Noncollinear magnetic orders in Fe/Cr superlattices

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We calculate in a full self-consistent way the noncollinear distribution of magnetic moments in Fe_5/Cr_n ($n=1$–6) superlattices by means of a d-band tight-binding model. Self-consistency is obtained on both magnitude and orientation of the moments: only the relative orientation $\Delta\varphi$ between the central moments of two adjacent Fe layers is fixed, the other moments being free to orientate. We find that, when $\Delta\varphi$ is varied from 0 to 180°, the total energy of the system behaves in accordance with the phenomenological *proximity magnetism model* proposed by Slonczewski only when the Cr thickness is not too small. For very thin Cr layers ($n < 2$), the behavior is totally different. © *1997 American Institute of Physics.* [S0021-8979(97)46208-4]

Theoretical studies of magnetic order assuming noncollinear arrangement are gaining much interest lately because of the many accurate calculation methods existing and the ever growing computational capacities. Calculations of noncollinear magnetic orders are however still mainly restricted to systems with high symmetry and few inequivalent sites. In most studies, the orientation of the noncollinear magnetic moments is fixed and the total energy of the system is compared to the collinear configuration or to other noncollinear ones.[1,2] On the other hand, Kübler *et al.*[3] have performed a self-consistent calculation, on magnitude and angle, of the magnetism of bulk γ–Fe–Mn, $RhMn_3$, and $PtMn_3$, based on the local approximation to the density functional theory. Noncollinear magnetic orders can also occur in more complex systems such as multilayered structures. Many experimental studies have shown the existence of noncollinear couplings between ferromagnetic layers separated by a nonmagnetic or an antiferromagnetic material.[4–7] Such noncollinear magnetic arrangements can be induced by structural defects at the interfaces, like terraces, and are important to understand the mechanism of exchange coupling. In those situations, the assumption in calculations of helical magnetic order can prove to be too restrictive[8] and the computation of the magnetic moments distribution self-consistently in both magnitude and orientation is necessary. However, the *ab initio* methods still require too long a computation time to consider complex systems with many inequivalent sites and the three components of the magnetic moment vectors. In order to compute a full noncollinear magnetic order, more approximate methods are required such as the semiempirical tight-binding method.

Here we determine the noncollinear magnetic order in Fe/Cr superlattices. The motivations to study such a system are the large amount of interesting experimental results on Fe/Cr systems. Noncollinear couplings between Fe layers separated by a Cr spacer have been first reported by Rührig *et al.*[4] in 1991, and have since been reported by other groups.[5,9] It is to be noted that whereas most groups have reported 90° couplings, Schreyer *et al.*[5] have observed an unexpected 50° couplings in Fe/Cr superlattices. We calculate the noncollinear distribution of magnetic moments in Fe_5/Cr_n superlattices (with $n=1$–6) as a function of the relative orientation $\Delta\varphi$ between the central moments of two adjacent Fe layers.

We use the tight-binding model restricted to d electrons in which the Hamiltonian is given in the basis of atomic orbitals. In the case of noncollinear magnetic orders, the quantization axis is different from site to site. To express both parts of the Hamiltonian in the same spin basis, it is necessary to rotate the local quantization axis according to a reference direction z, by applying a spin 1/2 rotation matrix on the exchange part of H. The magnetic moments are obtained self-consistently by means of the real-space recursive method[10] with the relation $\Delta_i = I_i . M_i$, where Δ_i is the band splitting, I_i is the effective exchange integral, and M_i the local magnetic moment on the site i. Self-consistency is imposed on both the magnitude and the angle of the moments. The iterative process for a noncollinear distribution of moments consists in calculating the three components $[M_{r,i}(p), M_{\theta,i}(p), M_{\varphi,i}(p)]$ of the new local magnetic moment $\mathbf{M}_i(p)$ in the three directions $u_{\mathbf{r},i}(p)$, $u_{\boldsymbol{\theta},i}(p)$, and $u_{\boldsymbol{\varphi},i}(p)$ of the local basis; the resulting moment $\mathbf{M}_i(p)$ defining the direction of the local quantization axis for the next iteration. Self-consistency is obtained when, for the $(p+1)^{\text{th}}$ iteration:

$$\max_i\{|M_{r,i}(p+1)-M_{r,i}(p)|\}<\epsilon$$

$$\text{and} \quad \max_i\{|M_{\theta,i}(p)|, |M_{\varphi,i}(p)|\}<\epsilon$$

with ϵ equal to 10^{-5}. Preliminary calculations with self-consistency on both θ and φ have shown that the magnetic moments always remain in the (001) plane, allowing thus to fix θ to $\pi/2$ and to leave φ free to orientate. The hopping integrals and the exchange and Coulomb effective integrals used in our calculations are chosen to reproduce satisfactorily *ab initio* results obtained with the full potential augmented plane waves (FLAPW) technique and have been used in previous studies.[8,11]

