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Preserved interfacial magnetism and giant antiferromagnetic exchange coupling in Co/Rh sandwiches

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Abstract. – The interlayer coupling in Co/Rh/Co sandwiches prepared by UHV evaporation has been investigated by means of magnetization and transport experiments. We found that the antiferromagnetic coupling strength for thin Rh layers is the largest ever obtained in magnetic systems, reaching approximately 34 erg/cm^2 for 5 Å thick Rh interlayer. This value is 7 to 8 times larger than the highest value previously observed in the Co/Ru system prepared under similar conditions. This unexpectedly large difference in the coupling strength between these two systems is mainly due to the magnetic nature of the interfaces. This is further supported by *ab initio* calculations of the magnetic moments in superlattices with mixed interfaces.

The discovery of the interlayer oscillatory exchange coupling between two ferromagnetic layers separated by a nonmagnetic spacer layer in magnetic multilayers has recently attracted considerable attention [1], [2]. All theories agree in describing the interlayer exchange coupling in terms of quantum interferences due to electronic confinement in the spacer layer, and that the oscillation periods are determined by extremal spanning vectors of the spacer Fermi surface (FS) [3], [4]. The strength and the phase of the coupling depend both on the geometry of the FS and on the reflection amplitudes for electrons scattered from the interfaces between the spacer layers and the magnetic layers [5]. Mathon *et al.* [3], by using detailed tight-binding calculations of the coupling, have shown that a mismatch between the ferromagnetic and spacer layer d bands, which increases with an increasing number of holes in the spacer, leads to a systematic variation of the coupling strength across the transition metal series. This is in agreement with experimental results reported by Parkin [6], which show that the exchange-coupling strength increases systematically from the 5d to 4d to 3d metals and exponentially with increasing number of d electrons along each period.

The Co/Ru system has been found to present a very large antiferromagnetic coupling strength, the largest ever found in metallic multilayers, of the order of 6 erg/cm^2 (for $t_{\mathrm{Ru}} = 6$ Å) for UHV grown superlattices [7]. Moreover, sputtered multilayers of Co/Ru multilayers have shown dramatic differences in their coupling strength, as revealed by the values obtained, respectively, by Parkin *et al.* [2] (5 erg/cm^2 for Ru thickness $t_{\mathrm{Ru}} = 3$ Å) and Bloemen *et* *al.* [8] (0.5 erg/cm^2 for $t_{Ru} = 4$ Å). These results show the sensitivity of the coupling strength to the preparation conditions, *i.e.* to the spacer layer structure and to the quality of the magnetic/nonmagnetic interfaces.

The crucial point of this letter is to show for the first time correlation between *the exchange coupling strength and the nature of the magnetic/nonmagnetic interfaces.* The large difference in coupling strengths between Co/Rh and Co/Ru for very thin spacer layers, "7-8 times larger in the Co/Rh system", makes these two systems particularly appropriate for this topic. Such large difference between these two systems cannot be attributed to the additional electron in the Rh "*d*" band but can only be explained by the nature of the magnetic/nonmagnetic interfaces. In fact, a strong reduction in the magnetism of the interfaces is observed and accounted for in terms of 2 Å thick magnetically dead Co at each interface for Co/Ru, while the magnetization is preserved at the Co/Rh interfaces. This result is further confirmed by first-principles electronic-structure calculations.

Co/Rh sandwiches were deposited on freshly cleaved and *in situ* heated (700 °C) mica substrates using an UHV e-beam evaporator. A 100 Å thick epitaxial fcc (111) Rh buffer layer was grown at 700 °C to provide a smooth and clean single crystalline surface. After cooling the substrate to 0 °C, Co and Rh layers were subsequently grown and covered by a 24 Å thick Rh protection layer. A series of up to twelve samples was prepared with fixed Co thickness ($t_{Co} = 32$ Å) and Rh thicknesses ranging from 5 Å to 30 Å.

RHEED patterns obtained during the growth and scanned over the whole surface reveal high crystalline quality and show a close-packed (111) fcc structure of both Co and Rh layers. Nuclear magnetic resonance (NMR) provided clear evidence of the (111) fcc structure of both Co layers with the main resonance line lying at 216 MHz. Details of the growth and structural characterization of the sandwiches are reported elsewhere [9].

