Evidence for a Direct *d-d* Hybridization Mechanism for the Interlayer Exchange Coupling in Epitaxial Co/Mn Multilayers

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The interlayer exchange coupling between successive Co layers and the magnetic ordering of the Mn spacer layers are investigated as a function of temperature in epitaxial Co/Mn multilayers. The interlayer coupling strength is shown to decrease steeply with increasing temperature and to vanish at the Néel temperature of the antiferromagnetically ordered Mn spacer. This result indicates that the interlayer coupling originates from a direct *d-d* exchange interaction at the Co-Mn interfaces and propagates through the magnetic ordering of the Mn layers via short range exchange interactions.

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The discovery of an exchange coupling between Fe films separated by thin Cr spacer layers [1] and its oscillatory behavior as the Cr thickness is varied [2] together with its relevance to giant magnetoresistance [3] have attracted considerable attention in metallic multilayers based on transition metal ferromagnets. An oscillatory exchange coupling, sometimes multiperiodic, has since been observed in a large number of systems containing transition as well as noble metal spacers [4]. Several theoretical approaches have been proposed to explain the origin of this phenomenon and the factors that determine the oscillation periods [4].

For paramagnetic metal spacers, it is now commonly accepted that the interlayer exchange coupling can be described by an RKKY-like model in which a long range interaction is mediated by the nearly free *sp* electrons of the spacer material [5]. Quantitative as well as qualitative predictions from this theory have been verified experimentally for systems with noble metal spacers [6]. Such an RKKY-like approach might not be fully appropriate when the spacer material is also a transition metal, especially when it is magnetically ordered. In that case, the magnetic behavior of the multilayered structures might indeed be mostly determined by the *d* states and more especially the interfacial *d-d* hybridization [7]. Experimental results on the Fe/Cr(100) [8] and Fe/Mn(100) [9] systems seem to corroborate this assessment. For both of them, it has been shown that the coupling oscillates with the two-monolayer period expected from the combination of a direct *d-d* interaction between the two elements and a magnetic ordering within the spacer consisting of ferromagnetically aligned (100) atomic planes coupled antiferromagnetically. However, it has been pointed out that the short-period oscillating coupling across Cr(100) spacers can also be explained in the RKKY picture [10], i.e., without invoking any magnetic ordering in Cr. It could arise from the particular nesting of the Cr Fermi surface along the [100] direction which interestingly is also at the origin of the spin-density wave antiferromagnetism in bulk Cr.

At present, the role played by the spacer antiferromagnetic (AF) ordering in the interlayer exchange coupling still remains an outstanding problem. For the relationship between the interlayer coupling and the magnetism of the spacer to be uniquely demonstrated, the coupling should be proved to vanish when the intervening layers become paramagnetic or at least to be strongly diminished above the spacer Néel temperature if another mechanism is to coexist with the discussed direct *d-d* hybridization mechanism.

In this paper we report the experimental discovery of such a strong interplay between the magnetism of the spacer layers and the exchange coupling between successive ferromagnetic layers in epitaxial Co/Mn multilayers. The AF ordering of the Mn spacer is evidenced by the existence of an interfacial exchange coupling between Co and Mn, which is manifested by a unidirectional (or exchange) anisotropy for the ferromagnetic layers and a nonvanishing high-field rotational hysteresis. The Néel temperature (T_N) of the buried Mn layers is determined from the thermal variation of this high-field rotational hysteresis. Strikingly, the exchange coupling between adjacent Co layers is shown to decrease steeply with increasing temperature and to be suppressed *above* T_N.

The samples are $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(t_{\mathrm{Mn}}\ \text{Å})]_{12}$ multilayers epitaxially grown at 260 K by ultrahigh vacuum *e*-beam evaporation. The overlayers were deposited on single crystal 150 Å thick hcp (0001) Ru buffer layers previously evaporated onto mica substrates at 950 K. All samples were covered with a 40 Å thick hc (0001) Ru capping layer. Studies by reflection high energy electron diffraction and x-ray diffraction of the structural properties of those multilayers with t_{Mn} larger than 12 Å showed that Co adopts the room temperature stable hcp (0001) structure, while Mn is stabilized in a sixfold in-plane symmetric metastable structure identical to the $Cu_2Mg(111)$ Laves phase which closely resembles the complex α-Mn phase (stable from 0 to 1000 K in bulk Mn) [11]. The chemical abruptness of the interfaces was ensured by the low temperature deposition as verified

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by ^{59}Co nuclear magnetic resonance experiments which revealed an intermixed region limited to less than two atomic layers [12].

