

Noncollinear magnetism caused by intermixing at ferromagnetic/antiferromagnetic interfacesE. Martínez,¹ R. Robles,² D. Stoeffler,³ and A. Vega¹¹*Departamento de Física Teórica, Atómica y Óptica, Universidad de Valladolid, E-47011 Valladolid, Spain*²*Department of Physics, Uppsala University, SE-75121 Uppsala, Sweden*³*Institut de Physique et de Chimie des Matériaux de Strasbourg (UMR C7504 CNRS-ULP), F-67034 Strasbourg, France*

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Fe-Cr alloy layers on Fe(001) substrates with different atomic environments are theoretically investigated through a self-consistent real-space tight-binding method parametrized to density functional theory and without collinear restriction for the spin directions. We show that the intermixing at the Cr/Fe interface, experimentally observed at the first stages of the Cr growth on Fe, originates noncollinear magnetic arrangements within the system and even induces magnetic walls in the Fe layer with a magnetic domain close to the interface. A different behavior is shown if steps are present at the interface; i.e., noncollinear magnetism becomes more localized and the induced magnetic wall in Fe vanishes.

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INTRODUCTION

The existence of noncollinear (NC) magnetic configurations is a well-known fact, and there is a large variety of systems where this magnetic arrangement can be observed. One example is the domain walls in which the magnetic moment direction changes from one orientation to the opposite one.¹ Another example is nanostructures of transition-metal (TM) elements of the middle of the series, like freestanding clusters of Cr, Mn, and Fe.^{2,3} NC magnetism can in general appear when there is magnetic frustration in a particular region of the system.^{4,5} Most of those systems belong to the so-called magnetic nanostructures, either due to their intrinsic size, like freestanding clusters, or to the size of the region in which magnetic NC effects can develop, like in constrained domain walls or in systems where magnetic frustration is associated with a structural defect.

In the last years, there has been a growing interest in NC magnetic nanostructures, which has been increased by recent experimental and theoretical advances. From the experimental side, the development of spin-polarized scanning tunneling microscopy and spectroscopy^{6,7} (SP-STM/STS) has been a great advance in magnetic characterization techniques at the atomic scale. The SP-STM/STS allows one to identify NC magnetic configurations in supported films and surfaces, something that has been a challenge for many years. On the theoretical side, quantum-mechanical spin-polarized calculations in low-dimensional systems have generally been confined to a global quantization axis. However, in recent years large efforts have been directed toward the implementation of codes able to deal with NC magnetism in both density functional theory (DFT) and semiempirical tight-binding (TB) methods. Within the DFT formalism, the magnetic NC degree of freedom has been implemented in all-electron^{8–10} and pseudopotential³ approaches. These methods provide a detailed description of the phenomenon, being able to deal even with intra-atomic NC magnetism.^{1,11} However, a high computational effort is required and therefore these methods have been applied to systems with very few nonequivalent sites, like very small freestanding TM clusters^{3,10} or ideal monolayers.¹ Calculation of extended systems with broken

symmetries is difficult not only due to the large number of inequivalent sites but also because, even having a certain periodicity from the structural point of view, consideration of NC magnetic configurations can impose much larger periods (magnetic periodicity) and, consequently, the unit cell to be considered can be even larger.

The semiempirical self-consistent real-space TB method has been widely used mostly in the collinear (C) framework with successful results for the study of complex TM nanostructures like clusters, surfaces, and interfaces. We have recently implemented the possibility of having a different spin-quantization axis for each site in the system, thus allowing one to study NC magnetic configurations without symmetry restrictions and providing at the same time a good degree of accuracy. The Hamiltonian is parametrized to DFT results, and the recursion method is used for self-consistently determining the electronic structure.^{4,12}

The Fe/Cr interface is a relevant system for its properties of technological interest, like magnetoresistance. In a recent work, we have investigated the NC magnetism of Cr films supported on a stepped Fe(001) substrate.⁴ This is a prototype of one of the magnetic NC kinds of systems mentioned above—i.e., a magnetically frustrated system. It is an interesting system from the fundamental point of view because in the same region competing tendencies to ferromagneticlike and antiferromagneticlike couplings can be found. This competition is the origin of the magnetic frustration and of the related NC effects which were observed in the most stable situation.⁴ In this system frustration was associated with the steps at the interface, so it is an example of an intrinsically NC situation caused by a structural defect. In that work we considered perfect interfaces between Fe and Cr. However, interdiffusion has been experimentally observed in the first stages of Cr growth on Fe at room temperature.¹³ The interdiffusion is a mechanism, different from the presence of structural defects like steps, that might also originate magnetic frustrations and related NC effects. To our knowledge, this possibility has not been investigated so far.

