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1992 J. Phys.: Condens. Matter 4 L495

(http://iopscience.iop.org/0953-8984/4/36/005)

J. Phys.: Condens. Matter 4 (1992) L495–L502. Printed in the UK

LETTER TO THE EDITOR

Interface scattering and the giant magnetoresistance of MBE-grown Co/Cu superlattices

M J Hall†, B J Hickey†, M A Howson†, C Hammond‡, M J Walker†, D G Wright‡, D Greig† and N Wiser†§

† Department of Physics, The University of Leeds, Leeds LS2 9JT, UK
‡ School of Materials, The University of Leeds, Leeds LS2 9JT, UK

Received 30 June 1992

Abstract. A study has been carried out of the effect of interface scattering on the magnitude of the giant magnetoresistance (MR) of MBE-grown Co/Cu superlattices. We find that increasing interface scattering by annealing the Co/Cu superlattice leads to a progressive *decrease* in the magnitude of the MR. In contrast to our results, it was recently reported that annealing *increases* the MR for Fe/Cr superlattices. An explanation is presented in terms of the spin dependence of interface scattering which accounts both for our data for the Co/Cu system, as well as for the opposite results obtained for the Fe/Cr system.

The properties of magnetic multilayers and superlattices are currently the subject of intense investigation, both experimentally [1–28] and theoretically [29–41]. One of the most interesting features of these sytems is that they exhibit a giant magnetoresistance (GMR), of order 10–100%, in comparatively small magnetic fields. Recently, there have been a number of experimental studies [10, 11, 14, 21–28] of the relationship between the properties of the superlattice interfaces and the magnitude of the GMR. These measurements show that for the Fe/Cr system [11, 23, 25], annealing the superlattice significantly *increases* the magnitude of the GMR. Moreover, this result holds regardless of whether the Fe/Cr superlattice is grown by MBE [11] or by sputtering [23] or by UHV evaporation [25].

We have extended our studies to the Co/Cu system. Recently, we reported [26] the first observation of a GMR for MBE-grown Co/Cu superlattices. We have now studied the effect of surface properties on the GMR by annealing our superlattices. We found that successive annealing led to a progressive *decrease* in the magnitude of the GMR, from initial values of about 15–20% to final values of only a few per cent. In contrast to the results obtained for the Fe/Cr system, in no case did annealing the Co/Cu superlattice produce an increase in the GMR.

Our experimental results can be understood in terms of electron scattering at the interface between the magnetic and the non-magnetic layers. Annealing a superlattice increases interfacial scattering, and this scattering is independent of the spin direction of the conduction electrons for the Co/Cu system. We shall see that this implies that

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

the GMR should decrease as a function of annealing, which is in agreement with our data for the Co/Cu superlattices.

Interface scattering also enables one to understand the completely different results that were obtained for Fe/Cr superlattices, for which the GMR was observed to increase upon annealing. For this system, one must take account of the strong spin dependence of electron scattering by Cr impurities in an Fe host [42, 43], which leads to spin-dependent scattering at the interface. Friedel [44] has shown that this spin dependence is caused by resonant scattering due to the virtual bound state of Cr in the d-band of ferromagnetic Fe. This spin-dependent scattering formed the basis for the successful calculation of Zhang, Levy and co-workers [31–34] of the GMR of Fe/Cr superlattices. We propose here that this resonant scattering, present in the Fe/Cr system but absent in the Co/Cu system, also provides the explanation for the opposite signs observed for the change in the magnitude of the GMR upon annealing these two systems.

Our sample was a superlattice of Co(15 Å)/Cu(7 Å) grown in a VG 80M MBE facility in which the base pressure was 3×10^{-11} mbar. The substrate was (110) GaAs which was annealed at 600 °C to achieve the RHEED streaks characteristic of surface reconstruction. A 500 Å buffer layer of Ge was then deposited at 500 °C at a rate of 0.16 Å s^{-1}. The first metallic layer of 15 Å of Co was deposited at 100 °C at a rate of 0.2 Å s^{-1}; it grew as (110) BCC. A 10 Å layer of Au(111) was then grown at a rate of 0.05 Å s^{-1}. The superlattice consisting of 20 bilayers of Co(15 Å) and Cu(7 Å) were then grown on top of this. We had previously found [26] that the use of the Au buffer layer is essential for producing good epitaxial superlattices of Co/Cu.