Figures 1(a) and 1(b) show the magnetization versus in-plane magnetic field curves obtained at 4.5 K by SQUID magnetometry after field cooling from room temperature (RT) in 80 kOe for two $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(t_{\mathrm{Mn}}\ \text{Å})]_{12}$ multilayers with different spacer thickness, namely, 32 Å [Fig. 1(a)] and 14 Å [Fig. 1(b)]. Both samples exhibit large shifts of their *M-H* loops along the field axis (310 and 250 Oe, respectively). These exchange bias fields H_{ex} are clear manifestations of the interfacial exchange coupling between the ferromagnetic (FM) Co layers and the Mn layers which are thus shown to be antiferromagnetically ordered at 4.5 K. Notice that the observation of such an exchange anisotropy is laid down by the condition that the anisotropy energy of the AF layers must exceed the interfacial coupling energy [13]. If this condition was not satisfied, in some regions of the samples at least, the antiferromagnet moments would not be anchored to the crystal lattice and would follow the motion of the ferromagnet magnetization resulting in centered hysteresis loops. Consequently, Mn in the $Cu_2Mg(111)$ Laves phase structure appears highly anisotropic.

Beyond the fact that they both exhibit exchange anisotropy, the two multilayers in which the Co magnetization lies preferentially in the plane show entirely different magnetic behaviors. The squareness of the hysteresis curve obtained for the $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(32\ \text{Å})]_{12}$ structure [Fig. 1(a)] reveals that the Co layers are either ferromagnetically coupled or uncoupled. In contrast, the high saturation field ($\sim$3.5 kOe) and the low remanent magnetization ($M/M_s \sim 0.3$ for $H = -H_{\mathrm{ex}}$) associated with the slanted *M-H* loop of the $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(14\ \text{Å})]_{12}$ structure [Fig. 1(b)] is the signature of a rather strong AF exchange coupling between adjacent Co layers across the intervening Mn [14]. The large coercive field $H_c = 260$ Oe, defined as the half-width of the loop at zero magnetization, disagrees with previous observations of small or almost zero coercivity for antiferromagnetically coupled multilayers [15]. It certainly originates from the large hysteresis losses that are often associated with the existence of FM and AF interfacial exchange coupling [13,16].

Contrary to those obtained at 4.5 K, the in-plane hysteresis loops measured at 320 K for the $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(32\ \text{Å})]_{12}$ [Fig. 1(c)] and $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(14\ \text{Å})]_{12}$ [Fig. 1(d)] multilayers are not shifted. This reveals that the blocking temperature T_B for which the exchange bias field of H_{ex} goes to zero is lower than 320 K in both samples. In addition, the two magnetization curves are now very similar to each other. Both of them exhibit small saturation fields (200–300 Oe) and remanent magnetization to saturation magnetization ratios of about 0.9. Since these features are also observed for simple $\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(t_{\mathrm{Mn}}\ \text{Å})$ bilayers grown under the same conditions, they can reasonably be attributed to small anisotropy effects. Consequently, we can assert that the transition from antiferromagnetically coupled to uncoupled Co layers occurs below 320 K in the $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(14\ \text{Å})]_{12}$ structure.

The complete variation of the AF exchange coupling between 4.5 and 320 K was determined for the $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(14\ \text{Å})]_{12}$ multilayer from the hysteresis loops measured by SQUID magnetometry [Fig. 2(a)]. The exchange coupling strength J_{AF} was calculated from the following expression:

$$J_{\mathrm{AF}} = t_{\mathrm{Co}} \int_0^{M_s} H\, dM\,, \tag{1}$$

with $t_{\mathrm{Co}} = 24$ Å and in which the in-plane anisotropy is assumed to be negligible. Up to 300 K, J_{AF} is seen to decrease almost linearly following a law

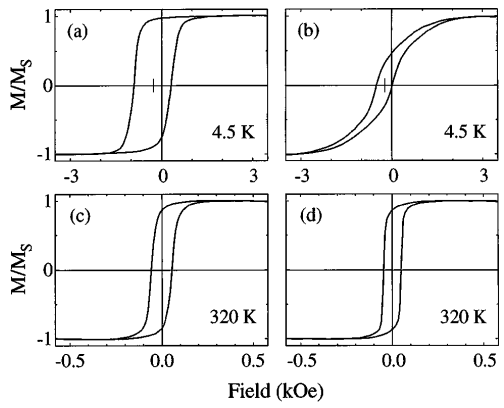

FIG. 1. In-plane hysteresis loops measured on (a),(c) $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(32\ \text{Å})]_{12}$ and (b),(d) $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(14\ \text{Å})]_{12}$ multilayers at (a),(b) 4.5 K (after field cooling in 80 kOe) and (c),(d) 320 K.

