Strong temperature dependence of the interlayer exchange coupling strength in Co/Cu/Co sandwiches

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We present a study of the temperature dependence of the interlayer coupling strength in Co(hcp)/Cu sandwiches. The thermal variation of the coupling has been studied between 20 and 300 K by superconducting quantum interference device magnetometry for the samples corresponding to the first and second maxima in the oscillation of the exchange coupling of a series of Co/Cu(t_{Cu})/Co trilayers. We find a very strong decrease of the exchange coupling strength between 20 and 300 K. We show that the existing theoretical models cannot explain this unusual strong decrease. However, we believe that this behavior could be due to the confinement of carriers of one spin orientation in the spacer layer. [S0163-1829(97)07830-2]

I. INTRODUCTION

Since the discovery of the exchange coupling between ferromagnetic layers separated by a nonmagnetic layer,[1] various models[2] have been proposed to explain its mechanism. As recently shown by Bruno,[3] in agreement with Stiles,[4] these various approaches (such as total-energy calculations, RKKY model, free-electron model, hole-confinement model, or Anderson model) can be considered as particular cases of the quantum well theory of the exchange coupling. In this general description, the interlayer magnetic coupling consists of quantum interferences due to spin-dependent reflections of Bloch waves at the paramagnet-ferromagnet interfaces.

The RKKY theory, considering the Fermi surface of the spacer layer, succeeded very early in predicting the oscillation periods of the coupling. In the general frame of the quantum well theory, the oscillation periods are related to the oscillation of the reflection coefficient at the magnetic/nonmagnetic interface and the results are the same as within the RKKY theory.

The various models have shown that the coupling strength J is governed essentially by the degree of band matching between the nonmagnetic and ferromagnetic metals. In the quantum well state model, this is expressed in terms of the spin asymmetry of the reflection of the electrons at the magnetic/nonmagnetic interfaces. However, a realistic evaluation of the exchange coupling strength requires accurate knowledge of the features of the interfaces.[5]

Several theoretical studies have focused on the temperature dependence of the interlayer exchange coupling.[6–8] A general trend is that the velocity of the electrons at the extremal points of the Fermi surface governs the temperature dependence.

Using ferromagnetic resonance between 10 and 300 K in the case of Co/Ru/Co trilayers, Zhang *et al.*[9] could recently confirm that the thermal variation of the exchange coupling strength roughly follows the relationship

$$J(T)=J_0\,\frac{T}{T_0}\Big/\sinh\left(\frac{T}{T_0}\right)$$

predicted by one-band model[6] and the free-electron model.[3] The characteristic temperature is given by

$$T_0=\frac{\hbar v_F}{2\pi k_S L},$$

where v_F is the Fermi velocity and L the spacer thickness. Although the above-mentioned model had to be somewhat modified, the variation of $J(T)$ could be well reproduced by only considering the features of the spacer Fermi surface. The Fermi velocity of Ru, of the order of 10^7 cm s^{-1}, is about an order of magnitude smaller than most nonmagnetic metals and this leads to a characteristic temperature T_0 of the order of 100 K. Such a value of T_0 is well suited to explain the strong variations of J with the temperature in Co/Ru structures.

It has been nevertheless found in other structures, such as Fe/Pd/Fe or Fe/Cu/Fe trilayers,[10] that the exchange coupling can also be very sensitive to temperature. The strong variation of the coupling with the temperature is unexpected within the models taking only into account the features of the Fermi surface of the spacer metal.

However, within the quantum well theory,[11] the thermal variation of J depends not only on the spacer Fermi surface, but also on the degree of confinement of magnetic carriers in the spacer quantum well, which is governed by the mismatch between the spacer and ferromagnet bands. It is demonstrated that the temperature dependence of J is very strong when the Fermi level lies near the top of the confining well.

In this paper we present experimental results on the exchange coupling in Co(hcp)/Cu sandwiches grown by UHV evaporation. We find a decrease of the order of 73% of the antiferromagnetic exchange coupling between 20 and 300 K for samples corresponding to the first and second maxima in the exchange coupling oscillation. We compare our results to the behavior expected from theories.

