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Dependence of giant magnetoresistance on Cu-layer thickness in Co/Cu multilayers: A simple dilution effect

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The dependence of the giant magnetoresistance in Co/Cu multilayers on Cu spacer layer thickness is shown to be surprisingly straightforward for multilayers comprised of thin Co layers. At 4.2 K the magnetoresistance decays simply as the inverse Cu spacer layer thickness, which we consider to be a result of *dilution* of the Co/Cu interfacial regions which give rise to the giant magnetoresistance effect. At 295 K there is an additional exponential decay whose decay length we attribute to volume scattering within the Cu layers. High-resolution cross-section transmission electron micrographs show a high degree of structural ordering within the Cu layers and across the Co/Cu interfaces, perhaps accounting for the long volume scattering lengths ($\simeq 300$ Å at 295 K) found within the Cu layers.

The discovery of "giant" magnetoresistance in a variety of metallic multilayers comprised of alternating layers of magnetic and nonmagnetic layers has generated widespread interest.[1-5] Of particular importance are sputtered multilayers of Co and Cu which exhibit by far the largest magnetoresistance values of any magnetic material near room temperature.[6-7] Many Cu-based multilayers containing magnetic layers other than Co also display significant magnetoresistance.[8-10] A variety of theoretical models[11-13] have been proposed to account for the magnetoresistance and, in particular, to account for the dependence of the magnitude of the magnetoresistance on the thickness of the magnetic and nonmagnetic layers.[14-16] In this paper we show that substantial magnetoresistance in Co/Cu multilayers persists for Cu layers up to more than 450 Å in thickness. We propose that this dependence of magnetoresistance on the Cu-layer thickness can be simply explained in terms of *dilution* of the magnetic/nonmagnetic interface regions and volume scattering within the interior of the Cu layers.

Multilayered structures were prepared by dc magnetron sputtering in a high-vacuum deposition system (base pressure of $\simeq 2\times10^{-9}$ Torr). Several series with as many as 19 samples were prepared sequentially using computer control of the substrate platform position and of the shutters located between the platform and each magnetron source. The high stability of the deposition sources over the time required to deposit an entire series of samples ensured that the relative variation in thickness of layers from one sample to another within a given series was precisely known. The absolute thicknesses were inferred, to within about ±5%, using thick calibration films made during each deposition series. The multilayers were grown at $\simeq 40\,^\circ$C in 3.3 mTorr Ar at $\simeq 2$ Å/sec on thin Ru buffer layers that were deposited upon chemically polished Si (111) wafers.

Previously, we have shown that the nature of the buffer layer strongly influences the structural integrity of Co/Cu multilayered films and consequently their magnetic and magnetotransport properties. In particular, we have shown that Fe buffer layers give Co/Cu structures with very flat Co and Cu layers.[6] This flatness is especially important with regard to the properties of films with thin Cu spacer layers. For thicker Cu layers (i.e., greater than about 15 Å) buffer layers other than Fe such as Co, Cr, or Ru give multilayer films with magnetotransport properties comparable to those films prepared with Fe buffer layers. For the present studies, primarily involving multilayers with thick Cu layers, buffer layers of thin Ru rather than Fe were deliberately chosen to avoid the possibility of contributions from the anisotropic magnetoresistance, and the magnetic moment of Fe to the magnetoresistance, and magnetization, respectively, of the films. The resistance of the films was measured using a low-frequency ac lock-in technique with pressure contacts in a standard 4-in-line geometry, and it was measured with both the sensing current and the applied magnetic field, in the plane of the film, with the field both orthogonal and parallel to the current.[17]

The multilayers studied in this work were of the form Si(111)/Ru(50 Å)/[Co(11 Å)/Cu(t_{Cu})]$_N$/Ru(15 Å) where $N=20$ for thinner Cu layers and 6 for thicker Cu layers. X-ray-diffraction studies showed that the films were polycrystalline and that both the Co and Cu layers were fcc with pronounced $\langle 111\rangle$ texturing. Significant x-ray intensity was also found at the (200) Bragg reflection. For otherwise similar structures the ratio of the integrated intensity of the (111) to (200) Bragg peaks, I_{111}/I_{200}, depended on the Cu layer thickness, t_{Cu}. For example, for $N=20$, as t_{Cu} was varied from $\simeq 3$ to $\simeq 25$ Å, I_{111}/I_{200} increased from $\simeq 10$ to $\simeq 16$ whereas for $N=6$, as t_{Cu} varied from $\simeq 50$ Å to $\simeq 500$ Å, I_{111}/I_{200} decreased from $\simeq 10$ to $\simeq 1$. Note that the full width at half maximum (FWHM) of the rocking curve of the (111) reflection typically decreased with increasing t_{Cu} ranging from $>20°$ for the thinnest Cu layers to $\simeq 10°$ for the thickest Cu layers studied. The microstructure of various samples were characterized by high-resolution cross-section transmission electron microscopy using a JEM-4000EX operated at 400 kV, with typical magnifications of 500 000 times. The samples were prepared for electron microscopy by mechanical polishing followed by ion-milling with 5 keV argon ions until perforation occurred.[18]