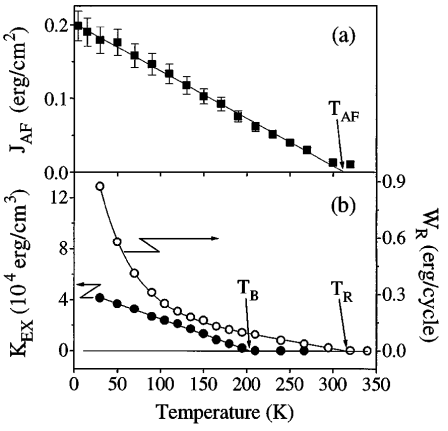

FIG. 2. Temperature dependence of (a) the interlayer coupling constant J_{AF}, (b) exchange anisotropy constant K_{ex} (solid circles), and high-field rotational hysteresis W_R (open circles) for a $[\mathrm{Co}(24\ \text{Å})/\mathrm{Mn}(14\ \text{Å})]_{12}$ multilayer.

of the form $J_{AF}(T) = J_{AF}(0)(1 - T/T_{AF})$ with $J_{AF}(0) = 0.20$ erg/cm^2 and $T_{AF} = 313 \pm 10$ K. Above 300 K, the experimental data deviate from the linear law and J_{AF} does not vanish at T_{AF}. However, this deviation can quite likely be ascribed to the anisotropy effects mentioned previously. Indeed, at high temperature, the in-plane anisotropy energy which contributes to the J_{AF} constant calculated from expression (1) is no longer negligible compared to the actual exchange coupling energy and therefore it artificially increases J_{AF}. In these conditions, even though J_{AF} does not drop to zero, T_{AF} is a good estimate for the temperature at which the AF exchange coupling disappears in the [Co(24 Å)/Mn(14 Å)]$_{12}$ multilayer.

In-plane magnetic torque measurements as a function of temperature were performed on the [Co(24 Å)/Mn(14 Å)]$_{12}$ sample in order to determine the Néel temperature of the buried Mn layers. For the experiments performed below RT, the sample was cooled down in a 10 kOe in-plane external field. The magnetic torque curves $\Gamma^{CW}(\varphi)$ and $\Gamma^{CCW}(\varphi)$ were then taken for both clockwise (CW) and counterclockwise (CCW) directions of rotation of an applied field, which was always sufficiently strong to fully saturate the Co magnetization. Figure 3(a) shows typical $\Gamma^{CW}(\varphi)$ and $\Gamma^{CCW}(\varphi)$ curves. Two torque contributions have to be considered in order to describe the measured $\Gamma^{CW}(\varphi)$ and $\Gamma^{CCW}(\varphi)$ curves. The first contribution is given by $\Gamma_{rev}(\varphi) = [\Gamma^{CW}(\varphi) + \Gamma^{CCW}(\varphi)]/2$ and arises from *reversible* changes of magnetization. The second is given by $\Gamma_{irr}(\varphi) = [\Gamma^{CW}(\varphi) - \Gamma^{CCW}(\varphi)]/2$ and results from *irreversible* reorientations of magnetic moments in the Mn layers with respect to the saturated rotating Co magnetization. This contribution is directly related to the rotational hysteresis W_R [Eq. (2)], whose persistence in magnetic fields very much larger than the Co saturation field (a) provides further strong evidence for the interfacial coupling between the FM Co, and AF Mn layers and (b) implies the existence of AF regions with unblocked magnetic moments [17]. Figures 3(b) and 3(c) show examples of $\Gamma_{irr}(\varphi)$ curves deduced for the [Co(24 Å)/Mn(14 Å)]$_{12}$ multilayer:

$$W_R = \int_0^{2\pi} \Gamma_{irr}(\varphi)\, d\varphi\,. \tag{2}$$

For all temperatures in the 20–340 K range, the $\Gamma_{rev}(\varphi)$ curves were successfully fitted to the following function:

$$\begin{aligned}\Gamma_{rev}(\varphi) = {} & K_{ex}\sin(\varphi - \varphi_{ex}) + 6K_4^{(1)}\sin[6(\varphi - \varphi_4)] \\ & + 6K_4^{(2)}\sin[12(\varphi - \varphi_4)] \\ & + 6K_4^{(3)}\sin[18(\varphi - \varphi_4)]\,,\end{aligned} \tag{3}$$

where φ is the in-plane angle of the Co magnetization, φ_{ex} is the angle of the external field applied during the cooling, and φ_4 is the angle of one of the $[11\bar{2}0]$-type in-plane easy axes of the Co layers. In this formula, the first and second terms are, respectively, the unidirectional anisotropy and magnetocrystalline anisotropy contributions to the torque, while the third and fourth terms correspond to higher order Fourier components of the second term. The presence of these high-order components reveals that some of the magnetic moments which are subjected to a sixfold symmetric anisotropy are not fully saturated. Since our experiments Co saturation was always ensured, part of the sixfold symmetric torque component must originate from the unblocked regions of the Cu_2Mg(111)-Mn layers, whose in-plane magnetocrystalline anisotropy matches that of the hcp (0001) Co layers. This is in excellent agreement with the 60°-periodic $\Gamma_{irr}(\varphi)$ curves in which the maxima, revealing six in-plane hard axes for Mn, occur for angles corresponding to the in-plane hard axes of Co.

