

Topological antiferromagnetism at Cr surfaces and interfaces

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(Received 20 January 1994)

The Hubbard tight-binding Hamiltonian is used to derive the local magnetic moments of vicinal Cr surfaces and interfaces. For Cr vicinal surfaces, the magnetic arrangement is a double-cell configuration which consists of monoatomic steps antiferromagnetically coupled to nearest-neighbor monoatomic steps. These results allow us to explain the apparently contradictory observations obtained by spin-resolved and angle-and-energy-resolved photoemission. For the Fe monolayer on vicinal Cr surfaces, a magnetic configuration with a periodicity of two steps is also obtained. As a consequence, the sign of the magnetization at the Fe overlayer oscillates from step to step. This result is in agreement with recent experiments using scanning-electron-microscopy with polarization analysis and magneto-optic Kerr effect.

The possibility of tailoring new materials with properties of technological interest like large magnetization and magnetic ordering as well as giant magnetoresistance has opened new prospects in surface physics.^{1,2} Several technical experiments are responsible for the intense activity in this field: (i) The new sample-preparation techniques which now permit the manufacture of devices with extraordinary accurate specifications. Molecular-beam epitaxy, sputtering, metal-organic chemical vapor deposition, and lithography are now available. (ii) The availability of sophisticated sample characterization techniques based on x-rays and ultraviolet photons from synchrotron sources, neutrons, and electron microscopes. However in some cases, particularly for Cr surfaces, controversial experimental results do not lead to a unified picture of surface magnetization. Besides this huge amount of experimental results, progress has been made in the calculation of a large variety of problems due to the increasing availability of fast, operationally inexpensive and numerically intensive computers.³ Calculations based on the full-potential augmented-plane-wave (FLAPW) method are expected to provide the best results but are usually restricted to systems where some periodicity remains present, as in the case of films or sandwiches. For more complex systems, like those concerned with the interlayer coupling in magnetic superlattices, methods like the linear-muffin-tin orbitals (LMTO)⁴ or the augmented spherical wave (ASW)⁵ have to be used. Those methods also can be used only in the case where two-dimensional (2D) periodicity remains. This is, however, not the case in real systems, which are usually very far from 2D periodicity at the surface or interface. High-Miller-index surfaces can sometimes be a reasonable approximation of the real world. In this case the 2D periodicity remains but with a much higher number of inequivalent sites as compared to low-index surfaces. For example, Unguris,

Celotta, and Pierce⁶ and Purcell *et al.*⁶ have displayed by using a wedge structure the existence of short-period antiferromagnetic exchange coupling between Fe layers separated by Cr layers. This was done by growing an Fe/Cr/Fe sandwich in which the Cr layer thickness varies linearly with a small angle. Therefore, with this wedge structure, the complete behavior as a function of Cr thickness could be investigated on a single sample. These Fe/Cr/Fe wedge systems cannot, however, be described by any of the *ab initio* methods mentioned above. Therefore, in order to circumvent the inherent difficulties concerning the study of these vicinal surfaces, we perform a real-space calculation by using a Hubbard tight-binding Hamiltonian.

In this paper, we compute the magnetic-moment distribution of Cr stepped surfaces. Since Allan's proposal⁷ of an enhanced layered antiferromagnetic structure (LAS), i.e., a sequence of (001) layers of alternating magnetization with the surface layer having a magnetic moment 3–4 times higher than in the bulk, considerable attention has been devoted to the determination of the magnetization at the surface of chromium. Meier, Pescia, and Schrieber⁸ through spin-resolved photoemission experiments (SRPES) and Carbone and Alvarado⁹ through spin-polarized low-energy electron diffraction have not detected any polarization at the surface of Cr(001). However, through angle- and energy-resolved photoemission experiments (ARPES), Klebanoff and co-workers¹⁰ have seen spin-split surface states. Clearly, the simple model of a ferromagnetic perfectly flat Cr(001) surface as assumed in the calculations^{1,11,12} cannot explain the results of both SRPES and ARPES experiments. It can explain ARPES but not SRPES, because this latest experiment which measured the net magnetization within the first 2–3 layers should exhibit a net magnetization in the LAS configuration.

