PHYSICAL REVIEW B VOLUME 49, NUMBER 2 1 JANUARY 1994-II

Langevin-like giant magnetoresistance in Co-Cu superlattices

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(Received 10 August 1993)

We present evidence for a new type of giant magnetoresistance in (111) cobalt-copper superlattices with atomically smooth interfaces. 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. The findings help to explain similarities in magnetotransport behavior with recently reported granular systems as well as differences with antiferromagnetically coupled multilayers.

Several groups[1–3] have reported giant magnetoresistance (GMR) effects in Co-Cu(111) superlattices grown by molecular-beam epitaxy (MBE) that are comparable to those observed in antiferromagnetically coupled Fe-Cr multilayers.[4] Whether the magnetoresistance (MR) behavior observed in Co-Cu(111) samples also originates from antiferromagnetic (AFM) coupling is somewhat unclear at this point because their magnetization seems to be predominantly ferromagnetic in character with only a small fraction of the sample showing indications of AFM coupling.[1,5] In another recent experiment[6] AFM coupling was not observed in Co-Cu(111) multilayers grown on Cu single-crystal substrates.

Sample defects have been invoked as a possible explanation of why AFM coupling may be masked in the Co-Cu(111) system. For example, it has been suggested that stacking faults[7] and pinholes[8] may lead to ferromagnetic bridging across neighboring layers. Well-controlled sample growth and detailed atomic-scale characterization are therefore crucial to understanding the magnetic behavior of these materials.

In this paper we present MR and magnetization results on (111) single-crystal Co-Cu superlattices, prepared by MBE techniques with atomically smooth interfaces. We observe the appearance of a new type of GMR, one which is not dependent on AFM coupling and is operative up to high magnetic fields. By careful control of the interfacial quality, and consequently the uniformity of the layering, we probe the role of the interfaces. In the limit of atomically smooth interfaces, our results suggest that the lowered dimensionality of the interfaces dominates the behavior rather than sample defects.

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 subsequent 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/sec, and Cu from a Knudsen cell at a rate of 0.33 Å/sec. Details of the growth are described in a previous publication.[9] The growth was monitored *in situ* by reflection high-energy electron diffraction (RHEED) using a charge-coupled device imaging and analysis system.[10] X-ray scattering performed after growth confirmed that the layer stacking was fcc in the (111) orientation.[9]

A crucial aspect of the interface characterization involved spin-echo NMR measurements of the local cobalt environment.[11] 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 3 Cu neighbors. The measured spectra therefore provide definitive evidence for atomically abrupt interfaces and pure fcc stacking in these samples. These results will be described in a subsequent publication.[12] Cross-sectional high-resolution-transmission-electron-microscopy (HRTEM) experiments were also carried out for the samples studied here. The results reveal that the superlattice interfaces are atomically flat with lateral crystal coherence length of several hundred Å, as illustrated by the micrograph shown in Fig. 1. Owing to the lack of contrast between Co and Cu atoms, the HRTEM images were deliberately defocused in order to show interference fringes due to the superlattice bilayers, as shown in the horizontal bands in Fig. 1. The flatness and continuity of these bands do not support the view that the growth of (111)-oriented samples is particularly prone to pinhole[8] formation.

We now focus on the GMR and magnetization data. 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 superconducting quantum interference device magnetometer. The measured saturation moments are within 5% of the bulk value for Co (1440 emu/cm^3). Both the magnetization and MR measurements were made for fields applied in the growth plane along the [110] and [112] directions of the superlattice.[13] Figure 2 compares the MR for a [Co(7.5 monolayer (ML))/Cu(5.5 ML)]$_{26}$ superlattice with its magnetization for fields applied along the [112] direction. Immediately it is apparent that the magnetization saturation field is more than 100 times smaller than that of the MR. In fact the MR is still changing significantly at the highest field we can achieve in our cryostat (5 T). Some

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authors have ascribed this high-field behavior of the MR to very strong AFM coupling.[1-3] In what follows we will provide an alternative explanation which resolves the discrepancy between the different saturation behaviors of the MR and the bulk magnetization.

