An alternate route to giant magnetoresistance in MBE-grown Co–Cu superlattices (invited)

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Co–Cu superlattices grown by MBE in the (111) orientation show weak or nonexistent interlayer exchange coupling, yet several groups have observed large high-field magnetoresistance signals in excess of 30%. In the present work, we address some of the questions relating to GMR and the interlayer coupling by studying samples with atomically abrupt interfaces, as probed by real-time RHEED techniques, HRTEM, and spin-echo NMR. We propose that the lowered dimensionality of the structure leads to an enhancement of the scattering of conduction electrons from paramagnetic interfaces obeying a Langevin-like saturation at very high fields, well beyond the switching field of the Co layers. Scaling between the GMR and thermopower measurements suggests that a spin-dependent density of states at the Co–Cu interfaces is responsible for the observed magnetotransport behavior in these samples, rather than antiferromagnetically coupled Co layers.

I. INTRODUCTION

The magnetotransport behavior of (111)-oriented Co–Cu multilayers grown by molecular beam epitaxy (MBE) has recently generated some controversy. A much debated issue concerns the appearance of giant magnetoresistance (GMR) in samples that show little[1–4] or no[5,6] evidence of antiferromagnetic (AFM) interlayer coupling. The latter is a well-established mechanism for GMR in magnetic multilayers *via* spin-dependent scattering; it is characterized by a sharp "knee" in the resistance versus field curve at the flip field of the counteraligned spins.[7]

A number of authors have put forward sample defects as a possible explanation why AFM may be masked in the Co–Cu (111) system. For example, it has been suggested that stacking faults[8] and pinholes[9] may lead to ferromagnetic bridging across neighboring Co layers. However, ferromagnetic bridging, by definition, suppresses AFM coupling, and it should suppress spin-dependent GMR effects as well. While no definitive evidence has yet been presented for such a mechanism, the scanning tunneling micrographs of de la Figuera *et al.*,[10] showing a tendency for island nucleation, graphically illustrate the need for careful growth studies as well as atomic-scale structural characterization.

In this paper we present an alternative description of the GMR in (111) Co–Cu MBE samples. Our approach is based on a comparison of magnetotransport and magnetization measurements on a series of carefully grown superlattices with atomically smooth interfaces. We observe what appears to be a new type of GMR, one that is not dependent on AFM coupling and is operative to very high magnetic fields. In the limit of atomically smooth interfaces, our results suggest that the lowered dimensionality of the magnetic/nonmagnetic interfaces in our samples is the key factor in the observed magnetotransport behavior rather than sample defects.

II. SAMPLE GROWTH AND CHARACTERIZATION

The samples in this research were grown by MBE on Ge-buffered (110) GaAs substrates. Buffer layers of 15 Å (110) bcc Co, followed by 20 Å (111) Au, were deposited on the Ge to initiate layer-by-layer superlattice growth in the (111) orientation. The superlattice samples typically consist of 30 bilayers. The pressure during superlattice growth was $<4\times10^{-10}$ mbar, and the substrate temperature was held at 150 °C. Co was deposited from an electron beam hearth at rates between 0.15 and 0.25 Å/s, and Cu from a Knudsen cell at a rate of 0.33 Å/s. The thickness variations of individual layers of Co and Cu were controlled to about 0.5 ML. Details of the growth are described in a previous publication.[11] The growth was monitored *in situ* by reflection high energy electron diffraction (RHEED) using a CCD imaging and analysis system.[12] X-ray scattering performed after growth confirmed that the layer stacking was fcc in the (111) orientation.[11]

A crucial aspect of the interface characterization involved spin-echo NMR measurements of the local cobalt environment.[13] Only two characteristic NMR peaks were observed, one of which corresponds to bulk fcc Co with 12 Co neighbors and the other to interfacial Co having three Cu neighbors (see Fig. 1). The results will be described in more

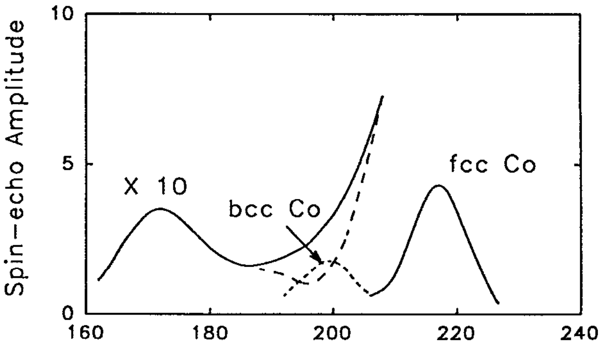

FIG. 1. Co spin-echo NMR spectra[14] at $T=2$ K in a Co–Cu superlattice $[\mathrm{Co}(7.5\ \mathrm{ML})/\mathrm{Cu}(3\ \mathrm{ML})]_{30}$. The interface peak has been magnified by a factor of 10. The dashed line shows the contribution of the 15 Å bcc Co buffer layer.