In theoretical models used till now, perfectly flat surfaces are assumed. In practice, however, terraces separated mostly by single steps cannot be avoided.¹³ This geometry can explain the absence of macroscopic magnetic moment at the surface of the Cr(001) compound (Fig. 1). In order to study this stepped surface, the simplest description consists of a double-cell configuration, periodically repeated, the average magnetic moment being zero. This type of configuration, called "topological antiferromagnetism" by Blügel, Pescia, and Dederichs,¹⁴ has been confirmed by means of scanning tunneling microscopy (STM) by Wiesendanger *et al.*¹⁵ However, Blügel, Pescia, and Dederichs¹⁴ wrote in their paper that the determination of the magnetization map of such steps "is out of the range of current supercomputers." To our knowledge, such first-principle calculations on those steps have not appeared up to now: this is probably due to the fact that these *ab initio* methods require the use of a supercell and thus the number of inequivalent sites which can be included in the calculation is limited. Therefore, to study stepped surfaces, an alternative approach must be found. In this work the spin-polarized densities of states are computed by using a tight-binding Hubbard Hamiltonian in the unrestricted Hartree-Fock approximation. For the last ten years, this approach has been used with success to describe surface, cluster, and thin-film magnetism.^{11,16,17} The calculations are realized in real space by the recursion method.

In this work we study the vicinal surfaces of Cr with index $(102n-1)$, with $n=1-4$. These surfaces can be seen as Cr(001) stepped surfaces, with a step height of one atomic layer and with n rows parallel to the direction [010]. Such a calculation has already been performed for Fe $(102n-1)$ stepped surfaces.¹⁹ As displayed in Fig. 1, the study of this topological antiferromagnetism needs to consider a double-cell configuration where the polarization in two nearest-neighboring steps is opposite in sign. Let us note that in the case of ferromagnetic steps¹⁸ a single cell is considered. Also, in the case of a V overlayer on Fe $(102n-1)$, the single-cell configuration is imposed by the ferromagnetic substrate. More surprisingly, in the case of a vanadium stepped surface single-cell and double-cell configurations have given different magnetic-moment distributions.²⁰ For Cr $(102n-1)$ vicinal surfaces only the double cell configuration exists because in the case of semi-infinite Cr systems the polarization

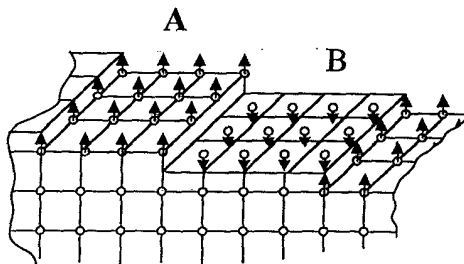


FIG. 1. Schematic view of the Cr $(102n-1)$ vicinal surface for $n=4$. The nearest-neighboring steps A and B present magnetic moments with opposite polarization.

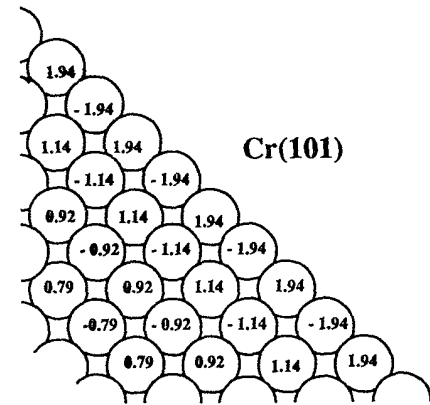


FIG. 2. Local magnetic moment μ (in units of μ_B) per atom for the semi-infinite (101) surface of Cr in the in-plane antiferromagnetic configuration.

should converge to the LAS after a small number of layers.

In this calculation we restrict ourselves to d orbitals and 12 levels of the continued fraction.¹⁹ The hopping integrals are of the canonical type and their values are adjusted in order to recover the bandwidth of Varma and Wilson.²¹ Local neutrality has been used in order to determine the spin polarization of a given atom, whereas the exchange integral J has been fixed in order to recover the bulk magnetic moment of Cr. In Figs. 2-6 we report the result obtained for $n=1-4$ and $n=\infty$ [001] crystallographic surfaces. To perform the numerical calculation we restrict ourselves to four layers of inequivalent atoms. In order to test this approximation, we have increased, in the Cr(103) case, the number of inequivalent layers up to six: practically no change appears in the surface layer (less than 1%). The total number of inequivalent atoms is $4n$ for a given cell A . Atoms of the nearest-neighboring cells B have just the opposite polarization (antiferromagnetic coupling). As an example, for the vicinal surface (107), computations have been performed on 32 inequivalent atoms. By changing the input values of the magnetic moments we have checked carefully that for each vicinal surface a unique magnetic-moment distribution is obtained.

