Response of magnetically frustrated nanostructures to external magnetic fields: Stepped Cr/Fe interfaces

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The response of typical magnetically frustrated systems to an external magnetic field is investigated in order to get insight on the qualitative trends and their origin at the electronic-structure level. Electronic-structure calculations are conducted for Cr overlayers of different thickness on the stepped Fe(001) substrate, using a self-consistent noncollinear tight-binding method in which the Hamiltonian has been extended to account for the interaction with the external field. The average magnetization at the Cr overlayers is obtained through the local magnetic moments distribution of the system at different external magnetic fields. The complex nonmonotonous behavior of the remagnetization process as a function of the external field is explained in terms of the local geometrical and chemical environment of the inequivalent Cr atoms which leads to local magnetic couplings having different response. In particular, we find minima at low fields due to the strength of the local antiferromagnetic couplings and changes in slope at higher fields associated to spin-flip transitions taking place at certain Cr atoms when the system is trapped in collinear magnetic configurations.

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I. INTRODUCTION

Transition-metal interfacial nanostructures are a matter of intense research, both experimentally and theoretically, due to their fundamental and technological relevance. The rich variety of magnetic behaviors that one may encounter depending on the constituent elements and the structural characteristics of the interface (smoothness, roughness, interdiffusion, …) can be used to design new magnetic devices, provided that one is able to understand and predict the underlying physics. A well-known example of this is the giant magnetorresistance (GMR) (Refs. 1 and 2) and its use in the development of GMR sensors and the read and write heads of the new generation of hard disks in computers.

Fe/Cr interfacial systems are the prototype of ferromagnetic (F) -antiferromagetic (AF) interfaces so they have been widely investigated. Both Fe and Cr crystallize in the body-centered-cubic structure and have quite similar lattice parameter which ensures a small lattice mismatch when grown together in the multilayer (ML) or overlayer (OL) configurations that are usually grown. The intrinsic magnetic properties of these interfacial Fe/Cr systems in the ML and OL configurations are now reasonably well understood. For instance, the oscillations of the interlayer exchange coupling between Fe slabs as a function of the thickness of the Cr spacer are explained,[3–7] and noncollinear magnetic arrangements have been characterized in ML and alloys[8–11] as well as in stepped interfaces due to magnetic frustrations.[12]

Defects like steps, roughness, or interdiffusion are usually present at the interfaces of transition-metal systems, and in the particular case of Fe/Cr interfaces, their effect on the intrinsic magnetic properties has been recently investigated.[12,13] In previous work, we found that magnetic frustration associated to those defects lead to the development of noncollinear magnetic arrangements which in general are not strictly localized around the defect. However, in order to deeply understand the magnetic nature of the system, not only the intrinsic properties have to be determined but also its response to external magnetic fields which, besides, are commonly used in the experimental characterizations, such as in the GMR studies or in determining hysteresis cycles.

The aim of the present work is to study the response of a typical magnetically frustrated system to an external magnetic field in order to get insight on the qualitative trends expected at the macroscopic level (averaged quantities) and their origin at the electronic-structure level. We have chosen the Fe/Cr stepped interface and, in particular, the OL configuration of a Cr film deposited on the stepped Fe substrate. We have conducted calculations, as a function of the Cr coverage (between one and two Cr monolayers), using the same noncollinear tight-binding (TB) model employed in our previous studies[12,13] but in which the Hamiltonian has been extended to account for the interaction with an external magnetic field. This model has been already applied to study the response of Mn films supported on the ideal Fe(001).[14] Since we are interested in general qualitative trends, we have taken an ideal system and made several approximations, as in our previous work:[14] calculations have been carried out at $T=0$ K in the mean-field approximation, we have not considered interdiffusion and structural relaxation.

The rest of the paper is organized as follows. In the next section, we give the details of our theoretical approach. In Sec. III, we present the results and the main conclusions are summarized at the end.