The temperature dependence of the exchange anisotropy constant K_{ex} is depicted in Fig. 2(b). As already reported for the NiFe/FeMn [18] and Fe_3O_4/CoO [19] systems, K_{ex} decreases linearly with increasing temperature, going towards zero at the blocking temperature $T_B = 203 \pm 5$ K. Unfortunately, it must be emphasized that T_B is only a lower limit for the Néel temperature of Cu_2Mg-Mn. Indeed, whereas T_N is the temperature at which the long range magnetic order disappears in the AF material, T_B is no more than the temperature above which the anisotropy energy of the AF Mn layers becomes, everywhere in the sample, smaller than the interfacial coupling energy. Furthermore, the persistence of some high-field rotational hysteresis at temperatures larger than T_B, as illustrated in Fig. 3(c), demonstrates unambiguously that the interfacial exchange coupling and then the AF ordering of Mn still exist above the blocking temperature.

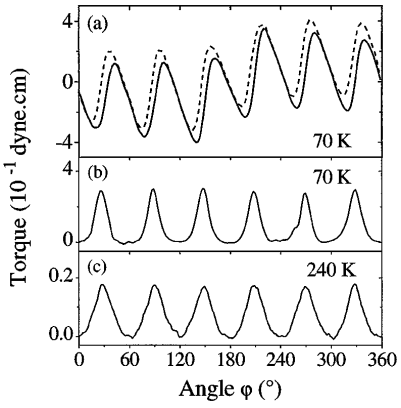

FIG. 3. In-plane torque curves on a [Co(24 Å)/Mn(14 Å)]$_{12}$ multilayer cooled in a 10 kOe magnetic field: (a) $\Gamma^{CCW}(\varphi)$ (solid line) and $\Gamma^{CW}(\varphi)$ (dashed line) curves at 70 K, $\Gamma_{irr}(\varphi)$ curves at (b) 70 and (c) 240 K.

Figure 2(b) shows the thermal variation of the rotational hysteresis measured in 10 kOe for the $[Co(24\ Å)/Mn(14\ Å)]_{12}$ multilayer. W_R exhibits a roughly exponential decrease up to 170 K and then a linear decay up to $T_R = 315 \pm 5$ K, where it is heading to zero. As a general rule for FM-AF coupled systems, $T_B \leq T_R \leq T_N$. Consequently, in the same way as T_B, T_R should only be considered as a lower limit for the Néel temperature. However, previous studies have shown that T_R is a good estimate of T_N [17] and, therefore, we conclude that the ordering temperature of the Cu_2Mg-Mn layers is certainly close to the temperature at which the rotational hysteresis vanishes $T_R = 315 \pm 5$ K.

In this study, we have clearly identified two distinct magnetic features: (i) an interfacial exchange coupling between Co and Mn layers which evidences the AF ordering of Mn with a Néel temperature close to $T_N = 315 \pm 5$ K and (ii) an interlayer exchange coupling between adjacent Co layers across Mn spacer layers which decreases steeply with increasing temperature and disappears around $T_{AF} = 313 \pm 10$ K. The almost perfect coincidence of T_N and T_{AF} highlights the strong interdependence between the interlayer exchange coupling and the magnetic order of the spacer material. Mn must be antiferromagnetically ordered for a coupling between Co layers to be exerted across it. This result demonstrates that the coupling between the magnetizations of two successive Co layers is entirely constructed from the strong interfacial interaction between Co and Mn and the AF ordering of Mn, which probably consists of ferromagnetic (111) atomic planes aligned antiferomagnetically with respect to each other in the [111] direction. Hence the interlayer coupling develops through the whole spacer by means of short range *d-d* exchange interactions. Surprisingly, no coupling due to a long range RKKY-like interaction seems to coexist with the exchange coupling arising from the direct *d-d* hybridization mechanism in the studied samples. Although the *d-d* hybridization mechanism is probably not in all layered films with antiferromagnetic transition metal spacer as predominant as it is in the present Co/Mn multilayers, our findings suggest that it should be taken into account when looking for the origin of interlayer exchange coupling in such systems.

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