The aim of the present paper is to demonstrate that the interdiffusion at the Cr/Fe interface, which is experimentally observed at the first stages of the Cr growth, can originate NC magnetic arrangements within the system and even mag-

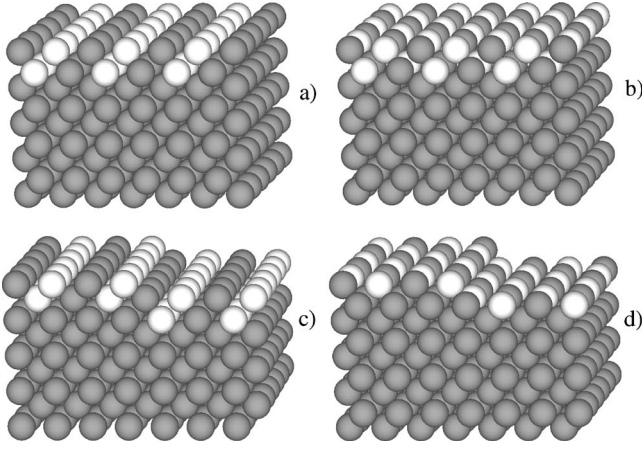


FIG. 1. Illustration of the investigated Fe-Cr mixed layers deposited on ideal (a),(b) and stepped (c),(d) Fe(001) substrates. Dark (light) spheres represent Fe (Cr) atoms.

netic domains in the Fe layers close to the interface. For this purpose, we have considered 50% Fe-Cr mixed monolayers and bilayers deposited on the Fe (001) substrate. We have analyzed two configurations: one in which the Fe and Cr atoms are arranged in adjacent lines [(2×1) cell] and another one in which they form a $c(2\times 2)$ arrangement. In both cases, no structural defects are present. Furthermore, we have also investigated a system in which both Fe-Cr mixing and steps at the interface coexist. In Fig. 1, three-dimensional views of all these systems are given.

THEORETICAL METHOD AND COMPUTATIONAL DETAILS

The magnetic moment distribution within the system is determined by self-consistently solving a TB Hamiltonian for the s , p , and d valence electrons in a mean-field approximation. The Hamiltonian can be split into a band term and an exchange term, which in the orthogonal $|i\alpha\rangle$ basis of atomic site i and orbital α and with the usual notation are

$$H = H_{band} + H_{exch}, \quad (1)$$

$$H_{band} = \sum_{\substack{i,j \\ \alpha,\beta}} [(\epsilon_{i\alpha}^0 + U_{i\alpha,j\beta}\langle\hat{n}_{j\beta}\rangle + Z_i\Omega_{i\alpha})\delta_{ij}\delta_{\alpha\beta} + t_{ij}^{\alpha\beta}(1 - \delta_{ij})] \times |i\alpha\rangle\langle j\beta| \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}, \quad (2)$$

$$H_{exch} = \sum_{i,\alpha} \left(-\frac{1}{2}J_{i\alpha}\mu_{i\alpha} \right) |i\alpha\rangle\langle i\alpha| \begin{bmatrix} \cos\theta_i & e^{-i\phi_i}\sin\theta_i \\ e^{i\phi_i}\sin\theta_i & -\cos\theta_i \end{bmatrix}. \quad (3)$$