II. THEORETICAL MODEL

Our semiempirical theoretical model is based on the fully self-consistent TB method[12,13] that we extended to account for the interaction of the spin moments with an external magnetic field.[14] Our TB method is parametrized through a mapping to the *ab initio* TB-LMTO[15] Hamiltonian in the first-order expansion of its TB form and in the Slater-Koster approximation, from which the two-center hopping integrals can be extracted. The spin-quantization axis is independent

in each site, thus allowing to describe noncollinear magnetism. The electronic charge distribution and the local magnetic moments in the system are obtained by self-consistently solving, using the Haydock's recursion method,[16] the resulting one-particle TB Hamiltonian for the *s*, *p*, and *d* valence electrons in a mean-field approximation (equivalent to the unrestricted Hartree-Fock approximation) which retains the main electronic exchange-correlation effects to account for the itinerant magnetism in transition-metal systems at $T=0$ K.

The Hamiltonian can be split into three terms: the spin-independent term H_{ind}, the spin-dependent term H_{mag}, and the term accounting for the interaction with the external magnetic field H_{Bext}. A localized orthogonal atomic-orbital basis $\{|i\alpha\rangle\}$ is introduced to represent in matrix form the Hamiltonian $H=H_{ind}+H_{mag}+H_{Bext}$, where

$$H_{ind}=\sum_{\substack{i,j\\ \alpha,\beta}}\left[(\epsilon_{i\alpha}^{0}\delta_{\alpha\beta}+U_{i\alpha,j\beta}\langle\hat{n}_{j\beta}\rangle+Z_i\Omega_{i\alpha}\delta_{\alpha\beta})\delta_{ij}+t_{ij}^{\alpha\beta}(1-\delta_{ij})\right]|i\alpha\rangle\langle j\beta|\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}, \tag{1}$$

$$H_{mag}=\sum_{i,\alpha,\beta}\left(-\frac{1}{2}J_{i\alpha\beta}\mu_{i\alpha}\right)|i\beta\rangle\langle i\beta|\begin{bmatrix}\cos\theta_i & e^{-i\phi_i}\sin\theta_i\\ e^{i\phi_i}\sin\theta_i & -\cos\theta_i\end{bmatrix}, \tag{2}$$

$$H_{Bext}=-\sum_{i,\alpha}\frac{g_s\mu_B}{\hbar}\vec{B}_i\vec{S}_i|i\alpha\rangle\langle i\alpha|=-\frac{1}{2}g_s\mu_B\sum_{i,\alpha}|i\alpha\rangle\langle i\alpha|\begin{bmatrix}B_i^z & B_i^x-iB_i^y\\ B_i^x+iB_i^y & -B_i^z\end{bmatrix}. \tag{3}$$

The hopping integrals $t_{ij}^{\alpha\beta}$ between orbitals α and β of different sites i and j are assumed, as usual, to be spin independent. The spin-independent diagonal elements $\epsilon_{i\alpha}^{0}+U_{i\alpha,j\beta}\langle\hat{n}_{j\beta}\rangle+Z_i\Omega_{i\alpha}$, are the sum of (i) the orbital level of the isolated atom $\epsilon_{i\alpha}^{0}$, (ii) the electrostatic level shift $U_{i\alpha,j\beta}\langle\hat{n}_{j\beta}\rangle$, accounting for the electronic charge redistribution which is parametrized by the Coulomb integral $U_{i\alpha,j\beta}$, and (iii) the crystal-field potential which, as a first approximation, reads $Z_i\Omega_{i\alpha}$, where Z_i is the local atomic coordination of site i and $\Omega_{i\alpha}$ is a potential which depends on the orbital character (localized like the *d* orbitals or delocalized like the *sp* ones).

H_{mag} describes the magnetic effects, $J_{i\alpha}$ being the exchange parameter, and $\mu_{i\alpha}$ the local magnetic moment, whose direction is given by the angles (θ_i,ϕ_i) defined in the spin-rotation matrix which is applied to the spin basis at each atomic site. Notice that, in the noncollinear framework, the spin-quantization axis is different at each site.

H_{Bext} describes the interaction of the local spin magnetic moments with the external magnetic field $\vec{B}_i$, g_s being the gyromagnetic factor, and μ_B the Bohr magneton. In the present work, we focus on the response to uniform external fields and, therefore, for the calculations it is pertinent to choose the direction of the external magnetic field (which is the same for all atomic sites) as the direction of the reference (laboratory) quantization axis *z*, to which the local spin-quantization axes are rotated. In such conditions, the matrix in Eq. (3) has a diagonal form since σ_x and σ_y Pauli matrices do not enter in the expression.

