RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 46, NUMBER 14 1 OCTOBER 1992-II

Giant magnetoresistance and enhanced antiferromagnetic coupling in highly oriented Co/Cu (111) superlattices

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(Received 14 May 1992; revised manuscript received 15 July 1992)

We demonstrate the presence of large oscillatory antiferromagnetic (AF) interlayer coupling and "giant" magnetoresistance in crystalline (111)-oriented Co/Cu superlattices grown on Pt buffer layers on (0001) sapphire substrates. The AF coupling strength is ≈4–5 times larger than previously reported for sputtered polycrystalline Co/Cu multilayers. However, a significant fraction of the superlattice (≈70%) remains ferromagnetically coupled. This may account for the comparatively low giant magnetoresistance values observed of ≈40% at 3.5 K and ≈26% at 295 K compared to values of > 120% at 4.2 K and > 65% at 295 K, respectively in sputtered structures. Similar crystalline (111) Co/Cu structures grown on Cu buffer layers on GaAs(110) show no evidence for antiferromagnetic coupling, although they are of nearly comparable structural perfection. The presence or absence of antiferromagnetic coupling in (111) Co/Cu superlattices appears to be a result of subtle structural imperfections giving rise to direct ferromagnetic coupling of neighboring Co layers.

Recently oscillatory indirect exchange coupling of Co layers via Cu spacer layers in sputtered (111) Co/Cu textured multilayers was reported.[1] This system is particularly interesting, since it displays very large or "giant" saturation magnetoresistance (MR) values at room temperature of more than 65%, larger than is found in any other metallic multilayer.[1–3] In contrast, several groups have reported the absence of evidence for antiferromagnetic (AF) coupling in Co/Cu multilayers or Co/Cu/Co sandwiches grown oriented along ⟨111⟩, although antiferromagnetic coupling has been found in single-crystal (100) Co/Cu/Co sandwiches.[4] The lack of observation of coupling in single-crystalline Co/Cu structures has led to speculations that the coupling observed in polycrystalline sputtered films may actually arise from grains oriented in directions distinct from ⟨111⟩.[5] This point of view appears to be buttressed by theoretical models of the coupling in Co/Cu, which suggest the coupling is stronger along ⟨100⟩ and ⟨110⟩ than along ⟨111⟩.[6,7] Nevertheless, a second highly plausible possibility is that AF coupling in ⟨111⟩-oriented single-crystal Co/Cu multilayers is present but obscured by direct ferromagnetic coupling resulting from structural defects, for example, magnetic bridges between adjacent Co layers.[1] The recent finding of antiferromagnetic coupling in highly textured, ultrahigh-vacuum- (uhv) evaporated Co/Cu multilayers on thick Au buffer layers on float glass supports the latter conjecture.[8] In this paper we compare the properties of crystalline Co/Cu (111) superlattices grown on a variety of buffer layers and substrates. We show that similar Co/Cu (111) multilayers of nearly comparable structural perfection, as evidenced from x-ray diffraction and reflection high-energy electron diffraction (RHEED) studies, exhibit widely differing antiferromagnetic coupling strengths and saturation magnetoresistance.

Figure 1(a) shows a comparison of the dependence of saturation magnetoresistance[9] on Cu spacer layer thickness for Co/Cu (111) textured films, prepared by dc magnetron sputtering, and for twinned crystalline (111) Co/Cu films deposited by uhv electron-beam deposition. The sputtered films were grown on 50-Å-thick Ru buffer layers on Si(111) wafers at ≈40°C. These films had structures 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. The crystalline films were grown in an uhv deposition system with a base pressure immediately after deposition of 1.5×10^{-10} Torr. These films were grown on chemically etched GaAs (110) wafers, and heat cleaned *in situ* at 580 °C. The ⟨111⟩ orientation of the Co/Cu superlattice was obtained by growing the superlattice on a buffer layer of 9-Å bcc Co followed by ≈55–60 Å of fcc Cu according to the pro-