Figure 1 shows low-magnification electron micrographs of a Si(111)/Ru(50 Å)/[Co(11 Å)/Cu(340 Å)]$_6$/Ru(15 Å) structure recorded deliberately at underfocus. The multilayer comprises six Co/Cu bilayers with 340-Å-thick Cu spacer layers. Very little compositional contrast is expected between layers since Co and Cu are close neighbors in the Periodic Table and have similar lattice constants. Nevertheless the Co layers are clearly visible in images taken at large underfocus values due to the presence of Fresnel fringes at the Co/Cu interfaces. The Co layers appear to be wavy with excursions from linearity on the order of 10 Å. The multilayer is polycrystalline with grains ranging in size from about 1000 to 4000 Å as measured parallel to the substrate. Large grains of various orientations are observed in different regions of the sample with most grains extending throughout the entire thickness of the multilayer ($\simeq$2500 Å).

Figure 2 shows a high-magnification image taken from the same sample at the optimum defocus condition. Cu(111) lattice fringes extending over large regions of sample are visible, and the positions of the Co layers are delineated by the periodic strain contrast. Selected area electron diffraction, as well as the high-resolution lattice imaging indicates that the majority of the grains are ⟨111⟩ oriented to within a few degrees of the Si(111) substrate. [X-ray-diffraction studies showed that the (111) rocking curve FWHM was only 10.1° for this sample.] Grains with other orientations are occasionally observed but are usually poorly oriented with respect to the substrate. The majority of the grains are well structured and uniform although scattered defects such as stacking faults and twin boundaries are present in some places.

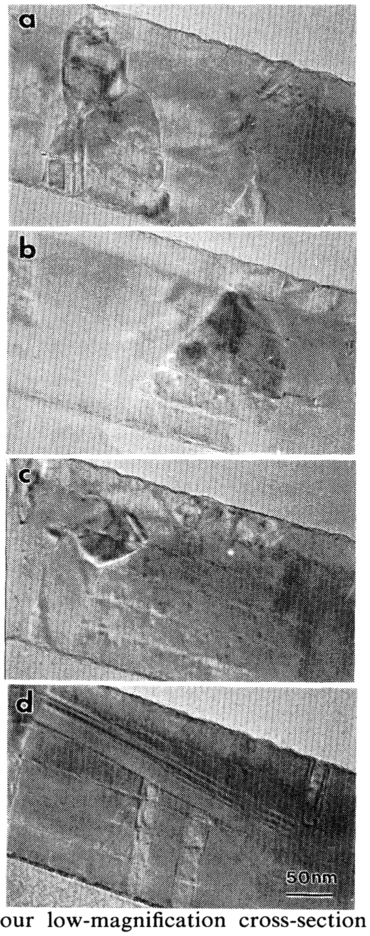

FIG. 1. Four low-magnification cross-section transmission electron micrographs showing different regions of a Co/Cu multilayer of form Si(111)/Ru(50 Å)/[Co(11 Å)/Cu(340 Å)]$_6$/Ru(15 Å). The micrographs were deliberately recorded at substantial underfocus condition to enhance contrast of separate Co and Cu layers. Six Co and six Cu layers are readily identified.

Figure 3 shows the dependence of the saturation magnetoresistance (MR) on Cu spacer layer thickness t_{Cu} as measured at temperatures of 295 and 4.2 K, where we define the saturation MR as the maximum change in resistance with field divided by the resistance at high field. Four well-defined oscillations in the magnitude of the saturation MR are evident as t_{Cu} is increased. This behavior is similar to that previously observed for comparable multilayers grown on Fe except that the magnitude of the MR at the first peak ($t_{\mathrm{Cu}} \simeq 9$ Å) is almost twice as high, $\simeq 100\%$, for multilayers grown on Fe.[6,7] The curves drawn on these figures are explained below.

