuno and Inomata [18] reported the presence of oscillatory coupling, whereas Schad *et al* [19] found no oscillatory behaviour.

In view of these conflicting results, we measured the strength of the exchange coupling also as a function of the Co layer thickness. No oscillations were observed.

In section 2, we describe the samples. Our experimental results are presented in section 3, as a function of both the Cu thickness and the Co thickness. The conclusions follow in section 4.

2. Growth and characterization of the samples

The Co/Cu samples were deposited by DC magnetron sputtering in a system with a base pressure of $\sim 5 \times 10^{-8}$ Torr. Growth rates of ~ 4 Å s^{-1} were achieved using a sputtering pressure of 3.0 mTorr of Ar gas. The Si(001) substrates were washed in organic solvents to remove grease; more aggressive cleaning techniques were avoided in order to preserve the very smooth native oxide layer on the surface. Samples were grown without buffers or caps and contained either twenty or fifty bilayer repeats. Low-angle x-ray reflectivity and diffuse scattering experiments were performed on station 2.3 at the Daresbury Synchrotron Radiation Source [20] with a slit-limited instrumental resolution of 40 arc seconds. High-angle diffraction scans were also performed at the SRS using a Soller slit detector collimator and on a conventional Bragg–Brentano powder diffractometer using Cu Kα radiation and a curved crystal graphite monochromator. The magnetoresistance was measured at room temperature using a standard four-probe DC technique, and magnetometry was performed at room temperature by means of the magneto-optic Kerr effect (MOKE).

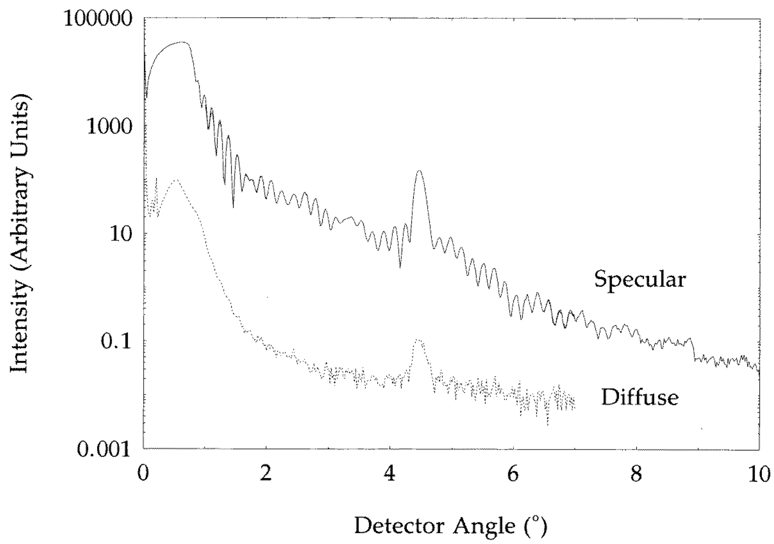

Figure 1. Low-angle reflectivity data for the Co/Cu multilayer taken with an x-ray wavelength of 1.48 Å. Clear Kiessig fringes extend out to about 8°, indicating the smooth nature of the surface. The modulation is caused by CuO as the layers are uncapped. There is a single Bragg peak visible at 4.5°. The diffuse scatter shows that the roughness is correlated throughout the multilayer stack.

Figure 1 shows the low-angle x-ray data for both the specular and diffuse components of the scatter for $[Co(10 Å)/Cu(10 Å)]_{20}$. The specular results show very clear Kiessig fringes extending up to about 8 degrees and a single multilayer Bragg peak. The clarity of the fringes is determined by the roughness of the top surface. The gradual rate of decrease in intensity with scattering vector observed here indicates that there is very little surface and interface roughness. Longitudinal (offset $\theta - 2\theta$) diffuse scans were measured using an offset of $-0.1°$. These results show very low diffuse scatter, again consistent with very little roughness. The presence of a clear Bragg peak in the diffuse scatter indicates that much of the roughness is *correlated* throughout the multilayer stack. Born wave analysis of transverse (θ) scans give a correlated roughness of 1.0 ± 0.5 Å. Another unusual feature of these samples is that the use of an Fe buffer generally makes the samples rougher (with a smaller GMR), as compared to the samples grown directly onto the native SiO_2 on the polished silicon surface.