The local densities of states (LDOS) are calculated through the retarded Green's function using the recursion method[16] in the real space which provides the local-orbital projected LDOS on an arbitrary local quantization axis through a rotation of the initial recursion vector in the spin space. We calculate electronic occupations and magnetic moments at each orbital and site of the system by integrating the LDOS up to the Fermi level, with the global charge neutrality condition. All the parameters of the TB model as well as the interlayer distances have been taken as in Ref. 12.

It is worth analyzing the orders of magnitude involved in the interaction with the external magnetic field. 1 T$=5.789\times10^{-5}$ eV$/\mu_B$: this means that extremely high external fields are needed to overcome typical antiferromagnetic couplings in transition metals such as Cr. This is expected to come out from our model, particularly if finite-temperature effects and interdiffusion are not taken into account. For an accurate quantitative description, these effects as well as the structural relaxation should be considered in the calculation. However, although our model tends to overestimates, for instance, the magnetic saturation fields, the general trends and their microscopic origin can be well understood. The same crystalline structure of Fe and Cr bulks (bcc) and their similar lattice parameters justify our not having considered structural relaxation in a first approximation (constant volume approximation and pseudomorphic growth). We note that a description by means of electronic-structure calculations is a step forward over classical phenomenological approaches. Although the Heisenberg model can also account for the spin reorientations and noncollinear magnetism in frustrated magnets, it is better suited to describe localized magnetism (like that of Gd) than to describe band (or itinerant) magnetism typical of transition-metal-based systems. The Heisenberg model uses several parameters for describing the magnetic behavior, like the exchange constants (between first and second neighbors) and the moduli of the localized magnetic moments (usually taken from the respective bulks). The main drawback when dealing with itinerant magnetism is that the

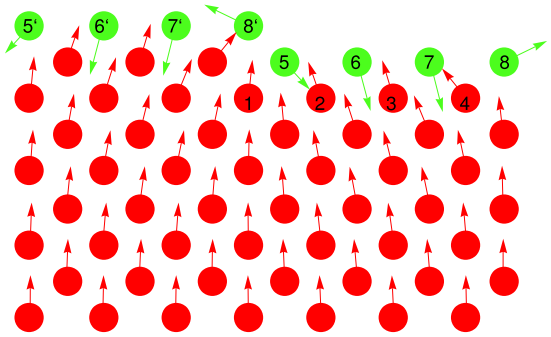

FIG. 1. (Color online) Ground-state noncollinear magnetic configuration of the Cr monolayer deposited on a stepped Fe(001) substrate (only a fraction of the system is represented). The arrows are proportional to the modulus of the local magnetic moments. No absolute direction for the magnetic moments exists since the spin-orbit interaction is not taken into account.

effect of the electronic delocalization (described in the TB method through the hopping integrals) is not taken into account and, therefore, the decrease (increase) in the local magnetic moments due to the increase (decrease) in atomic coordination is not accounted for. Besides, the typical reduction in the local magnetic moments occurring in the frustrated regions is not appropriately described. All those effects are present in our systems. Besides, an electronic-structure-based model has more predictive character.

III. RESULTS

As indicated in Sec. I, we have performed calculations for a Cr film deposited on the stepped Fe(001) substrate. We have considered different Cr coverages, starting from the monolayer and increasing row by row up to completing the bilayer, and terraces with a width of 4 atomic rows. The main reason to have chosen terraces of 4 atomic rows in width is that this width is large enough to contain surface and interface atoms with essentially all the possible local atomic coordinations that one may encounter in systems with larger terraces. In particular, these 4-atoms width terraces have atoms, at the central positions that resemble the surface atoms in the ideal (step-free) semi-infinite system as regards their atomic coordination and symmetry up to next-nearest neighbors. The intrinsic properties of these systems, that is, in the absence of external fields, were investigated by part of the authors[12] in order to explain the role played by the magnetic frustration associated to the structural defects (monatomic steps at the Fe/Cr interface) in the magnetic properties of the system. Due to the particular configuration of these systems, with an AF/F interface between Cr and Fe, together with the presence of monatomic steps, it is not possible to find a collinear configuration in which the natural couplings (AF between Cr-Cr and Cr-Fe, and F between Fe-Fe) are fulfilled in the whole system. Therefore, the local magnetic moments rotate in order to reach a more stable configuration, where the magnetic frustrations are reduced. The noncollinear ground-state configuration was consistent with stronger AF couplings between Cr-Cr nearest neighbors than between Cr-Fe ones. This noncollinear configuration is illustrated in Fig. 1 in the case of the supported Cr monolayer, and more details can be found in our previous paper.[12]