In Fig. 1, the magnetic order obtained in the Fe_5/Cr_6 superlattice with $\Delta\varphi=120°$, is shown as well as the definition of $\Delta\varphi$. We see that when $\Delta\varphi$ is different from 0°, the usual collinear order, that is the antiferromagnetic coupling between Fe and Cr and the layer-by-layer antiferromagnetic structure of Cr,[12–14] is perturbated and a noncollinear order is induced in order to minimize the frustrations. It is also to be noted that Fe does not remain perfectly ferromagnetic within a layer.

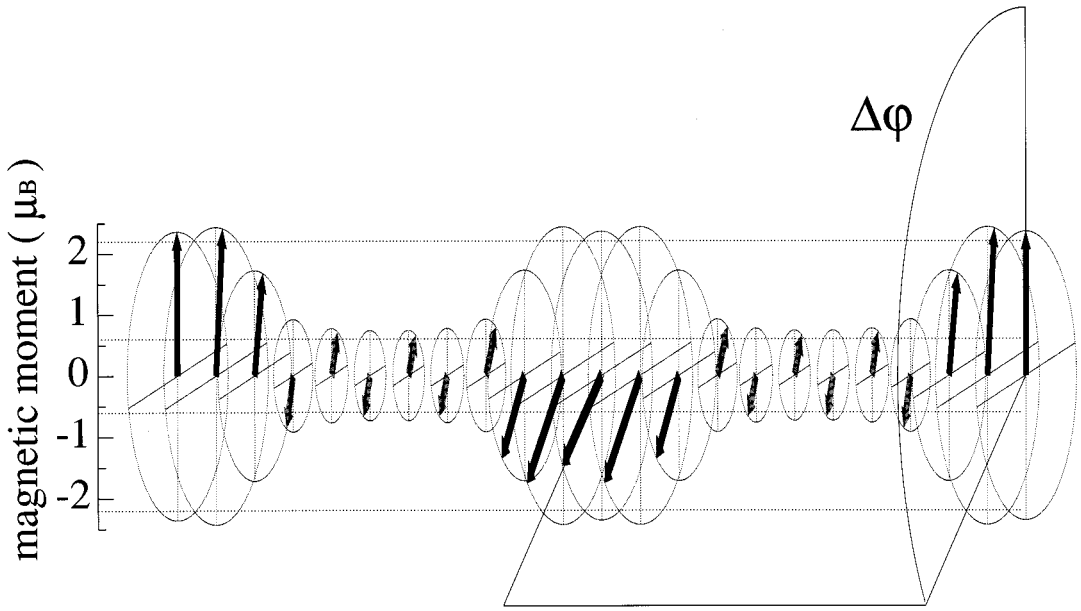

FIG. 1. Distribution of the magnetic moments obtained in the $Fe_5/Cr_6(001)$ superlattice with $\Delta\varphi=120°$. The black arrows correspond to Fe magnetic moments, the grey arrows to Cr magnetic moments, and $\Delta\varphi$ is defined as the relative orientation between the moments of the central atoms in two successive Fe layers. The radius of the circles is proportional to the magnitude of the moments. The horizontal dashed lines correspond to the bulk value of the Fe and Cr moments (respectively, 2.2 and 0.6 μ_B).

