Mean Free Path Effects on the Current Perpendicular to the Plane Magnetoresistance of Magnetic Multilayers

Didier Bozec, M. A. Howson, and B. J. Hickey
Department of Physics and Astronomy, E. C. Stoner Laboratory, University of Leeds, Leeds LS2 9JT, United Kingdom

Smadar Shatz and Nathan Wiser
Jack and Pearl Resnick Institute for Advanced Technology, Department of Physics, Bar-Ilan University, Ramat-Gan, Israel

E. Y. Tsymbal and D. G. Pettifor
Department of Materials, University of Oxford, Oxford OX1 3PH, United Kingdom
(Received 20 December 1999)

We have carried out an experimental and theoretical study of the magnetoresistance MR(H) in the CPP (current perpendicular to the planes) mode for two types of magnetic multilayers that differ only in the ordering of the magnetic layers: $[\mathrm{Co}(10\ \text{Å})/\mathrm{Cu}(200\ \text{Å})/\mathrm{Co}(60\ \text{Å})/\mathrm{Cu}(200\ \text{Å})]_N$ and $[\mathrm{Co}(10\ \text{Å})/\mathrm{Cu}(200\ \text{Å})]_N[\mathrm{Co}(60\ \text{Å})/\mathrm{Cu}(200\ \text{Å})]_N$. The series resistor model predicts that in the CPP mode MR(H) is independent of the ordering of the layers. Nevertheless, the measured MR(H) curves were found to be completely different for the two cases. Calculations based on a realistic band structure and the Kubo formula show that the results are a consequence of a long mean free path.

PACS numbers: 75.70.Cn, 73.40.–c, 75.70.Pa

The giant magnetoresistance exhibited by magnetic multilayers [1] continues to be a subject of great interest. The first measurements of the magnetic-field dependence of the magnetoresistance, MR(H), were performed in the CIP (current in the plane of the layers) mode for reasons of technical simplicity. Recently, however, experimental [2–6] and theoretical [7,8] interest has shifted to the investigation of MR(H) in the CPP (current perpendicular to the plane of the layers) mode. The CPP-mode resistance of magnetic multilayers is often analyzed in terms of the series resistor model [8], which predicts that the relevant length scale is the spin-diffusion length. However, we shall show here that, in certain limits, the important length scale for determining MR(H) is the electron mean free path.

As is well known, the giant magnetoresistance occurs in magnetic multilayers because the spin-up electrons and spin-down electrons have different scattering rates. If the electron does not flip its spin upon scattering, then the spin-up and spin-down electrons constitute two separate currents, with different resistivities, as if flowing in two parallel wires. In the CPP mode, the resistances of the different layers add in series [8]. Therefore, it would seem that two magnetic multilayers that differ only in the ordering of the layers would yield identical results for MR(H) in the CPP mode.

To test this idea, Chiang and co-workers at Michigan State University measured [9] CPP MR(H) for the two configurations $[\mathrm{Py/Cu/Co/Cu}]_N$ and $[\mathrm{Py/Cu}]_N[\mathrm{Co/Cu}]_N$ (denoted as "interleaved" and "separated" configurations, respectively), where Py is $\mathrm{Ni_{84}Fe_{16}}$. Although the expectation was that identical MR(H) curves would be obtained for the interleaved and the separated configurations, these workers found [9] that the two resulting MR(H) curves were completely different. They attributed the different MR(H) curves they observed for the two configurations to the short spin-diffusion length in Py. They had previously analyzed resistivity data within the framework of Valet-Fert theory [8] and obtained [10] for Py a spin-diffusion length of only 55 Å, thus implying significant mixing between the spin-up and spin-down electron currents. A similar interpretation is possible for the recent measurements of MR(H) on multilayers containing Fe as one of the magnetic layers [11], because the spin-diffusion length of Fe is not known.

In this work, we measured MR(H) for multilayers whose magnetic layers are known to have an unusually long spin-diffusion length. Nevertheless, our MR(H) curves are completely different for the interleaved and separated configurations. In addition, we present a new interpretation of the MR(H) curves which includes the effect of the electron mean free path. This is a significant departure from the accepted view of the CPP conductivity.