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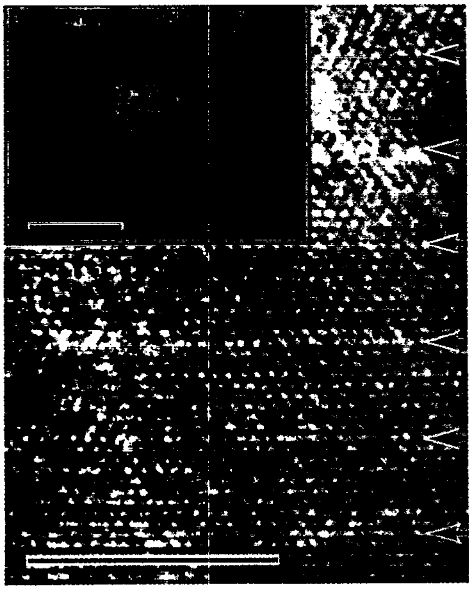

FIG. 2. Typical cross-sectional HRTEM micrograph along a [110] azimuth of $[\mathrm{Co}(7\ \mathrm{ML})/\mathrm{Cu}(3\ \mathrm{ML})]_{40}$. The variation in contrast along the growth direction, which is obtained by defocusing the image, corresponds to interference fringes due to the superlattice periodicity, as indicated by the arrows. Note that the defocused image shown here still exhibits atomic resolution, indicating high crystal coherence. The horizontal bar corresponds to 50 Å. Inset: the HRTEM image over a larger area. The horizontal bar corresponds to 100 Å. The straight horizontal contrast bands indicate flat superlattice layers.

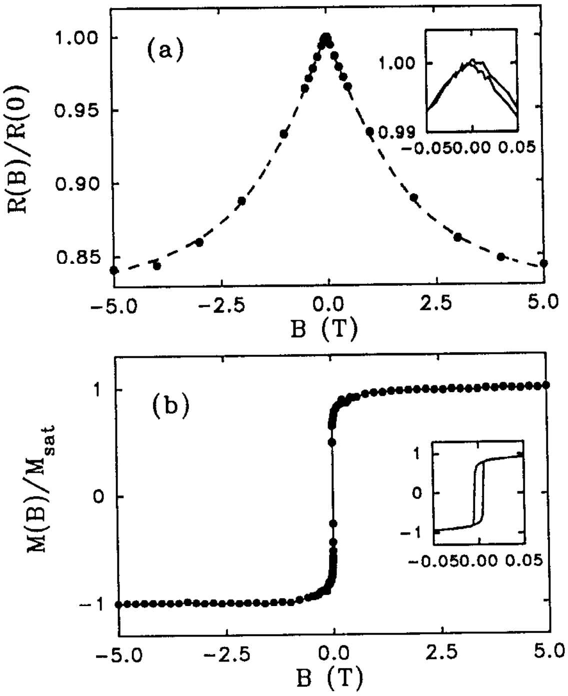

FIG. 3. (a) Ambient temperature magnetoresistance, $R(B)/R(0)$, vs field (Tesla) for a $[\mathrm{Co}(7.5\ \mathrm{ML})/\mathrm{Cu}(5.5\ \mathrm{ML})]_{26}$ superlattice. The dashed line is a theoretical fit to the Langevin function described in the text, where $N=480$ and $\beta=0.194$. Inset: low-field MR vs field dependence. (b) Magnetization curve for same sample. $M_{\mathrm{sat}}=1400\ \mathrm{emu/cm}^3$. Inset: low-field magnetization curve. $T=300$ K.

detail in a separate publication.[14] They provide two important insights on the microstructure of our MBE samples. First, a comparison of the intensities of the "bulk" and "interface" NMR peaks shows that a significant area of the Co–Cu interfaces is atomically abrupt. Second, the absence of an hcp stacking "shoulder"[13] on the high frequency side of the bulk fcc peak at 217 MHz places a lower limit on the stacking coherence of our samples. Coherent fcc stacking over at least 200 Å is inferred from these measurements. The NMR results therefore indicate a low density of stacking faults. This is contrary to the island-growth mechanism presented by Gradmann *et al.*,[8] where one would expect a large concentration of stacking faults.

With reference to the STM results[10] mentioned above, where Co growth was performed at an ambient temperature on a Cu(111) single crystal surface, we note some important differences with the growth of our samples. The main difference is that in our samples the lattice mismatch (~1.8%) is shared between the (thin) Cu layers and the Co layers. Neither layer fully relaxes during growth, so that coherent layer-by-layer deposition is promoted.[15] Also, the substrate temperature in our case is elevated in order to enhance the surface diffusion of the incident atoms. We find much improved growth compared to ambient temperature conditions.