Let us now discuss the response of those F/AF OL systems to a uniform external magnetic field. The magnetic field is applied along the direction of the magnetic moment of the Fe substrate, which defines now an absolute direction in the space.

For the Cr monolayer deposited on the stepped Fe(001) substrate, we show in Fig. 2 the evolution of the local magnetic moments (both the moduli and the orientation with respect to the magnetic field) of each inequivalent Cr atom in the system (illustrated in Fig. 1). As general trends, the local magnetic moments tend to align with the magnetic field, the local antiferromagnetic couplings being quite strong, as expected (their alignment with the field requires a huge value of the external field, B_A^0). A nonmonotonous behavior is found as a function of the external field and the details of this complex remagnetization process can be understood in terms of the different local geometrical and chemical environments of the Cr atoms.

The nearest neighborhood of the atoms of types 6 and 7 (equivalent to 6′ and 7′ by symmetry) is composed of four Fe atoms (types 2, 3 and 3, 4 in Fig. 1, respectively). Besides, Cr 6 and 7 are relatively far from the step. Therefore, in the absence of magnetic field, their magnetic couplings with the Fe atoms are essentially the natural one (AF). When we start applying the magnetic field, the magnetic moments of Cr 6 and 7 tend at first to align antiparallel to the field since they try to keep their AF coupling with the Fe moments

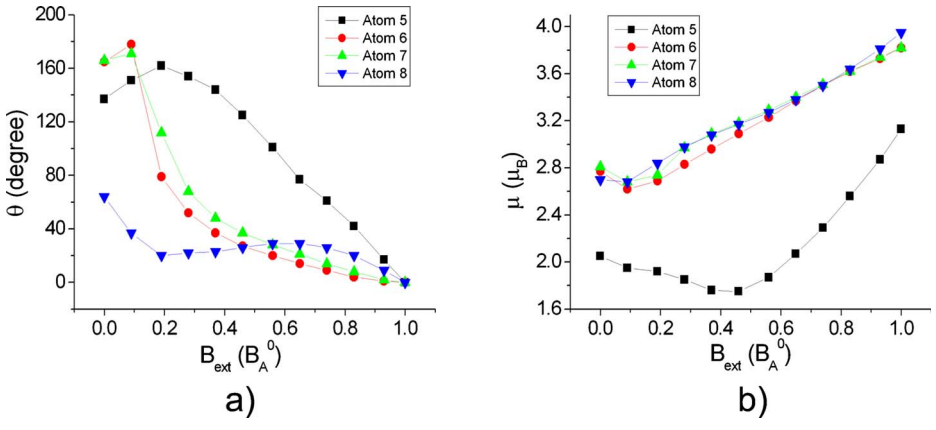

FIG. 2. (Color online) (a) Evolution of the magnetic moments of the atoms marked in Fig. 1 as a function of the external uniform magnetic field (in units of B_A^0, the field required to align all the magnetic moments in the direction of the external field). (b) The same as in (a) for the moduli of the magnetic moments.