For the present study of MR(H), we chose Co for the magnetic metal as Co has an extremely long spin-diffusion length. Measurements [12,13] yield a value of 450 Å or 600 Å or maybe [9] 1000 Å for the spin-diffusion length of Co. We used two different thicknesses, 10 and 60 Å, to achieve independent switching due to their different coercivities.

Measurements of MR(H) were carried out for $[\mathrm{Co}(10\ \text{Å})/\mathrm{Cu}/\mathrm{Co}(60\ \text{Å})/\mathrm{Cu}]_N$ and $[\mathrm{Co}(10\ \text{Å})/\mathrm{Cu}]_N[\mathrm{Co}(60\ \text{Å})/\mathrm{Cu}]_N$, where N is the number of repeats, and Cu-layer thickness was chosen to be 200 Å, sufficient to ensure that the magnetic layers were decoupled. The multilayers were grown in our VG-80M molecular beam epitaxy facility which has base pressure of typically 4×10^{-11} mbar. Our CPP measurements used the superconducting Nb electrode

 0031-9007/00/85(6)/1314(4)$15.00

technique, developed in collaboration with Pratt and coworkers [2]. The superconducting equipotential [3,4] ensures that the current is perpendicular to the layers. We used a SQUID-based current comparator, working at 0.1% precision to measure changes in the sample resistance of order 10 pΩ. Consistency between the interleaved and separated samples was enhanced by growing the two configurations during the same run for each value of N.

The data for MR(H) for $N = 4, 6, 8$ are presented in Fig. 1, where the squares and circles represent the interleaved and separated configurations, respectively. The peak values of MR that we obtained are typical of uncoupled multilayers with a small number of repeats. Our peak value of MR increased very substantially (from 20% to 33%) in going from $N = 4$ to $N = 8$.

There are several characteristic features of these data. (i) The most important feature is the striking difference between the MR(H) curves for the two configurations, both in shape and in magnitude. (ii) For each N, the maximum value of MR(H) is much larger for the interleaved configuration than for the separated configuration. (iii) The MR(H) curve for the interleaved configuration exhibits a single peak, whereas for the separated configuration, MR(H) is the superposition of two peaks, with the second being much broader and less well delineated than the first.

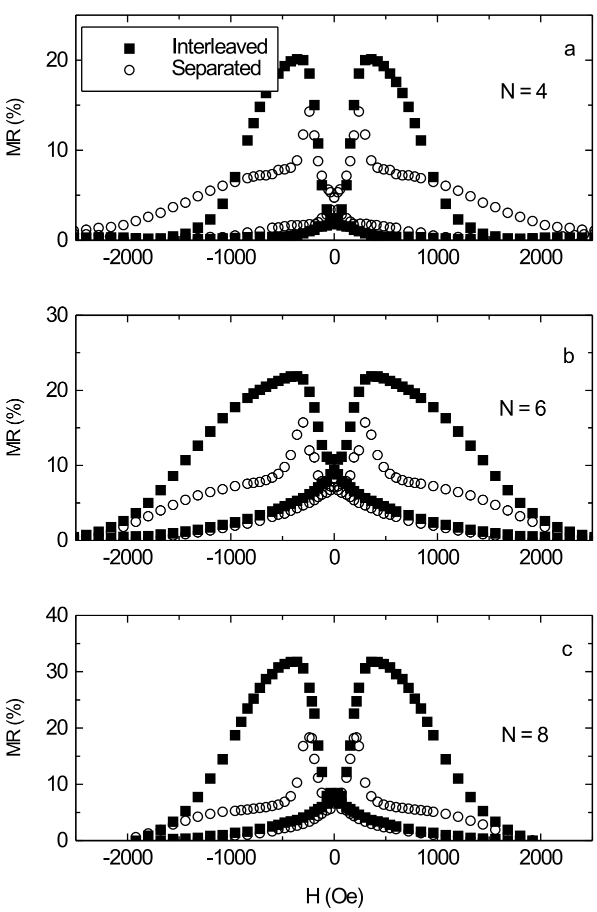

FIG. 1. Magnetic-field dependence of the magnetoresistance MR(H) for the interleaved (squares) and separated (circles) multilayers containing Co (10 Å) and Co (60 Å) as the two magnetic metals, for the indicated number of repeats.