II. STRUCTURE AND MAGNETORESISTANCE

A series of Co(24 Å)/Cu(t_{Cu})/Co(24 Å) sandwiches with t_{Cu} ranging from 3.2 to 29.6 Å was prepared by UHV evaporation on freshly cleaved mica substrates. A 112-Å-thick Ru

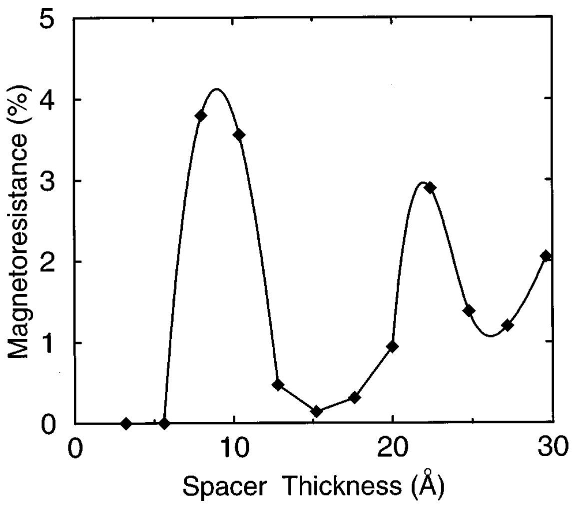

FIG. 1. Magnetoresistance of the Co(hcp)/Cu sandwiches as a function of the Cu interlayer thickness at room temperature. The field is applied along the film plane with the current parallel to the field. The line is only a guide to the eye.

buffer layer was grown at 700 °C in order to provide a flat and single-crystalline surface. After cooling down the substrate to −14 °C, a thin Cu layer was grown prior to the deposition of the Co/Cu sandwich. The latter was then covered with a thin Cu layer and finally a thin Ru cap layer. Reflection high-energy electron diffraction (RHEED) performed *in situ* has shown a close-packed structure with a sixfold symmetry in plane.[12] NMR has clearly evidenced the (00.1) hcp structure with good crystallographic quality of both Co layers for samples with spacer thickness from 3.2 to 25 Å.[13] Although hcp growth of Co on a Cu(111) surface is in fact expected, as shown by Hochstrasser *et al.*[14] Co has until now always been obtained with a fcc(111) structure in molecular-beam-epitaxy- (MBE-) grown superlattices or sandwiches, even when using a Cu(111) single-crystal substrate.[15,16] We believe that the thin Cu seed layer has allowed the occurrence of the hcp phase by providing a very flat surface with a small lattice mismatch for the first Co layer. Although the growth of the second Co layer occurs on a rougher surface than for the first one, the hcp stacking remains of good quality as can be seen from the main line shape (no fcc or stacking faults contribution) of the NMR spectra.[13]

Magnetoresistance (MR) measurements have been first performed at room temperature for all samples using the classical four-point method with the applied field in plane and parallel to the current. Figure 1 shows the variation of the MR ratio as a function of the Cu thickness. The oscillation period (about 13 Å) and the position of the first maximum (about 9 Å) are quiet close to the values usually observed on (111) Co/Cu multilayers. This shows that the hcp structure of the Co layers has no significant effect on both period and phase of the MR oscillation. The sandwiches with $t_{Cu}=8$ Å and $t_{Cu}=22$ Å have been chosen to study the temperature dependence of the coupling strength since they correspond to the maxima in the exchange coupling oscillation.