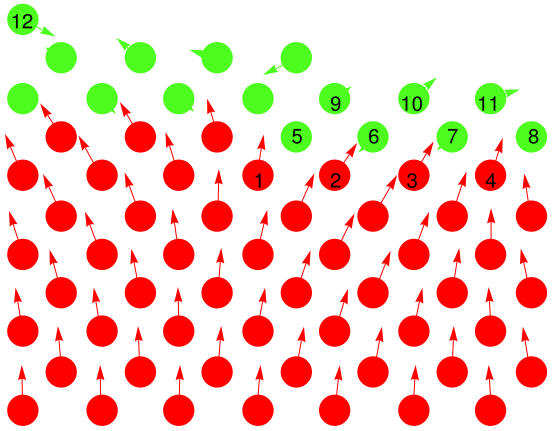

FIG. 3. (Color online) As in Fig. 1 for the Cr bilayer deposited on the Fe stepped substrate.

which rotate toward the field. This is reflected in the increase in the angle at low external fields up to values of approximately $0.1B_A^0$ [Fig. 2(a)]; the magnitude of the field up to this value is not yet enough to overcome the AF coupling between Cr and Fe. Keeping this AF coupling within this field range implies a reduction in the modulus of the local moment at those Cr atoms [Fig. 2(b)]. If we further increase the intensity of the applied magnetic field, we observe a marked change in this tendency and when the field is large enough, the moments of Cr 6 and 7 start rotating into alignment with the field. At the same time, they start increasing their moduli to converge to the magnetic saturation limit.

Cr atoms of types 5 and 8 of Fig. 1 behave in a markedly different way as compared with those of types 6 and 7. Atoms of type 5 are surrounded by four nearest Fe neighbors (types 1 and 2 in Fig. 1) and two nearest Cr neighbors (type 8′) while the Cr atoms of type 8 have as nearest neighbors two Fe atoms and two Cr ones. Therefore, AF Cr-Cr couplings are present now. In the ground-state configuration (without applied field), we can already see that the Cr-Cr couplings (atom 8 with 5′, and also 5 with 8′) are stronger than the Cr-Fe couplings since these Cr-Cr AF couplings are closer to the perfect antiparallel alignment (they are close to 180°) than the Cr-Fe ones (close to 90°). When applying the magnetic field, we can observe that higher fields are required to overcome the Cr-Cr couplings than to overcome the Cr-Fe ones. Thus, now up to fields of about $0.2B_A^0$, the magnetic moment of the Cr atoms of type 5 keeps aligned antiparallel to the field as a consequence of its strong coupling with the moments of atoms 8′ which easily rotate toward the field since they are close to 90° (a bit less) with respect to the field, thus the torque from the field on them being very strong [see Fig. 2(a)]. We see that the evolution of the orientation with the external field is now much slower than for Cr atoms of types 6 and 7. We conclude that the Cr-Cr AF couplings are stronger here than the Cr-Fe ones. The variation in the moduli of the local magnetic moments also shows this effect, as one can see in Fig. 2(b). An increase in the angles at low fields correlates with a decrease in the moduli. The moduli start increasing as soon as the moments start to rotate toward the field and depart from the local AF alignment, that is, when the field is large enough to overcome the antiferromagnetic exchange couplings, and they keep increasing to approach the magnetic saturation. In the supplementary material, the field-dependent local magnetic-moment distribution for this system is illustrated with an animation.[17]

Our results show that for a deep understanding of the remagnetization process of these systems as a function of the external field, it is necessary to analyze their behavior at the atomic scale and, in particular, the local geometrical and chemical environment which leads to the different local magnetic couplings having different response to the external field.

In order to enrich the discussion and to provide more information, we consider now different Cr coverages. We increase the Cr coverage row by row until reaching the deposited Cr bilayer, as illustrated in Fig. 3. One additional Cr row corresponds to the Cr atoms of type 9; for two additional Cr rows we will add types 9 and 10 and so on up to reaching the bilayer. As approaching the bilayer limit, Cr atoms of surface kind will coexist with Cr atoms of interface kind which have much lower magnetic moments. Besides, the number of different sort of magnetic couplings increases now and a detailed analysis becomes much more difficult than for the supported monolayer discussed above.