Resistance versus field curves at 4.2 K for four samples containing relatively thick Cu spacer layers of 70, 150, 300, and 425 Å respectively, are shown in Fig. 4. The resistance versus field curves are clearly hysteretic and exhibit peaks at positive and negative fields, $\pm H_m$. Magnetic hysteresis loops on the same samples shows that H_m closely corresponds to H_c, the switching field of the multilayers, where the net magnetization passes through zero. As the temperature is increased, for a given sample, H_m decreases, and it has significantly lower values at room temperature. As t_{Cu} is increased within a series of samples, H_m gradually increases. Note that H_m depends on the growth of the structures and, for example, varies with buffer layer material.

Finally, we note the dependence of the room-temperature resistivity of the multilayers, measured in nominally zero field, on the copper film thickness t_{Cu} as

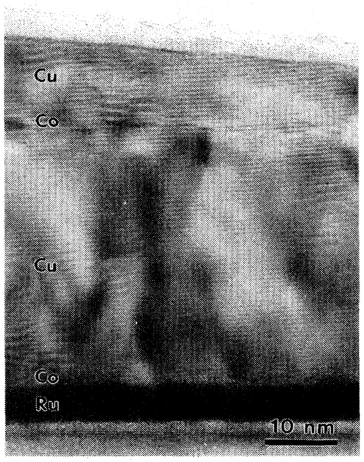

FIG. 2. High-magnification transmission electron micrograph, recorded in cross section at optimum defocus condition, of the same sample, Si(111)/Ru(50 Å)/[Co(11 Å)/Cu(340 Å)]$_6$/Ru(15 Å) imaged in Fig. 1.

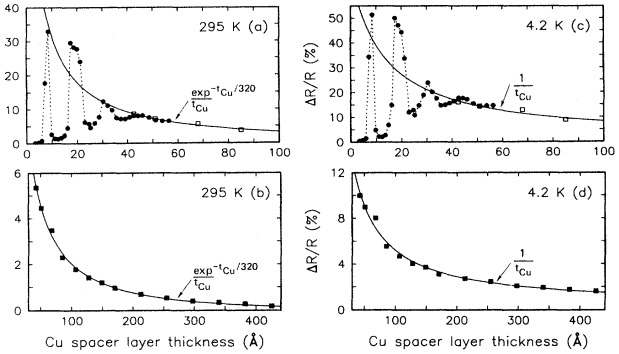

FIG. 3. Saturation magnetoresistance vs Cu spacer layer thickness for several series of multilayers of the form Si(111)/Ru(50 Å)/[Co(11 Å)/Cu(t_{Cu})]$_N$/Ru(15 Å). The number of Co/Cu periods, N, is 20 (solid circles) and 6 (open and closed squares). Data are shown for temperatures of (a) and (b) 295 K, and (c) and (d) 4.2 K. Note that since the MR increases with N, in (a) and (c) the $N=6$ data have been scaled by an empirical factor of 1.6 to make comparison with the $N=20$ data easier. Curves drawn through the data are of the form $1/t_{Cu}\exp[-(t_{Cu}/\lambda_{Cu})]$ at 295 K and $1/t_{Cu}$ at 4.2 K. Note the actual curves shown in the figure are of the exact form, $\Delta R/R=289/(4.3+t_{Cu})\exp[-(t_{Cu}/318)]$ and $\Delta R/R=0.28+554/(13+t_{Cu})$ at 295 and 4.2 K, respectively. The curves have been scaled by an empirical factor of 1.6 in (a) and (c).

shown in Fig. 5(a). These measurements were taken prior to placing the films in a magnetic field. Interestingly, oscillations of MR are clearly reflected as oscillations in the magnitude of the zero-field resistivity. As t_{Cu} becomes very large the resistivity approaches that measured on $\simeq 1000$ Å thick single Cu layers of $\simeq 3$ $\mu\Omega$ cm, whereas the resistivity of similar $\simeq 1000$ Å thick Co films was $\simeq 12$ $\mu\Omega$ cm. Figure 5(b) shows the dependence on t_{Cu} of the resistance ratio, $R_{295}K/R_{4.2}K$, measured in magnetic fields large enough to saturate the resistance of the multilayers.

The MR of these multilayers is related to a change in the relative orientation of the magnetic moments of successive Co layers with application of the field. For antiparallel alignment of the magnetic moments the resistance of the structure is increased relative to parallel alignment. The Co layers are magnetically exchange-coupled through the Cu layers. The coupling oscillates between ferromagnetic and antiferromagnetic coupling as the Cu layer thickness is increased, resulting in corresponding oscillations in the saturation MR.[4,7] The decreased MR at the first peak for multilayers grown on Ru as compared to multilayers grown on Fe is related to the extent of antiparallel alignment of the Co layers which is reduced in these structure. We attributed this particular result to direct ferromagnetic coupling of neighboring Co layers in some regions of the samples containing very thin Cu layers, possibly as a result of imperfections in the layering. For example, this could be a consequence of the waviness of layers that is visible in Fig. 1.

