Oscillatory Magnetic Exchange Coupling through Thin Copper Layers

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Confirming theoretical predictions more than 25 years old, we show that Co slabs are indirectly exchanged coupled via thin Cu layers with a coupling that alternates back and forth between antiferromagnetic and ferromagnetic. Four oscillations are observed with a period of $\simeq 10$ Å. Moreover, the antiferromagnetically coupled Co/Cu superlattices exhibit extraordinarily large saturation magnetoresistances at 300 K of more than 65%.

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Magnetic coupling between isolated $3d$ transition-metal ions in paramagnetic host metals via the spin polarization of the conduction electrons is well established.[1-3] In particular, various properties of spin glasses such as *Cu*Mn can be accounted for by an exchange coupling that oscillates in sign from ferromagnetic to antiferromagnetic depending on the separation of the magnetic ions.[3] The period of the oscillation is very short and for simple nearly-free-electron metals is of the order of the Fermi wavelength. However, in contrast, the nature of the exchange coupling between ferromagnetic slabs separated by paramagnetic transition metals has long been controversial. Both recent experiments[4] and studies dating back to the 1960s[5] have showed ferromagnetic coupling decaying monotonically with increasing separation of the magnetic slabs for a wide range of systems. While early models[6] purported to explain this nonoscillatory decay, later theories show rather generally that an oscillatory decay of the RKKY (Ref. 7) form is expected.[8] Recently, we observed oscillating magnetic coupling between Co layers separated by thin Ru layers and Fe or Co layers separated by Cr layers.[9] Ru, however, is a relatively complex $4d$ transition metal and Cr, one of the most unusual transition metals with a complicated antiferromagnetic spin-density-wave ground state.[10] In this paper we present evidence for an oscillatory indirect magnetic exchange interaction in one of the most nearly-free-electron-like transition metals, copper. This is directly manifested as giant oscillations in the saturation magnetoresistance of Co/Cu superlattice structures as the Cu layer thickness is varied. Indeed these structures show the largest saturation magnetoresistance values yet found in any ferromagnetic system, attaining values of more than 65% at room temperature. While the observed oscillation period is short at only $\simeq 10$ Å, nevertheless, the period is about twice as long as the Fermi wavelength of copper and thus surprisingly inconsistent with the expected period within the simplest RKKY model.

The structures were prepared in a dc magnetron sputtering system at an argon pressure of 3.25 mTorr at a deposition rate of 2 Å/sec. The films were grown on chemically etched Si(100) and Si(111) wafers at $\simeq 40\,^\circ$C. Series of up to twenty structures at a time were grown via computerized control of shutters. The films are polycrystalline with grain sizes of about 200 Å as determined from scanning-tunneling-microscopy images. Auger depth profiles of representative samples showed no significant ($\lesssim 1$ at.%) oxygen or carbon contaminants within the film structure, although as expected oxygen was found at the silicon/film interface. The layer thicknesses were primarily determined by using a surface profilometer to measure the thickness of nominally 1000-Å single-layer samples of the film constituents prepared at the same time. The superlattice periods determined in this way were in good agreement ($\simeq 10\%$) with those found from x-ray-diffraction studies. There is the possibility that strain in the film structures will systematically modify the actual thickness of the layers in the superlattice from the nominal values. X-ray-diffraction data show that both the Cu and thin ($\lesssim 20$ Å) Co layers are fcc and are predominantly (111) textured.

Copper-based superlattice structures have been extensively studied in the past.[11,12] Recently, evidence for antiferromagnetic coupling in single-crystal fcc (100) Co/Cu/Co superlattices[13,14] grown on Cu(001) and bcc (001) Fe/Cu/Fe trilayers[15] grown on Ag(001) has been found for Cu layers 5–8 and 10 monolayers thick, respectively. In each case a single antiferromagnetic region was observed with evidence for a crossover from[15] or to a ferromagnetic[14] region. We have prepared extensive sets of both Fe/Cu and Co/Cu superlattices for a wide range of Fe, Co, and Cu layer thicknesses. For the Fe/Cu structures we find antiferromagnetic coupling but only for structures containing ultrathin Fe layers a few angstrom thick. In contrast, the Co/Cu structures show evidence for antiferromagnetic coupling for Co layer thicknesses ranging from just 2.5 to more than 200 Å. For Co/Cu superlattices grown on 50-Å-thick Cu buffer layers with $\simeq 10$ Å Cu spacer layers, we find magnetic hysteresis loops similar to those reported for comparable single-crystal Co/Cu superlattices.[13] In particular, there is a large remanent magnetization in zero field which we