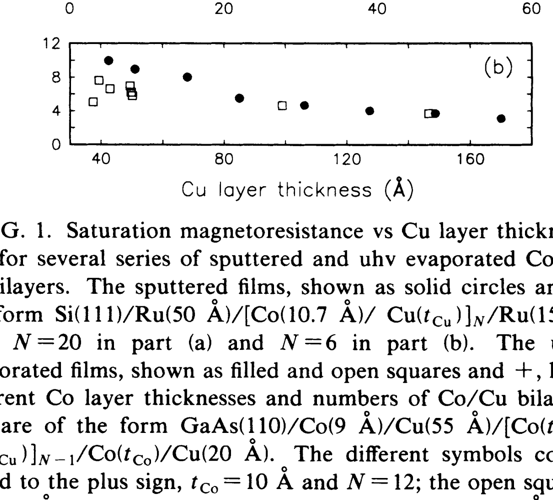

FIG. 1. Saturation magnetoresistance vs Cu layer thickness, t_{Cu}, for several series of sputtered and uhv evaporated Co/Cu multilayers. The sputtered films, shown as solid circles are of the form Si(111)/Ru(50 Å)/[Co(10.7 Å)/ Cu(t_{Cu})]$_N$/Ru(15 Å) with $N=20$ in part (a) and $N=6$ in part (b). The uhv-evaporated films, shown as filled and open squares and +, have different Co layer thicknesses and numbers of Co/Cu bilayers and are of the form GaAs(110)/Co(9 Å)/Cu(55 Å)/[Co(t_{Co})/ Cu(t_{Cu})]$_{N-1}$/Co(t_{Co})/Cu(20 Å). The different symbols correspond to the plus sign, $t_{\mathrm{Co}}=10$ Å and $N=12$; the open square, $t_{\mathrm{Co}}=21$ Å and $N=12$, and the solid square, $t_{\mathrm{Co}}=21$ Å and $N=6$.

cedures developed by Lamelas *et al.*[10] These structures were capped with ≈20 Å Cu. The structure was grown at 100 °C. Detailed structural studies established that the Co/Cu superlattice takes up a fcc structure twinned by a 180° rotation about the [111] axis.[11] As can be seen from Fig. 1, oscillations in the saturation magnetoresistance of the sputtered films are found as a function of Cu thickness similar to those previously reported for structures grown on Fe or Cu buffer layers.[1,3] Such oscillations in the saturation magnetoresistance correspond to oscillations between antiferromagnetic and ferromagnetic coupling. The magnetoresistance results from a change in the magnetic structure of neighboring Co moments from antiparallel to parallel as the field is changed. For thinner Cu layers, large magnetoresistance values are found at Cu layer thicknesses for which there is strong antiferromagnetic coupling. No such oscillations in magnetoresistance nor large magnetoresistance values are evident for the crystalline (111) Co/Cu structures grown on GaAs(110) shown in Fig. 1(a). Note that the largest MR values in sputtered Co/Cu multilayers of more than 65% at 295 K and 120% at 4.2 K are found for $t_{\mathrm{Cu}} \approx 8$–9 Å and in films grown using thin Fe buffer layers.[1,2] For similar multilayers grown on Ru buffer layers the MR values are about one half as large. However, for thicker Cu layers the MR values are comparable for multilayers grown on Fe and Ru.[12]