The most important distinction between these sputtered samples and those of other workers lies in the nature [6] of the crystalline texture revealed by high-angle x-ray scans. Under normal laboratory conditions, the high-angle (111) Co/Cu peak is often absent or very small. In our high-angle ($\theta - 2\theta$) diffraction data taken at the SRS, the (111) Cu/Co Bragg peak is small but well defined. The full-width at half-height maximum yields an average grain size of 140 Å. In figure 2, we show x-ray rocking curves in quasi-triple-axis geometry, for two

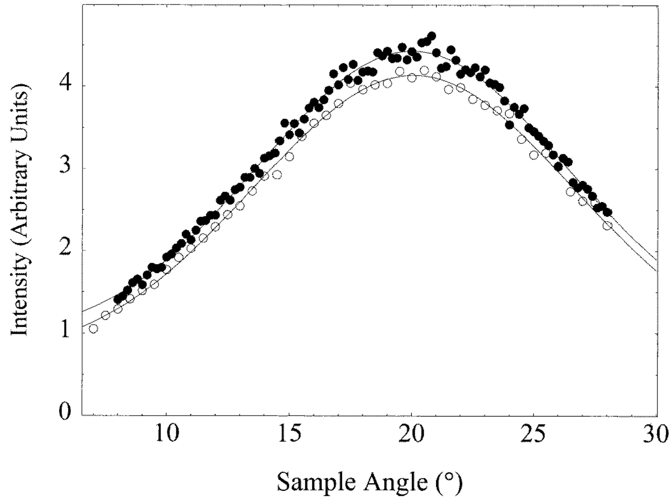

Figure 2. Transverse scans (rocking curves in quasi-triple-axis geometry) for two typical samples taken through the compromise Cu/Co(111) peak. The data have been corrected for the changing illumination area. The curves are fits to Gaussians with an FWHM of 16°.

typical samples. The spread of (111) oriented grains around the surface normal is about 16° FWHM. This explains the low intensity in the laboratory data. Our investigation of a large number of samples grown under different conditions showed that the period of the oscillation is independent of the sample crystallinity as measured by high-angle x-ray diffraction. We note the important difference between the texture of our samples and those discussed by Egelhoff and Kief [6].

A grain size of about 140–200 Å is fairly typical of both MBE and sputtered samples; it is the rocking curve width of our samples that is very unusual. Our previously reported MBE samples [21] had rocking curve widths much less than a degree, whereas these sputtered samples have widths of 16 degrees, which is to be compared with the two degrees reported for sputtered samples by most other workers. The large amount of disorder present in our sputtered samples means that the fine topological details of the Fermi surface are averaged out among the different grains.

3. Experimental results

3.1. Varying the thickness of the Cu layers

Figure 3 shows the room temperature magnetoresistance for multilayers having a constant 10 Å thickness of Co plotted as a function of the Cu thickness. The insets show longitudinal MOKE curves for various samples measured at the same temperature. The GMR data exhibit two clear peaks and a hint of a third peak, corresponding to regions of antiferromagnetic coupling separated by a region of ferromagnetic coupling. The period of this oscillation closely matches that reported previously for sputtered samples of Co/Cu, even though our samples differ from the others by virtue of their low crystalline texture.

The magnitude of the GMR is very high (rising to 130% at 4.2 K) and the coupling at the first antiferromagnetic peak has virtually zero remanence. The implication of these results is very important. To achieve good antiferomagnetic coupling, a large GMR and oscillations as