The evolution of the magnetic moment as $\Delta\varphi$ is varied from 0° to 180° is shown on Fig. 2 for two different Cr thicknesses: $n=6$ and $n=1$. The behavior of the moment obtained for $n=6$ is very similar to the one obtained for $n=5$, 4, 3, and 2, which are thus not shown here. The Fe moments are enhanced compared to the bulk value (2.20 μ_B) within a layer, but are reduced at the interface due to the *d*-band hybridization with Cr. The magnitude of the Fe moments remain almost constant when $\Delta\varphi$ is varied. All Cr moments are enhanced compared to their bulk value of 0.6 μ_B, but their magnitude decreases when $\Delta\varphi$ is very different from its value corresponding to the collinear order (180° when n is even, 0° when n is odd). The Cr moment behaves totally differently when the Cr spacer consists of only one monolayer ($n=1$). Whereas the behavior of the Fe moments do not change compared to the other cases ($n>1$), for $n=1$ the Cr moment is much more enhanced and is almost equal to the interfacial Fe moment (1.7 μ_B) in the collinear configuration. When $\Delta\varphi$ changes, the magnitude of the Cr moment decreases rapidly to a value slightly superior to its bulk value for $\Delta\varphi=180°$. On the other hand, the orientation of the moments behave in a similar way as a function of $\Delta\varphi$ whatever the value of n is (including for $n=1$): when $\Delta\varphi$ is small (or near 180° with even n), the angle varies linearly with $\Delta\varphi$. When $\Delta\varphi$ is larger than 90° (or smaller than 90° with even n), the moment rotates more slowly. It is to be noted that when n is odd, the angle of the central Cr atom varies perfectly linearly with $\Delta\varphi$ due to symmetry properties of the superlattice, the orientation of this moment being fixed *de facto* and being always orientated exactly in the direction of the bissector of $\Delta\varphi$. Such an effect is nonexistant when n is even.

Figure 3 gives the evolution of the total energy as a function of $\Delta\varphi$ for $n=6$, 2, and 1. As expected, the ground state occurs for the collinear order, that is with $\Delta\varphi=180°$ when n is even and $\Delta\varphi=0°$ when n is odd. The energy curves for $n=6$ and 2 have been fitted with the parabola $E=A_n(\Delta\varphi-\pi)^2$ on the [90°, 180°] interval. The values of the coefficients obtained are: $A_6=26.0$ and $A_2=25.7$ meV/crystallographic cell. For $n=6$, we find that the energy varies almost perfectly parabolically as a function of $\Delta\varphi$. This behavior is in accordance with a model recently proposed by Slonczewski,[15] the *proximity magnetism model*, which accounts for noncollinear exchange coupling between ferromagnetic layers separated by an antiferromagnetic spacer. This model is based on the antiferromagnetic structure of the spacer and on its thickness fluctuations. The exchange coupling energy is phenomenologically written: $E(\Delta\varphi)=J_+(\Delta\varphi)^2+J_-(\Delta\varphi-\pi)^2$ with $0\leqslant\Delta\varphi\leqslant\pi$ and where J_+ and J_- are two positive coefficients which, respectively, favor ferromagnetic and antiferromagnetic couplings. From the competition between both terms result noncollinear couplings, which can be different from 90° depending on the value of J_+ and J_-. In the case of a perfectly abrupt interface and an integer number of spacer monolayers, which is the case we have considered in our present calculations, one of the two coefficients vanishes: $J_+=0$ and $J_->0$ when the number of spacer layers is even (antiferromagnetic coupling), $J_-=0$ and $J_+>0$ when the number of spacer layers is odd (ferromagnetic coupling). The exchange coupling is thus expected to vary parabolically as a function of $\Delta\varphi$, as obtained in our calculation with $n=6$. Such an accordance with this model has also been found experimentally by Schreyer *et al.*[5] in Fe/Cr superlattices, indicating that the *proximity magnetism model* describes well the exchange coupling mechanism in such systems. The values they found for the exchange coupling coefficients are $J_+=5$ and $J_-=2$ mJ/m^2. Expressing our results in the same unit, we obtain $A_6=50.3$