The resistance of the samples was measured using a four-probe, low-frequency ac lock-in technique with spring-loaded gold-plated contacts. All magnetoresistance data were measured at 300 K, with the magnetic field applied along the film plane and parallel to the current direction. Magnetization measurements were performed using alternating gradient force and SQUID magnetometries. The saturation field was defined as the field at which the resistance or magnetization curve first deviates from the high field slope.

Figure 1 shows the variation of the saturation field (H_S) measured at room temperature (RT) as a function of the spacer layer thickness for both Co/Rh and Co/Ru systems. The most interesting result shown in fig. 1 is the very large saturation field, which is close to 160 kOe, observed for the sample with a Rh spacer layer thickness of 4.8 Å [10]. The saturation field reflects the maximum strength of the antiferromagnetic interlayer exchange coupling J_{AF} as $J_{AF} = H_S M_S t_{Co}/2$, where M_S and t_{Co} are the saturation magnetization and thickness of the magnetic layer, respectively. A model allowing bilinear, biquadratic and anisotropic terms [11] was unsuccessful to fit the rounded magnetization and magnetoresistance curves (insert of fig. 1) in the case of the Co/Rh system, which implies that a complicated mechanism takes place related to contributions due to the distribution of AF coupling strengths. In this case, J_{AF} represents the maximum coupling value that can be reached in the sample. With these considerations and on the basis of the experimental data, the J_{AF} value found for this sandwich ($t_{Rh} = 4.8$ Å) is equal to 34 erg/cm^2 at 300 K. The mean value of the exchange coupling, determined from the following expression:

$$\langle J_{AF} \rangle = t_{Co} \int_0^{H_S} (M_S - M)\mathrm{d}H \,,$$

is found to be smaller by a factor of two and remains, however, much larger than the values obtained in Co/Ru sandwiches.

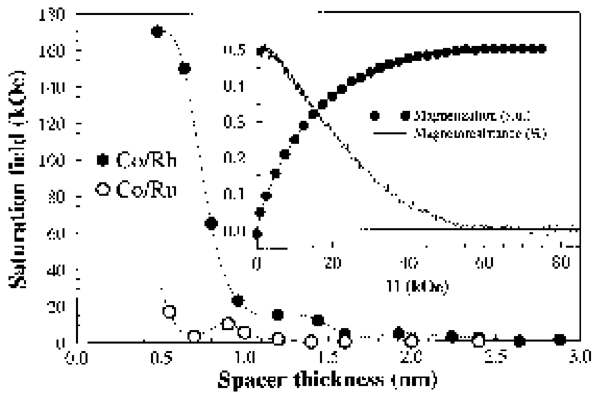

Fig. 1

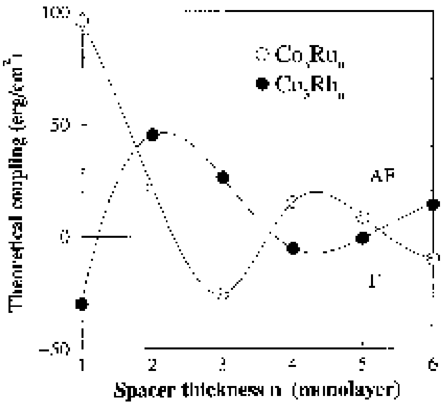

Fig. 2

Fig. 1. – Variation in the saturation field (H_S) of two series of twelve sandwiches, made respectively of Co/Rh and Co/Ru, as a function of Rh and Ru spacer layer thickness at 300 K. In the insert, magnetization and magnetoresistance curves for the Co(24)/Rh(7)/Co(24) sandwich are shown.

Fig. 2. – Variation in the theoretical exchange coupling as a function of the Rh and Ru spacer layer thicknesses.

Theoretical calculations predict much smaller differences on the exchange coupling strength between Co/Rh and Co/Ru systems, mainly related to the addition of only one "*d*" electron [3]. To have a more quantitative analysis of the exchange coupling for these systems, we have performed first-principles calculations based on the density functional formalism and the augmented spherical waves (ASW) method. We have calculated the exchange coupling for perfect hcp Co_3Ru_n and hcp Co_3Rh_n superlattices. Figure 2 shows the variation in the energy difference ΔE between ferromagnetic and antiferromagnetic arrangements as a function of the spacer layer thickness. In the calculated thickness range, ΔE shows similar period and amplitude, in contrast with the experimental data. It could be argued that the experimental difference in the coupling between Rh and Ru spacers is due to the fcc structure of the rhodium, while the previous calculations have all been performed with hcp structure. Additional calculations were then performed taking into account the fcc structure of the Co/Rh system [12]. As reported in table I, the energy values are even smaller in the fcc configuration. As a consequence, we conclude that the various theoretical approaches cannot reproduce the large experimental difference in the coupling between these two systems when perfect superlattices are considered.