H_{band} contains both the nondiagonal matrix elements (hopping integrals $t_{ij}^{\alpha\beta}$ between orbitals α and β of different sites i and j , which are assumed to be spin independent) and the spin-independent part of the diagonal matrix elements ($\epsilon_{i\alpha}^0 + U_{i\alpha,j\beta}\langle\hat{n}_{j\beta}\rangle + Z_i\Omega_{i\alpha}$), being the sum of (i) the local level $\epsilon_{i\alpha}^0$,

of (ii) the electrostatic level shift $U_{i\alpha,j\beta}\langle\hat{n}_{j\beta}\rangle$ accounting for the charge variations parametrized by the Coulomb integral $U_{i\alpha,j\beta}$, and of (iii) the crystal field potential $Z_i\Omega_{i\alpha}$, where Z_i is the local atomic coordination of site (i). H_{exch} describes the magnetic part through the exchange parameter $J_{i\alpha}$ multiplied by the magnitude of the local magnetic moment, $\mu_{i\alpha}$, whose direction is given by the angles (θ_i, ϕ_i) in the spin-rotation matrix. The local densities of states (LDOS) are calculated using the recursion method¹⁵ in real space, which provides the partial LDOS projected on an arbitrary local quantization axis through a rotation of the initial recursion vector in the spin space. We calculate the local electronic occupations and magnetic moments at each site by integrating the LDOS up to the Fermi level, with the global charge neutrality condition. More details of this procedure can be found in Ref. 4 and references cited therein.

All the parameters of the TB model have been taken as in Ref. 4. Using this parametrization, the effects of reduced coordination due to the surface and the Cr-Fe hybridization are taken into account in the hopping and exchange integrals of Cr and Fe by extracting them from a fit to *ab initio* TB-LMTO (Ref. 14) results for an ideal Cr monolayer deposited on Fe(001). No relaxation has been included in the geometries of the studied systems. Although the local relaxations may be important at surfaces, in the case of Fe and Cr this is less likely since both elements have a similar structure in the bulk (bcc for Fe a very close to bcc for Cr) with a very similar lattice constant. On the other hand, the aim of this paper is to explore the general behavior of a Fe-Cr system when some intermixing is present, with focus on a possible NC magnetic arrangement. In order to accurately describe the experimental situation, not only the relaxation should be included, but also the possible nonuniform mixing, the presence of local defects, etc. All these situations are beyond the scope of this work.

RESULTS AND DISCUSSION

Before presenting the results for the systems given in Fig. 1, it is worth noticing that in case of 50% Fe-Cr single monolayers deposited on the Fe (001) substrate, no magnetic frustrations exist since it is possible to have a “natural” magnetic coupling between moments of nearest-neighbor atoms in the whole system—that is, all Cr-Cr and Fe-Cr nearest neighbors with antiparallel moments and all Fe-Fe ones with parallel ones. In those cases, only the C magnetic solution exists. Let us analyze the situation for magnetically frustrated systems.

In Fig. 2, we show the magnetic map corresponding to the most stable C arrangement of the 50% Fe-Cr mixed bilayer with Fe and Cr atoms arranged in adjacent lines [(2×1) cell]. For simplicity, we do not report the values of all local magnetic moments in the system, although the arrows representing local moments are proportional to them. This system presents magnetic frustrations of some Cr-Cr first-nearest-neighbor pairs of atoms due to the strong antiparallel magnetic coupling between the moments of Fe and Cr first nearest neighbors. In the C framework, the only way for the system to overcome this situation is to reduce the modulus of some local magnetic moments in the frustrated region. This