The sample was cleaved into several pieces, each of which was annealed for one hour at a different temperature. The superlattice was characterized using RHEED *in situ* and x-ray analysis *ex situ*. Typical RHEED patterns for this sample were published in [26]. Low-angle x-ray scans for a sample before and after annealing are shown in figure 1.

The scan for the unannealed sample (curve (a)) is dominated by Kiessig fringes at two different frequencies, with a periodicity of 0.17° and about 4°, respectively. The former corresponds to the overall thickness of the superlattice, whereas the latter is due to the gold cap. These fringes mask the low-angle Bragg peak at around 4°. Curve (b), which corresponds to the sample after annealing at 250 °C, is qualitatively similar to curve (a). This implies that the integrity of the interfaces is maintained upon annealing at 250 °C. The dramatic change in the x-ray scan after annealing the sample at 350 °C (curve (c)) indicates a loss of interfacial integrity due to annealing at the higher temperature.

The resistivity was measured with the current and magnetic field along the GaAs [112] direction since this was the direction along which the sample could easily be cleaved into a long rectangular shape. The magnetoresistance was previously found [26] to be independent of the angle between the current and the magnetic field.

Figure 2 shows the magnetoresistance of the samples for various annealing temperatures. It is seen that the saturation magnetoresistance progressively decreases as the annealing temperature is raised, whereas the functional form and the saturation field remain unchanged.

It is generally agreed that the GMR observed in magnetic superlattices and multilayers results from the antiferromagnetic coupling between neighbouring magnetic layers. In the antiferromagnetic configuration, electron scattering is on average independent of its spin direction as it travels through successive layers. Applying a

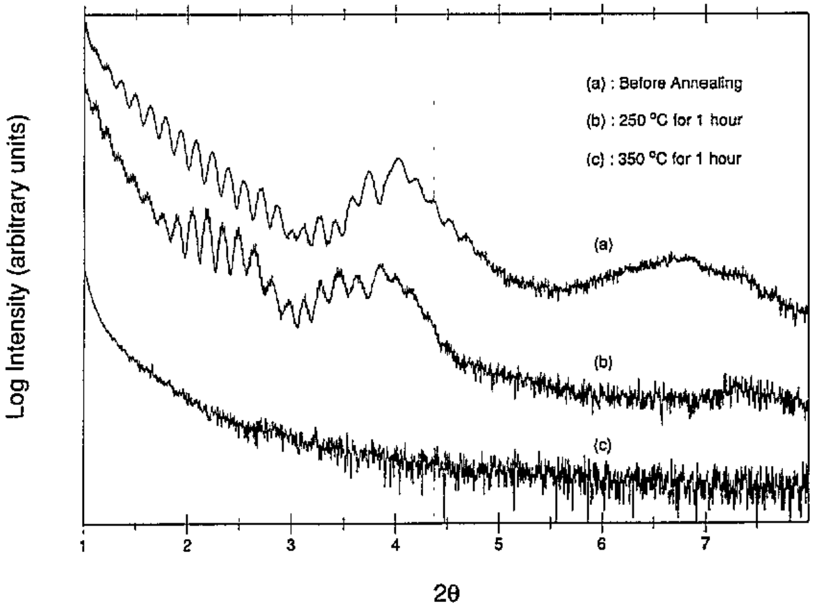

Figure 1. Low-angle x-ray scans for a Co/Cu superlattice at different annealing temperatures.