To understand the origin of these differences, we compare experimental results of the magnetization with calculations of the magnetic moments for superlattices including atomic mixing at the interfaces. Figure 3 shows the variation in saturation magnetization per unit surface of Co, $M_S t_{Co}$, *vs.* cobalt thickness (t_{Co}) for both mica/Ru(150 Å)/Co(t_{Co})/Ru(50 Å) and mica/Rh(150 Å)/Co(t_{Co})/Rh(50 Å) single magnetic layers. The linear decrease in magnetization with decreasing t_{Co} is expressed by linear functions which intercept the abscissa at thicknesses of 4 Å and 0 Å for Ru/Co/Ru and Rh/Co/Rh single layers, respectively. This indicates that a strong reduction in the magnetization is observed in the Co/Ru system while no variation is detected in the Co/Rh case. Such reduced magnetization is a consequence of reduced magnetic moments of the cobalt atoms at the interfaces, which cannot be ascribed to sharp interfaces [13], but necessarily to the existence of dilute alloys at the Co/Ru interfaces. Alloying at the interfaces is consistent with NMR data, which have shown that intermixing at the

Table I. – *Experimental* $J_{\rm exp}$(erg/cm^2) *and theoretical* $J_{\rm th}$(erg/cm^2) *magnitude of the antiferromagnetic exchange coupling for* Co/Ru *and* Co/Rh *systems in both fcc and hcp configurations.*

	$J_{\rm th}$(erg/cm^2)	$J_{\rm exp}$(erg/cm^2)	Thickness (Å)	Reference
Co/Ru (hcp)	–	5 (4.2 K)	3	[6] (multilayers)
	–	0.5 (300 K)	4	[8]
	–	4.5 (300 K)	5	[12]
	95	–	2	This work
Co/Rh (hcp)	44	–	4	This work
Co/Rh (fcc)	28	–	4	This work
	–	34	5	This work
	–	1.6	7.9	[9]

Co/Ru interfaces occurs over three monolayers [11]. As a consequence, the magnetization profile can be approximated by two nonmagnetic atomic planes per Co layer, *i.e.* one magnetically dead Co monolayer per interface. In contrast, the experimental evidence of preserved magnetization in the Co/Rh system is not necessarily related to the existence of sharp interfaces. Arguments for diffused interfaces in the Co/Rh system are given by NMR data [9] as well as similarities between the phase diagram of CoRu and CoRh alloys [14].

Self-consistent band structure calculations of the magnetic moments on each site of the superlattices have been performed. We use the first-principle Augmented Spherical Wave technique and the Local Spin Density Approximation (LSDA) formalism for treating exchange and correlation of a many-body electron system. The periodic cell contains 32 nonequivalent atoms (4 atoms per in-plane cell) and the band structure was computed with 288 k-points in the irreducible part of the Brillouin zone. The accuracy on the magnetic-moment values was estimated to be better than $0.01\mu_{\rm B}$ [15]. To provide a proper comparison with the experimental results, we used mixed interfaces on the basis of the linear profile determined by NMR measurements on Co/Ru sandwiches [11] with the following structure:

$$\mathrm{Co/Co/Co/Co_{0.75}M_{0.25}/Co_{0.5}M_{0.5}/Co_{0.25}M_{0.75}/M/M}, \quad \text{where} \quad \mathrm{M = Ru\ or\ Rh}\,.$$

Figure 4 reports the magnetic-moment distribution as a function of the position of all inequivalent atoms in the stack. At this stage, several comments can be made:

i) The pure atomic cobalt planes are differently perturbed by the proximity of the interfaces. The magnetic moments of cobalt atoms having Ru neighbours are slightly reduced from the bulk value, while those of Co with Rh neighbours are preserved.

ii) For the first (I) with 75 at%Co, second (II) with 50 at%Co and third (III) with 25 at%Co mixed CoRu planes, large reduction in the cobalt magnetic moments approximately of the order of 12% (plane I), 40% (plane II) and 95% (plane III) is obtained. The situation is completely different in the case of Co with Rh neighbour. In contrast to the Ru case, the cobalt atoms keep their magnetic moments close to the bulk value for all Rh concentrations, even for plane III.

iii) In addition, the Rh atoms are highly polarized with magnetic moments up to $0.65\mu_{\rm B}$ for plane I and remain relatively large with fewer cobalt neighbours ($0.25\mu_{\rm B}$ for plane III). In contrast, the induced magnetic moment of the Ru atoms in plane I ($0.3\mu_{\rm B}$), consistent with the presence of three first cobalt neighbours, decreases very rapidly when the number of cobalt atoms decreases to reach zero for plane III.

