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Interface roughness, magnetic moments, and couplings in $(A)_m/(Cr)_n(001)$ superlattices (A = Fe, Co, Ni)

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We use the real-space tight-binding method to determine the magnetic moments and the *interlayer* magnetic couplings (IMC) of $(A)_m/(Cr)_n$ (A = Fe, Co, Ni) superlattices (SL) with perfect or diffuse A/Cr interfaces. For perfect SL, the IMC oscillate with a short period in agreement with experiment. For the same SL with an interface roughness coming from interfacial ordered compounds, the occurrence of interfacial frustrated Cr-Fe bonds and of Cr magnetic-moment instability can lead to a strong reduction of the Cr moments even if n is large ($n \simeq 30$) and also to a damping and even the disappearance of the previous IMC oscillations. Therefore, the Cr spacer is not—as often assumed—a passive medium transmitting indirect RKKY-like interactions.

The magnetic interactions between thin ferromagnetic (F) layers $(A)_m$ (A = Fe, Co, Ni) separated by nonmagnetic or antiferromagnetic (AF) transition-metal (TM) spacers $(B)_n$ are extensively studied in relation to the possible use of magnetoresistive and magneto-optical devices. Observations of AF interlayer magnetic couplings (IMC) have been reported on the $(Fe)_m/(Cr)_n$ system and for Cr thicknesses of a few atomic monolayers (ML).[1] More recently, long-ranged oscillatory IMC have been observed in multilayers obtained by sputtering, the oscillatory period being surprisingly about the same ($\lambda \simeq 10$ Å) for all the TM in which couplings were observed.[2] However, the dependence of the IMC,[3] and of the corresponding magnetoresistances[4] on the crystallographic quality of the superlattices (SL) and on the interface roughness has been pointed out. In this respect, the most remarkable results have been obtained for Fe/Cr/Fe(100) sandwiches.[3] IMC oscillations with a period of about 10 Cr ML were observed for room-temperature growth. However, well-ordered sandwiches grown at higher temperature ($T_s \simeq 80\,^\circ$C) and with a better crystallographic quality exhibit oscillations with a period of 2 Cr ML for thick Cr layers, both oscillations being observed for the thinnest Cr layers. Such short-ranged oscillations in Fe/Cr have also been reported by other authors.[5]

The IMC can be determined theoretically from $\Delta E_{F-AF} = E_F - E_{AF}$. E_F (E_{AF}) is the total energy when successive $(A)_m$ layers are F (AF) ordered. An *ab initio* calculation for the $(Fe)_3/(Cr)_n(001)$ SL (Ref. 6) showed that the electronic structure of the Cr spacer is strongly perturbed by the F layers. However, IMC much larger than experimentally observed and a crossover from F ($n < 3$) to AF ($3 < n < 7$) couplings were found. Then, a systematic study of the SL electronic structure was done using a real-space tight-binding (TB) calculation.[7] It is in agreement with the previous one for the magnetic-moment distributions in $(Fe)_m/(Cr)_n$ SL, but it predicts in these SL oscillating IMC with *a period of 2 Cr ML*. These oscillations also have been obtained more recently by atomic sphere approximation and linear muffin-tin orbital calculations.[8] They are similar to those which have been observed for "perfect" SL in Ref. 3, but they are still larger than obtained experimentally.

The aim of this paper is to study the electronic structure of $(A)_m/(Cr)_n$ SL (A = Fe, Co, Ni) and, more precisely, to (i) determine the role of the magnetic order and the importance of the magnetic perturbation induced in the Cr spacer by flat or rough A interfaces, (ii) understand its physical origin in terms of magnetic frustration, and (iii) relate it to the behavior of the IMC and investigate the sensitivity of the IMC (order of magnitude and periodicity) to the nature of the interfaces *at the atomic scale*. We study the role of interdiffusion in Fe/Cr SL by introducing interfacial ordered compounds (IOC). With rough interfaces it is not possible, in general, to satisfy all the AF-nearest-neighbor Cr-Cr and Cr-Fe interactions. The interfacial Cr atoms being close to a magnetic-nonmagnetic instability,[9] it is energetically less expensive to suppress (or strongly decrease) the Cr interfacial moment than to keep a frustrated magnetic arrangement. As usual in Cr,[10] this local perturbation propagates in the Cr spacer far from the defect, so that the IOC can completely change the magnetic-moment distribution and the IMC *from the magnetic frustrations they introduce.*