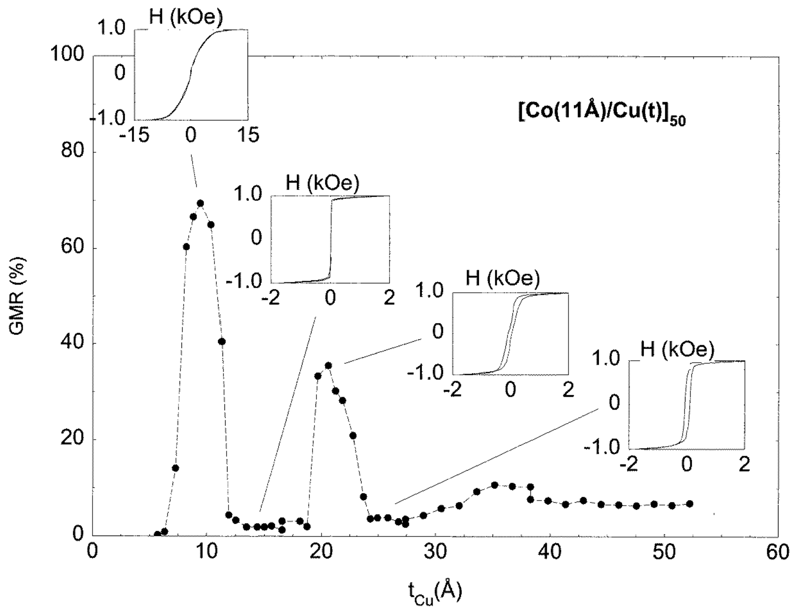

Figure 3. The GMR as a function of the thickness of the Cu layer. Each inset shows a MOKE image corresponding to the indicated sample. There are clear oscillations with a period of about 10 Å, showing distinct regions of antiferromagnetic and ferromagnetic coupling.

a function of the spacer thickness, it is only necessary to have well defined layers. The origin of the oscillations in these samples is the RKKY interaction and the aliasing effect, with the aliasing period given by $2\pi/\Lambda = [2k_F - n2\pi/d]$, where n is an integer and d is the spacer thickness increment (interlayer spacing for a crystal).

3.2. Varying the thickness of the Co layers

Our experimental data show no oscillations as a function of the Co thickness. The magnitude of the GMR does not depend on the magnitude of the coupling, but the interlayer exchange coupling strength can be determined by measuring the magnetic field that saturates the magnetoresistance. The analysis proceeds by writing the energy per unit area of a magnetic layer,

$$E = -\mu_0 MHt \cos\theta - J\cos\Theta \tag{1}$$

where Θ is the angle between the magnetizations M of neighbouring magnetic layers, θ is the angle between the magnetic field H and the magnetization, t is the thickness of the magnetic layers and J is the exchange coupling energy per unit area, with negative values of J corresponding to antiferromagnetic coupling.

Note that there is no anisotropy term in (1). Magnetometry measurements of the samples showed no sign of anisotropy for any thickness of the magnetic layer. Moreover, the low texture revealed by high-angle x-ray scans indicates that there is little evidence to support the existence of in-plane anisotropy, despite the presence of a magnetic field during growth.

A straightforward analysis of (1) shows [22] that the magnetic field that saturates the magnetization, and hence the magnetoresistance, is given by

$$H_s = 4J/\mu_0 Mt. \tag{2}$$

Therefore, the measured saturation field H_s gives directly the strength of the exchange coupling energy J.

There is a further point to the analysis. Many years ago, Néel [23] discussed what he picturesquely called the 'orange-peel effect', relating to the wavy structure that forms as thin magnetic layers increase in thickness. The 'orange-peel effect' has since been studied by a number of workers [24, 25]. The orange-peel structure is a two-dimensional sinusoidal wave, characterized by roughness amplitude σ and wavelength λ. Atomic-force-microscopy measurements confirm the presence of such surface roughness for the thicker magnetic layers and the x-ray analysis of the samples showed that much of this roughness is correlated throughout the multilayer stack.

The effect of this wavy structure is to generate magnetic poles on neighbouring magnetic layers which couple ferromagnetically, thus reducing the effective strength of the antiferromagnetic exchange coupling. A clear exposition of the effect and the explicit formulae are given in [25]. For the present analysis, it is sufficient to note that the effective coupling J_{eff} can be written

$$J_{eff} = J - J_{op}t^2 \tag{3}$$

where J is the true antiferromagnetic exchange coupling energy and J_{op} is the correction term due to the orange-peel effect. Inserting (3) into (2) yields the final expression, which is to be compared with the data,

$$H_s t = (4/\mu_0 M)(J - J_{op}t^2). \tag{4}$$

The values of t and H_s are measured, M is given by its handbook value (1.4 MA m^{-1} for Co at room temperature) and the values of J and J_{op} are determined by fitting to the data. In figure 4, we plot the experimental values of $H_s t$ as a function of t^2 for the Co/Cu multilayers, with each point representing a different sample. The accuracy with which we can define the relative layer thicknesses in a series of samples is determined by x-rays to be 0.5 Å.