III. TEMPERATURE DEPENDENCE OF THE EXCHANGE COUPLING

The temperature study of the interlayer exchange coupling of the samples Co(24 Å)/Cu(8 Å)/Co(24 Å) and Co(24 Å)/Cu(22 Å)/Co(24 Å) has been carried out between 20 K and room temperature by use of superconducting quantum interference device (SQUID) magnetometry with fields up to 80 kOe. The qualitative evolution of the exchange coupling strength can be seen in Fig. 2, which presents the magnetization loops measured at 20, 150, 200, and 250 K for the sandwich with $t_{Cu}=8$ Å. The important decrease of the saturation field [and also of the area between the $M(H)$ and the $M=M_{sat}$ lines] with increasing temperature evidences the strong sensitivity of the coupling strength to temperature. Using the relationship $J_{AF}=H_{sat}M_{sat}t_{Co}/2$, holding for sandwiches (where H_{sat} is the saturation field, M_{sat} the saturation magnetization, and t_{Co} the magnetic layer thickness), the exchange coupling strength has been evaluated as a function of temperature as shown in Fig. 3. The decrease of 73% of J

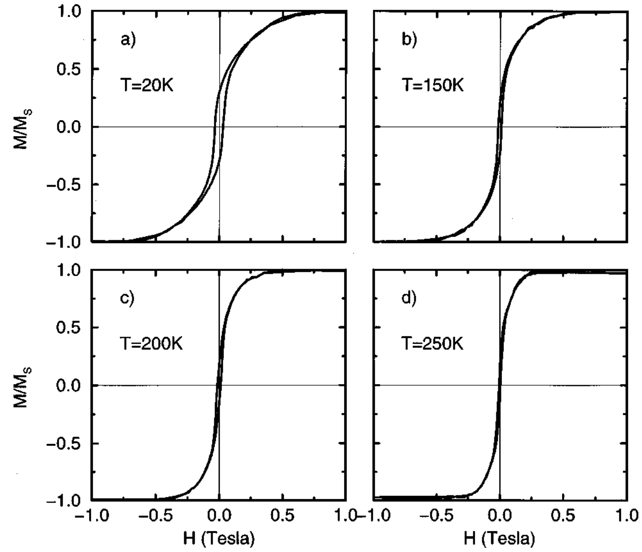

FIG. 2. Magnetization loops obtained by SQUID magnetometry for the Co(24 Å)/Cu(8 Å)/Co(24 Å) sandwich at (a) 20 K, (b) 150 K, (c) 200 K, and (d) 250 K.

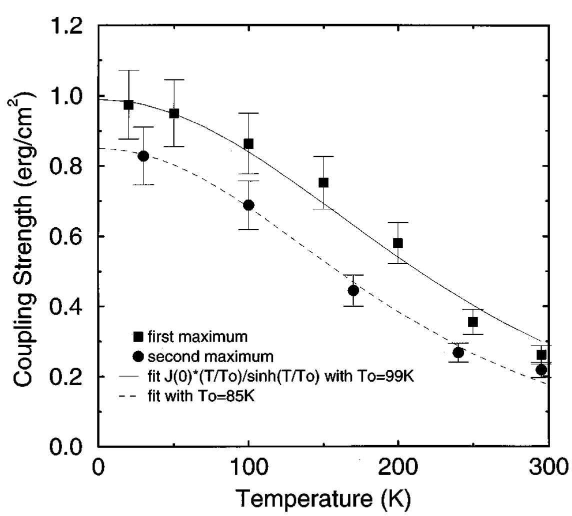

FIG. 3. Evolution with temperature of the antiferromagnetic coupling strength for the Co(24 Å)/Cu(8 Å)/Co(24 Å) (squares) and the Co(24 Å)/Cu(22 Å)/Co(24 Å) (circles) sandwich. The lines correspond to the fit with the relevant expression in the Edwards model.