The field dependence of the MR shown in Fig. 2(a) can be described accurately by a Langevin-like saturation function, $1-\beta[\coth\alpha - 1/\alpha]$, 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. 2(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, most likely at the interfaces between Co and Cu layers, is responsible for the GMR in our samples. We note the possibility that the Cu conduction electron band could itself become partially polarized in proximity to the Co layers.[14] Interfacial "loose" spins have also been proposed in order to explain the origin of biquadratic coupling.[15]

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 towards unity at $T=0$, presumably as more and more of the interface spins lock into the (saturated) bulk Co magnetization, as shown in Fig. 3. 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. The value of β, corrected for electron-phonon scattering, is practically temperature independent showing that the temperature dependence is controlled by coupling to the bulk ferromagnetic moment. We note in passing that these findings make contact with recent observations of GMR in annealed granular Co-Cu films.[16] In these systems the field dependence of the GMR is also Langevin-like.

The field-dependent MR results discussed here point to a new mechanism for GMR that depends on the magnetism of the interfaces, not the ferromagnetic spins in the Co layers, since the Co layers are already fully saturated at low fields. GMR based on AFM coupling[4,17] is not present in the results discussed here, since it requires one-to-one correlation between MR and magnetization. In the sputtered samples, which typically have somewhat diffuse interfaces, it describes the MR rather well. Consequently, the MR curve for sputtered samples exhibits clear saturation that correlates with the bulk magnetization.[18]

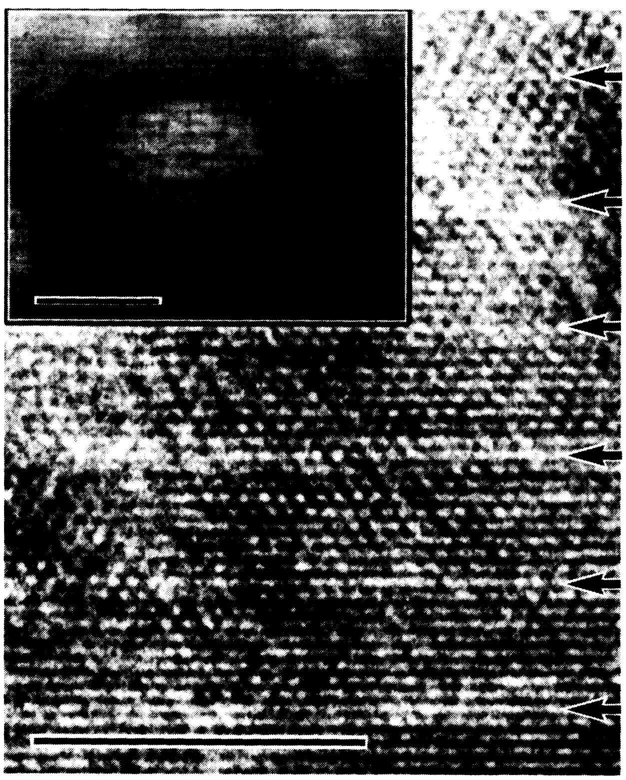

FIG. 1. Typical cross-sectional HRTEM micrograph along [110] azimuth of $[Co(7\ ML)/Cu(3\ ML)]_{40}$. The variation in contrast along the growth direction, which is obtained by defocusing the image, corresponds to interference fringes due to the superlattice periodicity, as indicated by the arrows. Note that the defocused image shown here still exhibits atomic resolution indicating high crystal coherence. The horizontal bar corresponds to 50 Å. Inset: HRTEM image over a larger area. The horizontal bar corresponds to 100 Å. The straight horizontal contrast bands indicate flat superlattice layers.

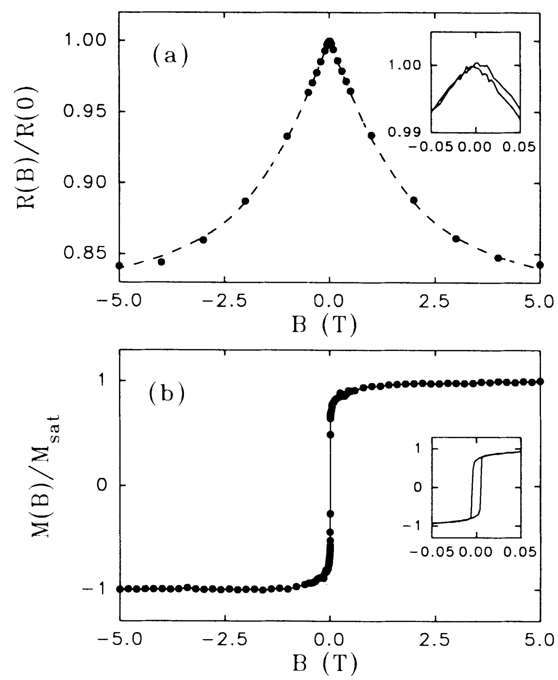

FIG. 2. (a) Ambient temperature magnetoresistance, $R(B)/R(0)$, vs field (Tesla) for a $[Co(7.5\ ML)/Cu(5.5\ ML)]_{26}$ superlattice. The dashed line is a theoretical fit to the Langevin function described in the text, where $N=480$ and $\beta=0.194$. Inset: low-field MR vs field dependence. (b) Magnetization curve for same sample. $M_{sat}=1400\ emu/cm^3$. Inset: Low-field magnetization curve. $T=300$ K.