Regarding the magnitude of the resistivity, at saturation, ρ lies in the range 4–7 $\mu\Omega$ cm with the value for the interleaved configuration being a bit higher (10%–15%) than for the separated configuration for each N. The resistance itself lies in the range 11–18 nΩ.

To ensure that the differing results for MR(H) for the two configurations are not due to differences in their magnetic properties, the magnetization as a function of field was measured for each sample. We found that the two configurations yield the same magnetization and the magnitudes of the saturation fields correspond closely to the saturation fields of MR(H).

The key to understanding these data is the following. Kinetic theory arguments show that the electron mean free path is far longer than the thicknesses of the magnetic layers. Therefore, the potential "felt" by the electron is the combined potential of several layers. One cannot speak of the resistivity of a single Co layer, but rather it is a property of pairs of neighboring magnetic layers that determines the resistivity. For such a case, the contribution of the spin-direction-dependent resistivity depends [14] on the cosine of the angle θ_{ij} between the moments of neighboring (denoted i and j) magnetic layers.

All the features of the MR(H) curves listed above can be understood in terms of this idea.

(i) For the interleaved configuration, the neighboring magnetic layers are Co layers of *different* thickness, and hence the maximum angle θ_{ij} is *large*, whereas for the separated configuration, the neighboring magnetic layers are Co of the *same* thickness (except for one boundary layer), and hence the maximum angle θ_{ij} is *small*. Since MR(H) depends on this angle, there is no reason to expect MR(H) to be the same for the two configurations.

(ii) It also follows that MR(H) will be larger for interleaved multilayers than for separated multilayers, because the angle θ_{ij} is larger for the former configuration.

(iii) For the interleaved configuration, there is only *one* angle θ_{ij} that is relevant, namely, the angle between the moments of the *different* (10 and 60 Å) neighboring magnetic layers. Therefore, there will be only one peak, as the angle θ_{ij} becomes progressively larger, passes through a maximum at the saturation field of the Co (60 Å) layer and then becomes smaller as the Co (10 Å) layer also saturates. By contrast, for the separated configuration, there are two angles θ_{ij} that are relevant, namely, the angle between neighboring moments for *each* of the two sets of magnetic layers. As each of these two angles passes through its maximum, a peak will be obtained for MR(H), leading to *two* overlapping peaks, with each maximum occurring at the value of the magnetic field that corresponds to the appropriate coercive field.

If the spin-diffusion length is very long, it is known [8] that a simple expression is obtained for MR(H). Applying the phenomenological theory of Wiser [15] readily yields that the magnetoresistance due to an ij pair of neighboring magnetic layers is

$$MR_{ij}(H) = c_{ij}[1 - \cos\theta_{ij}(H)]^2. \qquad (1)$$

For our samples, there are three parameters c_{ij} corresponding to the three different types of neighboring pairs of magnetic layers: $i = j = 1$; $i = j = 2$; $i = 1, j = 2$, where 1 refers to Co (60 Å) layers and 2 refers to Co (10 Å) layers. We determined the values of the parameters c_{ij} by fitting to the MR(H) data.

For the interleaved configuration, the magnetic field dependence of the angle $\theta_{1,2}$ is determined as follows. The magnetization increases linearly with field, and the magnetization is proportional to the cosine of the angle between the magnetic moment and the field. Therefore, $\cos\theta_1$ and $\cos\theta_2$ are each linear in the field, but with different coefficients. Equation (1) contains $\cos\theta_{1,2} = \cos(\theta_1 - \theta_2)$. Expanding the cosine gives the required field dependence.