As the Cu spacer layer thickness is increased in the sputtered Co/Cu multilayers, the magnitude of the magnetic coupling decreases rapidly and consequently the oscillations in magnetoresistance fade away. Nevertheless enhanced magnetoresistance compared to the anisotropic magnetoresistance of the individual Co layers persists for Cu layers more than 500 Å thick. The magnetoresistance in these cases is related to the possibility of magnetic domains in neighboring Co layers being aligned antiparallel to one another for certain field ranges. Indeed, the resistance of these structures is maximized close to the coercive field of the Co layers where the net magnetization passes through zero and there is the greatest likelihood of antiparallel magnetic domains in successive Co layers. Figure 1(b) compares saturation magnetoresistance data for sputtered and uhv-evaporated (111) Co/Cu multilayers comprised of thicker Cu spacer layers. The structures contain the same number of Co layers. Although the uhv-evaporated structures contain thicker Co layers, the magnitude of the magnetoresistance is not very sensitive to the Co layer thickness for thicker Cu layers. As can be seen from the figure, the dependence on Cu layer thickness and the magnitude of the magnetoresistance are similar in both cases for Cu layers thicker than ≈80 Å. These results suggest that for sufficiently thick Cu layers the Co layers in the films evaporated on GaAs can switch independently in a manner similar to the sputtered films but that when the Cu layer thickness is reduced the Co layers become increasingly ferromagnetically coupled, preventing the Co layer magnetic moments from rotating with respect to each other. Moreover, the similarity of saturation magnetoresistance values for films containing thicker Cu layers, suggests that the magnitude of the giant MR effect is approximately the same for both uhv-evaporated and sputtered films, for the same degree of antiparallel interlayer alignment of the Co layers.

Figure 2(a) shows an x-ray θ–2θ diffraction pattern for a typical (111) Co/Cu sample corresponding to the uhv-evaporated structures shown in Fig. 1. These data were collected on a Blake Instruments double-crystal diffractometer equipped with a GaAs(100) analyzer crystal using Cu K_α radiation. Apart from the strong (220) GaAs Bragg peak [and leading edge of the corresponding (440) peak near $2\theta = 100°$] the only significant diffraction intensity is found at the (111) and (222) fcc Co/Cu Bragg peaks and their respective weak superlattice satellite peaks. No diffraction intensity is found at the (110) or (100) fcc Co/Cu peaks, thus demonstrating the absence of such crystallites. The inset to the figure shows the rocking curve through the (111) Co/Cu Bragg peak. The narrow width of this peak at half maximum, $\Delta\omega \approx 0.78°$, indicates that the sample is highly oriented along the [111] direction.

Figure 2(b) shows x-ray diffraction data for an uhv-evaporated Co/Cu superlattice grown epitaxially on a Pt(111) seed layer prepared at 600 °C on (0001) sapphire wafers. The superlattice was grown at ≈0 °C and capped with a thin (≈30-Å) Pt overcoat also deposited at ≈0 °C. *In situ* RHEED and low-energy electron-diffraction studies showed the Co/Cu superlattice had a highly oriented [111] growth axis epitaxially aligned with the Pt buffer and substrate with sixfold symmetry. The sixfold symme-

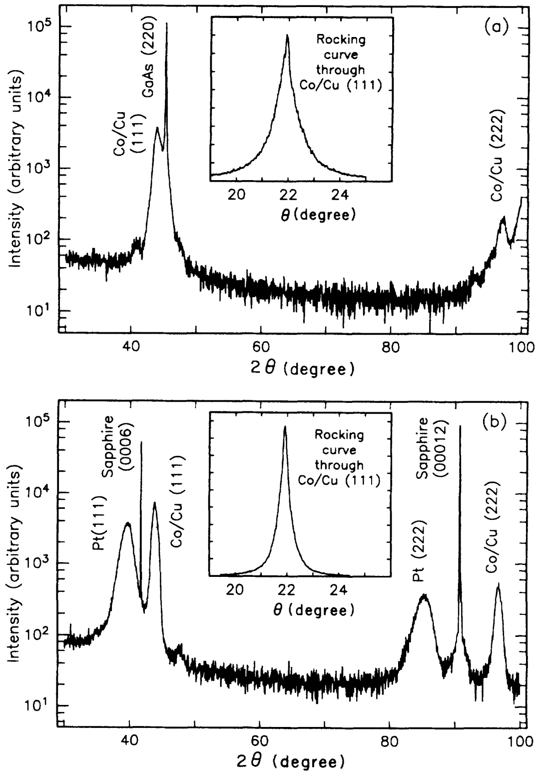

FIG. 2. x-ray θ–2θ diffraction pattern and (inset) rocking curve through the fcc Co/Cu (111) Bragg peak for two uhv-evaporated Co/Cu superlattices of the form (a) GaAs(110)/Co(9 Å)/Cu(55 Å)/[Co(21 Å)/Cu(8 Å)]$_{11}$/Co(21 Å)/Cu(20 Å) and (b) sapphire (0001)/Pt(30 Å)/[Co (16 Å)/Cu(9 Å)]$_{13}$/Co(16 Å)/Pt(30 Å).