The high quality of our MBE samples is also illustrated by cross-sectional high-resolution transmission electron microscopy (HRTEM) (see Fig. 2). Note that the atomic layers are coherent over several hundred Å, which again is consistent with our x-ray diffraction and RHEED experiments. The flatness and continuity of the layers in this micrograph does not support the view that the growth of (111)-oriented samples is particularly prone to pinhole formation.

The MR measurements were made using the standard dc four-point probe technique with the field applied parallel to the current (longitudinal MR). The magnetization was measured in a commercial SQUID magnetometer and the measured saturation moments are within 10% of the bulk value for Co. Both the magnetization and MR measurements were made for fields applied in the growth plane along the [110] and $[11\bar{2}]$ directions of the superlattice.[16]

III. RESULTS AND DISCUSSION

A. Comparison of magnetization and magnetoresistance

Figure 3 compares the MR for a $[\mathrm{Co}(7.5\ \mathrm{ML})/\mathrm{Cu}(5.5\ \mathrm{ML})]_{26}$ superlattice with its magnetization for fields applied along the $[11\bar{2}]$ direction. Immediately it is apparent that the magnetization is largely saturated by 500 Oe, whereas the MR is still changing significantly at 50 000 Oe, the highest field we can achieve in our cryostat. Moreover, the magnetization is clearly ferromagnetic in character, with no evidence for a flip-field in the MR curve, such as is seen in the case of AFM-coupled Fe–Cr.[7] In what follows we will provide an alternative explanation that resolves the discrepancy between the different saturation behaviors of the MR and the bulk magnetization.

The field dependence of the MR shown in Fig. 3(a) can be described accurately by a Langevin-like saturation func-

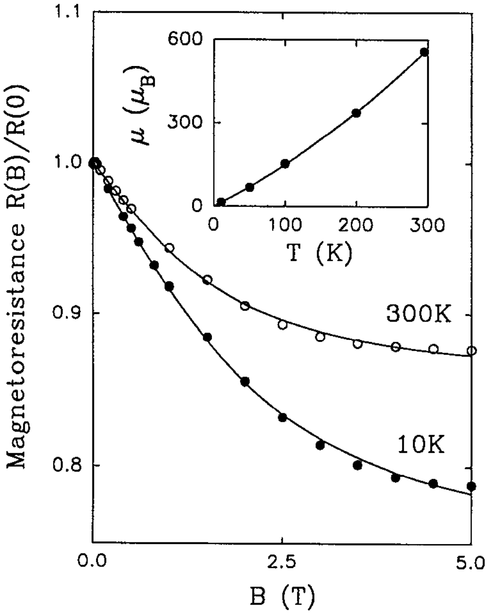

FIG. 4. Field dependence of MR at two different temperatures for [Co(7.5 ML)/Cu(5.5 ML)]$_{30}$. The solid lines are fits to the Langevin function described in the text. Inset: the number of correlated spins as a function of temperature.

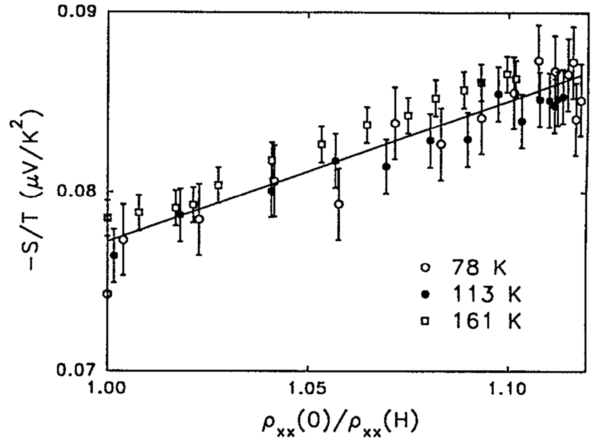

FIG. 5. Scaling behavior of magnetothermopower and magnetoresistance at various temperatures for [Co$_{15\,Å}$/Cu$_{10\,Å}$]$_{30}$.

tion, 1-β(coth $\alpha - 1/\alpha$), where $\alpha = N\mu_0 B/k_B T$, β, and N are fitting parameters and μ_0 is the Bohr magneton. The dashed line in Fig. 3(a) is a fit to the experimental data using the Langevin function. This specific field dependence suggests that scattering from an assemblage of paramagnetic spins, mostly likely at the interfaces between Co and Cu layers, is responsible for the GMR in our samples. We note here that the Cu conduction electron band becomes partially polarized in proximity to the Co layers.[17] Interfacial "loose" spins have also been proposed in order to explain the origin of biquadratic coupling.[18]

It is interesting to point out here that the character of the paramagnetism is not that of isolated spins; rather, we find that there are substantial correlations, akin to a superparamagnetic layer. We envision the paramagnetic spin arrangement as forming small patches of correlated spins of size $\sqrt{N}$. The values of N returned by the Langevin fit at ambient temperature are roughly 500, falling approximately linearly toward unity at $T=0$, as shown in Fig. 4. The enhanced exchange interactions between the interfacial spins and the ferromagnetic Co spins at low temperatures perhaps give rise to the observed decrease of N. We note in passing that these findings make contact with observations of GMR in annealed granular Co–Cu films.[19] In these systems the field dependence of the GMR is also Langevin-like, presumably reflecting the important role of interface states in these systems as well.