Let us first summarize the results we obtained for perfect SL. Details of the real-space TB recursion method we use here are reported elsewhere.[7] The magnetic-moment distributions of *perfect* bcc(001) $(Fe)_5/(Cr)_n$ (Fig. 1), $(Co)_3/(Cr)_n$, and $(Ni)_6/(Cr)_n$ superlattices are characterized in all cases by a central magnetic defect which occurs in the Cr layers when the interlayer magnetic arrangement is incompatible with the natural AF Cr order and the strong interfacial Cr-A coupling, i.e., for an AF (F) interlayer magnetic arrangement and odd (even) n values. This defect occurs from the *frustration* of the Cr-Cr AF couplings induced by the Fe layers and is characterized by *nearly vanishing* Cr moments. However, the A/Cr interfaces are clearly different for the three situations we considered: (i) for A = Fe, the interfacial Fe (Cr) moments are reduced (enhanced) as compared to their bulk values and are AF coupled; (ii) for A = Co, the slightly reduced Co and the slightly enhanced Cr magnetic moments at the interface are ferromagnetically coupled; and (iii) for A = Ni, the Ni (Cr) magnetic moments

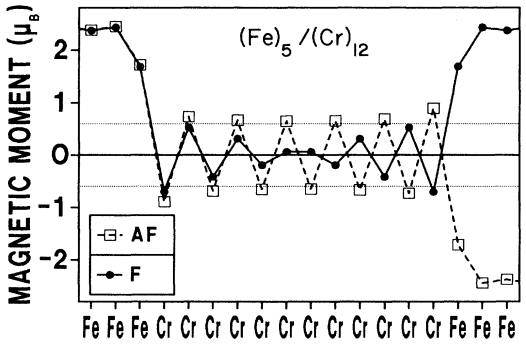

FIG. 1. Magnetic-moment distributions for $(Fe)_5/(Cr)_{12}$ superlattices in the ferromagnetic (F) (solid line) and antiferromagnetic (AF) (dashed line) interlayer magnetic arrangements.

are strongly reduced (enhanced) and are ferromagnetically coupled. Nearly identical results are obtained using either TB and local-spin-density-approximation (Refs. 6–8) calculations. For $(Fe)_m/(Cr)_n$ and $(Ni)_m/(Cr)_n$ the IMC are found to oscillate rapidly with the parity of n (Fig. 2) for all n values. For large Cr thicknesses ($n \geq 8$) it can be split into an interfacial energy γ and a defect energy γ_d ($\gamma = 183$ meV, $\gamma_d = 11$ meV in Fe/Cr SL).[7] The oscillating IMC is then due to the energy of the magnetic defect induced in the Cr layer via the strong interfacial AF Cr-A coupling. For $(Co)_3/(Cr)_n$, ΔE_{F-AF} is negative for $1 \leq n \leq 5$ with a large value for $n = 1$, the oscillations of ΔE being recovered only for larger thicknesses ($n \geq 8$): the Co-Cr F coupling is too weak[11,12] to obtain such oscillations for $n \leq 6$. Finally, note that these periodic oscillations are due to the AF order of Cr: they disappear for a nonmagnetically ordered spacer with $I(Cr) = 0$ and are thus strongly temperature dependent. However, they remain important for the Néel temperature of Cr, the disappearance of the Cr magnetic moments occurring only

