Giant magnetoresistance in antiferromagnetic Co/Cu multilayers

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We report giant values of saturation magnetoresistance in sputtered antiferromagnetic Co/Cu multilayers containing thin Co and Cu layers 8–10 Å thick. We discuss the key importance of the buffer layer in controlling the growth of flat Co and Cu layers. As shown by cross-section transmission electron microscopy high-quality structures are found for growth on Fe buffer layers. Such structures display saturation magnetoresistance at 300 K of more than 65% with saturation fields of $\simeq 10$ kOe. These values are several times larger than previously found for any magnetic material at room temperature.

Recently very large saturation magnetoresistance was observed in single crystal[1,2] and polycrystalline[3,4] Fe/Cr multilayers. The magnetoresistance is intimately linked to a change in the magnetic structure of the multilayers with magnetic field. In zero field adjacent Fe layers are aligned antiferromagnetically with respect to one another.[5,6] In relatively small magnetic fields the Fe layers reorient themselves parallel to the field and the resistance correspondingly decreases and saturates when the net magnetization saturates. Understanding the origin of this unusual giant magnetoresistance is complicated by the possibility of magnetism or magnetic ordering within the Cr layers (see, for example, Ref. 7). Indeed, theoretical models that have been proposed require the conduction electrons within the magnetic layers to undergo spin-dependent scattering from Cr atoms dissolved in the Fe layers at the Fe/Cr interface.[8,9]

Here we report the observation of even larger giant magnetoresistance in a second multilayer system, sputtered polycrystalline Co/Cu. The field dependence of the magnetization of these samples provides evidence for antiferromagnetic coupling of the Co layers consistent with recent neutron diffraction experiments on single crystal Co/Cu superlattices.[10] Thus, a common origin of the giant MR to that in Fe/Cr is suggested. However, unlike Cr, which is a transition metal, Cu is a noble metal with no unfilled $3d$ bands at the Fermi level, simplifying theoretical treatments although out of the scope of existing models. We find saturation magnetoresistance values in sputtered Co/Cu multilayers of more than 65% and 115% at 295 and 4.2 K, respectively, compared to corresponding values of only 13 and 80% in Fe/Cr. The dramatically higher values at room temperature in Co/Cu, more than five times higher than previously observed in any magnetic system, suggest this new material may be potentially useful in magnetic field sensor devices.

The Co/Cu multilayers were grown in a computer controlled high vacuum dc magnetron sputtering system with a base pressure of $\simeq 2\times10^{-9}$ Torr. Series of up to 19 samples were sequentially deposited at $\simeq 50$ °C in 3.25 mT argon at 2 Å/s. X-ray diffraction studies showed that the films are polycrystalline and that for the films of interest here both the Co and Cu layers are fcc with (111) texturing. The samples were prepared on polished and chemically etched Si(100) wafers. It was found that the growth of the multilayers is significantly affected by the nature of buffer layers grown between the silicon wafer and the multilayer.

Figure 1 shows room temperature resistance[11] versus field data for four identical $[\mathrm{Co}(10\ \text{Å})/\mathrm{Cu}(9\ \text{Å})]_{16}$ multilayers grown on 50 Å Cu or Fe buffer layers with 50 Å Fe or Cu capping layers. The shape of the resistance curves is distinctly different depending on the buffer layer. However, comparison of the magnitude of the magnetoresistance for different samples requires careful consideration of the effect of the buffer and capping layers. In particular, since the Cu buffer/capping layers are much more conducting than Fe layers of the same thickness the sensing current is shunted to a greater extent through Cu than Fe layers. Thus, shunting of the current away from the multilayer will trivially reduce the observed magnetoresistance as can be seen in Fig. 1 by comparing samples deposited on the same buffer layer but with Cu rather than Fe capping layers. However, the extent of shunting should be approximately the same for the two samples with Fe/Cu and Cu/Fe buffer/capping layers that contain the same total thicknesses of Fe, Cu, and Co layers. Since the sample grown on the Fe buffer layer has a much larger magnetoresistance it is clear that the nature of the buffer layer has an important effect on the physical properties of the film. Magnetic hysteresis curves for these same samples are shown in Fig. 2. Fields much larger than the coercive field of the Co layers are required to saturate the magnetization consistent with antiferromagnetic coupling of adjacent Co layers. Moreover, for the samples grown on Fe the remanent magnetization in zero field corresponds to the additional magnetization expected from the Fe. This means that the net moment of the Co layers in zero field is very small, such that they are almost perfectly antiferromagnetically aligned. In contrast, the samples with Cu buffer layers display much larger remanent magnetizations indicating only partial antiferromagnetic alignment of the Co layers. The data on the latter samples are similar to those

