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Finite size scaling effects in giant magnetoresistance multilayers

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Abstract
We report on the findings of a study conducted on Co/Cu giant magnetoresistance multilayers with a wide variation in the number of bilayer repeats at temperatures from 4.2 to 300 K. The total film thickness varied from 77 to 2700 Å. When the data were treated within a Fuchs–Sondheimer approximation for finite size effects at any one temperature an excellent agreement with the observed variation in giant magnetoresistance ratio was found. Over ∼50 bilayers were required before the full giant magnetoresistance is observed. Unexpectedly, the length scale on which the GMR rises was found to be *the same* at all temperatures measured. This leads to unphysical temperature dependences in the Fuchs–Sondheimer mean free paths. Subsequent structural characterization showed that this length scale is related to the vertical coherence length of the grains within the sample. The same length scale was found for other related multilayer systems grown in the same chamber. The breakdown of the Fuchs–Sondheimer model is discussed in terms of modern theories of phase-coherent transport in the giant magnetoresistance: full giant magnetoresistance is only obtained when electrons can coherently sample a large enough number of layers.

1. Introduction

It is now a commonplace that the giant magnetoresistance (GMR) is observed in multilayered materials of a 3d ferromagnet and a non-magnetic metallic spacer layer: specifically, large effects are only found in well-band-matched materials combinations such as Fe/Cr or Co/Cu. Despite the huge body of work on the GMR in recent years [1–3], there are still discrepancies between the proposed theoretical models and certain observations. The simplest models, such as those set out at the time of the discovery of GMR [4, 5], treat the current as being carried by independent populations of spin ↑ and ↓ electrons, which provide parallel conduction channels. An essential feature of these models is that there is spin dependent scattering in the magnetic layers or at the ferromagnet–spacer interfaces. The GMR then arises from spin filtering effects, when layer moments are parallel one spin current can flow freely, whilst when they are antiparallel both spin channels are partly blocked.

GMR is commonly measured in two different experimental geometries: those with current in the plane (CIP) and current perpendicular to the plane (CPP). The CPP geometry is in general more tractable theoretically [6]. Measurements on the other hand are usually performed in the much less experimentally demanding CIP geometry. In this case the electrons do not flow from one layer to the next in a simple fashion, and finite size effects must be correctly taken into account due to the extreme thinness of the multilayer and the layers it comprises.

The importance of the mean free path, ℓ, in interpreting the resistivity of layered metallic structures was previously pointed out by Gurvitch [7]. In many transition metal superlattices the higher resistivities of these metals lead to mean free paths much shorter than individual layer thicknesses, allowing the application of a parallel resistor network to calculate the total resistivity. It was assumed that ℓ would be truncated at the interfaces. The interfaces are treated as entirely diffusive with zero transparency. It is easy to see that this will lead to zero CIP GMR, as both spin channels are completely scattered and there is hence no difference between them.

A semiclassical theory of the GMR was given by Dieny [8], extending the formalism of Camley and Barnas [9]. Finite size effects are accounted for using Fuchs–Sondheimer boundary conditions [10, 11] when solving the Boltzmann equation by assigning a spin dependent mean free path to each layer. The interfaces now possess some finite transparency. A number of experiments have focused on attempting to measure the mean free paths of individual layers within this formalism. Gurney *et al* measured the spin ↑ and inferred the spin ↓ mean free path in Co at room temperature as 55 and 6 Å respectively, by the use of a spin valve GMR trilayer with a conducting back layer [12]. More recently Shakespear *et al* have measured the temperature dependence of the majority spin mean free path in $Co_{90}Fe_{10}$; it is found to drop from roughly 100 Å at low temperatures to about 30 Å at room temperature. In these experiments the impact of averaging over all electron incidence angles was neglected, so that a meaningful quantitative comparison with these values is difficult. Strijkers *et al* measured the spin ↑ ℓ in Co to be 75 Å at 300 K [14].