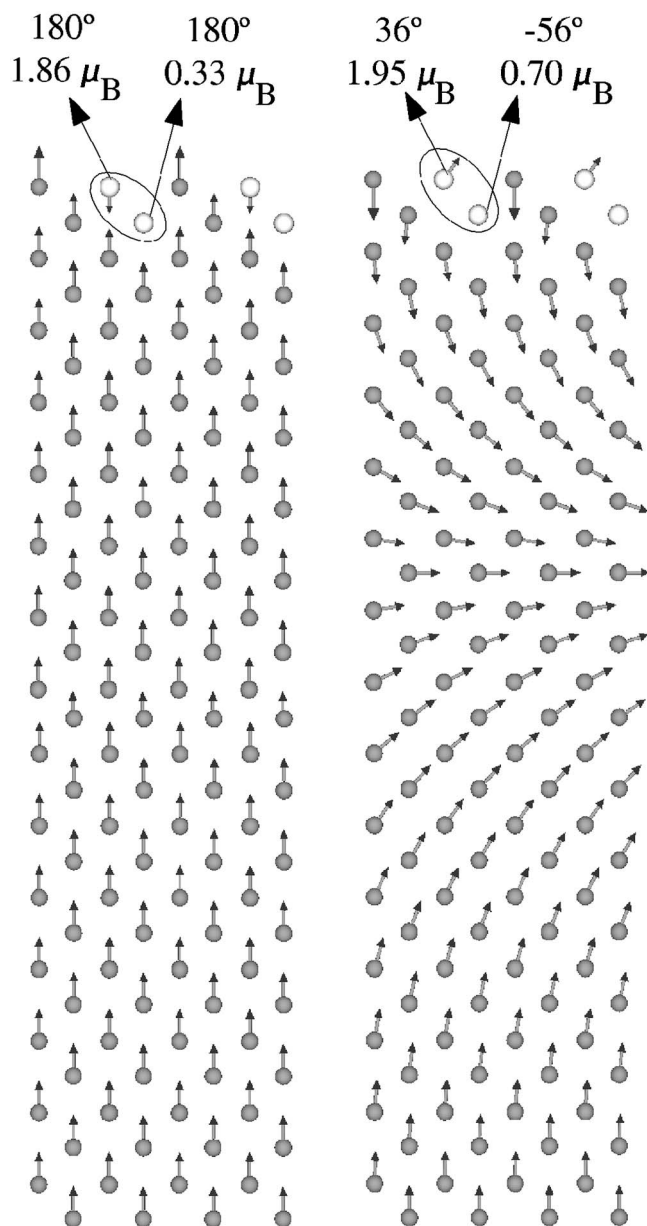


FIG. 2. Left: collinear solution and ground-state noncollinear solution for the 50% Fe-Cr mixed bilayer of Fig. 1(a). Only a portion of the system is illustrated as a projection on the (010) plane. The magnetically frustrated sites are marked with ellipses and their magnetic moment (absolute value and angle respect to the Fe bulk) are given.

effect is remarkable on one of the Cr sites displaying a moment of only $0.33\mu_B$, half the value of the local moment in bulk Cr, whereas its local moment should be enhanced due to the loss of coordination caused by the proximity of the surface and to the hybridization with the surrounding Fe atoms. When the C restriction is removed, the system has the additional degree of freedom of rotating the magnetic moments and a NC solution (Fig. 2) appears. Now the frustration effect is locally reduced and distributed over many more atoms. Actually, the extension of the frustration effect is surprising, reaching well below the surface (more than 20 layers). The Fe atoms close to the surface are antiferromag-

netically coupled to deeper Fe layers (Fe bulk); thus a domain wall is formed in Fe until the bulk orientation is recovered. It is worth pointing out that couplings between Fe-Fe neighbors are close to 0° , so it can be said that the NC solution resembles the best compromise to the natural coupling in the whole system. In the frustrated region, the local moments increase in the NC solution with respect to the collinear one: in particular, the strongly frustrated Cr site in the C solution doubles now its magnetic moment (see Fig. 2). The local magnetic moments being larger in the NC arrangement, the magnetic energy is also larger and, consequently, the NC solution becomes more stable than the C one. Here, the NC solution results in a net perpendicular component of the magnetization over the first surface layers with respect to the deeper Fe layers, something that could be confirmed experimentally. The NC solution has been carefully checked. First, the number of layers included in the calculation has been increased until the solution was stabilized. Second, several starting points have been tried, getting the same solution in all cases. In particular, it is important to remark that a small perturbation of the collinear solution leads to the NC one, revealing its instability.

The same discussion is valid for the 50% Fe-Cr mixed bilayer with Fe and Cr atoms arranged in a $c(2 \times 2)$ configuration (see Fig. 3). In this case a double domain wall is formed and the perturbation extends even further below. Also here a perpendicular component of the magnetization of the first layers with respect to the Fe bulk develops in the NC arrangement. This constitutes further evidence of the collective character of magnetism.