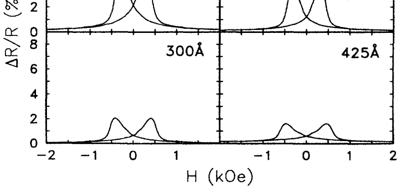

FIG. 4. Resistance vs field curves for four Co/Cu multilayers of the form Si(111)/Ru(50 Å)/[Co(11 Å)/Cu(t_{Cu})]$_6$/Ru(15 Å) with Cu spacer layer thicknesses t_{Cu} of 70, 150, 300, and 425 Å.

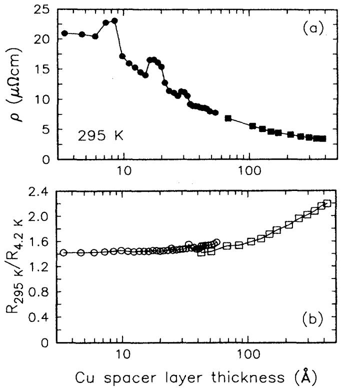

FIG. 5. Dependence on Cu spacer layer thickness t_{Cu} of (a) the zero-field resistivity at 295 K and (b) the high-field resistance ratio $R_{295}K/R_{4.2}K$, for multilayers of the form, Si(111)/Ru(50 Å)/[Co(11 Å)/Cu(t_{Cu})]$_N$/Ru(15 Å), with $N=20$ (solid and open circles) and 6 (closed and open squares).

When the Co layers are strongly ferromagnetically coupled via the Cu layers, the magnetic moments of neighboring Co layers remain parallel for all fields. These multilayers exhibit very small MR, corresponding to the bulk anisotropic magnetoresistance (AMR) of the Co layers themselves. For example, the sample with a Cu spacer layer $\simeq 3.5$ Å thick, displayed an AMR (Ref. 19) of $\simeq 2\%$ at 4.2 K and $\simeq 0.6\%$ at 295 K. The AMR becomes progressively smaller as the proportion of Co to Cu is decreased in the multilayer, with increasing t_{Cu}. Except for very thin Cu layers, all the multilayers showed MR enhanced compared to their corresponding AMR, suggesting a degree of antiparallel alignment of the Co layers was present in most structures. As shown in Fig. 3, the amplitude of the oscillations in MR becomes progressively smaller as t_{Cu} is increased. Concomitantly the field strength at which the multilayers exhibit maximum resistance increases from approximately zero at the first peak[6,7] to a value determined by the coercive field H_c of the magnetic layers for thicker Cu layers (see Fig. 4).

As the Cu layer thickness is increased above $\simeq 60$ Å the MR decays. The functional dependence of MR on t_{Cu} is surprisingly simple, particularly at low temperatures. For example, as shown by the hyperbolic line sketched in Fig. 3(d), the MR decays roughly as $1/t_{Cu}$ at 4.2 K. Since the giant MR phenomenon is predominantly determined by scattering at the Co/Cu interfaces[20,21] this functional dependence can be rationalized in terms of *dilution* of the interfacial scattering regions as the measuring current, which is parallel to the layers, is shunted through the bulk of the Cu layers away from the interfacial regions.

Scattering within Cu layers will clearly diminish the flow of electrons between Co layers. Such scattering is expected to be determined by *volume* scattering within the interior of the Cu layers. This can be described by a scattering length, λ_{Cu}, which should be related to the mean free path in thick Cu layers, where the contribution from scattering at the surfaces of the film is small and scattering within the interior of the film is predominant. Taking into account both this volume scattering and the dilution effect mentioned above, for sufficiently thick Cu layers where most of the current is carried by the Cu layers, one would thus expect that the MR would vary as $\simeq 1/t_{\mathrm{Cu}}\exp[-(t_{\mathrm{Cu}}/\lambda_{\mathrm{Cu}})]$. For MR data taken at 295 K, as shown by the line drawn in Fig. 3(b), this particular functional form closely describes the dependence of MR on t_{Cu} for Cu-layer thicknesses varying from 50 to more than 500 Å, giving a value for λ_{Cu} of $\simeq 320$ Å. Such a long mean free path[22] is consistent with the high degree of structural ordering within the individual Cu layers as revealed by the TEM micrographs shown in Figs. 1 and 2. Moreover micrographs taken on a series of structures with different Cu-layer thicknesses showed that in each case the grain size was several times the Co-Cu bilayer thickness so that it is not surprising the data in Fig. 3 can be described by a mean free path which is independent of the Cu layer thickness. Note that, as described in the figure caption, the data in Fig. 3 were fitted with an equation of the exact form $a+b/(t_s+t_{\mathrm{Cu}})\exp[-(t_{\mathrm{Cu}}/\lambda_{\mathrm{Cu}})]$: the small corrections can be qualitatively understood as accounting first for a contribution from the AMR (a), and second for shunting of part of the current through the thickness t_s of the Ru and Co layers.

