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Calculated angular dependence of the interlayer couplings in Fe/Cr superlattices having imperfect interfaces

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Abstract

We present calculations of the non-collinear magnetic structure in Fe/Cr superlattices having imperfect interfaces modeled by considering atomic steps in the Cr layers and Fe/Cr interfacial ordered compounds. The interlayer couplings are obtained directly from self-consistent tight binding band structure calculations. We show that the bilinear–biquadratic expression for the coupling energy fits nicely the calculated interlayer couplings curves.

Keywords: Electronic structure; Exchange coupling—bilinear–biquadratic; Interface roughness; Multilayers—metallic

This last decade, considerable effort has been devoted to enhance the theoretical description of the magnetic configuration at the atomic scale in more and more complex nanostructured systems combining angular and magnitude degrees of freedom of the magnetic moment. When non-collinear magnetic structures are considered in metallic systems, the itinerant character of the transition metals requires a band structure description. Such an approach is needed to describe the interlayer exchange coupling in metallic multilayers in particular when the spacer layer, separating two ferromagnetic layers, is itself magnetically ordered, like in the Fe/Cr case, where magnetic frustrations play a major role on the overall properties. The method we use describes correctly the Cr magnetism in Fe/Cr multilayers for the range of Cr thickness we consider here even if it does not include the SDW. Frustrations due to imperfections are present whatever the thickness is since they originate from the interfaces and can extend over a significant fraction of the spacer layer [1,2]. Up to now, only self-consistent collinear solutions have been considered resulting in an overestimation of the frustration energy and limiting the determination of the interlayer coupling to the relative energy between parallel and antiparallel magnetic states [1,2].

The study of Fe/Cr multilayers needs to include interfacial imperfections being at the origin of non-collinear magnetic features [3,4] as well as strong reductions of the Cr local magnetic moments [5]. This can only be done with simplified electronic structure descriptions (like the real space tight binding approach) allowing to build large unit cells containing a few tens of non-equivalent atoms. Our method is completely equivalent to ab initio ones for the determination of total energy differences in Fe/Cr systems as shown by the comparison between our results (Fig. 2 in Ref. [6]) to the recently ab initio one published by the group of Dresden (Figs. 2 and 3 in Ref. [7]) obtained with spiral magnetic states in the Cr spacer. This comparison shows a very good quantitative agreement between both results : they obtain a total energy difference of 40.8 meV per unit cell for 6Fe/6Cr superlattices whereas we obtained 41.2 meV for 5Fe/5Cr (a more recent calculation for 5Fe/6Cr gives 37.8 meV per unit cell). However, since they do not allow the direction of the magnetic moments to relax, they obtain only a bilinear contribution to the interlayer coupling energy. Details on the calculation method can be found in previous papers [8,9]. In order to reduce the computer time, we assume

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that all magnetic moments remain in the same plane and we build the smallest possible unit cells by considering straight line [0 1 0] steps and ordered compounds in a doubled in plane cell. Our calculations remain nevertheless at the limit of what can be done with the most recent generation of massively parallel computers.

As previously reported for perfect interfaces [6], the calculated interlayer coupling energy $E_C(\Delta\theta_M)$ between the magnetisation of successive Fe layers follow the bilinear–biquadratic expression

$$E_C(\Delta\theta_M) = -J_1 \cos(\Delta\theta_M) + J_2 \cos^2(\Delta\theta_M), \quad (1)$$

where J_1 and J_2 are, respectively, the bilinear and biquadratic coupling constants and $\Delta\theta_M$ is the angle between the magnetisation of the two successive Fe layers. For small Cr thickness n (smaller than 11 atomic layers (AL)), collinear magnetic states are obtained for $\Delta\theta_M = 0^\circ$ and 180°. This is related to the vanishing of the inner Cr magnetic moments when frustration increases. On the contrary, for n larger than 12 AL, $E_C(\Delta\theta_M)$ follows the parabolic expression first evidenced by Slonczewski [4]

$$E_C(\Delta\theta_M) = C\left(\frac{\Delta\theta_M - \Delta\theta_{min}}{\pi}\right)^2, \quad (2)$$

where C and $\Delta\theta_{min}$ are, respectively, the coupling constant and the angle corresponding to the energy minimum. This shows clearly that, for perfect interfaces, nearly spiral magnetic states occur in the Cr spacer for n large enough.