The results obtained for the different coverages are depicted in Fig. 4 and in the supplementary material, we illustrate the field-dependent local magnetic moments distribution with an animation for each coverage.[17] In order to further explore the strength of the Cr-Cr coupling, let us focus on the Cr atoms of type 5. The modulus of their magnetic moment in the absence of external field decreases when increasing the Cr coverage. The moment of Cr 5 decreases from $2\mu_B$ in the supported monolayer [Fig. 2(b)] to about $0.8\mu_B$ in the bilayer, approaching the Cr-bulk value [Fig. 4(d)]. This effect is the logical consequence of increasing the coordination number and the loss of the surface character. This atom is also affected in a different way, depending on the coverage, when applying a magnetic field. As one can observe in Fig. 4 (left panels), the field required to rotate the magnetic moment of atom 5 into alignment is markedly higher than in the Cr ML discussed previously (see Fig. 2). We can take as a reference Fig. 4(a) in which a single Cr row is added. The Cr atoms of type 5 have now four Cr nearest neighbors. The required magnetic field to rotate the local magnetic moment is now much larger, due to the fact that this field must overcome now more Cr-Cr couplings and the couplings in the nearest neighborhood of Cr 5 are close to the natural ones. Similar analysis can be done for the other atoms. Another interesting feature is that for Cr coverages larger than the monolayer, and particularly for those closer to the bilayer limit, the system is trapped in collinear magnetic configurations for certain ranges of the external field. This can be observed in the left panel of Fig. 4 where, for those field ranges, the angle of each Cr moment is either 0° or 180°. When the systems are trapped in those collinear magnetic configurations, only the modulus of the local magnetic moments changes with the external field until spin-flip transitions take place. We note that these steps in the angular distribution when spin flips take place, correlate with abrupt changes in the modulus of the moment of the corresponding