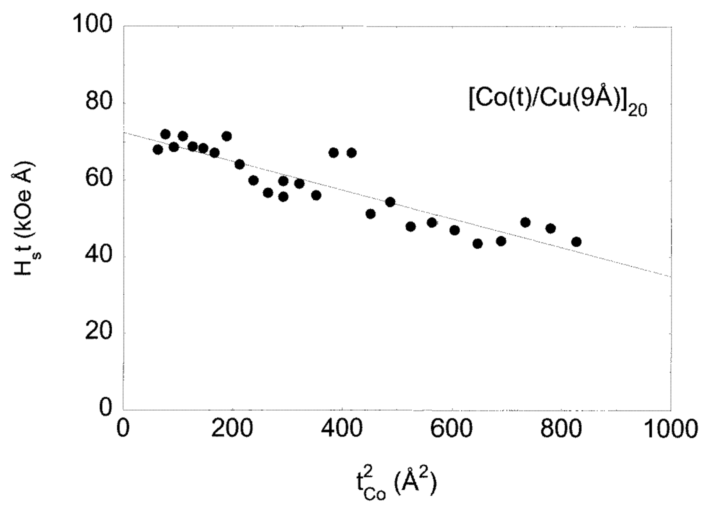

Figure 4. Saturation field H_S times the thickness of the Co layers as a function of the square of the thickness for Co/Cu multilayers. The straight line is the best fit to the data.

The straight line in figure 4 yields a reasonable fit to the data. The intercept and slope of the line determine J and J_{op}, respectively. A thickness-independent value of J_{eff} would yield a horizontal line, which is clearly at variance with the data, implying that the orange-peel effect is significant.

Oscillations as a function of Co thickness are not apparent in the data. The absence of oscillations might be explained by the work of Barnas [26]. He found that for a band splitting of 2 eV, the oscillations are so severely damped that only two large peaks are visible below 10 Å and virtually none above.

We can confirm the reliability of our data by comparing our derived value of J with that of previous workers. From the intercept of figure 4, we obtain

$$J \approx 0.25 \text{ mJ m}^{-2} \tag{5}$$

which is in agreement with the values previously reported for Co/Cu multilayers by Mosca *et al* [1] (≈ 0.3 mJ m^{-2}) and by Parkin *et al* [1] (≈ 0.15 mJ m^{-2}).

From the slope of figure 4, we obtain

$$J_{op} \approx 1.3 \times 10^{13} \text{ J m}^{-4}. \tag{6}$$

This value of J_{op} is consistent with the following values for the wavelength λ and the roughness amplitude σ of the orange-peel wave:

$$\lambda \approx 140 \text{ Å} \qquad \sigma \approx 12\% \text{ of bilayer thickness.} \tag{7}$$

These values are consistent with both the synchrotron x-ray scans and the atomic force microscope imaging of the top surface of our samples.

4. Conclusions

We have shown that a sufficient condition for long-period oscillations in the exchange coupling of Co/Cu multilayers is *well defined layer thicknesses*, even without a complicated Fermi surface. For poorly textured samples, where the Fermi surface is essentially spherical, oscillations have been observed in the exchange coupling as a function of the Cu thickness with a period of 10 Å. These samples have a room-temperature GMR of ~70% at the first antiferromagnetic maximum at a magnetic field of 0.6 T. Adding Fe buffer layers *reduces* the magnitude of the GMR and makes the samples rougher.

We also measured the strength of the exchange coupling as a function of the thickness of the magnetic layers, with no sign of oscillations being observed.

Acknowledgments

It is a pleasure to thank Mark Howson, Christian Lehmann and Matthew Wormington for very useful discussions. C H Marrows was supported by a University of Leeds Henry Ellison Scholarship, T P A Hase was supported by an EPSRC CASE studentship with Daresbury Laboratory and N Wiser was supported by a grant from the UK–Israel Science and Technology Research Fund.