For the field dependence of the separated samples the MR passes through maxima at the coercive field of each thickness of Co. We assumed a parabolic form for the field dependence of each of the angles $\theta_{1,1}$ and $\theta_{2,2}$, whose maximum was taken as a fitting parameter.

The calculated results for the interleaved and separated configurations for $N = 8$, together with the data, are presented in Figs. 2a and 2b. The agreement between the calculated curves and the data is evident.

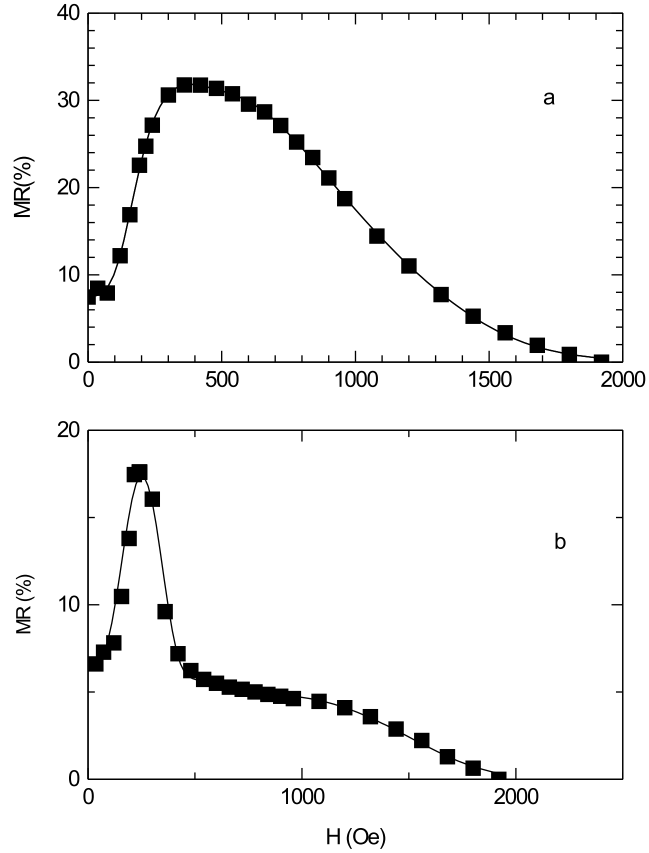

FIG. 2. Comparison between the calculated curves and the data points (squares) for MR(H) for $N = 8$ for the interleaved configuration (a) and for the separated configuration (b).

Calculations of the conductance have been performed using the Kubo formula [16] within a real space approach [17] which is convenient for layered systems. In this approach we consider a disordered Co/Cu(001) multilayer consisting of four layers of Co and three layers of Cu, each layer being of 10 monolayer (ML) thickness, and connected to the two perfect semi-infinite Cu(001) leads. The electronic structure of the multilayer and the leads is treated using a realistic multiband tight-binding model accounting for s, p, and d orbitals with their full hybridization and spin polarization [18]. First, we find the matrix elements of the surface Green's function for the semi-infinite leads, which can be expressed in terms of the Green's function for the bulk metal [19]. Then, the Co/Cu multilayer is constructed by adding disordered layers onto the left lead. The disorder is introduced as a random variation of the on-site atomic energies of the Co and Cu atoms with a uniform distribution of standard deviation 0.6 eV [18]. This gives the bulk resistivity of 4.64.6 $\mu\Omega$ cm for Cu and 14.3 $\mu\Omega$ cm for Co, which are reasonable. We note that the model takes into account interface scattering due to disorder introduced at the potential step between the Co and Cu layers. As was shown recently [20], this model explains quantitatively both the bulk and interface resistance of Co/Cu spin valves measured *in situ* during deposition. The Green's function of the added layers is recalculated at each step recursively by solving numerically the respective Dyson equation [17]. Once the sample has been fully constructed, the last layer is bonded to the right lead in order to obtain the Green's function $\hat{G}(E_F)$ of the full system, which enters the expression for the conductance:

$$\Gamma = \frac{\hbar}{\pi a^2} \langle \mathrm{Tr}[\mathrm{Im}\hat{G}(E_F)\hat{J}\,\mathrm{Im}\hat{G}(E_F)\hat{J}]\rangle . \qquad (2)$$