More recent theories treat the entire multilayer as a single entity. There are calculations of both CPP and CIP GMR using a fully realistic s–d hybridized band structure in both infinite multilayers [15] and spin valve trilayers [16]. Recent theoretical results [16, 17] show that the individual layer properties are interdependent and the measured GMR is a property of the entire multilayer. Other models of phase-coherent transport in these structures [18] also require us to take a more holistic view of the properties of the multilayer.

In this paper we report experimental results where we have measured the finite size effects on GMR in multilayers over a wide temperature range. We have chosen to study these effects in the archetypal GMR system of Co/Cu multilayers. Through the use of this well-characterized and widely studied system we are able to make a number of points of general validity. We have varied the number of bilayers deposited in multilayers at the first antiferromagnetic (AF) coupling peak in the oscillation with Cu thickness [19]. We have chosen to carry out the GMR measurements in the CIP geometry in order to make use of Fuchs–Sondheimer theory [10, 11] in analysing the data. However we will see that the use of the Fuchs–Sondheimer model, despite describing the size dependence of the GMR quite accurately, leads to an unphysical temperature dependence for the mean free path.

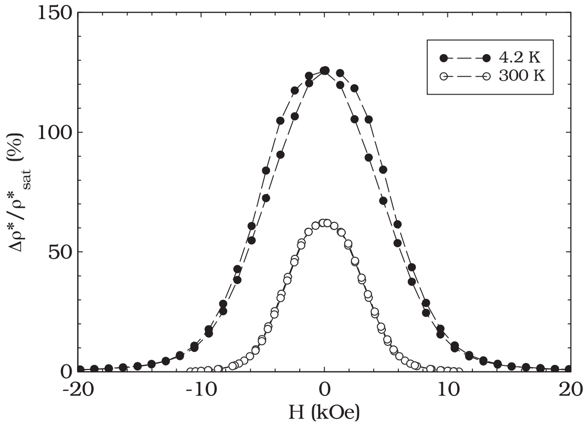

Figure 1. The giant magnetoresistance of the {Co(7.5 Å)/Cu(8 Å)} × 100 multilayer at 300 and 4.2 K. The parabolic shape of the peak indicates near-perfect antiferromagnetic coupling.

2. Sample preparation techniques

A series of samples were prepared by dc magnetron sputtering onto pieces of (001) oriented Si wafer under ultrahigh vacuum (UHV) conditions. Typical deposition rates were 3 Å s^{-1}. All the samples were prepared in a single vacuum cycle to minimize variations in preparation conditions. The only variable in the run was the number of bilayers of Co/Cu deposited, N, varied from 5 to 175. We have found that the use of buffer or capping layers is not necessary to achieve high growth or sample longevity, and thus the samples are entirely described by a simple Co(7.5 Å)/Cu(8 Å) bilayer repeated N times. We shall report on a detailed study of this set of samples. Grazing incidence x-ray reflectivity spectra confirmed that the bilayer period, Λ, was 15.5 Å, and varied from sample to sample with a standard deviation of only 0.3 Å. As discussed below, high angle x-ray diffraction was also performed to investigate the crystallographic structure of the samples, using the same x-ray equipment, which provides Cu Kα radiation with a wavelength of 1.541 Å. Previous experiments have shown that the layering quality and interface smoothness is very good in such samples [20]. In the later part of the paper we shall also briefly examine similar samples grown on 100 Å thick Cr buffer layers, and also Ru(14 Å)/{Co(27 Å)/Ru(14 Å)} × N multilayers, prepared under the same conditions.

3. Results and discussion

In figure 1 we show the room temperature and liquid He temperature GMR curves for the {Co/Cu} × 100 sample, measured by a standard four contact dc method. The shape of the curve is close to parabolic around zero field, without any sign of a pointed peak. This indicates good quality AF coupling. This shape is seen in all the samples except those with a very small number of bilayer repeats ($N = 5, 10$). Magneto-optic Kerr effect (MOKE) measurements confirm that the remanence of all but the thinnest multilayers is negligibly small. High resolution cross-sectional transmission electron micrographs of such samples [21] show that the first three or four layers are relatively poorly defined, and polarized neutron reflectometry suggests that they are ferromagnetically coupled [22], and hence contribute little to the GMR. Similar results have been found by another group [23], where the first few layers act as a buffer for the remainder of the sample.