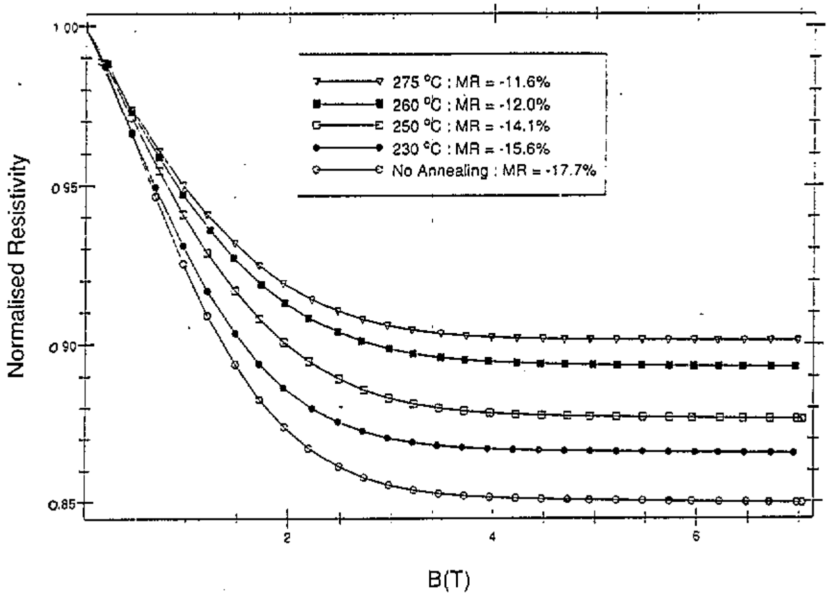

Figure 2. Magnetoresistance at 4.2 K of a Co(15 Å)/Cu(7 Å) superlattice annealed at various temperatures.

saturation magnetic field to align the superlattice into the ferromagnetic configuration then leads to a strong dependence of electron scattering on spin direction (assuming no spin mixing, as is appropriate at low temperatures). It is easy to show [38] that such a system exhibits a GMR whose magnitude depends on the ratio of the scattering probability of the spin-up electrons to that of the spin-down electrons. We have attributed [26] our observed GMR for MBE-grown Co/Cu superlattices to antiferromagnetic coupling between neighbouring Co layers. The large saturation field exhibited in figure 2 is a characteristic signature of such antiferromagnetic coupling. More detailed information about the magnetic structure of the superlattices can be obtained from magnetization measurements. The results of such measurements will be reported in a separate publication.

A theoretical analysis of the GMR in terms of an equivalent network of resistors has been given by Edwards *et al* [36] who emphasized bulk scattering of the electrons in the magnetic host. Based on the marked spin dependence of the d-band density of states at the Fermi energy of the magnetic layers, Edwards, Mathon and co-workers [35–38] have derived a GMR whose magnitude is in good agreement with the largest measured values as a function of layer thickness.

Now consider the scattering of an electron at the interfaces between the Co and the Cu layers of the superlattice. Since electron scattering at the Co/Cu interfaces is 'catastrophic' and independent of the bulk density of states, interface scattering is independent of the direction of the electron spin and hence serves to reduce the magnetoresistance (whose magnitude, divided by the resistance in the saturation magnetic field, will be denoted by MR). The more interface scattering, the smaller the value of MR. Because annealing the Co/Cu superlattice *increases* the proportion of interface scattering relative to bulk scattering, it follows that annealing should *decrease* MR. This is indeed what we observe for Co/Cu superlattices (see figure 2).

These ideas can be made quantitative by including interface scattering in the resistor network model of Edwards *et al* [36]. (An excellent review of this model has been given by Mathon [38].) One adds additional 'resistors' in the network for each spin direction to represent interface scattering, and then repeats the Edwards *et al* calculation of MR. The details will be reserved for a separate publication, and we report here only the final result:

$$\mathrm{MR} = \frac{\left[\frac{1}{2}p_\mathrm{b}\beta_\mathrm{b}(\alpha_\mathrm{b}-1) + p_\mathrm{i}(1+r)(\alpha_\mathrm{i}-1)\right]^2}{\left[p_\mathrm{b}\beta_\mathrm{b}(\alpha_\mathrm{b}+r/\beta_\mathrm{b}) + 2p_\mathrm{i}(1+r)(\alpha_\mathrm{i}+1)\right]\left[p_\mathrm{b}\beta_\mathrm{b}(1+r/\beta_\mathrm{b}) + 4p_\mathrm{i}(1+r)\right]} \tag{1}$$

where r is the ratio of the thicknesses of the Cu and the Co layers, β_b equals unity for the Co/Cu system [36, 38], and p_b and p_i represent the proportion of the electron scattering that takes place in the bulk and at the interfaces, respectively ($p_\mathrm{b} + p_\mathrm{i} = 1$).