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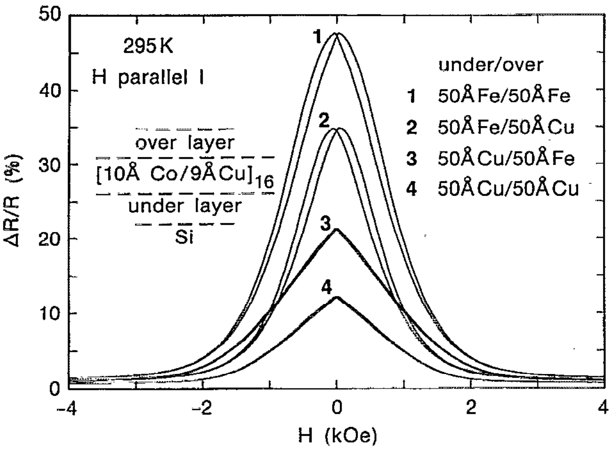

FIG. 1. Room-temperature resistance vs field curves for four samples of the form Si(100)/buffer layer/[10 Å Co/9 Å $Cu]_{16}$/capping layer with 50-Å-thick buffer and capping layers of respectively (1) Fe and Fe, (2) Fe and Cu, (3) Cu and Fe, and (4) Cu and Cu.

found previously for molecular beam epitaxy deposited Co/Cu multilayers grown on Cu(100) single crystals.[10]

The microstructures of the two samples shown in Fig. 2 were characterized in cross section using transmission electron microscopy. The samples were prepared in the standard manner[12] by mechanical polishing followed by ion milling with 5 keV argon ions to perforation. A JEM-4000EX high-resolution electron microscope operated at 400 kV was used for observation, with typical magnifications of 500 000 times.

Observations by high-resolution electron microscopy showed that the structural morphology of the two films was considerably different. These differences were more apparent in electron micrographs such as those shown in Fig. 3 which were recorded at substantial underfocus conditions (i.e., at -2000 Å rather than the optimum defocus of -480 Å). Figure 3 (a) shows that reaction of the Cu buffer layer with the Si substrate has occurred, in some places to depths of greater than 50 Å, whereas there is no