The dependence of the GMR on the number of bilayers, N, was measured for many temperatures between 4.2 and 300 K—we show data for a selection of these temperatures

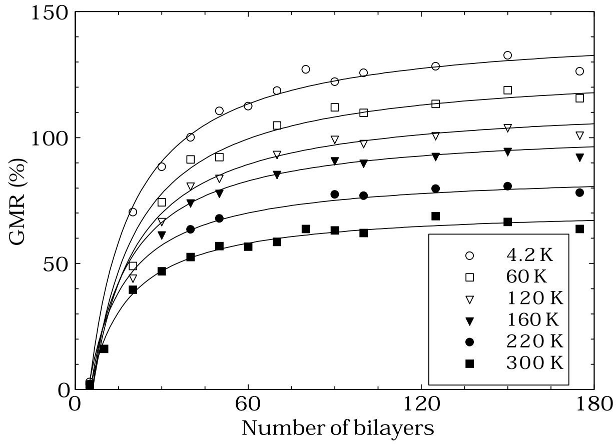

Figure 2. The giant magnetoresistance ratio as a function of number of bilayers N, measured at several temperatures between room temperature and 4.2 K. The data shown are for selected temperatures in the overall range measured. The solid lines represent fits from within the Fuchs–Sondheimer model as described in the text—only the mean free paths ℓ_0 and ℓ_{sat} were free fitting parameters.

across this range in figure 2. The GMR increases rapidly at first, and approaches asymptotically a limiting value for $N \to \infty$. In the Fuchs–Sondheimer approximation in a thin film when one assumes that all boundary scattering will be diffusive the measured effective resistivity ρ^* can be written as

$$\rho^* = \rho\left(1 + \frac{3}{8}\frac{\ell}{t}\right) \tag{1}$$

for a film of thickness t which has bulk scattering leading to a resistivity ρ and mean free path of ℓ. The first of the two terms represents bulk scattering from e.g. defects and phonons, whilst the second term represents additional scattering due to the surfaces. We define the GMR ratio in the usual way as

$$\mathrm{GMR} = \frac{\Delta\rho^*}{\rho^*_{sat}} = \frac{\rho^*_0}{\rho^*_{sat}} - 1. \tag{2}$$

Both the zero field and high field resistivities (ρ^*_0 and ρ^*_{sat} respectively) show finite size effects at any one temperature with characteristic bulk resistivities and mean free paths according to equation (1). The bulk resistivities ρ_0 and ρ_{sat} were determined from straight line fits of $\rho^* t$ against t using the Fuchs–Sondheimer expression of equation (1). These were then used to constrain the fits to equation (2) shown in figure 2 to extract the mean free paths ℓ_0 and ℓ_{sat} at each temperature. (These are the overall mean free paths in the two states, averaged over the spin channels in the appropriate manner.) The GMR for an infinite multilayer is found to be 147% at 4.2 K, dropping to 72% at 300 K. It is surprising that even for multilayers comprising in excess of 100 bilayers the GMR still seems to be limited by finite size effects.

The parameters determined from these fits are shown in figures 3 and 4 for all temperatures measured. (We obtain the same fits, within the error bar, if we only fit to higher N samples where the remanent fraction is smaller than 20%.) Both ρ_0 and ρ_{sat} show typical metallic behaviour (figure 3), increasing linearly with temperature above about 50 K and with a Bloch–Grüneisen type higher power dependence below that temperature. The difference $\Delta\rho$ shows only a weak reduction as the temperature warms. The slightly smaller temperature coefficient of resistance in the antiparallel state can be explained due to the possibility of interband transitions giving

