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Spin engineering: Direct determination of the Ruderman-Kittel-Kasuya-Yosida far-field range function in ruthenium

S. S. P. Parkin

IBM Research Division, Almaden Research Center, 650 Harry Road, San Jose, California 95120-6099

D. Mauri

IBM Storage Systems Products Division, 5600 Cottle Road, San Jose, California 95193

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Oscillations in both *ferromagnetic* and antiferromagnetic indirect exchange coupling through Ru are directly measured. Ferromagnetic coupling is determined by *spin engineering* multilayered structures comprised of trilayers of $Ni_{80}Co_{20}$/Ru/$Ni_{80}Co_{20}$ in which one of the $Ni_{80}Co_{20}$ layers is strongly antiferromagnetically pinned to a Co layer. Oscillations in antiferromagnetic exchange coupling are measured in companion $Ni_{80}Co_{20}$/Ru multilayers. The dependence of the exchange coupling strength on Ru thickness is well described by the Ruderman-Kittel-Kasuya-Yosida far-field range function, providing a direct measurement of this function through a transition metal.

Exchange coupling of ferromagnetic transition-metal layers through nonferromagnetic metallic layers has become a field of intense interest recently.[1-9] Oscillations in the antiferromagnetic (AF) interlayer exchange coupling between thin Fe, Co, Ni, or Ni alloy layers separated by a variety of spacer layers such as Cr and Ru,[6] as well as Cu,[8-10] have been found as the spacer layer thickness is increased. A variety of theoretical models have been proposed to account for these results.[11-15] Some of these models including those based on space quantization of electrons of different spin types in the spacer layer[13] conclude that the interlayer exchange coupling strength oscillates through zero changing sign back and forth from antiferromagnetic to ferromagnetic (*F*) in a manner similar to that predicted in the well-known Ruderman-Kittel-Kasuya-Yosida[16] (RKKY) model. However, in other models it is argued that although the magnitude of the interlayer coupling oscillates, it actually remains[14] or may remain[15] of the same sign, either antiferromagnetic or ferromagnetic, independent of the spacer-layer thickness.

Presently the experimental situation is unclear, partly because it is more difficult to measure ferromagnetic than antiferromagnetic interlayer exchange coupling. The latter is easily determined in multilayered or sandwich structures from the field dependence of the magnetization. The AF interlayer exchange coupling is simply related to the strength of the magnetic field required to overcome the AF coupling between successive magnetic layers so as to align all the magnetic layers parallel to one another. However, clearly this method cannot determine the coupling if it is ferromagnetic since the magnetic layers will prefer to be aligned parallel to one another even in zero field. The first observation of oscillatory AF interlayer exchange coupling[6] was carried out on Fe/Cr and Co/Ru multilayers using this method. Unfortunately the high coercivity of the individual Fe or Co layers obscures whether or not the antiferromagnetic coupling actually goes to zero.

A number of other more sophisticated techniques have been used to study interlayer exchange coupling. These include Brillouin light scattering (BLS),[17,18] ferromagnetic resonance[5] (FMR), and spin-polarized low-energy electron diffraction (SPLEED).[2,19] In BLS and FMR the coupling strength is deduced from its effect on the measured frequency of excited spin-wave modes. Although coupling strengths of both signs can be found, such data can be complicated to interpret and the sensitivity of such techniques is limited to relatively large interlayer exchange coupling strengths. SPLEED has been used in the following manner. By taking advantage of its extreme surface sensitivity the direction of magnetization in remanence of the topmost layer of a previously magnetized asymmetric sandwich structure is determined relative to that of the lower layer. However, since the measurement is restricted to zero field the magnitude of the interlayer coupling cannot be determined and the existence of ferromagnetic coupling can only be inferred.

In this paper we use a method to directly determine the strength of ferromagnetic exchange coupling through metallic spacer layers sandwiched between soft ferromagnetic layers by pinning the magnetization of one of the magnetic layers antiparallel to the applied magnetic field. The pinning is accomplished by an additional magnetic layer strongly antiferromagnetically coupled to the back of one of the soft layers through a second thin metallic layer. Paradoxically the magnetic moments of the two soft layers become antiparallel on application of a field. We find evidence for both ferromagnetic interlayer exchange coupling and oscillations in the strength of this coupling for ruthenium spacer layers.