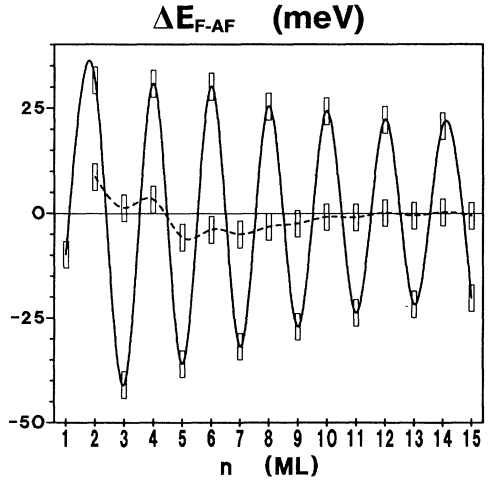

FIG. 2. Total-energy difference $\Delta E_n = E_n(F) - E_n(\mathrm{AF})$ for the crystallographic cell and per interface atom as a function of the Cr thickness n for $(Fe)_5/(Cr)_n(001)$ superlattices in case (a) (solid line) and in case (d) (dashed line). The size of the rectangles gives an idea of the estimated errors.

above the SL ordering temperature.[13]

Experimentally, the interdiffusion leads to interfacial alloys and compounds. Our real-space method is particularly suited to study the modification induced on the interfacial magnetism and the IMC by IOC. This is because the computer time does not dramatically increase with the size of the AF cell as in *ab initio* methods. This method allows us to control easily the convergence of the iterative calculation: self-consistency has been assumed to be achieved for the kth step of the iteration when the total energy $E[k]$, the charge-transfer distribution $\Delta N_i[k]$, and the magnetic-moment distribution $M_i[k]$ verify

$$|E[k] - E[k-1]| < 10^{-5}\ \mathrm{eV},$$

$$\max_i\{|\Delta N_i[k] - \Delta N_i[k-1]|\} < 10^{-5}e,$$

and

$$\max_i\{|M_i[k] - M_i[k-1]|\} < 10^{-5}\mu_B.$$

Here, we restrict our study to $A = \mathrm{Fe}$ and to $m = 5$ SL to ensure we have at least two pure Fe ML in the Fe layers. For illustration, we consider below different SL with typical interfaces corresponding to different interdiffusion ranges δ: (a) $\delta = 0$ ML, $(Fe)_5/(Cr)_n$ perfect superlattices without interdiffusion; (b) $\delta = 1$ ML, $(Fe)_4(Fe_{50\%}Cr_{50\%})/(Cr)_{n-1}(Fe_{50\%}Cr_{50\%})$; (c) $\delta = 2$ ML, $(Fe)_3(Fe_{50\%}Cr_{50\%})(Fe_{50\%}Cr_{50\%})/(Cr)_{n-2}(Fe_{50\%}Cr_{50\%})(Fe_{50\%}Cr_{50\%})$; (d) $\delta = 2$ ML, $(Fe)_3(Fe_{75\%}Cr_{25\%})(Fe_{25\%}Cr_{75\%})/(Cr)_{n-2}(Fe_{25\%}Cr_{75\%})(Fe_{75\%}Cr_{25\%})$; (e) $\delta = 3$ ML, $(Fe)_2(Fe_{75\%}Cr_{25\%})(Fe_{50\%}Cr_{50\%})(Fe_{25\%}Cr_{75\%})/(Cr)_{n-3}(Fe_{25\%}Cr_{75\%})(Fe_{50\%}Cr_{50\%})(Fe_{75\%}Cr_{25\%})$ with $n = 1,2,3$ to 15. Case (a) is treated as those described previously, cases (b) and (c) are treated with a $\sqrt{2}\times\sqrt{2}$ cell in the (001) plane, and cases (d) and (e) are treated with a 2×2 cell in the layer's plane. Figure 3 presents the IOC of case (d).