The previous results confirm that NC solutions can be originated by Fe-Cr intermixing at the first stages of Cr growth on Fe due to the magnetic frustrations that arise. However, as illustrated by the two considered cases, these solutions are highly sensitive to the nature of the Fe-Cr mixing. In order to check this sensitivity, additional calculations have been performed. For the same system geometry—i.e., without steps—we have considered a different mixing (75% of Cr in the surface layer and 25% in the subsurface one). In this case the frustration is smaller than in the previous ones. The most stable solution that we have obtained is a C one, in which the Cr atoms in the surface layer couple antiferromagnetically to Fe, whereas Cr atoms in the second layer are ferromagnetically coupled to Fe. The latter atoms are the only source of magnetic frustration. However, the magnetic moment in these atoms is noticeable larger than in the previous cases ($0.52\mu_B$ compared to the previous $0.33\mu_B$) and the frustration is not enough to drive a NC arrangement. Actually, although several initial NC configurations have been tested, we have not been able to converge any NC solution. Consequently, if during the early stage of the growth regions with different types of mixing coexist together with regions without mixing, the competition would cancel the NC driving force and the deep magnetic wall would vanish, making its observation particularly improbable. Such situations are extremely difficult to investigate directly even with our method.

Let us now investigate a system where both intermixing and monoatomic steps are considered. The first is the origin of the NC driving force of the deep magnetic wall, while the

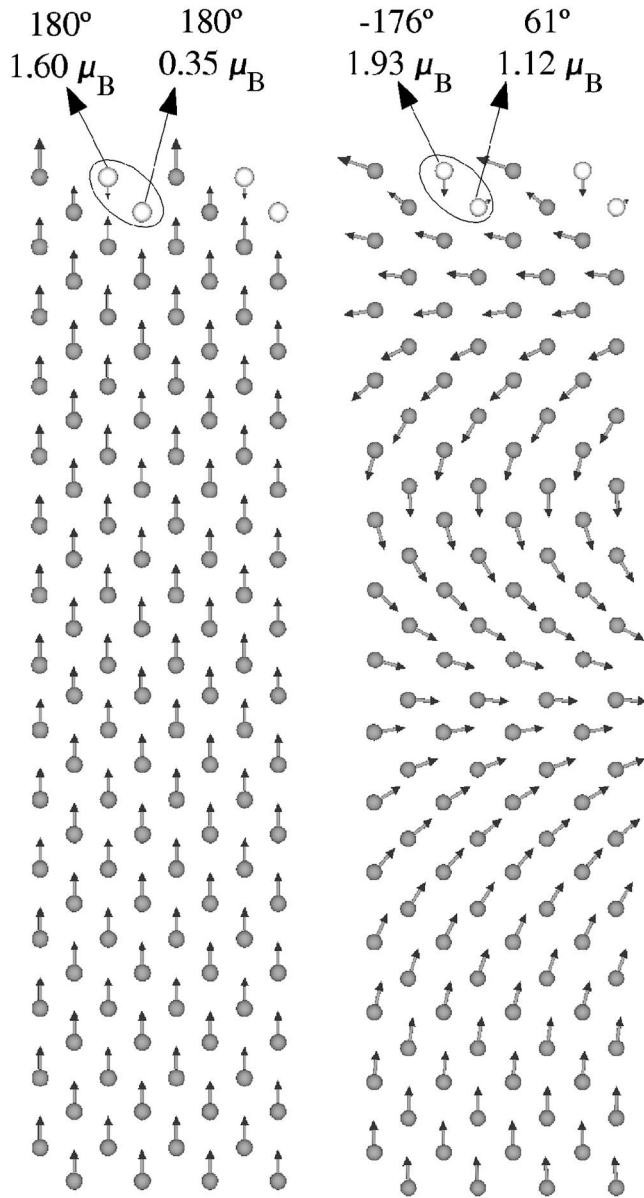


FIG. 3. As in Fig. 2 for the 50% Fe-Cr mixed bilayer of Fig. 1(b).

second introduces (i) a small phase shift between the walls coming from each terrace and (ii) additional magnetic frustration. In our previous work,⁴ we demonstrated that such structural defects at Fe-Cr interfaces produce magnetic frustration and NC magnetic arrangements. Therefore, two sources of frustration will coexist now and a more complex NC picture is expected.