try indicates two possible domain orientations for the (111) growth related by a 180° rotation. The x-ray data also show the superlattice is single crystalline, oriented along [111]. The width of the rocking curve through the Co/Cu fcc (111) reflection is only $\approx 0.43°$ wide. While the rocking curve is somewhat narrower than for the (111) Co/Cu sample deposited on GaAs(110) shown in Fig. 2(a), both samples prepared by uhv deposition have rocking curves 25–50 times narrower than for the sputtered Co/Cu multilayers of Fig. 1. Notwithstanding the similar structural perfection of the superlattices grown on GaAs and sapphire shown in Fig. 2, these superlattices exhibit very different magnetotransport properties. Figure 3 shows resistance versus field curves at 295 and 3.5 K for a typical sample grown on sapphire. Substantial magnetoresistance and very large antiferromagnetic interlayer coupling is found. The maximum antiferromagnetic coupling strength observed in such structures was ≈ 1.7 erg/cm^2 at ≈ 5 K, approximately four to five times larger than the largest coupling strength observed in sputtered Co/Cu multilayers.[13] However, in contrast to sputtered Co/Cu films grown on Fe buffer layers,[2] magnetization versus field curves—see, for example, the inset in Fig. 3—show incomplete antiferromagnetic coupling with large remanent magnetizations in low fields of about 70% of the saturated magnetization.[14] This suggests that a large part of the uhv-evaporated sample remains ferromagnetically coupled. Since the antiferromagnetic coupling strength corresponds to only a few percent of the magnetic exchange couplings within the Co layers themselves, a tiny number of, for example, pin holes through the Cu layers, perhaps at grain or twin boundaries, could easily result in large ferromagnetic coupling sufficient to overwhelm the much weaker indirect antiferromagnetic coupling. This may occur either over small regions or over the whole sample depending on the distribution and nature of the magnetic bridges. In particular, if the bridges are far apart compared to widths of possible domain walls, then there may be portions of the sample that remain AF coupled, while other portions are ferromagnetically bridged. Under these circumstances, the antiferromagnetic coupling strength derived from the fields required to saturate the resistance or magnetization may not be sensitive to the presence of the ferromagnetically coupled regions. On the other hand, the magnetoresistance is expected to scale approximately with the fraction of the sample that is antiferromagnetically coupled. The largest saturation magnetoresistance observed in uhv-evaporated superlattices grown on sapphire (see Fig. 3) was 40% at 3.5 K and 26% at 295 K. However taking into account the fact that only 30% of the sample is AF coupled, we conclude that if there were 100% AF coupling, the saturation magnetoresistance of the uhv-evaporated Co/Cu superlattices would be approximately 3.3 times higher and thus comparable to the largest MR found in sputtered Co/Cu multilayers.[2]

Figure 4 shows the dependence of saturation magnetoresistance on Cu layer thickness for a series of uhv-evaporated samples prepared on (0001) sapphire. The Cu layer thickness was inferred from x-ray-diffraction studies and electron microprobe determination of the Co-Cu composition. Figure 4 shows evidence for the first antiferromagnetic oscillation at a Cu thickness of ≈ 8–9 Å, as found for sputtered Co/Cu multilayers [see Fig. 1(a)]. As the Cu thickness is further increased, the MR becomes very large for Cu layer thicknesses of ≈ 35 Å. Although the data in Fig. 4 suggest the possibility of additional oscillations in MR, related experiments on (111) Co/Cu multilayer wedges,[15] uhv evaporated under similar growth conditions, show a smooth variation in MR with Cu thickness with evidence for a single MR oscillation corresponding with the first AF peak. The position of this peak is similar to that shown in Fig. 4. For thicker Cu layers a broad maximum in MR of comparable magnitude to the MR at the first AF peak is found, centered at > 70 Å, for measurements at 4.2 K. However, at room temperature the MR is almost independent of Cu layer thickness for Cu layer thicknesses ranging from ~ 20 Å to > 75 Å.