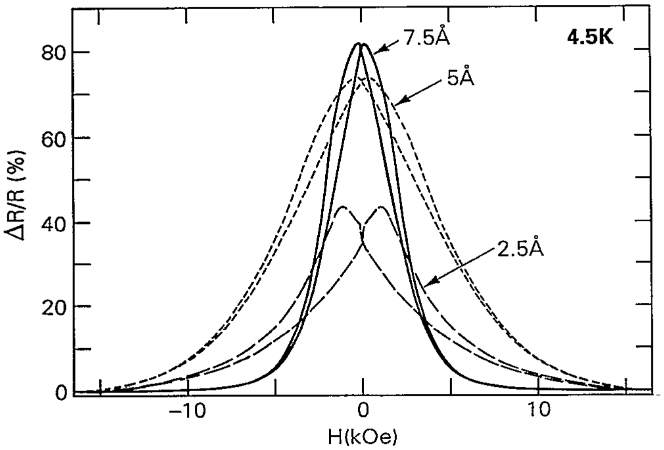

FIG. 1. Transverse magnetoresistance vs in-plane field for three superlattices of the form Si/Fe(40 Å)/[Co(t_{Co})/Cu(9.3 Å)]$_{16}$/Cu(19 Å) for Co layer thicknesses of 2.5, 5, and 7.5 Å at 4.5 K.

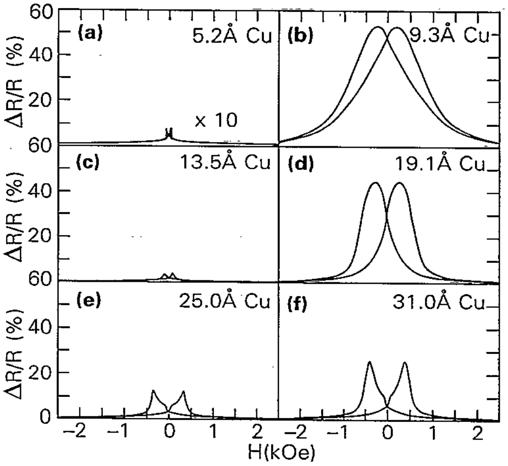

FIG. 2. Transverse magnetoresistance vs in-plane field curves for six representative samples from a series of superlattices with structures Si/Fe(40 Å)/[Co(10 Å)/Cu(t_{Cu})]$_{16}$ for Cu layer thicknesses varying from 5.2 to 31 Å.

suggest indicates partial contact of successive Co layers because of rough interfaces. For perfect antiferromagnetic coupling between identical magnetic layers, one expects almost no remanent magnetization as seen in Fe/Cr (Refs. 9, 16, and 17) and Co/Cr and Co/Ru (Ref. 9) antiferromagnets. Remarkably, we have found that by growing otherwise identical Co/Cu superlattice structures on Fe buffer layers ($\gtrsim$40 Å thick) the remanent magnetization of the Co layers becomes practically zero, indicative of almost 100% antiferromagnetic coupling. Indeed, x-ray and cross-section transmission-electron-microscopy studies give evidence for well-defined layers with Fe buffer layers but poor layering for Cu buffer layers.[18]