between 20 and 300 K for the sandwiches with $t_{Cu}=8$ Å and $t_{Cu}=22$ Å is huge compared to the values observed by other groups[17,18] in (111) Co/Cu systems. The strong sensitivity of the antiferromagnetic coupling strength to temperature may be due to the nature of the Co(hcp)/Cu interfaces in our sandwiches. However, one cannot exclude that the effective anisotropy contributes significantly to the observed temperature dependence of the energy of these Co/Cu sandwiches. Indeed, in these sandwiches the Co layers have a hcp structure with a large magnetocrystalline anisotropy (5.5 erg/cm^2 for bulk Co) and are temperature dependent. For this reason we have performed torque measurements for several temperatures (between 20 and 300 K) on Co/Cu sandwich with the same Co thickness as the sample presented in this study, but with the Cu thickness of 1.6 nm (corresponding to the minimum of the coupling) to get rid of the exchange coupling.[19] The results show that the effective anisotropy is strong at 300 K with the value $K_{eff}=-1.2$ erg/cm^2, indicating that the magnetization lies in the film plane. The K_{eff} decreases with decreasing the temperature and reaches the value $K_{eff}=-0.65$ erg/cm^2 at $T=20$ K, which is still strong enough to maintain the magnetization in the film plane. This result is in agreement with the well-known variation of the magnetocrystalline anisotropy of the bulk Co from 5.5×10^6 erg/cm^3 at room temperature to approximately 9×10^6 erg/cm^3 at low temperature,[20] which is strong, but not sufficient to counterbalance the shape anisotropy (13×10^6 erg/cm^3) and to switch the magnetization out of the film plane. On the basis of these results, the anisotropy term has been neglected in the calculation and the whole temperature effect has been attributed to the change of the exchange coupling.

To confirm such a strong temperature dependence of the coupling, magnetoresistance measurements have been performed on the sample with $t_{Cu}=8$ Å at 4.2 and 300 K. The MR curves reported in Fig. 4 support the strong temperature dependence of the interlayer coupling strength, with saturation fields of, respectively, 8 and 2.5 kOe. The MR value at room temperature reaches 4%, indicating a good crystallographic quality of the samples. However, the unexpected small MR value observed at 4.2 K (about 1%) can be explained by the shunting effect in the relatively thick Ru buffer layer.

The calculated J_{AF} represent the maximum coupling values that can be reached in our samples. Indeed, the shape of the magnetization curves (Fig. 2) suggests that the coupling is not homogeneous in the samples and a biquadratic component in the coupling can be expected. Thus we have used a magnetization model (presented elsewhere[21]) to fit magnetization loops, adding a biquadratic coupling to the classical bilinear term. The magnetization loops have never been well reproduced by combining bilinear and biquadratic coupling terms. We conclude the presence of a distribution of independent magnetic behaviors from areas larger than the lateral magnetic coherence length (L_{Co}) of the Co layers.

As already mentioned in the Introduction, the theoretical models[7,6] predict that the temperature dependence of the coupling is governed by the velocity of carriers at the stationary points of the spacer Fermi surface. The dominant temperature factor is indeed

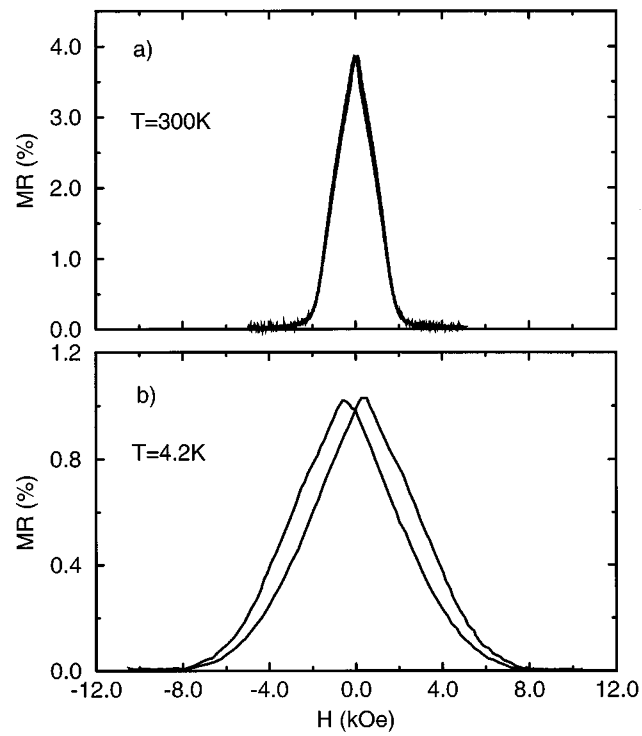

FIG. 4. Magnetoresistance curves for the Co(24 Å)/Cu(8 Å)/Co(24 Å) sandwich at (a) 300 K and (b) 4.2 K. The saturation fields of respectively 2.5 and 8 kOe confirm the strong temperature dependence of the antiferromagnetic coupling strength.