The samples were prepared in a high-vacuum sputtering system ($\approx 10^{-9}$ Torr) from high-purity dc magnetron sources. The deposition rate was 2 Å/sec in 3.3×10^{-3} Torr argon. The samples were deposited at approximately 50 °C on chemically polished Si(100) wafers. Deposition rates were monitored by quartz-crystal microbalances. Layer thicknesses were determined from the measured thicknesses of nominally 1000-Å-thick calibration films made at the same time as the multilayers. Up to 19 samples were prepared sequentially via computer control of source shutters and substrate platform. Selected samples were studied *ex situ* by Auger sputter depth profile analysis and no evidence for oxygen or carbon contamination above the Auger detection limits ($<\simeq 1\%$) was found.

Ruthenium is used for the spacer layer in the experiments discussed here. Ru forms an ideal spacer material since oscillatory antiferromagnetic interlayer exchange coupling is found for Ru sandwiched between Co layers[6] as well as between Fe, Ni, $Ni_{80}Fe_{20}$, and $Ni_{80}Co_{20}$ layers or any combination of these magnetic materials.[20] As discussed below by taking advantage of the widely varying strengths of antiferromagnetic exchange coupling between these different magnetic layers and the different degree of AF exchange coupling in the limit of ultrathin Ru layers we can *spin engineer* in a highly predictable manner structures with complex magnetic arrangements of the layers.

For these experiments coupling between $Ni_{80}Co_{20}$ layers was studied since layers of this material are readily saturated in low fields of a few oersted. Multilayers of the form shown schematically in Fig. 1 were prepared. The interlayer exchange coupling constant, J_{12}, is measured between two $Ni_{80}Co_{20}$ layers, of thickness, t_F, separated by a Ru spacer layer of variable thickness, t_S. The essential architecture of the structure is that, additionally, one of the $Ni_{80}Co_{20}$ layers, *F* I, is antiferromagnetically coupled via a second thin Ru layer of thickness, t_P, of just a few angstroms, to a third magnetic layer, in this case cobalt. The coupling between Co and $Ni_{80}Co_{20}$ via Ru is several times larger than the coupling between two $Ni_{80}Co_{20}$ layers via Ru for equivalent Ru thicknesses.[20] Moreover Co is AF coupled to $Ni_{80}Co_{20}$ in the limit of ultrathin Ru layers with a coupling strength that rapidly increases as the Ru-layer thickness is decreased to the point ($\simeq 3$ Å) at which direct coupling through pin holes in the Ru layer overwhelms the AF coupling. Consequently, the $Ni_{80}Co_{20}$ layer, *F* I, is extremely strongly antiferromagnetically coupled to the Co layer. In contrast, the AF coupling between the $Ni_{80}Co_{20}$ layers in the same limit is very small. Finally, the thickness of the Co layer is chosen such that the magnetic moment of the Co layer is approximately equal to the sum of the magnetic moments of the two $Ni_{80}Co_{20}$ layers. Under these circumstances, neglecting anisotropy, the net moment of the structure will be approximately zero in zero field for ferromagnetic J_{12} (see Fig. 1).

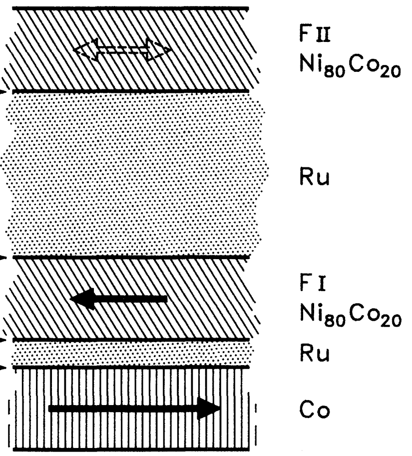

FIG. 1. Schematic diagram of sample structure. The exchange coupling, J_{12}, between two $Ni_{80}Co_{20}$ layers is measured by pinning the moment of one of the $Ni_{80}Co_{20}$ layers (*F* I) antiparallel to a Co layer. The moment of the Co layer is set equal to the sum of the moments of the two $Ni_{80}Co_{20}$ layers.