Up to now we have only considered the coupling energies for perfect interfaces due mainly to computer time limitation. The speed of the convergence and the access to recent powerful massively parallel computers allow to determine directly the interlayer coupling for large cells taking into account interfacial imperfections. As usual in such kind of studies, we increase the in plane cell and include Fe and Cr atoms in the same atomic layer. Mixed interfaces are modeled by $c2 \times 2$ and 2×2 in plane chemical cells [1,2]. The $c2 \times 2$ cell corresponds to a $Fe_{0.5}Cr_{0.5}$ mixed interfacial atomic layer and the 2×2 one corresponds to two $Fe_{0.75}Cr_{0.25}/Fe_{0.25}Cr_{0.75}$ mixed interfacial atomic layers. Atomic steps, separating Cr and Fe terraces corresponding to a periodic repetition of λ Cr and λ Fe atomic rows, at one interface are built with a $2\lambda \times 1$ in-plane cell; these cases are denoted by the terraces size $TS = \lambda + \lambda$. Finally, with a $2\lambda \times 2$ in plane cell, we can consider simultaneously both interfaces; these cases are denoted by $TS = (\lambda + \lambda) \times 2$.

The coupling energy for a given value of $\Delta\theta$ is calculated by maintaining fixed the angle for all Fe atoms of the atomic layer in the middle of the Fe layers (see Fig. 1 of Ref. [6]). We define $\Delta\theta_M$ by the angle between the two successive magnetisation resulting from

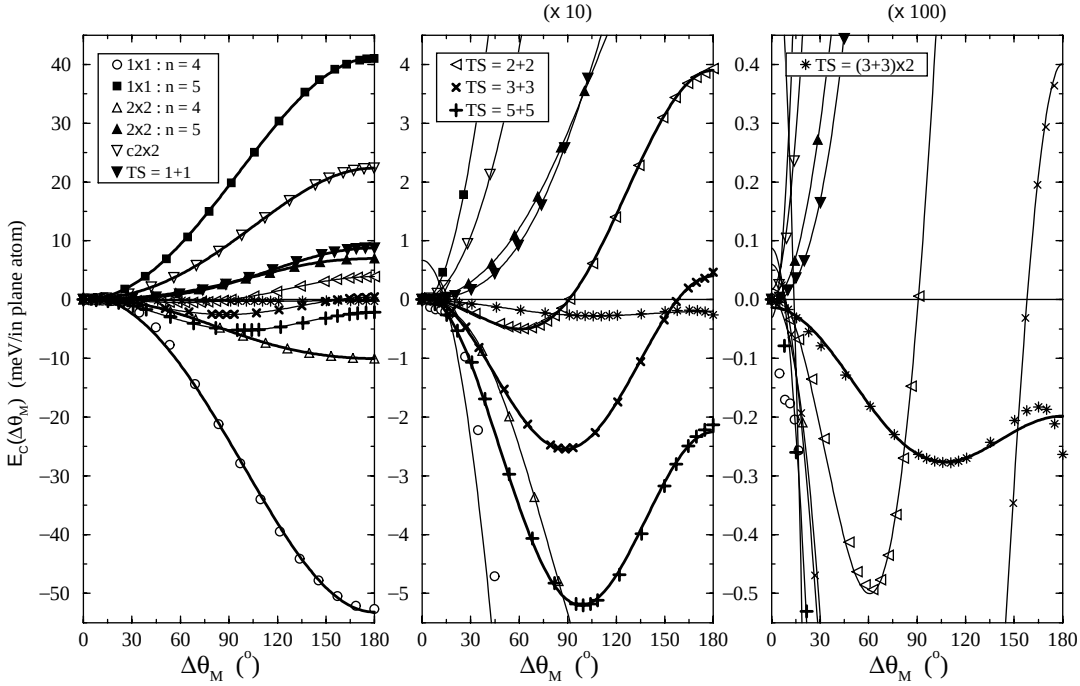

Fig. 1. Calculated (*symbols*) and fitted (*solid lines*) coupling energies E_C as a function of $\Delta\theta_M$ for all Fe/Cr multilayers considered in this work having a Cr thickness between 4 and 5 atomic layers. The two graphs at right correspond to a zoom of the graph at left for small energies.