Side by side with sapphire substrates, selected structures were grown on GaAs(100), MgO(100), Si(111), or SrTiO$_3$(110), in some cases employing alternative seed

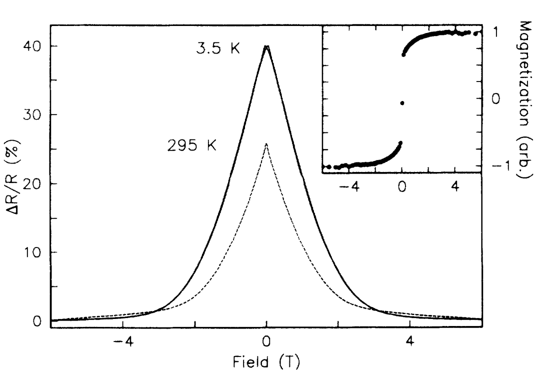

FIG. 3. Resistance vs in-plane field curves measured at 3.5 and 295 K for an uhv-evaporated Co/Cu superlattice of the form, sapphire(0001)/Pt(30 Å)/[Co(11 Å)/Cu(9 Å)]$_{30}$/Co(11 Å)/Pt(30 Å). The inset shows the normalized film magnetization vs in-plane field at 295 K. The saturation magnetization per Co volume is, within experimental error ($\approx 10\%$), that of bulk Co.

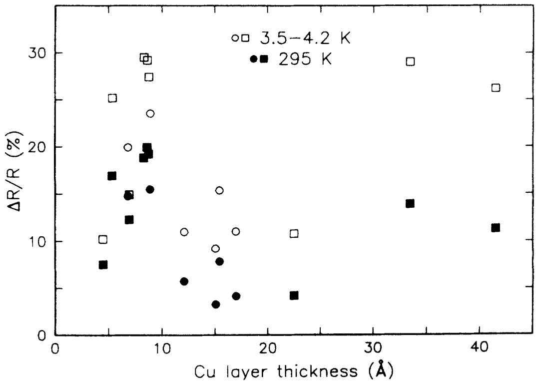

FIG. 4. Saturation magnetoresistance Cu spacer layer thickness, t_{Cu}, for a series of Co/Cu superlattices of the form sapphire(0001)/Pt(30 Å)/[Co(16 Å)/ Cu(t_{Cu})]$_{13}$/Co(16 Å)/Pt(30 Å), open (3.5 K) and filled (295 K) squares—resistance saturated by applying fields of up to 6 T; open (4.2 K) and filled (295 K) circles—resistance measured in fields of up to 16 kOe and almost completely saturated.

layers of thin Fe or Pt, and growth temperatures as low as 10 °C. This enabled the epitaxial growth of superlattices oriented along ⟨111⟩, ⟨110⟩, and ⟨100⟩, but typically these films showed little evidence for substantial AF interlayer coupling. However, growth of structures on GaAs(111)/Ag(40 Å)/Pt(30 Å) did lead to structures with AF coupling strength comparable to companion superlattices grown on sapphire but with lower MR.