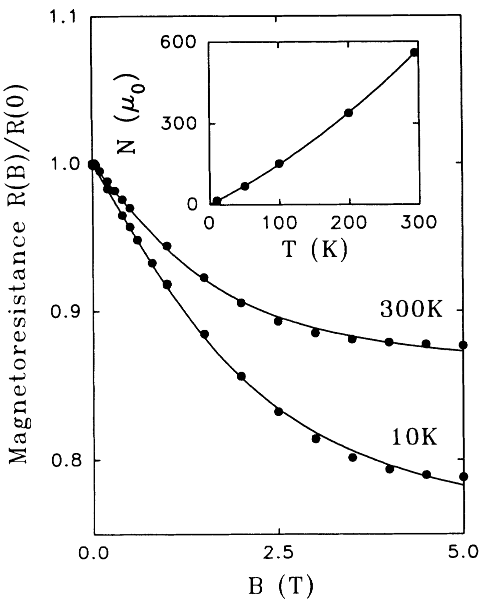

FIG. 3. Field dependence of MR at two different temperatures for [Co(7.5 ML)/Cu(5.5 ML)]$_{30}$. The solid lines are fits to the Langevin function described in the text. Inset: Number of correlated spins as a function of temperature.

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 interfacial roughness, as indicated by modulated RHEED patterns, gives rise to a less prevalent high-field Langevin-like MR. One can envisage regions of the sample which 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[5] on (111) Co-Cu superlattices grown on sapphire. Magnetic neutron scattering on our samples has so far revealed no evidence for coupling, consistent with the mechanism described above. We believe the presence of AFM coupling and the absence of high-field MR in sputtered films are the result of atomically rough interfaces. The observed GMR in such samples arises from antiferromagneticaly coupled layers.

In summary we have demonstrated what we believe is a different 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 spin-polarized interfacial electronic states. We hope that our results will stimulate additional theoretical work in this area.

We acknowledge useful discussions with R. Merlin, A. Ehrlich, D. M. Edwards, J. Mathon, and W. Pratt. We also acknowledge the HRTEM assistance provided by B. Demczyk and J. Mansfield of the University of Michigan Electron Microbeam Analysis Laboratory. Funding for this work was provided by U.S. ONR Grant No. N00014-92-J-1335.

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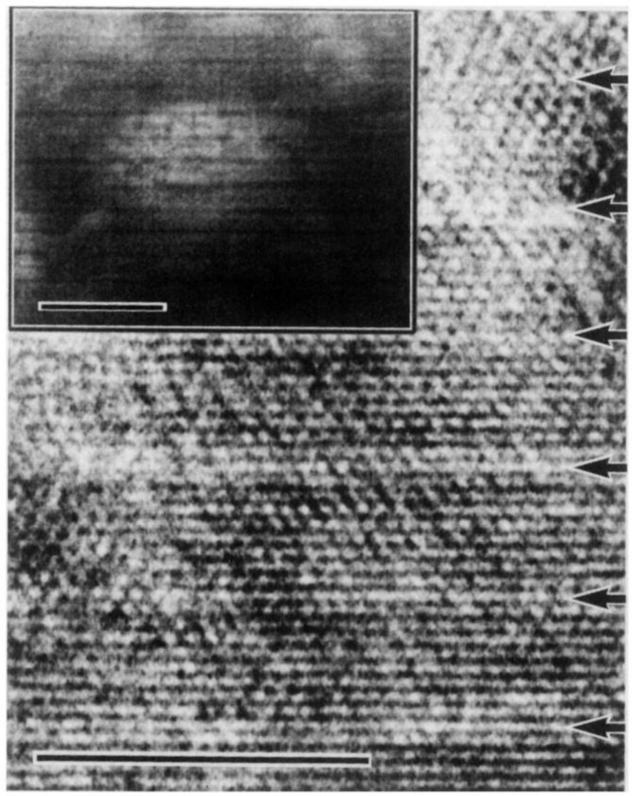

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