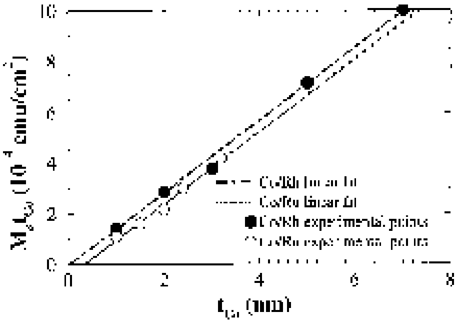

Fig. 3

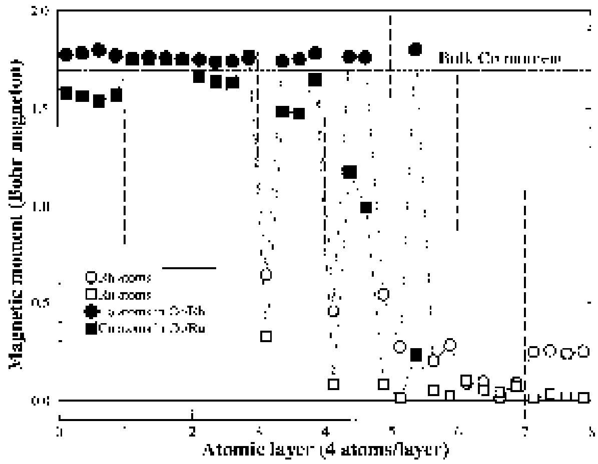

Fig. 4

Fig. 3. – Variation of the measured saturation magnetization per unit surface of Co, $M_S t_{Co}$, with Co layer thickness t_{Co} for both mica/Ru (150 Å)/Co (t Å)/Ru (50 Å) (open circles) and mica/Rh (150 Å)/Co (t Å)/Rh (50 Å) (solid circles) Co single layers at $T = 300$ K.

Fig. 4. – Evolution of the magnetic moment as a function of the position of the Co atoms in the following stack: $Co/Co/Co/Co_{0.75}M_{0.25}/Co_{0.5}M_{0.5}/Co_{0.25}M_{0.75}/M/M$, with M = Ru or Rh from *ab initio* calculations. The dashed line corresponds to the bulk Co moment value.

The strong difference between the magnetic nature of the Co/Rh and Co/Ru interfaces is probably the reason for the unexpectedly large difference in the interlayer exchange coupling strength between these two systems. Indeed, as we mentioned in the introduction, all theories describe the interlayer exchange coupling in terms of spin asymmetric electronic confinement in the spacer layer [3], [4]. The strength of the coupling is determined by the spin asymmetry $\Delta r = [r^{\uparrow} - r^{\downarrow}]/2$ of the electron wave function reflection amplitudes at each potential step [4] (the electrons of one spin type, the majority electrons of the Co are more strongly reflected). In the preasymptotic regime and in the case of polarization in the spacer layer—which is the case under consideration in this paper—band matching between two bulk-like wave functions cannot be assumed because of the absence of bulk-like states within the spacer layer. However, same ingredients can explain the strength of the exchange coupling in terms of quantum well states which relates the size of the coupling to the spin asymmetry of the scattering potential steps $\Delta V = [\delta V^{\uparrow} - \delta V^{\downarrow}]/2$, δV^{σ} being the potential step for the σ spin direction. One can assume that the coupling gets stronger when $|\Delta V|$ is larger, even if the coupling results from interference between different reflected contributions. Each monolayer of the intermixed zone introduces supplementary potential steps (PS) which can be defined as PS1(X/Plane III), PS2(Plane III/Plane II), PS3(Plane II/Plane I) and PS4(Plane I/Co) with X = Ru or Rh. These potential steps add spin-independent (via the average potential $\langle \delta V \rangle = (\delta V^{\uparrow} + \delta V^{\downarrow})/2$) and spin-dependent (via ΔV) contributions to the confined wave function. Within a tight-binding approach, the potential V can be evaluated for each intermixed plane by $V^{\uparrow}(V^{\downarrow}) = V(\mathrm{X}) + x \times V(\mathrm{Co})/(J/2)\langle M \rangle$, where x is the Co concentration, $\langle M \rangle$ the average local magnetic moment and J the effective exchange integral (which is on the order of 1 eV). As a consequence, the spin asymmetry in the potential step PSn is directly related to the average local magnetic moment variation by: $\Delta V = (J/2)(\langle M \rangle_{n+1} - \langle M \rangle_n)$. The calculation betrays a much stronger polarization of Rh, and no loss in the cobalt magnetic moment, thus indicating the stronger impact of an asymmetry in the electron confinement in the Co/Rh spacer. In contrast, for the Co/Ru system, reflections at PS1 have no spin asymmetry, are poorly spin asymmetric at PS2 and highly spin asymmetric at PS3 and PS4, whereas for the Co/Rh system, reflections at all potential steps are highly spin asymmetric. Finally, since the reflections at PS2, PS3 and PS4 occur on the transmitted fraction of the wave function, there is a significant reduction in the contribution to the exchange coupling from PS1 to PS4 in the case of Co/Ru.