As first found for single-crystal Fe/Cr multilayers[16,19] and subsequently in similar polycrystalline structures,[9] antiferromagnetic Fe/Cr structures show anomalously large negative magnetoresistance. The resistance of the structure is intimately related to the magnetic state of the structure and saturates when a field large enough to orient all the magnetic layers in the structure parallel to one another is applied. We find that coupled Co/Cu superlattice structures form a second giant magnetoresistance system with saturation magnetoresistance values even larger than those found in Fe/Cr. Figure 1 shows typical magnetoresistance data for three structures of the form Si/Fe(40 Å)/[Co(t_{Co})/Cu(9.3 Å)]$_{16}$/Cu(19 Å) for Co layer thickness of 2.5, 5, and 7.5 Å at 4.5 K. The data are taken with the sensing current and the magnetic field in the plane of the film and arranged orthogonal to one another. The magnetoresistance is referenced to the saturation value of the resistance at high field. Even larger magnetoresistance values are obtained for superlattice structures containing more bilayers and higher resistance capping layers. As the Co layer thickness is increased above $\simeq$10 Å the magnitude of the saturation magnetoresistance decreases approximately as the inverse cobalt layer thickness. For this reason detailed studies of the dependence of exchange coupling on Cu thickness were carried out for structures containing thin Co layers. Figure 2 shows representative transverse magnetoresistance versus in-plane field data at 4.5 K for six Co/Cu superlattices containing 10-Å Co layers with Cu layer thicknesses ranging from 5.2 to 31 Å. As shown in the figure the magnitude of the magnetoresistance clearly oscillates from small to large values with increasing Cu layer thickness. Figure 3 shows a compliation of data for more than forty samples. Four well-

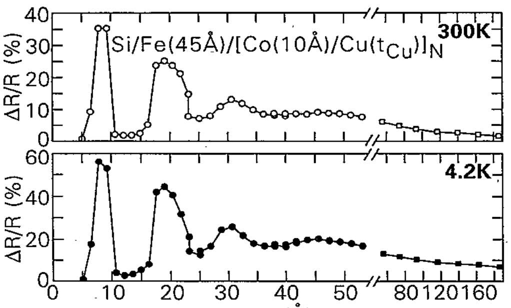

FIG. 3. Dependence of saturation transverse magnetoresistance on Cu spacer layer thickness for a family of related superlattice structures of the form Si/Fe(45 Å)/[Co(10 Å)/Cu(t_{Cu})]$_N$. An additional Cu layer was deposited on each film structure such that the uppermost Cu layer was $\simeq$55 Å thick. The number of bilayers in the superlattice, N, is 16 for t_{Cu} below 55 Å (●,○) and 8 for t_{Cu} above 55 Å (■,□). Values of $\Delta R/R$ are highly reproducible, within $\simeq \pm 5\%$ of $\Delta R/R$, as evidenced by multiple sample points at $t_{\mathrm{Cu}}=25$, 38, 40, and 42 Å.

defined oscillations in the saturation magnetoresistance are seen at both 4.2 and 300 K. Similar oscillations with the same period are seen for structures containing thicker Co layers and for asymmetric structures containing alternating thick and thin Co layers. In each case the period of the oscillation is about 10 Å but the period increases slightly with increasing Cu thickness. Perhaps not surprisingly the width of the oscillations also increases. The interlayer exchange coupling decreases rapidly as the Cu layer thickness is increased, but remarkably large magnetoresistance values persist for Cu layers more than 200 Å thick, well beyond the point at which the oscillations are washed out. Indeed, as the Cu thickness is increased, as shown in Figs. 2(e) and 2(f), the shape of the resistance curves change such that for thicker Cu layers the resistance displays sharp peaks at $\pm H_c$, where H_c is the coercive field of the Co layers. A likely explanation is that magnetic domains in adjacent Co layers, randomly arranged with respect to one another, give rise to significant amounts ($\simeq 50\%$) of antiparallel Co regions when the magnetization of the structure passes through zero at $\pm H_c$. Indeed, "uncoupled" Co layers in Co/Au/Co and Co/Cu/Co trilayers, purposely manipulated to have their magnetic moments arranged antiparallel, show enhanced magnetoresistivity although these effects are much smaller than reported here.[20,21]