Here $\langle \cdots \rangle$ denotes averaging over disorder configurations, a is the distance between the individual layers, and the local current operator $\hat{J}$ takes the form

$$\hat{J} = \frac{ie}{\hbar} \sum_{ij\alpha\beta} (\mathbf{r}_{jl+1} - \mathbf{r}_{il})\mathbf{n}[h_{\beta jl+1,\alpha il}\,|\beta, j, l + 1\rangle\langle\alpha, i, l\,| - h_{\alpha il,\beta jl+1}\,|\alpha, i, l\rangle\langle\beta, j, l + 1\,|], \qquad (3)$$

where $\mathbf{n}$ is a unit vector in the direction perpendicular to the planes, $|\alpha, i, l\rangle$ is the α orbital of the atom which lies within the layer l at the in-plane site i and which has coordinate $\mathbf{r}_{il}$, and $h_{\alpha il,\beta jl+1}$ are tight-binding hopping integrals between planes l and $l + 1$. The conductance is calculated using a grid of four k-points in the full Brillouin zone and is averaged over six random configurations of disorder. Figure 3 shows the resistance versus thickness of the Co/Cu multilayer: the resistance of the full

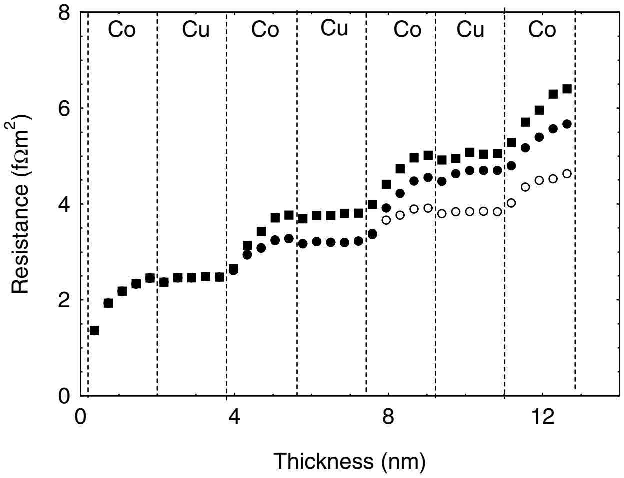

FIG. 3. Calculated resistance of the Co/Cu(001) multilayer which contains 10 ML of Co and 10 ML of Cu within each layer. The graph is plotted as a function of total thickness of the multilayer for the parallel magnetizations (open symbols) and for the antiparallel magnetizations (full symbols) within the interleaved (squares) and separated (circles) configurations.

structure is determined by the rightmost points in this figure. As is obvious from the figure, the result is in agreement with the experiments: giant magnetoresistance (GMR) for the interleaved configuration is higher than GMR for the separated configuration.

There are a few important points which follow from our modeling. (i) With increasing disorder in the Co/Cu multilayer the difference in GMR between the interleaved and separated configurations becomes smaller and eventually disappears when the mean free path becomes much less than the layer thicknesses. This is due to the fact that in this limit the series resistor model becomes justified. (ii) We find no difference in GMR between the interleaved and separated configurations within a single band tight-binding model with spin- and layer-dependent disorder, which determines different scattering rates for the majority and minority spin channels and for various layers. This is in agreement with the model which is based on a free-electron band structure for all layers [8]. (iii) The difference in GMR for the interleaved and separated configurations appears, however, when separate layers within the multilayer have different band structures like those in the Co/Cu multilayer. In this case, because of the change in the electron potential at the interfaces and the repeating Co and Cu layers quantum-well states are created within the multilayer. As was shown in Ref. [21], at film thicknesses comparable to or less than the mean free path, the series resistor model breaks down due to the reduced number of conducting channels within the potential-well structure. (iv) The calculation reproduces the important feature of the experiments, namely, an increase in the difference GMR between the interleaved and separated configurations with increasing the number of the Co/Cu bilayers.