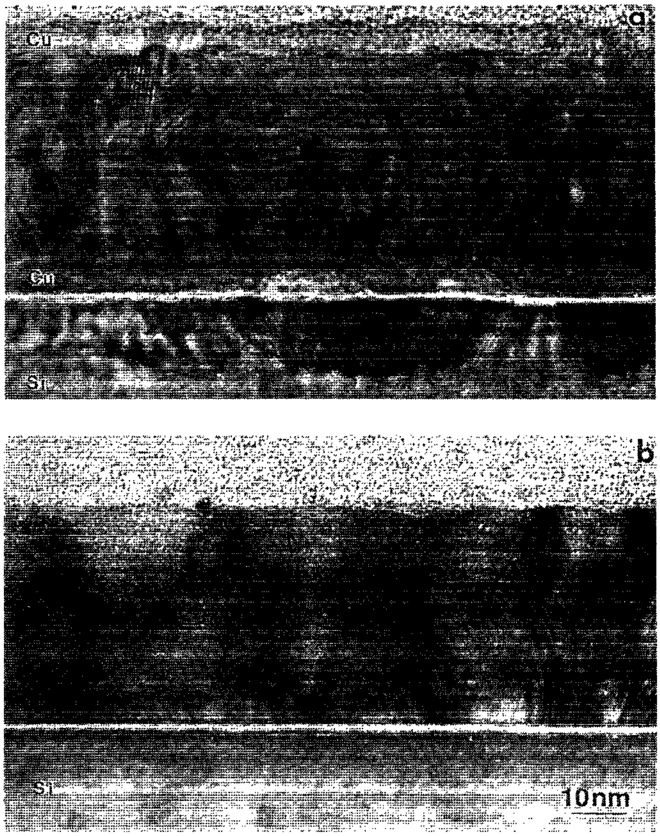

FIG. 3. Cross-section transmission electron micrographs of (a) Si(100)/50 Å Cu/[10 Å Co/9 Å $Cu]_{16}$/50 Å Fe and (b) Si(100)/50 Å Fe/[10 Å Co/9 Å $Cu]_{16}$/50 Å Cu. The micrographs are taken at an underfocus condition to enhance contrast of the Co and Cu layers.

sign of any reaction in the case of the Fe buffer layer [Fig. 3(b)]. Both micrographs display indications of diffraction contrast, more apparent in low-resolution images, that imply the occurrence of coherent columnar growth through several bilayers. Moreover, considerable waviness and buckling of the layers is visible in Fig. 3(a), unlike the comparatively smooth and flat layers in Fig. 3(b). High-magnification images of the Fe-based structure, recorded close to the optimum defocus, confirmed the existence of the columnar growth, although the well-ordered layer growth was no longer visible.

These observations are consistent with the properties of the films described above. The rumpling of the Co and Cu layers shown in Fig. 3(a) will result in partial ferromagnetic alignment of adjacent Co layers, possibly via direct contact of successive Co layers. Another possibility is that local variations of the Cu layer thickness result in variations in the magnetic coupling which is extremely sensitive to the Cu layer thickness, oscillating back and forth from antiferromagnetic to ferromagnetic coupling with a period of only $\simeq 10$ Å.[13] Thus we conclude that the samples grown on Fe buffer layers that show much less rumpling have larger magnetoresistance values resulting from the more complete antiferromagnetic alignment of the Co layers. We also note that the remanent magnetization in structures grown on Cu buffer layers is simply a result of poor structural integrity and not a consequence of mag-

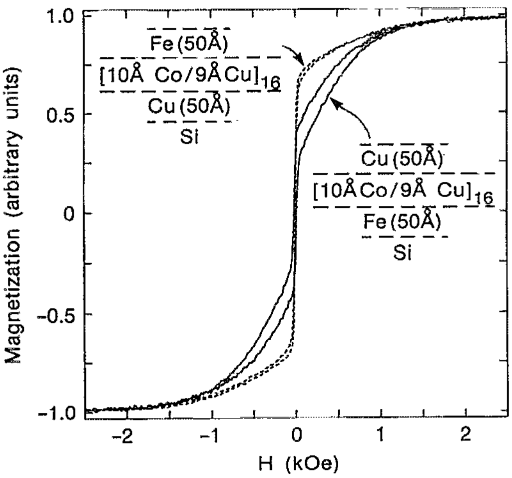

FIG. 2. Room-temperature in-plane magnetization vs field curves for samples 2 and 3 of Fig. 1.