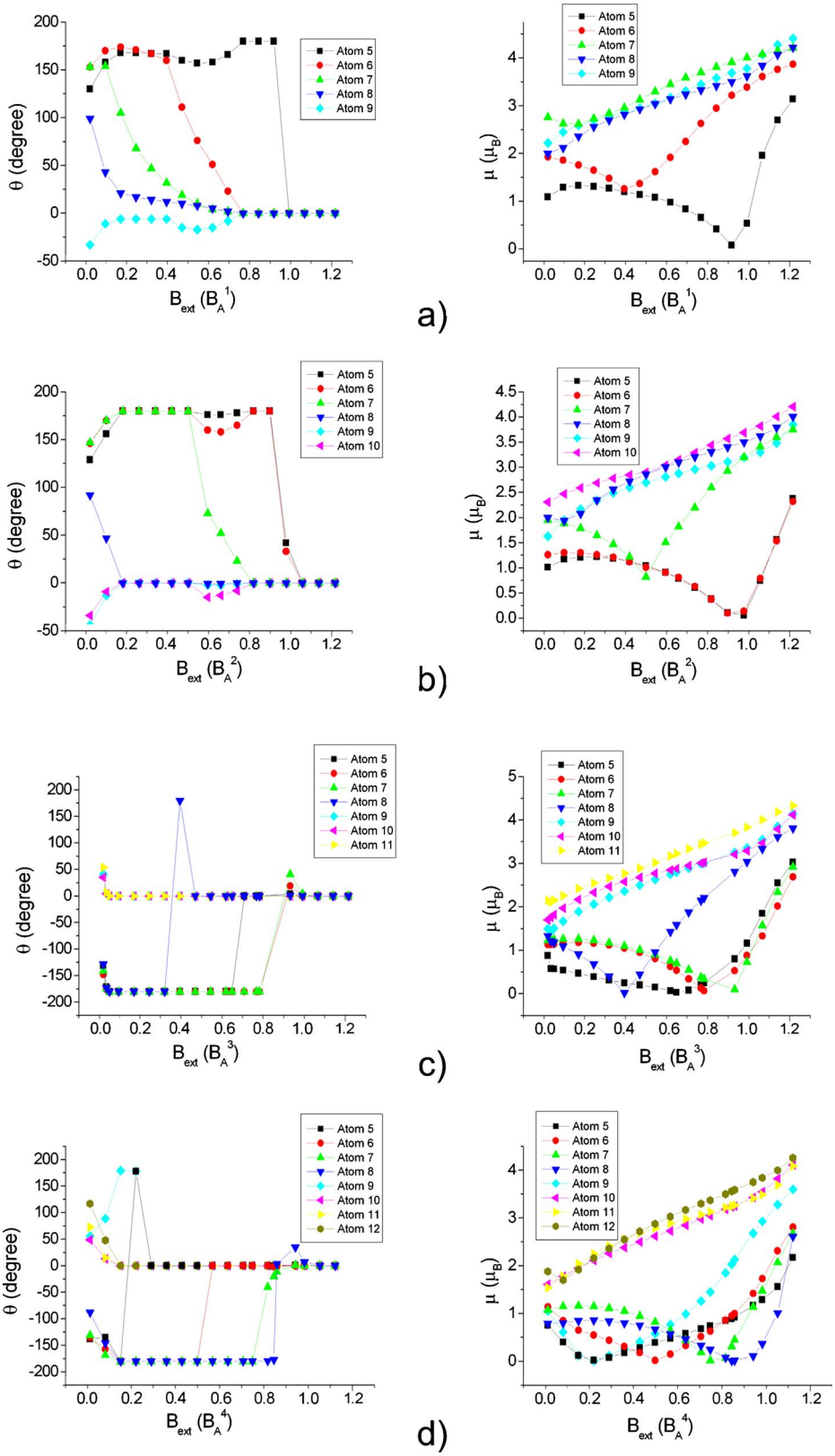

FIG. 4. (Color online) As in Fig. 2 for the atoms marked in Fig. 3 when increasing the coverage from 1 to 2 Cr ML: (a) one Cr row added, (b) two rows, (c) three rows, and (d) four rows (this case corresponding to the Cr bilayer).

atom (see right panel in Fig. 4). These details can be also observed in the supplementary material.[17] The existence of plateaus in the remagnetization process, associated with trapping in collinear magnetic configurations, was also obtained in our previous study of Mn overlayers on Fe(001),[14] and has been experimentally observed in nanostructures with antiferromagnetic couplings like through the Faraday rotation measurements of Kojima *et al.* for the chromium spinel oxide Cr_2O_4, that is a prototype of geometrically frustrated antiferromagnets.[18]

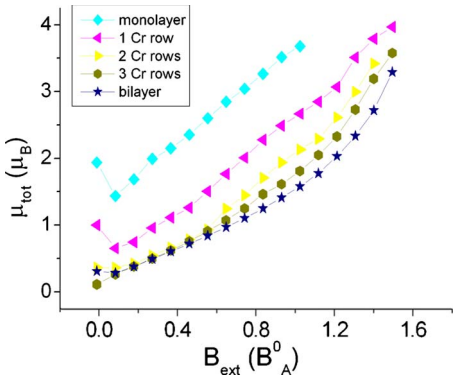

FIG. 5. (Color online) Evolution of the average magnetic moment as a function of the external field for each coverage from 1 to 2 ML.

Finally, in Fig. 5 we show the remagnetization process averaged over the Cr film for the different thicknesses, which is obtained by averaging the field-dependent local magnetic moments of the inequivalent Cr atoms for each of the five Cr coverages investigated. In Fig. 6, we plot the longitudinal and transversal components with respect to the external field. The modulus decreases as increasing the Cr coverage due to the presence of more coordinated Cr atoms at the interface which contribute to the average with much lower magnetic moments than the surface Cr atoms. For the modulus, we obtain an oscillation with a minimum at low fields, which tends to disappear as increasing the Cr coverage, together with a nonlinear behavior for larger fields, with marked changes in the first derivative (the slope). The trapping in collinear configurations is clearly reflected in the transversal component which goes to zero (at those fields, the change in modulus is reflected in the longitudinal component). These changes in slope also tend to disappear as increasing the Cr coverage. These trends can be only understood in terms of the complex behavior at the atomic scale discussed above. The minimum at low fields is due to the strength of the local AF couplings which rotate as a whole at low fields, and changes in the slope at higher fields are due to spin-flip transitions taking place at certain Cr atoms when the system is trapped in collinear magnetic configurations. Larger widths for the terraces are expected to give different dependencies of the remagnetization process (the average quantities) because the central atoms will contribute with more weight to the average behavior. However, provided that we have determined the magnetic response of the atoms with the different possible environments already for the 4-atoms width terraces, we did not find necessary to carry out the study for larger terraces.