$$\frac{T}{T_0}\Big/\sinh\left(\frac{T}{T_0}\right),$$

with $T_0=\hbar v_F/2\pi k_B L$, where $v_F=(1/\hbar)\partial E/\partial kz$ at the neck and belly of the Fermi surface and L is the spacer thickness. For Cu, v_F is of the order of 1.57×10^8 cm s^{-1} (free-electron model), leading to a theoretical characteristic temperature $T_{0\text{th}}$ of about 2400 K. This value is in fact well above the characteristic temperature $T_{0\,\text{exp}}=99$ K [first antiferromagnetic (AF) maximum] or $T_{0\,\text{exp}}=85$ K (second AF maximum) we find, by fitting the experimental results with the function

$$J(T)=J_0\frac{T}{T_0}\Big/\sinh\left(\frac{T}{T_0}\right)$$

see Fig. 3. Even when using the precise Fermi velocity at the neck, $v_F^*=0.67\times10^8$ cm s^{-1} determined by the de Haas–van Alphen effect,[22] which leads to $T_{0\text{th}}^*=1020$ K, the model is still unable to reproduce the strong decrease of the exchange coupling strength with increasing temperature.

The huge difference between experimental and theoretical T_0 values clearly shows that the observed decrease of the coupling strength is much stronger than expected from this theory. While in this model only the spacer Fermi surface is relevant, Cullen and Hathaway[8] assumed that the temperature dependence is due to the disordering of the ferromagnet moments. However, for sandwiches with 12 monolayers in one ferromagnet like in our case, the decease is weak, with a $T^{3/2}$ behavior at low temperatures, followed by a quasilinear decrease at higher temperatures. To explain what happens in our samples, it is likely that both spacer and ferromagnet

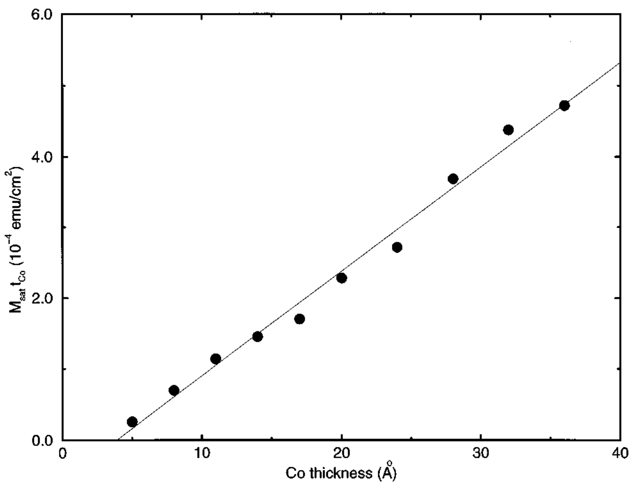

FIG. 5. Variation of the measured saturation magnetization per unit surface of Co, $M_S t_{Co}$, with Co layer thickness t_{Co} for the Co(t_{Co})/Cu(15.2 Å)/Co(t_{Co}) sandwiches.

layers have to be considered. A recent model of temperature dependence of J which depends on matching of ferromagnet and spacer bands in direction perpendicular to the layers[11] seems to concur with this hypothesis. The calculation performed on (001) Co/Cu predicts a strong temperature dependence when magnetic carriers of one spin orientation are fully confined in spacer potential well of finite depth. They also show that this temperature dependence is relatively stronger for thicker Cu spacer layers. Such results agree very well with our experimental measurements.