For case (a) we find the well-known oscillations. For cases (b) and (c) we obtain similar oscillations for large thicknesses, but they are slightly shifted towards the negative values for thin Cr layers ($n \leq 9$). Their magnitude is also reduced by a factor of 2 in comparison with (a): this is due to reduced magnetic moments in the Cr layer. However, apart from this, the magnetic-moment distributions for the two inequivalent [100] lines of sites are similar to those obtained for (a) either for $n-1$ and $n+1$ Cr ML [case (b)] or for $n-2$ and $n+2$ Cr ML [case (c)].

The Cr/Fe/Cr sandwiches with the (b) or (c) Cr/Fe interfaces do not exhibit, in their magnetic ground states, significative changes of the magnetic-moment distributions in comparison to those with perfect interfaces. This is because in both cases there are no frustrated magnetic bonds. However, when the Fe magnetic moments are reversed with respect to those of the Cr layers, Cr-Fe frustrated bonds are induced on the Cr interfacial atoms and a magnetic defect is located at the interface, the corresponding Cr atoms having once more a nearly vanishing magnetic moment. Therefore, the rough interface pins the magnetic defect which occurred, for flat interfaces, in bulk Cr, between the neighboring interfaces. In this model, the Cr/Fe exchange anisotropy comes from the energy of this defect ($\gamma = 0.18$ erg/cm^2).

For case (d) (Fig. 2) we obtain the most interesting result: the oscillations are damped and the IMC presents a

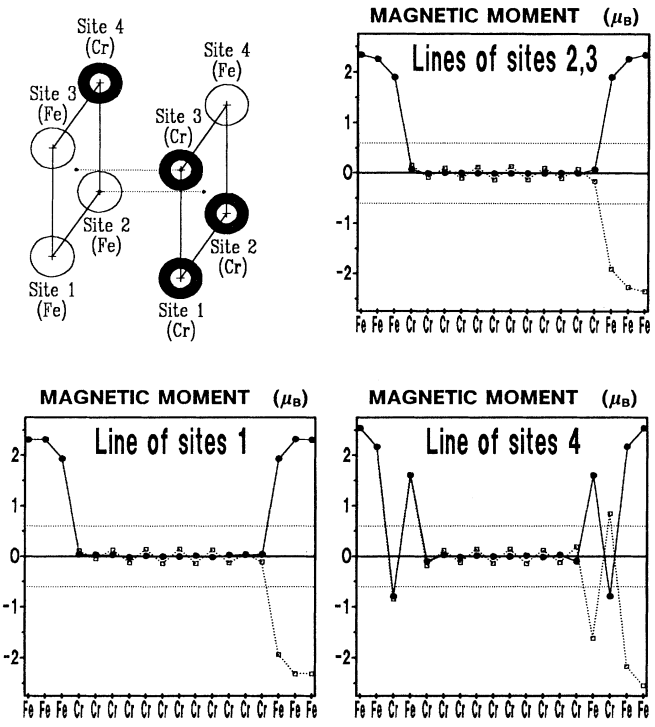

FIG. 3. Schematic representation of the interfacial ordered compound in case (d) (see text) and magnetic-moment distributions in the ferromagnetic (F) (solid lines) and antiferromagnetic (AF) (dotted lines) interlayer magnetic arrangements along the three inequivalent lines of sites.

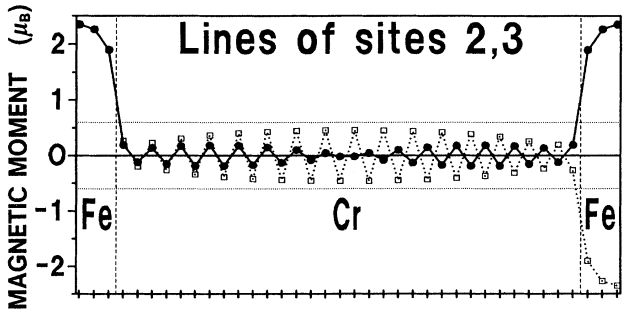

FIG. 4. Magnetic-moment distributions in case (d) for $(Fe)_5/(Cr)_{30}(001)$ superlattices in the ferromagnetic (F) (solid line) and antiferromagnetic (AF) (dotted line) interlayer magnetic arrangements.