It is interesting to note, as shown in Fig. 3(a) and 3(c), that extrapolations of the fitted MR curves to smaller t_{Cu} pass through the middle of the MR oscillations. For thick Cu layers where the indirect exchange coupling between the Co layers is negligible, the resistance of the multilayer is related to the extent to which electrons originating from magnetic domains in one Co layer are scattered in magnetic domains of the opposite sign in neighboring Co layers. The probability will be related to the arrangement of magnetic domains in each Co layer and will be highest for fields close to H_c. Nevertheless it will clearly be much less than unity. The MR will thus be reduced relative to the situation when the Co layers are arranged perfectly antiparallel to one another for some field range, which thereby accounts for the increased MR at the oscillation peaks compared with extrapolations from thicker Cu layers.

Finally, it should be mentioned that the enhanced MR is present only for structures containing at least two Co layers separated by Cu spacer layers. Several structures were grown that contained single Co layers of 10 Å thickness sandwiched between much thicker Cu layers. For these films the MR was not enhanced compared with the AMR for the Co layer. Thus, the MR observed in the multilayered films must be related to electrons propagating from one Co layer though the Cu layers to the neighboring Co layers.

In summary we have shown that the dependence of magnetoresistance on the Cu spacer layer in Co/Cu multilayers can be ascribed to two factors. First, as the Cu spacer layer thickness is increased the Co/Cu interfacial regions are *diluted* by shunting of current through the interior of the Cu layers. Second, scattering within the Cu layers give rise to an exponential decay of the magnetoresistance with Cu layer thickness, for thicker Cu layers. The scattering length associated with this decay at 4.2 K was found to be considerably longer than the largest Cu thickness examined, so that the MR was observed to vary inversely with the Cu layer thickness. At 295 K a decay length of greater than about 300 Å was found. Although this value may seem surprisingly long for sputtered polycrystalline films, high-resolution cross-section transmission electron micrographs confirmed that the films posses a high degree of structural coherence, particularly within the Cu spacer layers, with typical grain sizes, measured laterally, of several thousand Å, and along the multilayers of several bilayer periods.

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*Also at the Department of Physics.

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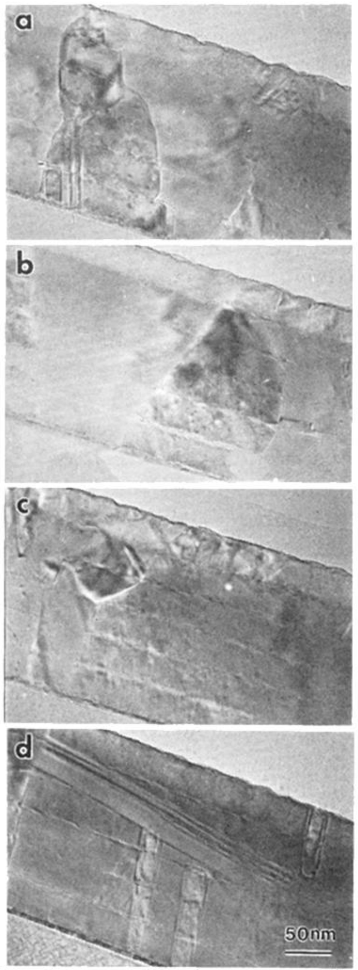

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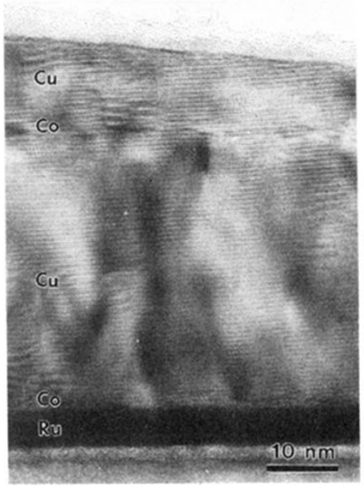

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