All these theoretical considerations give a strong support to the observation of a stronger coupling strength in Co/Rh than in Co/Ru, the differences being due to a larger spin asymmetry in the scattering potential steps at the most contributing planes of the intermixed interfaces.

In summary, we have evidenced a giant indirect antiferromagnetic coupling of the order of 34 erg/cm^2 in fcc (111) epitaxial Co/Rh sandwiches. By using magnetization measurements and *ab initio* calculations for the Co/Rh and the Co/Ru sandwiches, we have provided evidence of a correlation between the magnetic nature of the magnetic/nonmagnetic interface and the interlayer exchange coupling strength. We have shown that the sharper the magnetic nature of the interface, the higher the electronic confinement in the spacer layer and, consequently, the larger the interlayer exchange coupling.

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REFERENCES

[1] Grünberg P., Schreiber R., Pang Y., Brodsky M. B. and Sower H., *Phys. Rev. Lett.*, **57** (1986) 2442.

[2] Parkin S. S. P., More N. and Roche K. P., *Phys. Rev. Lett.*, **64** (1990) 2304.

[3] Mathon J., Villeret M., Edwards E. D. and Muniz R. B., *J. Magn. & Magn. Mater.*, **121** (1993) 242.

[4] Bruno P., *Phys. Rev. B*, **52** (1995) 411.

[5] Stiles M. D., *Phys. Rev. B*, **48** (1993) 7238.

[6] Parkin S. S. P., *Phys. Rev. Lett.*, **67** (1991) 3598.

[7] Ounadjela K., Muller D., Dinia A., Arbaoui A., Panissod P. and Suran G., *Phys. Rev. B*, **45** (1992) 7768.

[8] Bloemen P. J. H., van Kestern H. W., Swagten H. J. M. and de Jonge W. J. M., *Phys. Rev. B*, **50** (1994) 13505.

[9] Zoll S., Dinia A., Gester M., Stoeffler D., Jay J. P., van den Berg H. A. M., Ounadjela K. and Ousset J. C., to be published.

[10] Experiments at high fields have been performed using magnetization and magnetoresistance measurements which both agree very well for saturation field values. Moreover, on Co/Rh and Co/Ru samples which exhibit ferromagnetic exchange coupling, the only contribution to the MR curves originates from the anisotropic magnetoresistance saturating at 500 Oe.

[11] Zoll S. *et al.*, *J. Magn. & Magn. Mater.*, **156** (1996) 231; van den Berg H., Zoll S., Ounadjela K., Stoeffler D. and Dinia A., to be published.

[12] Stoeffler D., *J. Magn. & Magn. Mater.*, **165** (1997) 62.

[13] Dinia A. and Ounadjela K., *J. Magn. & Magn. Mater.*, **146** (1995) 66.

[14] Massalski T. B., *Binary Alloy Phase Diagram*, Vol. **2**, second edition (ASM International) 1990.

[15] Stoeffler D., Ounadjela K., Sticht J. and Gautier F., *Phys. Rev. B*, **49** (1994) 299.