Table 1
Coupling constants J_1, J_2, C (meV/in plane atom) obtained by fitting the calculated interlayer magnetic couplings curves displayed in Fig. 1. $\Delta\theta_M$(min) corresponds to the position of the energy minimum

1×1	$n = 4$	$n = 5$	$n = 10$	$n = 11$	$n = 20, 21$
J_1	−26.9	20.6	−15.4	15.2	$C_{20} = 29.0$
J_2	−2.23	1.15	5.40	4.58	$C_{21} = 26.8$
$\Delta\theta_M$(min)(°)	180	0	180	0	180/0
		2×2: $n = 4$	2×2: $n = 5$	$c2 \times 2$	TS = 1 + 1
J_1		−5.15	3.55	11.2	4.40
J_2		0.35	0.65	1.76	1.67
$\Delta\theta_M$(min)(°)		180	0	0	0
TS		2 + 2	3 + 3	5 + 5	(3 + 3) × 2
J_1		1.94	0.17	−1.17	−0.09
J_2		1.99	2.77	4.03	0.16
$\Delta\theta_M$(min)(°)		60	88	99	107

the calculation at fixed $\Delta\theta_M$. The substitution of $\Delta\theta$ by $\Delta\theta_M$ introduces a slight distortion in the $E_C(\Delta\theta_M)$ curves varying mainly their curvature and affecting directly the coupling constants listed in Table 1 obtained by fitting the calculated values (Fig. 1) with expressions 1 and 2. Fig. 1 shows $E_C(\Delta\theta_M)$ for all cases considered for n varying between 4 and 5 AL. The most remarkable result is that for a Cr thickness smaller than 12 atomic layers, all calculated coupling curves fit nicely expression 1. For perfect interfaces (first line of Table 1), the coupling constants fluctuate significantly for small Cr thickness with a surprising negative J_2 for $n = 4$ whereas for the largest thickness they become similar in magnitude. In agreement with the experiments [10], we found J_2 nearly equal to $|J_1|/3$ but the coupling strength is overestimated by at least one order of magnitude. The introduction of interfacial mixing at one interface (second line of Table 1) reduces significantly the coupling strength by a factor 3 to 4 and the energy minimum still corresponds to a collinear magnetic solution since $J_2 < |J_1|/2$. For imperfections having a larger periodicity (third line of Table 1) than these last cases, the energy minimum occurs for non collinear magnetic states with even J_2 larger than $|J_1|$. When the terrace size λ increases for TS $= \lambda + \lambda$, (i) J_1 diminishes rapidly and becomes negative for λ larger than 3 atomic rows and (ii) J_2 increases. This shows clearly that the coupling strength which was determined directly differs completely from the value deduced by averaging over values calculated for perfect interfaces. However, with these stepped interfaces, the coupling strength remains significantly higher than the experimental one. It is only for the last system we considered, with TS $= (3 + 3) \times 2$, that we obtain extremely small coupling constants (Table 1) around 0.1 meV/in plane atom demonstrating that both kind of imperfections are required for a realistic simulations. To our knowledge, it is the first time that a direct band structure calculation in Fe/Cr has been done taking into account both imperfections evidenced experimentally [10,12] and giving coupling strengths values similar to experiment. The coupling strength experimentally found by Heinrich et al. [10] are $|J_1| = 0.35$ and $J_2 = 0.13$ meV/in plane atom. The coupling strength experimentally found by Schreyer et al. [11] are $J_+ = 2.5$ and $J_- = 1.0$ meV/in plane atom (+ and − corresponding, respectively, to an odd and even number of Cr atomic layers).

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References

[1] D. Stoeffler, F. Gautier, Phys. Rev. B 44 (1991) R10389.
[2] J. Magn. Magn. Mater. 156, (1996) 114.
[3] A. Schreyer, et al., Phys. Rev. Lett. 79 (1997) 4914.
[4] J.C. Slonczewski, J. Magn. Magn. Mater. 150 (1995) 13.
[5] K. Mibu, et al., Phys. Rev. Lett. 84 (2000) 2243.
[6] C. Cornea, D. Stoeffler, Europhy. Lett. 49 (2000) 217.
[7] B. Yu. Yavorsky, et al., Phys. Rev. B 62 (2000) 9586.
[8] C. Cornea, D. Stoeffler, Comput. Mater. Sci. 10 (1998) 245– 249.
[9] C. Cornea, Thesis of the University Louis Pasteur, Strasbourg, April 1999, unpublished.
[10] B. Heinrich, J.F. Cochran, T. Monchesky, R. Urban, Phys. Rev. B 59 (1999) 14520.
[11] A. Schreyer, et al., Phys. Rev. B 52 (1995) 16066.
[12] A. Davies, et al., Phys. Rev. Lett. 76 (1996) 4175.