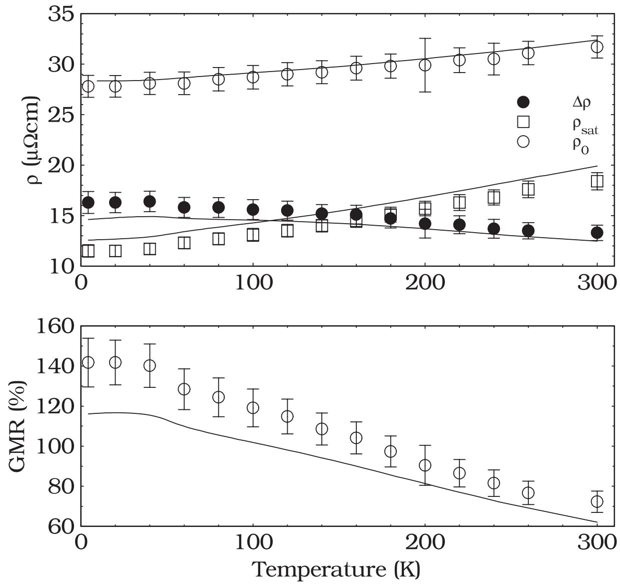

Figure 3. The bulk resistivities determined from the Fuchs–Sondheimer model as a function of temperature. Values for the zero field and saturated configurations of the multilayer are shown. Also shown are the quantities $\Delta\rho$ and the GMR, as defined in the text. The solid lines show data for the $N = 100$ sample, measured as upward sweeps in temperature at fixed field: either $H = 0$ or 20 kOe. All the same trends are reproduced, but $\rho^*_{\rm sat}$ is slightly higher, and hence $\Delta\rho^*$ and the GMR ratio, slightly lower than in the bulk limit determined from the fits. This is to be expected on the basis that there are still weak finite size effects in this multilayer.

rise to extra conductivity channels [15]. The drop in GMR with rising temperature is therefore mostly due to a rising $\rho_{\rm sat}$ in the denominator of the GMR ratio. The implication is that phonon scattering is the most significant of the various possible mechanisms that might reduce the GMR. All these curves look more or less as one might anticipate. The solid lines in the figure show the actual results for fixed-field temperature sweeps for the $N = 100$ sample. The saturated resistivity is a little higher, and the GMR a little lower, than the fitted values due to finite size limitations, but the overall trends can be clearly seen to be reproduced.

On the other hand the apparent behaviour for the mean free paths looks rather odd (figure 4). The mean free path in the saturated state shows a weak temperature dependence, getting slightly shorter as the temperature rises. One would expect this on the basis of a higher inelastic scattering rate as more excitations are introduced thermally into the system. On the other hand the mean free path in the zero field state must get *longer* with rising temperature in order for the model to continue to fit the data. The much larger error bars for ℓ_0 are due the fact that the overall length scale on which the GMR rises is much more sensitive to the longest length scale involved, reflected by a greater sensitivity in the fitting function to $\ell_{\rm sat}$.

It is clear that product of ρ and ℓ at any given temperature is not a constant, as might naively be thought. This is not unexpected after a brief consideration of even the most elementary theory. The well-known Drude expression for the conductivity is $\sigma = \frac{ne^2\tau}{m^*}$, with n the number density of carriers, e the electronic charge, m^* the effective mass and τ the relaxation time. Expressing the mean free path as $\ell = v_{\rm F}\tau$, with $v_{\rm F}$ the Fermi velocity we obtain $\rho\ell = \frac{m^* v_{\rm F}}{ne^2}$. Apart from e all these quantities are derived from the electronic band structure of the material. This is expected to undergo significant changes as we go from parallel to antiparallel alignment of adjacent layer moments [15]. Thermal smearing of the Fermi surface can also destroy