Figure 2 shows magnetic hysteresis loops for four structures of the form Si/Ru (85 Å)/[Co (15 Å)/Ru (6 Å)/$Ni_{80}Co_{20}$ (15 Å)/Ru(t_S)/$Ni_{80}Co_{20}$ (15 Å)]$_5$. The $Ni_{80}Co_{20}$ layers and Co layers are each $\simeq 15$ Å thick and t_S was varied from 3 to 33 Å. The structures contain five identical repeats of the five-layer unit shown in Fig. 1 separated from each another by a thick Ru layer, $\simeq 85$ Å thick. There is negligible exchange coupling through Ru layers more than $\simeq 60$ Å thick.[20] The Ru-layer thickness of $\simeq 6$ Å between the pinning Co layer and the $Ni_{80}Co_{20}$ *F* I layer was deliberately chosen such that the field required to align these layers was readily obtained with a small electromagnet. As shown in Fig. 2 this field is $\simeq 8$ kOe. The magnetic hysteresis loops are consistent with the expected spin arrangement shown in Fig. 1 and directly give evidence for ferromagnetic J_{12} for Ru-layer thicknesses near 3, 13, and 26 Å. In particular, as shown in Fig. 2, for

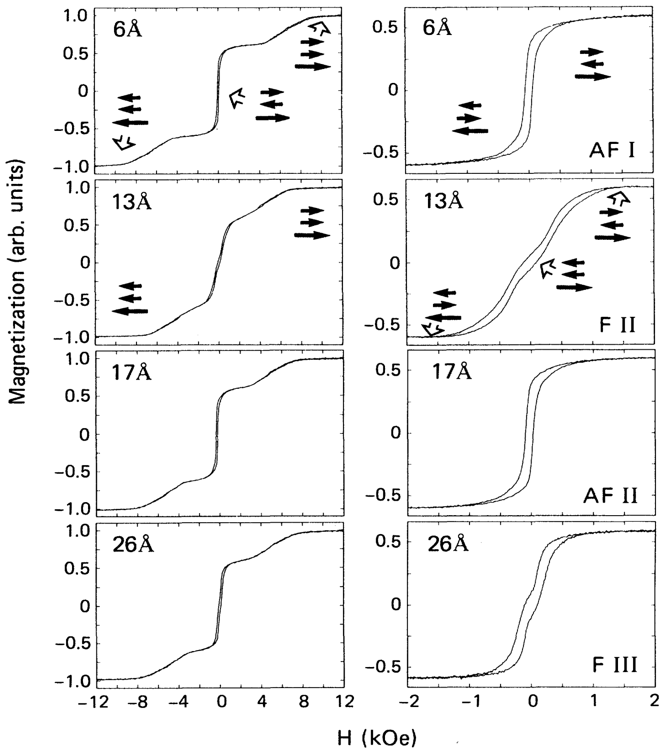

FIG. 2. Magnetization vs field curves for four samples of the form Si/Ru (85 Å)/[Co (15 Å)/Ru (6 Å)/$Ni_{80}Co_{20}$(15 Å)/Ru(t_S)/$Ni_{80}Co_{20}$ (15 Å)]$_5$ for $t_S = 6$, 13, 17, and 26 Å. The low-field data is shown in more detail on the right-hand side of the figure. Data are shown for representative samples from the first and second antiferromagnetic regions and second and third ferromagnetic regions. The arrangement of the moments of the Co and $Ni_{80}Co_{20}$ *F* I and *F* II layers as the field is varied is shown schematically for *F* and AF coupled samples.