Figure 5(a) shows the temperature-dependent resistance of the sample of Fig. 3 in zero field and at 5.5 T, a field much larger than the maximum saturation field. Figure 5(b) shows the difference, ΔR, between these two curves. The temperature dependence of ΔR is weak, in agreement with previous work on sputtered Co/Cu multilayers.[1] However, in contrast to the results on sputtered Co/Cu, ΔR decreases with decreasing temperature. The most likely explanation is the increased shunting of the sensing current through the relatively conducting Pt buffer and overlayers in these structures as the temperature is reduced. Indeed identical structures of the form, sapphire(0001)/Pt(t_{buffer})/[Co(16 Å)/Cu(7 Å)]$_{13}$/Co(16 Å)/Pt(30 Å) prepared on Pt buffer layer thicknesses, t_{buffer} of 30 and 300 Å, showed significantly different temperature dependences of $\Delta R/R$. For $t_{\mathrm{buffer}}=30$ Å, $\Delta R/R$ varied little from 11.0% at 300 K to 12.0% at 4.2 K, whereas for $t_{\mathrm{buffer}}=300$ Å, $\Delta R/R$ varied from 11.9% at 300 K to only 2.7% at 4.2 K even though the resistance decreased by only a factor of 2.0 in the former case but by a factor of 11.5 in the latter. Increased shunting of the current through the Cu spacer layers, whose resistance is expected to decrease significantly more than that of the Co layers as the temperature is decreased, may also lead to decreased magnetoresistance at lower temperatures.

In summary we have shown that highly oriented (111) Co/Cu superlattices epitaxially grown on Pt buffer layers on (0001) sapphire display substantial saturation magnetoresistance, exceeding 40% at 3.5 K, and very large indirect antiferromagnetic interlayer coupling strengths. These properties oscillate with Cu-layer thickness in a manner similar to that for sputtered polycrystalline Co/Cu multilayers. However, uhv-evaporated (111) Co/Cu superlattices of similar structural perfection grown epitaxially on GaAs show little evidence for substantial AF coupling. Moreover, for those structures displaying AF coupling, the degree of antiferromagnetic coupling is incomplete, about 30%. These results suggest that the presence of AF coupling in these uhv-evaporated superlattices is readily obscured by the possibility of structural defects, which result in direct ferromagnetic coupling between successive Co layers. The very large AF coupling strengths observed in the AF coupled regions of the superlattice as compared with those observed in sputtered multilayers suggest that the coupling strength is increased as the degree of crystalline orientation is increased. Such conclusions are inconsistent with conjectures that there is no coupling in [111] oriented Co/Cu multilayers.

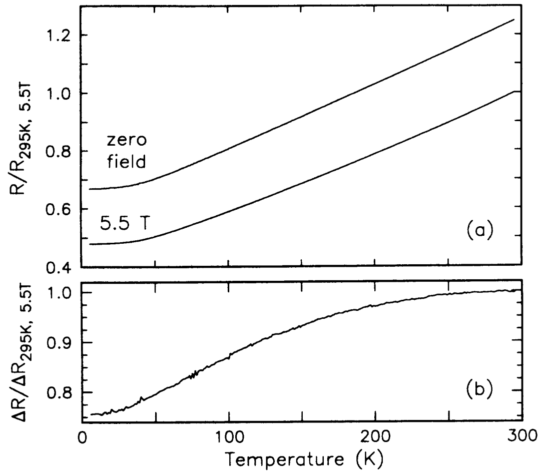

FIG. 5. (a) Resistance vs temperature in zero field (upper curve) and 5.5 T (lower curve) normalized to 1 at 295 K and 5.5 T, for the sample shown in Fig. 3. (b) Difference between the two curves of (a) normalized to 1 at 295 K.

ACKNOWLEDGMENTS

We thank K. P. Roche for technical support and D. Alvarez and G. Gorman for x-ray characterization. We thank C. Chappert and F. Herman for useful discussions.

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[9] Resistance measurements were made with a standard low-frequency lock-in technique using a four in-line contact geometry with gold-plated pressure contacts. The magnetoresistance is measured with respect to the sample resistance in high magnetic field. The saturation magnetoresistance is the highest resistance in the experimental field range normalized with respect to the resistance at the highest magnetic field used.

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[12] For these experiments we chose to grow the sputtered films on nonferromagnetic Ru rather than magnetic Fe buffer layers to avoid the latter complicating the interpretation of MR and magnetization studies.

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[14] The incomplete, yet large, AF coupling is more readily observed in MR than in magnetization measurements. Where it is not possible to make MR studies, e.g., for growth on single crystals or very thick buffer layers, partial AF coupling might be overlooked.

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