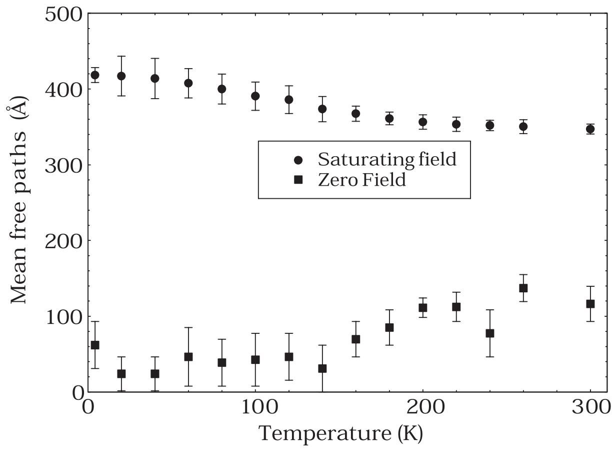

Figure 4. The mean free paths (ℓ_0 and ℓ_{sat}) determined from the Fuchs–Sondheimer model as a function of temperature. Values for the zero field and saturated configurations of the multilayer are shown.

fine features, so it is perhaps not too surprising that $\rho\ell$ should vary somewhat. Hence the temperature dependence of the system at high fields is not a major cause for concern.

The anomalous behaviour of ℓ_0 casts doubt on the whole procedure of analysing this data within a Fuchs–Sondheimer framework. This is in spite of the fact that it correctly describes the finite size dependence of the resistivity at all the temperatures considered, and hence gives remarkably good fits to the data as shown in figure 2 for a fixed temperature. A lengthening mean free path with a rising resistivity must be dismissed as unphysical. The reason for this behaviour can be seen when the presentation of the data in figure 5 is considered. Here we show the same type of data as in figure 2, but for all temperatures measured and in normalized form. It is immediately apparent that the overall length scale on which the GMR rises towards its maximum value is *independent* of temperature. This is contrary to our initial expectations that a cooler system would have longer mean free paths and hence require more bilayers before the GMR is no longer finite size limited. Since the overall length scale on which the GMR rises does not change as the temperature is varied, when one of the Fuchs–Sondheimer mean free paths shortens, the other must get longer in order to stop any overall variation. It is apparent that some sort of scaling of the finite size effects with temperature is occurring in this system. Similar scaling behaviour has also been observed in the Co/Ru multilayer system [24, 25] where weak coupling at the bottom of the stack is not found: even the first trilayer is strongly AF coupled. This is another reason for not ascribing the result in Co/Cu to the AF coupling improving as the multilayer stack height is increasing.

At this point it is worth digressing for a moment to compare the values we obtain for the mean free path with others in the literature. First principles calculations of spin dependent resistivities and GMR by Butler *et al* [26] give values of ∼200 Å for the antiparallel and ∼10 000 Å in the parallel state for Co/Cu multilayers of different thicknesses. This leads to GMR ratios in excess of 2000%. Comparison with our data shows that the estimate of ∼10 $\mu\Omega$ cm extra resistivity in the antiparallel state is in roughly correct, but the neglect of phonon and defect scattering leads to a saturated resistivity orders of magnitude too small, inflating the GMR ratio. Plaskett and McGuire performed a similar study to ours in Co/Cu samples with only weak antiferromagnetic coupling [27], and found mean free paths of 470 Å in the saturated state and 150 Å in the weakly antiparallel state. The $\rho\ell$ product in this work varied by more than a factor of 2. The dependence of GMR on the number of Co/Cu bilayers

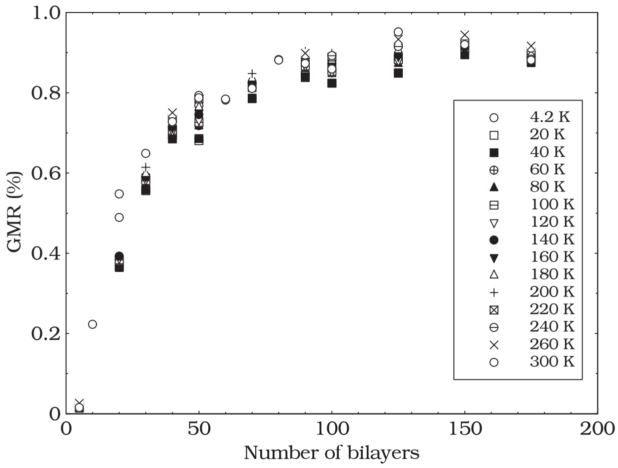

Figure 5. The GMR normalized to the 'bulk' GMR ratio determined from the resistivities presented in figure 3 as function of the number of bilayer repeat units N. The data sets for different temperatures collapse onto a common curve.