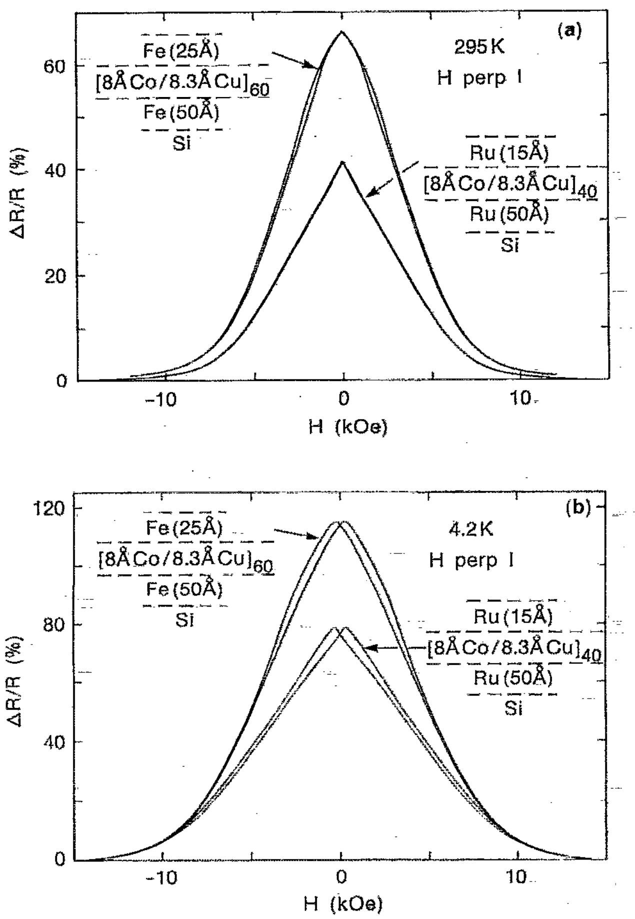

FIG. 4. Resistance vs field curves for two multilayers, Si(100)/50 Å Fe/[8 Å Co/8.3 Å Cu]$_{60}$/50 Å Cu and Si(100)/50 Å Ru/[9 Å Co/8.3 Å Cu]$_{40}$/15 Å Ru at (a) 300 and (b) 4.2 K.

netic anisotropy as invoked to account for similar data on MBE-grown Co/Cu single crystals.[14]

The growth of Co/Cu multilayers was studied on several other buffer layers. Surprisingly, although highly perfect Co/Ru multilayers can be grown on Ru buffer layers on Si,[3] Co/Cu multilayers grown on Ru display properties intermediate between those grown on Cu and those grown on Fe. Magnetoresistance data are shown for a typical structure in Fig. 4 and compared to a Co/Cu structure engineered to give very large magnetoresistance. As shown in Fig. 4, large values of magnetoresistance are possible in structures grown on Ru layers. However, the triangular shape of the resistance curve at low fields is similar to that for structures prepared on Cu buffer layers indicative of incomplete antiferromagnetic alignment of the Co layers. The data on the Si(100)/50 Å Fe/[8 Å Co/8.3 Å Cu]$_{60}$/50 Å Cu multilayer shown in Fig. 3(b) demonstrate that $\Delta R/R$ exceeding 65% at 295 K and 115% at 4.2 K are possible in Co/Cu multilayers. The magnetoresistance is maximized for thin Co and Cu layers approximately 8 Å thick and for about 60 bilayers. Large numbers of bilayers minimize shunting effects through the buffer and capping layers but multilayers grown with more than 60 bilayers have slightly reduced saturation magnetoresistance values. In contrast to Fe/Cr multilayers, the magnetoresistance in Co/Cu has a weak temperature dependence. Thus, while at room temperature the magnetoresistance of Co/Cu multilayers is four to five times larger than in Fe/Cr structures, at helium temperatures the magnetoresistance is just 50% higher. The weaker temperature dependence for Co/Cu compared to Fe/Cr is consistent with smaller spin-flip scattering in Cu than in Cr layers.

In summary, we report giant magnetoresistance in antiferromagnetic Co/Cu multilayers. Values of saturation magnetoresistance exceeding 65% are found at room temperature and 115% at 4.2 K with saturation fields of $\simeq 10$ kOe for structures containing $\simeq$8-Å-thick Co and Cu layers. Lower saturation fields of $\simeq 300$ Oe are obtained for thicker Cu layers of $\simeq 17$ Å. The magnetoresistance is also lower but still substantial, exceeding $\simeq 34\%$ at 300 K. These are by far the largest saturation magnetoresistance values yet found in any magnetic system. The magnitude of the magnetoresistance for thin Co and Cu layers is intimately related to the structural integrity of these layers. For growth on silicon wafers the novel use of an Fe buffer layer was shown to be crucial to the growth of high quality layered structures.

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