In Fig. 4 we plot the most stable C and NC solutions of the 50% Fe-Cr mixed bilayer with Fe and Cr atoms arranged in adjacent lines supported on a stepped Fe (001) substrate. The magnetic frustration is also located in the Cr sites. Although the mechanism to release the frustration is essentially the same as before, now it develops in a different way. The magnetic domains are associated with the steps, with components of the magnetization at the surface layers perpendicular to the bulk magnetization, but with opposite orientation from one step to the next (Fig. 4). This symmetry

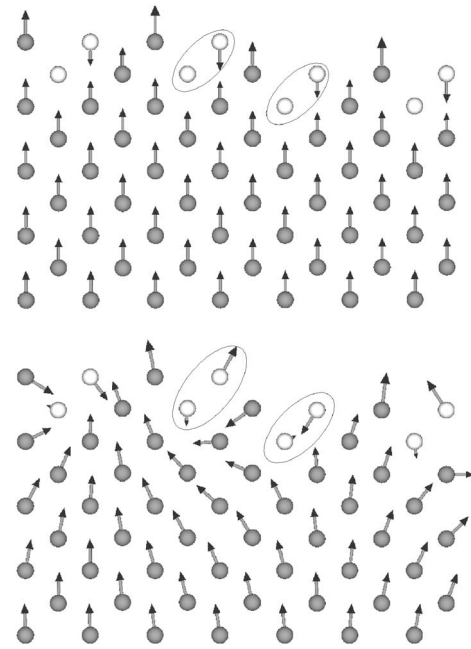


FIG. 4. As in Fig. 2 for the 50% Fe-Cr mixed bilayer supported on a stepped Fe(001) substrate [Fig. 1(c)].

vanishes the perpendicular component for the whole system, which constitutes an important difference compared to the flat surfaces. The other main difference is that the effect extends fewer than ten layers below the surface and the unexpected deep magnetic wall found for the ideal surfaces disappears.

Finally, if the mixed Fe-Cr layer has a $c(2 \times 2)$ arrangement with steps at the interface, frustration effects appear already in the first supported monolayer and the azimuthal angle of the local moment is not the same for all sites, in contrast with the previous cases. In order to illustrate the magnetic moments distribution now, we plot in Fig. 5 the C and ground-state NC solutions, respectively, in three views, which allows us to visualize both the local chemical environment around the structural defect and the azimuthal angle of the local moment. The magnetic frustration is located in the Fe-Cr nearest-neighbor pairs at the steps, whose local moments are coupled parallel (see top view of Fig. 5). This frustration is reduced in the NC solution. The Fe-Cr interaction is stronger than the Cr-Cr one since the first is closer to its natural coupling (180°) than the second. A magnetic domain structure also exists at the first Fe layers below the interface, but in this case the perpendicular components of the magnetization are small. The overall NC magnetic arrangement is more complex than in the other systems investigated here, since the azimuthal angle of the local moments is now playing a role. As in the previous case, no deep magnetic wall is present and the NC effect only extends a few layers below the surface.

Therefore, we can conclude that the simultaneous presence of intermixing and monoatomic steps suppresses the deep magnetic wall and strongly limits the extension of the NC region. Thus, in a realistic system, a different behavior can be expected close to a step or in a flat surface, and this different behavior could be experimentally observed. In gen-