was also studied by van den Berg *et al*, who found that the GMR did not saturate until ∼50 bilayer repeats were deposited [28]. They concluded only that the minority mean free path in Co was much greater than the 114 Å used by Dieny for NiFe [8]. Both these experiments estimated the values of ℓ by increasing the size of a multilayer structure which can be thought of as homogeneous on the scale of the mean free path, as we have done. The length scales measured are much longer than the mean free paths which are measured for individual layers in the experiments described in [12, 13] and [14].

An important question that these results beg is to ask what sets the scaling length that controls the behaviour in figure 5. The physical microstructure of the sample is temperature invariant at these low temperatures and this is an obvious place to search for different aspects that will develop in the same way as the GMR with N. We have a performed detailed structural characterization of these samples—and related sets—by means of x-ray scattering and plan-view and cross-sectional transmission electron microscopy. No meaningful correlation between the rising GMR ratio and the N dependence of quantities such as layer roughness, lateral grain size, and mosaic spread could be found [25]. However the out-of-plane (vertical) grain size, determined from the Debye–Scherrer broadening of the crystallographic high angle x-ray diffraction peaks [29] was the only quantity we could find that does show some degree of correlation. This is shown in figure 6, which we will now discuss.

In the top panel (a) of figure 6 we show the 4.2 and 300 K data for our set of Co/Cu multilayers that we have just attempted to analyse in terms of the Fuchs–Sondheimer model. We have found that Cr is a good buffer layer for promoting GMR in Co/Cu multilayers [25], and a room temperature curve for multilayers grown on a 100 Å thick buffer of this material is also shown, scaled to the maximum GMR obtained for these samples. This also falls on the same curve when the 100 Å thick Cr buffer is discounted. We also plot our results for Co/Ru multilayers [24], which also scale in the same way. The solid line is a fit of a rising exponential function $1 - \exp(t/\zeta)$, the length scale ζ for which is found to be 440 ± 10 Å. This choice of fit is purely empirical: the curve fits the data rather well, and gives a convenient measure the length scale.

Full details of our x-ray analysis may be found in [25], here we shall restrict ourselves to giving the most relevant points. In the lower panel (b), we show the scaled vertical coherence

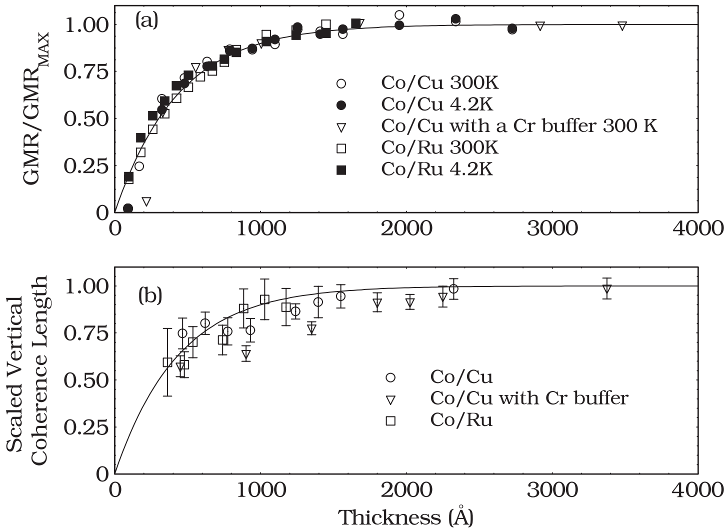

Figure 6. (a) The scaled variation of the GMR in three multilayer systems with total multilayer thickness. The multilayers are {Co(7.5 Å)/Cu(8 Å)} × N, Cr(100 Å)/{Co(7.5 Å)/Cu(8 Å)} × N, and Ru(14 Å)/{Co(27 Å)/Ru(14 Å)} × N. All the data collapse onto a common curve. The solid line is an exponential function $1 - \exp(t/\zeta)$ fitted simultaneously to all the data in the plot, the length scale for which is $\zeta = 440 \pm 10$ Å. (b) The variation of the vertical coherence length ξ with total multilayer thickness for the same three multilayer systems, as determined from the FWHM of Lorentzian squared fits to the (111) high angle x-ray Bragg reflections. The solid line is not a fit, but is the same exponential curve that is shown in panel (a). In both panels we do not include the Cr buffer layer thickness in the total.