Another explanation for the strong decrease of the exchange coupling strength lies along the same line and is related to the magnetic nature of the Co/Cu interfaces. We have measured the magnetization loops for a series of Co(t_{Co})/Cu(15.2 Å)/Co(t_{Co}) sandwiches. Figure 5 shows the variation in saturation magnetization per unit surface of Co, $M_s t_{Co}$, versus the cobalt thickness (t_{Co}). The linear decrease in magnetization with decreasing t_{Co} is expressed by a linear function which intercepts the abscissa at a thickness of about 4 Å. This indicates that about 2 Å of Co at each interface are magnetically dead at room temperature, due to intermixing. Such a dead layer would round off the potential well giving it a profile. When the temperature is decreasing, the dead layer is expected to become thinner, making consequently the well sharper than at room temperature. The evolution of the potential well shape with temperature could modify the confinement of the carriers and thus lead to a strong temperature dependence of the coupling.

IV. CONCLUSION

In conclusion, we have found an unexpected strong decrease of the interlayer coupling strength in Co(hcp)/Cu sandwiches. We expect this behavior to be due to the change of the potential well with temperature at the Co(hcp)/Cu interfaces. The theories existing up to now are not able to explain such a behavior. However, a model where the temperature dependence is not only governed by the spacer Fermi surface, but also by the ferromagnet, seems likely to explain our results.

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

[2] For a review, see *Ultrathin Magnetic Structures II*, edited by B. Heinrich and J. A. C. Bland (Springer-Verlag, Berlin, 1994), p. 45.

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

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

[5] S. Zoll, A. Dinia, D. Stoeffler, M. Gester, H. A. M. van den Berg, A. Herr, R. Poinset, and H. Rakoto, J. Magn. Magn. Mater. **165,** 442 (1997).

[6] D. M. Edwards, J. Mathon, R. B. Muniz, and M. S. Phan, Phys. Rev. Lett. **67**, 493 (1991).

[7] P. Bruno and C. Chappert, Phys. Rev. Lett. **67**, 1602 (1991).

[8] J. R. Cullen and K. B. Hathaway, Phys. Rev. B **47**, 14 998 (1993).

[9] Z. Zhang, L. Zhou, P. E. Wigen, and K. Ounadjela, Phys. Rev. Lett. **73**, 336 (1994); Phys. Rev. B **50**, 6094 (1994).

[10] Z. Celinski, B. Heinrich, J. F. Cochran, W. B. Muir, A. S. Arrot, and J. Kirschner, Phys. Rev. Lett. **65**, 1156 (1990); Z. Celinski and B. Heinrich, J. Magn. Magn. Mater. **99**, L25 (1991).

[11] J. d'Albuquerque e Castro, J. Mathon, M. Villeret, and A. Umerski, Phys. Rev. B **53**, 13 306 (1996).

[12] N. Persat and A. Dinia, J. Magn. Magn. Mater. **156**, 371 (1996).

[13] N. Persat, A. Dinia, J. P. Jay, C. Mény, and P. Panissod, J. Magn. Magn. Mater. **156**, 335 (1996).

[14] M. Hochstrasser, M. Zurkirch, M. Erdudak, E. Wetli, and D. Pescia, J. Magn. Magn. Mater. **148**, 4 (1995).

[15] W. F. Egelhoff, Jr. and M. T. Kief, Phys. Rev. B **45**, 7795 (1992).

[16] M. T. Johnson, R. Coehoorn, J. J. de Vries, N. W. E. McGee, J. aan de Stegge, and P. J. H. Bloemen, Phys. Rev. Lett. **69**, 969 (1992).

[17] S. S. P. Parkin, R. Bhadra, and K. P. Roche, Phys. Rev. Lett. **66**, 2152 (1991).

[18] F. Petroff, Ph.D. thesis, Université Paris-Sud Orsay, 1992.

[19] A. Dinia, N. Persat, and H. Danan (unpublished).

[20] W. Suchsmith and J. E. Thompson, Proc. R. Soc. London, Ser. A **225**, 362 (1954).

[21] S. Zoll, H. A. M. van den Berg, K. Ounadjela, D. Stoeffler, and A. Dinia, J. Appl. Phys. **79**, 2601 (1996).

[22] B. Lengeler, W. R. Wampler, R. R. Bourassa, K. Mika, K. Wingerath, and W. Uelhoff, Phys. Rev. B **15**, 5493 (1977).