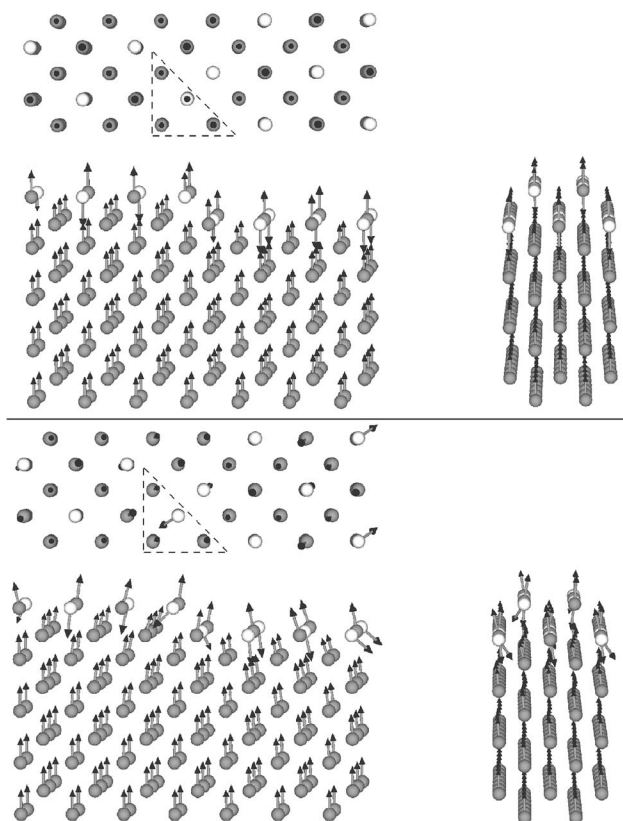


FIG. 5. Three views of the collinear and noncollinear solutions for the 50% Fe-Cr mixed monolayer [Fig. 1(d)]. The dashed line marks the frustrated Fe-Cr nearest-neighbor pairs.

eral, the results for the stepped surfaces confirm the sensitivity of the NC solutions to the details of the system.

SUMMARY

In summary, using a self-consistent real-space TB method parametrized to DFT calculations and without collinear restriction for the spin direction, we have studied the magnetic properties of 50% Fe-Cr mixed layers deposited on an Fe (001) substrate. Our results demonstrate that noncollinear magnetic effects can be originated by Fe-Cr intermixing at the first stages of Cr growth on Fe. The noncollinear effects are understood as a consequence of the magnetic frustrations, which are reduced when the local moments are not restricted to the same global spin-quantization axis. We have also considered the case in which both intermixing and monoatomic steps at the interface coexist in the same sample. We have seen that there is a different behavior of the component of the net magnetization perpendicular to the magnetic moments of the deeper Fe layers. While on the stepped surfaces this component is zero, on a surface without steps it has a nonvanishing value. These predictions could be experimentally confirmed, and we hope they will stimulate further experimental works.

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- ¹K. Nakamura, Y. Takeda, T. Akiyama, T. Ito, and A. J. Freeman, *Phys. Rev. Lett.* **93**, 057202 (2004).
- ²N. Fujima, *Eur. Phys. J. D* **16**, 185 (2001).
- ³T. Oda, A. Pasquarello, and R. Car, *Phys. Rev. Lett.* **80**, 3622 (1998).
- ⁴R. Robles, E. Martinez, D. Stoeffler, and A. Vega, *Phys. Rev. B* **68**, 094413 (2003).
- ⁵U. Schlickum, N. Janke-Gilman, W. Wulfhekel, and J. Kirschner, *Phys. Rev. Lett.* **92**, 107203 (2004).
- ⁶S. Heinze *et al.*, *Science* **288**, 1805 (2000).
- ⁷O. Pietzsch, A. Kubetzka, M. Bode, and R. Wiesendanger, *Science* **292**, 2053 (2001).
- ⁸L. M. Sandratskii and P. G. Guletskii, *J. Phys. F: Met. Phys.* **16**, L43 (1986).

- ⁹J. Kübler, K. H. Höck, and J. Sticht, *J. Appl. Phys.* **63**, 3482 (1988).
- ¹⁰D. Hobbs, G. Kresse, and J. Hafner, *Phys. Rev. B* **62**, 11556 (2000).
- ¹¹L. Nordström and D. J. Singh, *Phys. Rev. Lett.* **76**, 4420 (1996).
- ¹²C. C. Cornea and D. C. A. Stoeffler, *J. Magn. Magn. Mater.* **198–199**, 282 (1999).
- ¹³A. Davies, J. A. Strosio, D. T. Pierce, and R. J. Celotta, *Phys. Rev. Lett.* **76**, 4175 (1996).
- ¹⁴O. K. Andersen and O. Jepsen, *Phys. Rev. Lett.* **53**, 2571 (1984).
- ¹⁵R. Haydock, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1980), Vol. 35, p. 215.