The field-dependent MR results discussed here point to a new mechanism for GMR that depends on the scattering associated with interfacial magnetic states, not the ferromagnetic spins in the Co layers, since the Co layers are already fully saturated at low fields. The traditional spin-dependent scattering mechanism, as it has been discussed previously in the context of AFM coupling, is not present in the results discussed here, since it requires one-to-one correlation between MR and magnetization of the magnetic layer.

In light of the above discussion, what are the consequences for interlayer coupling? Due to enhanced scattering by the interfacial paramagnetic layers, the exchange interactions between the neighboring magnetic Co layers are significantly weakened. Our results also reveal that atomically "rougher" interfaces (i.e., samples showing broadened RHEED streaks indicative of short in-plane coherence) have decreased values of the high-field MR. These studies are preliminary at this point, and a fuller account will be reported in a subsequent publication. In the context of roughness, one can envisage regions of the sample that have smooth terraces, together with regions where islands have nucleated (e.g., Stranski–Krastanov growth). This may explain the observation of mixed coupling in recent polarized neutron scattering measurements[4] on (111) Co–Cu superlattices grown on sapphire.

B. Magnetothermopower measurements

In order to shed further light on the origin of the GMR in these superlattices, we have carried out a study of the magnetothermopower as a probe of spin-dependent density of states effects.[20] While the MR shows no apparent dependence on the Co magnetization (see Fig. 3), our measurements reveal a clear scaling relationship with the thermopower at temperatures where diffusion thermopower is dominant ($T>50$ K). Figure 5 illustrates the scaling, which is of the form $S(H)/T \sim \rho_{xx}(0)/\rho_{xx}(H)$. Scattering of conduction electrons due to the spin-dependent density of states of the unfilled d bands at the Fermi level leads to precisely this form of scaling.[20] A more detailed account of this work is presented in a separate publication.[21]

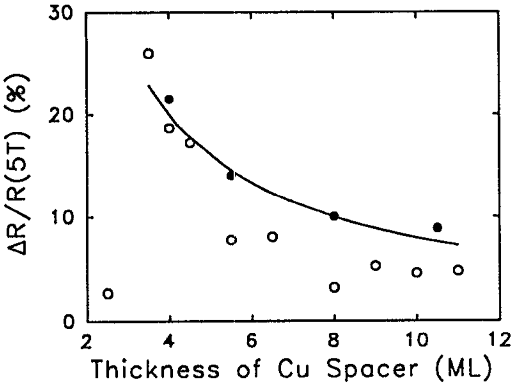

FIG. 6. Spacer layer thickness dependence of MR for Co–Cu (111) superlattices (●, this work; ○, Hall *et al.*[22]). $T=5$ K. The solid line is a fit to the $1/t_{\mathrm{Cu}}$ dependence discussed in the text.

C. Dependence of GMR on Cu layer thickness

Oscillations in the GMR as a function of nonmagnetic spacer layer thickness are often taken as a signature of antiferromagnetic exchange coupling. In the present work we have argued that the GMR in our samples originates not from AFM coupling, but from interfacial states that are spin dependent. We should not then expect to see oscillatory GMR behavior, but rather a monotonic decrease of the spin-dependent interface scattering contribution as the spacer layer thickness increases. This is precisely what we observe. Our MR data plotted in Fig. 6, along with previously published results[22] on MBE samples also grown on (110) GaAs substrates, show a $1/t_{\mathrm{Cu}}$ thickness dependence consistent with the dominant role of the interfaces in these samples. It is interesting that the monotonic increase of MR is truncated abruptly at a spacer layer thickness of approximately 5 Å (~2 ML). This thickness corresponds to a layer structure in which each Cu atomic layer is in atomically close proximity to Co. In this case, the MR falls to normal bulk-like values, suggesting that the spin-dependent density of states effects are quenched in the limit of very thin spacer layers.

IV. SUMMARY

We have demonstrated what we believe is a new type of high field MR mechanism, which results from scattering of conduction electrons from paramagnetic Co–Cu interfaces. Our observations call for a more thorough treatment of the band offsets at the interfaces, including $s-d$ hybridization, and for a better understanding of the interfacial scattering of the conduction electrons. We hope that our results will stimulate additional theoretical work in this area.

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