IV. SUMMARY

Using a self-consistent noncollinear tight-binding method in which the Hamiltonian has been extended to account for the interaction with the external field, we have calculated the electronic structure of Cr overlayers of different thickness on the stepped Fe(001) substrate as a function of the external field. This is a typical magnetically frustrated system due to the ferromagnetic/antiferromagnetic interface with the monatomic steps. We have considered different Cr coverages, starting from the monolayer and increasing row by row in the [010] direction up to completing the bilayer. The magnetic field is applied along the direction of the magnetic moment of the Fe substrate, which defines an absolute direction in the space.

A nonmonotonic behavior of the magnetic moments is found as a function of the external field and the details of this complex remagnetization process can be understood in terms of the different local geometrical and chemical environments of the Cr atoms. An increase in the angles is found at low fields and correlates with a decrease in the moduli. The moduli start increasing as soon as the moments start to rotate toward the field and depart from the local AF alignment, that is, when the field is large enough to overcome the antiferromangetic exchange couplings which are very strong, as expected. Larger external fields are required to overcome the antiferromagnetic Cr-Cr couplings than the Cr-Fe ones which give support to the stronger character of the Cr-Cr couplings.

For Cr coverages larger than the monolayer, and particularly for those closer to the bilayer limit, the system is trapped in collinear magnetic configurations for certain ranges of the external field, as it has been observed in other geometrically frustrated antiferromagnets.[18] When the systems are trapped in those collinear magnetic configurations, only the modulus of the local magnetic moments changes with the external field until spin-flip transitions take place. We note that these steps in the angular distribution when spin

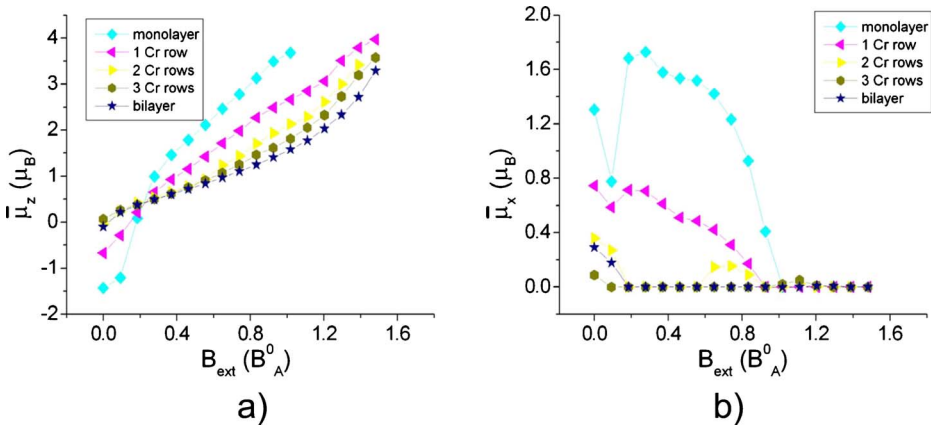

FIG. 6. (Color online) Evolution of the (a) longitudinal and (b) transversal components of the average magnetic moment as a function of the external field for each coverage from 1 to 2 ML.

flips take place, correlate with abrupt changes in the modulus of the moment of the corresponding atom.

For the modulus of the average magnetic moment within the whole Cr overlayer, we obtain an oscillation with a minimum at low fields, which tends to disappear as increasing the Cr coverage, together with a nonlinear behavior for larger fields, with marked changes in the slope. The trapping in collinear configurations is clearly reflected in the transversal component of the average moment which goes to zero (at those fields, the change in modulus is reflected in the longitudinal component). These trends can be only understood in terms of the complex behavior at the atomic scale discussed above. The minimum at low fields is due to the strength of the local AF couplings which rotates as a whole at low fields, and changes in the slope at higher fields are due to spin-flip transitions taking place at certain Cr atoms when the system is trapped in collinear magnetic configurations.

This is the first step in the understanding of the qualitative behavior of magnetically frustrated systems in the presence of external magnetic fields. Temperature effects and structural relaxation should also be considered for a quantitative description. We hope that the present study will stimulate further experimental work to confirm our predictions.

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