References

[1] Parkin S S P, More N and Roche K P 1990 *Phys. Rev. Lett.* **64** 2304
Parkin S S P, Bhadra R and Roche K P 1991 *Phys. Rev. Lett.* **66** 2152
Petroff F, Barthélémy A, Mosca D H, Lottis D K, Fert A, Schroeder P A, Pratt W P Jr, Loloee R and Lequien S 1991 *Phys. Rev.* B **44** 5355
Purcell S T, Folkerts W, Johnson M T, McGee N W E, Jager K, aan de Stegge J, Zeper W B and Grünberg P 1991 *Phys. Rev. Lett.* **67** 903
Mosca D H, Petroff F, Fert A, Schroeder P A, Pratt W P Jr and Loloee R 1991 *J. Magn. Magn. Mater.* **94** L1

[2] Coehoorn R 1991 *Phys. Rev.* B **44** 9331
[3] Bruno P and Chappert C 1991 *Phys. Rev. Lett.* **67** 1602
Bruno P 1993 *J. Magn. Magn. Mater.* **121** 248
[4] See various contributions in Heinrich B and Bland J A C (eds) 1994 *Ultrathin Magnetic Structures* vol II (Berlin: Springer)
[5] Mathon J 1991 *Contemp. Phys.* **32** 143
Youssef J B, Bouziane K, Koshkina O, Le Gall H, El Harfaoui M, El Yamani M, Desvignes J M and Fert A 1997 *J. Magn. Magn. Mater.* **165** 288
[6] Okuno S N and Inomata K 1993 *Phys. Rev. Lett.* **70** 1711
Parkin S S P, Chappert C and Herman F 1993 *Europhys. Lett.* **24** 71
Bobo J-F, Hennet L and Piécuch M 1993 *Europhys. Lett.* **24** 139
Leng O, Cros V, Schäffer R, Fuss A, Grünberg P and Zinn W 1993 *J. Magn. Magn. Mater.* **126** 367
Briner B and Landolt M 1994 *Phys. Rev. Lett.* **73** 340
[7] Bruno P, Kudrnovsky J, Drchal V and Turek I 1996 *Phys. Rev. Lett.* **76** 4254
Bruno P, Kudrnovsky J, Drchal V and Turek I 1997 *J. Magn. Magn. Mater.* **165** 128
[8] Bruno P 1994 *Phys. Rev.* B **49** 13 231
[9] Parkin S S P 1991 *Phys. Rev. Lett.* **67** 3598
[10] Bobo J-F, Baylac B, Hennet L, Lenoble O, Piecuch M, Raquet B and Ousset J-C 1993 *J. Magn. Magn. Mater.* **121** 291
[11] Egelhoff W F Jr and Kief M T 1992 *Phys. Rev.* B **45** 7795
[12] Miranda R 1993 *Phys. Scr.* T **49** 579
[13] Kano H, Kagawa K, Suzuki A, Okabe A, Hayashi K and Aso K 1993 *Appl. Phys. Lett.* **63** 2839
[14] Barnas J 1992 *J. Magn. Magn. Mater.* **111** L215
[15] Bruno P 1993 *Europhys. Lett.* **23** 615
[16] Qiu Z Q, Pearson J and Bader S D 1992 *Phys. Rev.* B **46** 8659
[17] Bloemen P J H, Johnson M T, van de Vorst M T H, Coehoorn R, de Vries J J, Jungblut R, aan de Stegge J, Reinders A and de Jonge W J M 1994 *Phys. Rev. Lett.* **72** 764
[18] Okuno S N and Inomata K 1994 *Phys. Rev. Lett.* **72** 1553
[19] Schad R, Beliën P, Barnas J, Verbanck G, Potter C D, Gladyszewski G, Moshchalkov V V and Bruynseraede Y 1996 *J. Magn. Magn. Mater.* **156** 341
[20] Tanner B K, Joyce D E, Hase T P A, Pape I and Grundy P J *Adv. X-ray Anal.* at press
[21] Hase T P A, Pape I, Tanner B K, Laidler H, Ryan P A and Hickey B J 1997 *Proc. Int. Conf. on Magnetism (Cairns, 1997)* at press
[22] Barthélmy A, Fert A, Baibich M N, Hadjoudj S, Petroff F, Etienne P, Cabanel R, Lequien S, Nguyen Van Dau F and Creuzet G 1990 *J. Appl. Phys.* **67** 5908
[23] Néel L 1962 *C.R. Acad. Sci., Paris* **255** 1545
Néel L 1962 *C.R. Acad. Sci., Paris* **255** 1676
[24] Yelon A 1971 *Physics of Thin Films* vol 6 (New York: Academic) pp 205–300
Rijks T G S M, Coehoorn R, Daemen J T F and de Jonge W J M 1994 *J. Appl. Phys.* **76** 1092
[25] Kools J C S 1995 *J. Appl. Phys.* **77** 2993
[26] Barnas J 1992 *J. Magn. Magn. Mater.* **111** L215