these Ru-layer thicknesses the magnetic hysteresis loops at low fields exhibit a characteristic shape requiring the application of a field of up to 1.3 kOe to reach the intermediate plateau in magnetization found in all the samples. This plateau at approximately half the total moment of the structure is consistent with parallel alignment of *F* II and the Co layer. For intermediate Ru thicknesses the plateau is attained in much smaller fields determined by the magnetic coercivity of the magnetic layers, consistent with antiferromagnetic J_{12} (see Fig. 1). The magnitude of the AF coupling was directly measured from the saturation field of a second series of simple bilayer multilayers of the form $[\mathrm{Ni_{80}Co_{20}}\ (30\ \text{Å})/\mathrm{Ru}(t_{\mathrm{Ru}})]_{20}$. The strength of the ferromagnetic and antiferromagnetic interlayer exchange coupling is thus given, respectively, by $2n_i|J_{12}| = H_S M t_F$, where H_S is the field required to attain the plateau in the spin-engineered structures and complete saturation in the bilayer multilayers. The coefficient n_i is 1 and 2, respectively, for these different structures, since each $\mathrm{Ni_{80}Co_{20}}$ layer is coupled to just one $\mathrm{Ni_{80}Co_{20}}$ layer in the spin-engineered multilayers, but two in the bilayer multilayers (neglecting end effects in the latter[7]).

Values of J_{12} determined from the saturation field as described above (corrected for coercivity) are plotted versus Ru-layer thickness for both series of structures in Fig. 3. The exchange coupling is clearly demonstrated to oscillate through zero. Moreover as shown in Fig. 3 the dependence of J_{12} is well described by a RKKY-like exchange coupling of the form, $J_{12} \propto \sin(\phi + 2\pi t_{\mathrm{Ru}}/\lambda_F)/t^p_{\mathrm{Ru}}$, where $p \simeq 1.8$ and $\lambda_F \simeq 11.5$ Å. The value of p is in good agreement with theoretical predictions of 2 for the planar geometry.[21] Although the value of λ_F is much longer than the Fermi wavelength for Ru, λ_F will be determined by the detailed shape of the Fermi surface[22] which will inevitably give rise to longer length scales.

These measurements provide a direct determination of the RKKY range function in a transition metal. Previously, NMR studies of dilute magnetic impurities in nonmagnetic metallic hosts have been used to indirectly measure the RKKY range function.[23,24] However, these studies are limited by interference from the randomly spaced impurities.

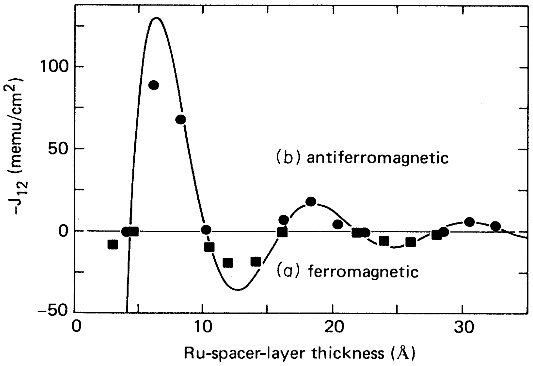

FIG. 3. Interlayer exchange coupling strength, J_{12}, for coupling of $\mathrm{Ni_{80}Co_{20}}$ layers through a Ru spacer layer. J_{12} is defined per unit area of the interface and is determined from magnetization curves of structures of the form (a) Si/Ru (85 Å)/[Co (15 Å)/Ru (6 Å)/$\mathrm{Ni_{80}Co_{20}}$ (15 Å)/Ru(t_S)/$\mathrm{Ni_{80}Co_{20}}$ (15 Å)]$_5$ for *ferromagnetic* coupling, and (b) Si/Ru (105 Å)[$\mathrm{Ni_{80}Co_{20}}$ (30 Å)/Ru(t_{Ru})]$_{20}$/Ru (105 Å) for *antiferromagnetic coupling.* The data points are shown as (a) squares and (b) circles. For each structural type only (a) *ferromagnetic* or (b) *antiferromagnetic* coupling can be measured. Data points are not shown for structures for which no coupling could be determined. The solid line corresponds to a fit to the data of an RKKY form as described in the text.

In summary, by taking advantage of the different strengths and phase of indirect exchange coupling through Ru we have spin-engineered magnetic layered structures to directly measure both the antiferromagnetic and ferromagnetic coupling through ruthenium. The exchange coupling is shown to oscillate about zero changing sign back and forth from ferromagnetic to antiferromagnetic. The dependence of the strength of the coupling on Ru thickness is well described by a RKKY-like range function. These results are in disagreement with recently proposed models of exchange coupling through transition metals,[14] but are in agreement with standard RKKY (Ref. 21) and related theories.[13]

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