The key quantities in (1) are α_b and α_i which denote, respectively, the ratio of spin-down electron scattering to spin-up electron scattering in the bulk (subscript $_\mathrm{b}$) of the magnetic layers and at the interfaces (subscript $_\mathrm{i}$). For the Co/Cu system, Edwards *et al* find that the value $\alpha_\mathrm{b} = 8$ yields good agreement with the GMR data and that this value accurately reflects the strong spin dependence of the density of states in ferromagnetic Co. Since interface scattering is independent of the direction of the electron spin, $\alpha_\mathrm{i} = 1$.

If we ignore interface scattering in (1) by setting $p_\mathrm{i} = 0$ and $p_\mathrm{b} = 1$, we recover the result [45] of Edwards *et al* [36]. The only free parameter in (1) is p_b, whose value we have chosen to make MR equal to the measured magnetoresistance for the unannealed superlattice, which is represented by the open circle in figure 3.

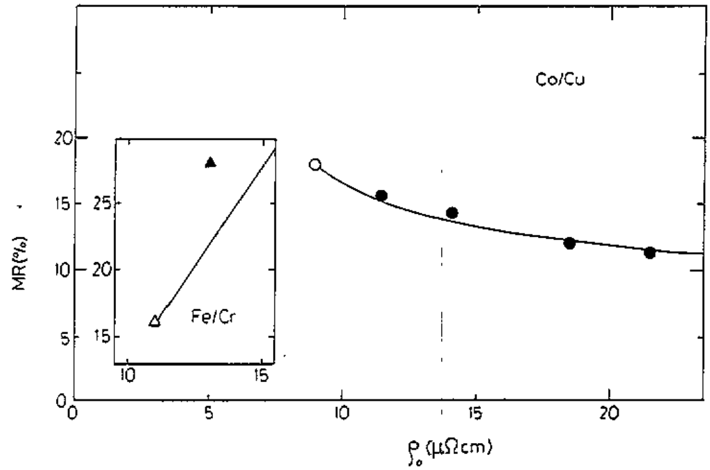

Figure 3. The value MR of the normalized magnetoresistance as a function of the residual resistivity ρ_0 upon annealing a Co/Cu superlattice and an Fe/Cr superlattice (in the inset). The open and full symbols represent the data for the unannealed and for the annealed superlattice, respectively. The curves have been calculated according to equation (1).

The full circles in figure 3 represent the values of MR and the resistivity ρ_0 measured after annealing the Co/Cu superlattice for one hour at 230 °C, at 250 °C, at 260 °C and at 275 °C. Annealing the superlattice has two effects. First, it increases the overall electron scattering which increases ρ_0. Second, annealing increases the ratio of interface scattering to bulk scattering, which increases p_i at the expense of p_b. Since $\alpha_i < \alpha_b$ for the Co/Cu system, increasing p_i *decreases* the overall spin dependence of electron scattering and hence *decreases* the value of MR. Therefore, the curve in figure 3, calculated according to (1), shows a monotonic decrease in MR as a function of ρ_0, in agreement with the data.

The data of figure 3 refer to superlattices that have undergone 'gentle' annealing, with the annealing temperature below 280 °C. At such low annealing temperatures, the structural integrity of the interface is maintained, and the effect of annealing is to cause diffusion of the Cu atoms into the neighbouring Co layer and vice versa. Part of this diffusion is restricted to the interface and part penetrates into the bulk. This distinction is not completely clear-cut, but it seems reasonable to assume that most of the additional electron scattering caused by annealing is due to interface scattering.