length at the principal crystallographic reflection of the sample, found at $2\theta = 43.49°$ for Co/Cu, $2\theta = 43.79°$ for Co/Cu on Cr, and $2\theta = 43.33°$ for Co/Ru. These appear to all be fcc (111) reflections. No other reflections were observed in any case. Good fits to these Bragg peaks were obtained using a Lorentzian squared lineshape, and the full width at half-maximum was used to determine the vertical grain size (coherence length ξ) using the Scherrer formula. This grain size rises with N and tends towards asymptotically ∼200 Å for the Co/Cu multilayers, slightly less (∼150 Å) for the Co/Ru. (Rocking curves give mosaic spreads of ∼20° for Co/Cu and ∼30° for Co/Ru, with a Gaussian distribution of domain directions, and almost no variation with N.) The values of ξ obtained, scaled to the value to which they are asymptotically rising, are what is plotted in panel (b) of figure 6. The solid line is the same exponential that is plotted in panel (a), an empirical fit to the scaled GMR data. It describes the general trend of the $\xi(t)$ data rather well.

4. Conclusions

There are two main conclusions that can be drawn from these results. The first is entirely empirical and is that a large number of bilayers are required to realize a large GMR in Co/Cu multilayers. Those groups which have reported the largest GMR ratios in this system have all used 50 or more bilayer repeats [20, 30, 31]. This is an entirely experimental result. Nevertheless it has important implications for the theory—it shows that the minimum length scale on which the intrinsic GMR of a these multilayers can be discussed is of order hundreds of Å. This casts doubt on the validity of models which assign local properties to each layer. It is better to think of the multilayer in terms of an effective medium for conduction. Other experimental results also indicate that one cannot accurately take a reductionist view of the GMR in terms of the properties of individual layers [32]. Advanced theories are able to reproduce the most salient features of these other experiments [33–35].

Secondly there is a single temperature independent length scale for the GMR, which is revealed by the scaling behaviour seen in figure 5. It is difficult to relate this length scale to the details of a microscopic theory describing a complex multiband system such as the multilayers in question, but our structural analysis suggests that the GMR is related to the vertical coherence length of the crystallographic grain structure in the multilayer, namely the distance that an electron can travel normal to the layers before scattering at a grain boundary. This determines the number of interfaces or layers that an electron can sample in a coherent manner. In our highest GMR multilayers this structural coherence length is $\sim$200 Å (as determined by Bragg reflections of x-rays with a wavelength roughly equal to that of an electron at the Fermi level in a metal). We propose the following interpretation of our results: at all the temperatures measured the spin $\uparrow$ mean free path is truncated by scattering at grain boundaries or other defects, and the inelastic (phonon or magnon) scattering rate never rises to be large enough to be the dominant effect for this spin. This may occur at higher temperatures, but experiments in this regime are complicated by the onset of structural changes in the sample as it anneals [36]. It has been shown that when discussing the spin polarization of the conductivity the appropriate length scale is the elastic mean free path, whilst the resistivity of the sample is also affected by the inelastic scattering [37]. The polarization gives rise to $\Delta\rho$ which is seen to decay only weakly with temperature. However the temperature dependent inelastic scattering will lead to a higher overall resistivity, reducing the GMR. We would argue that the electron wavefunctions, at least for spin $\uparrow$ carriers are phase coherent over an entire grain at all the temperatures examined here. It is perhaps unsurprising that a classical theory using Fuchs–Sondheimer boundary conditions should not give a fully realistic description of such a complex system. Further experimental and theoretical investigations are required to fully characterize the various fundamental length scales in the GMR.

Acknowledgments

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