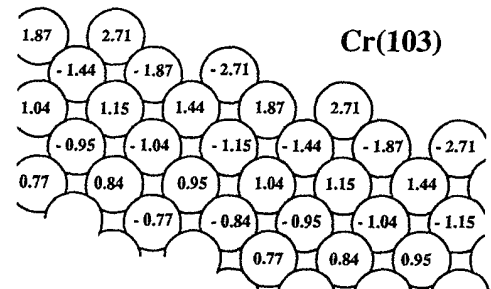


FIG. 3. Local magnetic moment μ (in units of μ_B) per atom for the vicinal (103) surface of Cr in the double-cell configuration.

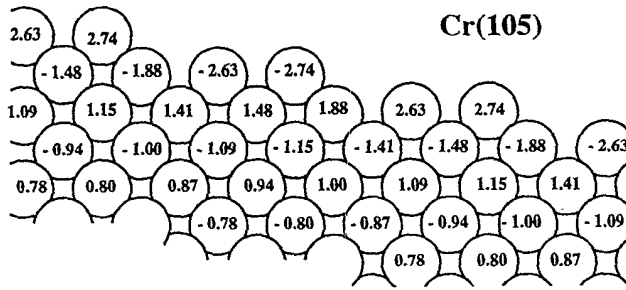


FIG. 4. Local magnetic moment μ (in units of μ_B) per atom for the vicinal (105) surface of Cr in the double-cell configuration.

The results can be discussed in relation to the simple coordination rule which states that the value of the magnetic moment of a given atom is directly related to the number of its neighboring atoms: the higher the number of neighboring atoms, the lower the magnetic moment. As already discussed by Victora and Falicov,¹¹ the magnetic moment for the crystallographic face (101) is lower as compared to the (001) one. The atom at the step edge on the vicinal surface bears the highest magnetic momentum whereas the kink atom bears the lowest one. These two results are in qualitative agreement with the coordination rule. The other atoms in the surface plane have magnetic moments whose values are nearly equal to those of the (001) surface because their coordination numbers are very similar to the (001) case.

The double-cell configuration has been already obtained in the case of vanadium surfaces²⁰ and seems to be present in the case of a monolayer of Fe on Cr(102n-1) vicinal surfaces (Fig. 7). We have first solved the case of Fe/Cr(001). In this simple case there is a clear tendency of the Fe atoms to align their magnetic moments antiparallel relative to their Cr neighbors. This is in fact not new and has been already obtained both theoretically¹¹ within the tight-binding formalism and experimentally²² in *in situ* magnetometer measurements during growth of ultrathin Cr films on Fe(001). On the other hand, Dorantes-Davila *et al.*²³ have determined, within the tight-binding formalism, the magnetic polarization of 1-5 layers of Fe on Cr(001). In all cases the Cr moments are aligned antiparallel to the Fe moments at the interface. In order to be sure that the magnetic-moment distribution obtained is unique we have used different sets of input values. In all cases we have obtained changes of

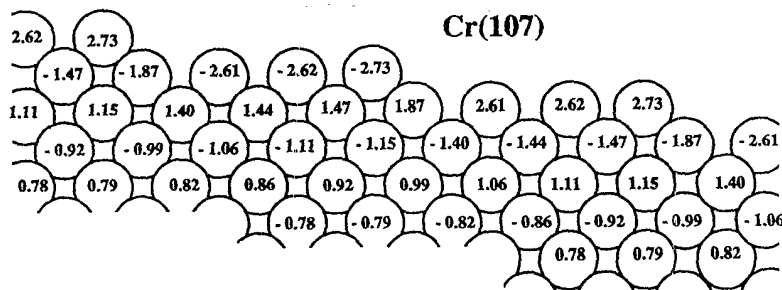


FIG. 5. Local magnetic moment μ (in units of μ_B) per atom for the vicinal (107) surface Cr in the double cell configuration.