crossover from AF to F between $n=4$ and 5. This is due to the nearly vanishing magnetic moments in the Cr spacer (Fig. 3): only 75% of the bulk Cr moment is recovered for $n=30$ ($M_{\text{center}}\simeq 0.45\mu_B$) (Fig. 4). The corresponding sandwiches are characterized by long-ranged magnetic perturbations induced by the Fe/Cr interface in the Cr layers. These perturbations are themselves induced by a large number of frustrated Fe-Cr bonds (3 Fe-Cr for 75% of the interfacial Cr atoms) and by the corresponding disappearance of the Cr moments. Moreover, the nearly vanishing magnetic moments in the SL for large n values come from the interaction between the two perturbations induced by the neighboring interfaces. Finally, for case (e) we obtain an intermediate state between (b) and (d); the perturbation in the Cr spacer is less extended, as confirmed also by sandwich calculations. To summarize, the interface roughness at the atomic scale, the corresponding magnetic frustration, and the Cr magnetic-moment suppression induced long-ranged perturbations in the Cr spacer which contribute to reduce or even to kill the rapid oscillations.

In conclusion, we have shown that the electronic structure of the Cr spacer is strongly dependent on the interfacial couplings, on the interface roughness, and on the spacer's magnetic order. The short-period oscillations which have been observed for Fe/Cr SL are thought to result from the energy of a magnetic defect. They occur at low temperature for all Cr-based F/AF SL, but only for those with "flat" interfaces which do not introduce too many frustrated CrFe magnetic bonds. In the itinerant magnetism scheme we use here, the instability of the Cr magnetic moment with respect to its local environment —which is interpreted in terms of frustrated Cr-Cr and Cr-Fe bonds—is an essential phenomenon to understand the F/AF interfacial magnetism, the nature of the defects, the exchange anisotropy, and the IMC in sandwiches, bilayers, and SL. Such interfacial effects are usually neglected. In most of the models developed up to now[14–17]—see, for example, the work by Wang and Levy[14] for the Fe/Cr SL—the IMC result from the coupling between A "impurities" located in neighboring $(A)_m$ layers.[15] These impurities are assumed to be isolated, their electronic structure is approximated by Friedel-Anderson virtual bound states and they interact via the unperturbed *paramagnetic* spacer's band states.[14] Therefore, these models do not use a consistent picture of the interfacial and spacer's magnetism. They neglect the frustration effects we study here and they do not consider the band structure of the $F(A)_m$ layers[18] that we take fully into account. However, they can be used qualitatively to describe the asymptotic form of the IMC ($n\rightarrow\infty$) for nonmagnetic spacers, a limit for which the accuracy of the present approach is not sufficient.

Note finally that if we have shown that the interface roughness can change qualitatively the general features of the IMC oscillations, the interpretation of the long-period oscillations remains a challenge since a detailed study of the crystallographic and chemical nature of the interfaces has not been done. Even in the "pair models" we recalled previously,[15–17] the magnetic couplings result from the superposition of oscillations whose periodicities are $2\pi/D_F^i$, where D_F^i are Fermi-surface (FS) diameters related to the various sheets of the FS: they are thus *a priori* strongly dependent on the nature of the spacer and on the crystallographic plane from which the SL is built. Moreover, even if long-ranged oscillations can be obtained from an effect of *vernier* between the distance d and D_F^i [$2\pi/dD_F^i\simeq 1.11$ for Cu/Co SL (Ref. 15)] it is hard to believe that such an effect is always efficient and more especially for TM whose FS are complicated.

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