The magnetization of the Co layers is in plane at room temperature and the moment of the layers is close to that of bulk Co. Magnetization loops show oscillations in saturation field consistent with the magnetoresistance data. From such magnetization loops it is not possible to measure ferromagnetic coupling nor to distinguish ferromagnetic coupling from the absence of any coupling. However, we have recently shown that the coupling does change sign, oscillating from antiferromagnetic to ferromagnetic, by using specially engineered sandwich structures. One of the magnetic layers in the sandwich is pinned, either by direct exchange coupling to an antiferromagnetic $Fe_{1-x}Mn_x$ layer or by indirect exchange coupling through an ultrathin Ru layer to an additional magnetic layer.[22] For structures containing identical magnetic layers of thickness t_F and saturation magnetization M_s the saturation field H_s is given by $H_s = -4J_i/M_s t_F$, where J_i is the antiferromagnetic interlayer exchange coupling strength. From the largest saturation field at the peak of the first antiferromagnetic oscillation we estimate $J_i \simeq 0.15$ erg/cm^2 in Co/Cu, about 30 times smaller than the largest effect in Co/Ru (Ref. 9) and more than 10 times smaller than that found in Fe/Cr,[9,16] but approximately the same size as that reported in single-crystal Fe/Cu/Fe trilayers.[15] The period of the oscillation is about 20% smaller than that of Co/Ru and about half that of Fe/Cr.[9] There appears to be no obvious correlation between these periods and those expected from the Fermi surfaces[23] of the bulk metals within RKKY theory.[7] For Cu, note that most wave vectors spanning the belly of the Cu Fermi surface[23] would give rise to much shorter oscillation periods than we find, whereas wave vectors spanning the [111] necks of the Cu Fermi surface would give rise to longer oscillation periods, closer to that observed. One might speculate that the band structures of the thin spacer layers are significantly different from the corresponding bulk material. However, the fact that we observe periodic oscillations out to thirty or more Cu layers makes this unlikely. As shown in Fig. 4 the interlayer exchange coupling is rather insensitive to temperature decreasing by only 20% between 4.2 and 400 K for a Si/Fe(40 Å)/[Co(10 Å)/Cu(9.3 Å)]$_{16}$/Cu(19 Å) structure. Moreover, although the saturation magnetoresistance $\Delta R/R$ drops substantially over the same temperature range, this is simply accounted for by the temperature dependence of the high-field resistance. The absolute change in sheet resistance $\Delta R_\square$ with field is almost independent of temperature. These results indicate that the magnetic exchange coupling and the giant magnetoresistance are electronic in origin and therefore insensitive to temperature, but a detailed theory is currently lacking.

In summary, we have presented evidence of the first observation of long-range oscillations in the indirect magnetic exchange coupling of ferromagnetic slabs via Cu. Oscillations with a period of $\simeq 10$ Å are observed for Cu layers up to 50 Å thick. The existence of such oscillations is consistent with simple RKKY models and with related models including possible space quantization of the electrons in the copper layers.[24]

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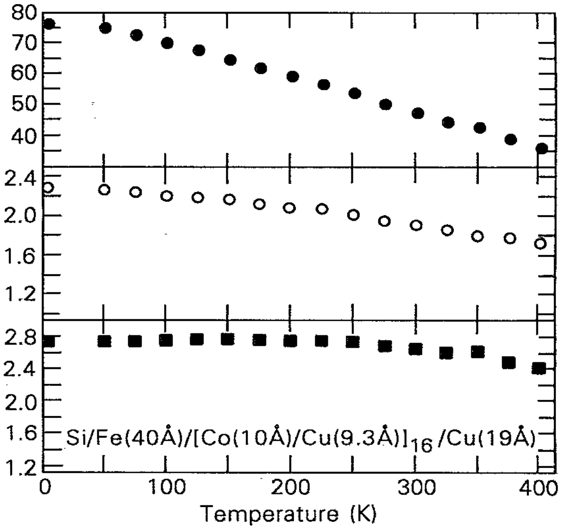

FIG. 4. Temperature dependence of the saturation magnetoresistance, saturation field, and sheet magnetoresistance for the superlattice Si/Fe(40 Å)/[Co(10 Å)/Cu(9.3 Å)]$_{16}$/Cu(19 Å).

tometer and we thank him and many colleagues at the Almaden Research Center for very useful discussions.

Note added.–Since we submitted this paper we have received several preprints concerning exchange coupling in Fe/Cu and Co/Cu multilayers complementary to the work described in this Letter.[25-28]

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