In conclusion, we have shown that the CPP-mode MR(H) curves can be explained quantitatively, for both the interleaved and the separated configurations, by taking into account that the electron mean free path is much longer than the thickness of a layer. Previously it was thought that the spin-diffusion length was the only relevant length scale in CPP GMR.

It is a pleasant duty to acknowledge that this research was supported by grants from the U.K.-Israel Science and Technology Research Fund and the U.K.-EPSRC. We appreciate discussions with C. H. Marrows and A. Carrington. D. B. thanks the University of Leeds for financial support, and E. T. thanks Hewlett-Packard for financial support.

[1] M. N. Baibich, J. M. Broto, A. Fert, Van Dau F. Nguyen, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988); S. S. P. Parkin, Phys. Rev. Lett. **71**, 1641 (1993); R. E. Camley and J. Barnas, Phys. Rev. Lett. **63**, 664 (1989); J. Mathon, Contemp. Phys. **32**, 143 (1991); S. Y. Hsu, A. Barthelemy, P. Holody, R. Loloee, P. A. Schroeder, and A. Fert, Phys. Rev. Lett. **78**, 2652 (1997); J. A. Borchers, J. A. Dura, J. Unguris, D. Tulchinsky, M. H. Kelley, C. F. Majkrzak, S. Y. Hsu, R. Loloee, W. P. Pratt, Jr., and J. Bass, Phys. Rev. Lett. **82**, 2796 (1999).
[2] W. P. Pratt, Jr., S.-F. Lee, J. M. Slaughter, R. Loloee, P. A. Schroeder, and J. Bass, Phys. Rev. Lett. **66**, 3060 (1991).
[3] N. J. List *et al.,* J. Magn. Magn. Mater. **148**, 342 (1995).
[4] N. J. List *et al.,* Mater. Res. Soc. Symp. Proc. **384**, 329 (1995).
[5] S.-F. Lee *et al.,* J. Magn. Magn. Mater. **118**, L1 (1993).
[6] S.-F. Lee *et al.,* Phys. Rev. B **52**, 15 426 (1995).
[7] H. E. Camblong, S. F. Zhang, and P. M. Levy, Phys. Rev. B **47**, 4735 (1993).
[8] T. Valet and A. Fert, Phys. Rev. B **48**, 7099 (1993); A. Fert and S.-F. Lee, Phys. Rev. B **53**, 6554 (1996).
[9] W.-C. Chiang, Q. Yang, W. P. Pratt, Jr., R. Loloee, and J. Bass, J. Appl. Phys. **81**, 4570 (1997).
[10] S. D. Steenwyk, S. Y. Hsu, R. Loloee, J. Bass, and W. P. Pratt, Jr., J. Magn. Magn. Mater. **170**, L1 (1997).
[11] D. Bozec *et al.,* Phys. Rev. B **60**, 3037 (1999).
[12] L. Piraux *et al.,* J. Magn. Magn. Mater. **156**, 317 (1996).
[13] L. Piraux, S. Duboix, A. Fert, and L. Belliard, Eur. Phys. J. **4**, 413 (1998).
[14] J. L. Gittleman, Y. Goldstein, and S. Bozowski, Phys. Rev. B **5**, 3609 (1972).
[15] N. Wiser, J. Magn. Magn. Mater. **159**, 119 (1996).
[16] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
[17] P. A. Lee and D. S. Fisher, Phys. Rev. Lett. **47**, 882 (1981).
[18] E. Yu. Tsymbal and D. G. Pettifor, Phys. Rev. B **54**, 15 314 (1996).
[19] J. Pollmann and S. T. Pantelides, Phys. Rev. B **18**, 5524 (1978).
[20] W. E. Bailey *et al.,* Phys. Rev. B **61**, 1330 (2000).
[21] E. Yu. Tsymbal and D. G. Pettifor, Phys. Rev. B **61**, 506 (2000).