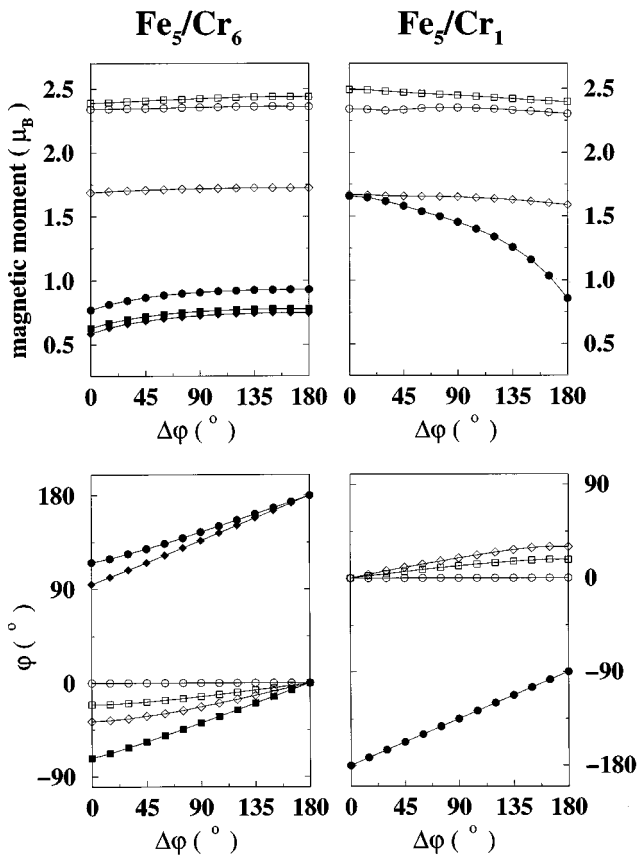

FIG. 2. Variation of the magnitude and the angle of the magnetic moments as a function of $\Delta\varphi$, in the $Fe_5/Cr_6(001)$ and $Fe_5/Cr_1(001)$ superlattices. The angles φ are given relative to the central Fe atom. The empty symbols correspond to Fe sites, and the filled symbols to Cr sites. The circles correspond to the central sites, the squares to the intermediate sites, the diamonds to the interfacial sites.

and $A_5=60.5$ mJ/m^2, which correspond, respectively, to J_- and J_+. Those values are more than ten times larger than the experimental ones. Such a discrepancy between calculated and measured coupling strengths is well known.[16]

When the thickness of Cr is reduced, the energy curves does not vary perfectly parabolically anymore. This is shown on Fig. 3 for $n=2$, and a similar behavior is also found for $n=5$, 4, and 3. The curves deviate significantly from the parabola when $\Delta\varphi$ is very different from its collinear order value. For $n=1$, the behavior is totally different. The energy curve is not monotonous anymore. The energy increases very rapidly and then decreases to a local minimum for $\Delta\varphi\simeq 90°$. The origin of such a behavior is not known so far. It could be due to both the large disorientation between the Fe and the large Cr interfacial magnetic moment and the limitation of our *d*-band model. The energy being very small ($\sim$5 meV/crystallographic cell), such a behavior might be altered when taking into account the ''*sp*'' electrons as well.[17]

The discrepancy with the *proximity magnetism model* when the Cr thickness is small (except $n=1$) can be explained by the assumptions made in the model. It is indeed assumed: (i) the magnitude of all moments remains constant, (ii) the Fe layers remain ferromagnetically ordered within a layer, and (iii) only small perturbation of the collinear magnetic order occurs. All three assumptions are clearly not satisfied when n is small and when $\Delta\varphi$ is larger than 90° for even values of n and when $\Delta\varphi$ is smaller than 90° for odd values of n.

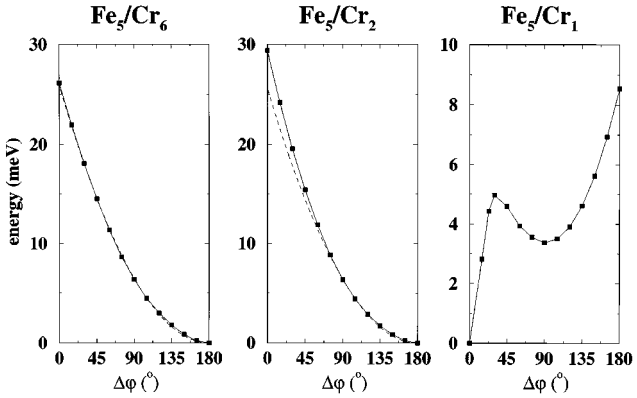

FIG. 3. Variation of the total energy per crystallographic cell as a function of $\Delta\varphi$, in the $Fe_5/Cr_6(001)$, $Fe_5/Cr_2(001)$, and $Fe_5/Cr_1(001)$ superlattices (squares and solid line). The dashed line corresponds to the fit of the curves by the parabola $E=A(\Delta\varphi-\pi)^2$, on the [90°,180°] interval.

As perspective to this study, it appears interesting to consider a more realistic interface with structural defects. In particular, from the presence of a monoatomic step, at one interface would result in the competition of both term in the expression of the exchange energy and a noncollinear coupling that should be displayed in the ground state. The distribution of the magnetic moments near such a defect is also interesting in order to understand the magnetic ordering of the Cr moments in the spacer.

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