Thus far, we have been discussing the Co/Cu system. The Fe/Cr system is very different because of a phenomenon that is not present in Co/Cu superlattices. This is the strong spin-dependent resonant scattering of electrons in ferromagnetic Fe caused by Cr impurities in the Fe layer [44]. The contribution of this spin-dependent interface scattering to the GMR has been analysed in detail by Zhang, Levy and co-workers [31–34] who find that a value [46] of $\alpha_i = 12$ gives good agreement with the GMR data for Fe/Cr superlattices.

To obtain the bulk value, α_b, we turn to measurements [42, 43] of the spin dependence of electron scattering by Cr impurities in bulk ferromagnetic Fe. These measurements yield $\alpha_b = 2.7$ according to Dorleijn and Miedema [42] or $\alpha_b = 6$ according to Fert and Campbell [43]. Regardless of which experimental value for α_b

proves to be more reliable, one sees that $\alpha_i > \alpha_b$. This implies that *interface scattering* is the major source of spin-dependent electron scattering for the Fe/Cr system, as has previously been emphasized [15, 31–34].

We now consider the effect of annealing an Fe/Cr superlattice. As already noted, annealing the superlattice increases the ratio of interface scattering to bulk scattering. Therefore, for the Fe/Cr system, annealing should *increase* the spin-dependent scattering, implying an *increase* in the magnitude of MR. This has been observed [11] for Fe/Cr superlattices.

These ideas regarding the Fe/Cr superlattice can be made quantitative by again referring to equation (1). Unfortunately, not all the material parameters of the Fe/Cr system are accurately known. However, one can show that agreement with experiment is possible by inserting into (1) the most favourable values of the material parameters within the range of the experimental options. This yields the curve shown in the inset of figure 3, where the open and full triangles represent the data [11] for the unannealed and annealed Fe/Cr superlattice, respectively. The agreement between theory and experiment appears satisfactory.

We wish to emphasize that the most significant result of our work is not the agreement between the calculation and the data shown in figure 3. Rather, our main point is that the *same* expression (1) for MR yields a *decrease* in MR upon annealing a Co/Cu superlattice but an *increase* upon annealing an Fe/Cr superlattice (if one includes the resonant spin-dependent interfacial electron scattering in Fe/Cr). This is the experimental 'anomaly' that required explanation.

Finally, we consider 'vigorous' annealing. When the annealing of the superlattice is carried out at a sufficiently high temperature, the structural integrity of the interface is undermined, leading to a sharp increase of spin-independent scattering. As already discussed, this is confirmed by the x-ray scans displayed in figure 1. For the Co/Cu superlattices, we indeed found that 'vigorous' annealing (at higher temperatures) produced a more rapid decrease in MR with increasing ρ_0 than did 'gentle' annealing (at lower temperatures).

For the Fe/Cr system, this phenomenon is particularly interesting, for the following reason. Equation (1) predicts that gentle annealing should initially lead to an *increase* in MR, but that vigorous annealing will eventually lead to a *decrease* in MR. These results are in accord with the data [11, 23, 25].

In conclusion, we find that annealing the superlattice is a useful tool for studying the effect of interface scattering on the magnitude of the GMR. In particular, the presence (for Fe/Cr) or absence (for Co/Cu) of spin-dependent electron scattering at the interfaces provides the explanation both for the initial increase in the value of MR upon annealing an Fe/Cr superlattice, as well as the decrease in MR upon annealing a Co/Cu superlattice.

We are greatly indebted to both the Magnetism and Magnetic Materials Initiative of the SERC and to the University of Leeds for their strong support of the MBE project. The skilled technical assistance provided by A A M Croxon and J Turton is also greatly appreciated.

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[45] Our α corresponds to their ratio α/β in equation (5) of [36] and equation (19) of [38]. We have

made this change in notation to facilitate the discussion of the Fe/Cr superlattices.

[46] The value they choose for their parameter $p = 0.55$ corresponds to $\alpha = 12$, according to equation (19) of [34].