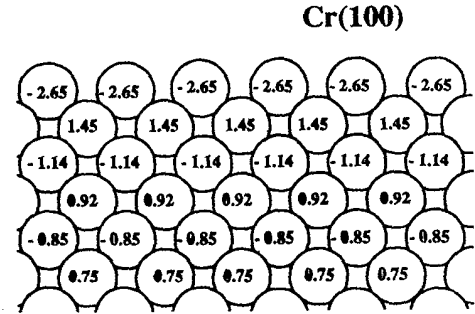


FIG. 6. Local magnetic moment μ (in units of μ_B) per atom for the semi-infinite (100) surface of Cr in the layered antiferromagnetic configuration (each plane is ferromagnetic whereas is coupled antiferromagnetically to its nearest-neighbor step).

sign of some local magnetic moments leading to the result reported in Fig. 7.

The present result is an important step for the explanation of the two-period oscillation in the exchange coupling of Fe/Cr/Fe(001) with a Cr wedge structure Unguris, Cellota, and Pierce⁶ and Purcell *et al.*⁶ have performed respectively scanning electron microscopy with polarization analysis and magneto-optic Kerr effect experiments to show that, for an extremely well-ordered Cr interlayer, oscillations with a period of two Cr layers were observed. In the calculation, we have replaced the wedge structure by a vicinal (107) surface. The results obtained are in agreement with those reported in Ref. 6 where the polarization of Fe changes sign from step to step. In this particular geometry, the oscillatory polarization of Fe arises simply from the antiferromagnetic coupling of the Cr substrate. In the experiments, the thickness of the Fe overlayer is about 2 nm and the length of the step (or order of μm) is much larger than the size of the steps studied here. However, previous studies²³ indicate that the Fe remains ferromagnetically ordered with the bcc configuration when the Fe thickness increases. Thus the experimental system should present magnetic domains with alternate magnetization, due to the interface with Cr.

The results obtained with a tight-binding Hubbard Hamiltonian in the unrestricted Hartree-Fock approximation in the case of high-index Cr surfaces can explain the discrepancies between SRPES experiments, which cannot detect any magnetic moment, and ARPES experiments, which displays spin-split states. The topological antiferromagnetism proposed by Blügel, Pescia, and

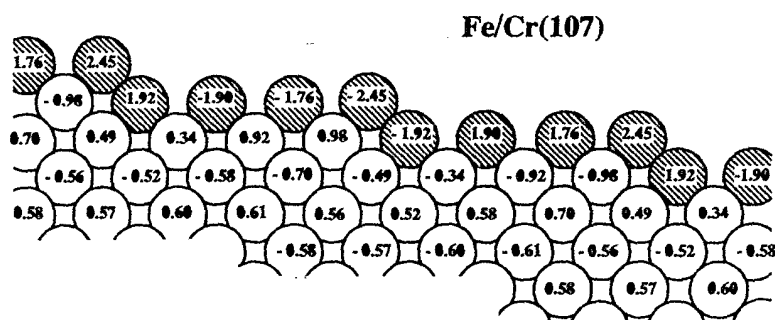


FIG. 7. Local magnetic moment μ (in unit of μ_B) per atom for an Fe monolayer on Cr(107) substrate. The white circles represent the Fe atoms and the dashed circles the Cr atoms.

Dederichs¹⁴ and calculated here, shows that the mean magnetic moment of the double-cell configuration is strictly zero: this is enough to explain SRPES experiments. However, the local polarization is not zero, so that ARPES can depict spin-split states. Moreover, Wiesendanger *et al.*¹⁵ were able to detect by means of STM the change of polarization when going from one step to the next. The magnetic moments on the step are, however, not all equal, but their values can be explained simply by the coordination-rule criterion, so that the atom with the lowest coordination (step-edge atom) has the highest magnetic moment. As discussed by Wiesendanger *et al.*,²⁴ in their study of magnetite, STM can also be a highly useful tool in the study of the topographic and magnetic surface structure of magnetic materials down to the atomic level. The Cr atom at the step edge has a magnetic moment 40% higher than the kink atom. We wonder if this difference may be detected by STM. Of course, contrary to the case of magnetite where atoms of different chemical type are present and where the crys-

tallographic arrangement is more anisotropic, here, only one chemical type of atom is present.

In summary the local magnetic distribution has been explicitly calculated for vicinal surfaces of Cr. Also, two-step periodicity is obtained for the Fe/Cr(107) system as a consequence of the antiferromagnetic order characteristic of the Cr substrate. Therefore, the sign of the magnetization at the Fe overlayer oscillates from step to step. For large step length, in-plane ferromagnetic coupling is obtained at each step, an exception being the atoms at the kink of the step. The present result gives support to the fact that the short oscillatory period of the spin polarization of the Fe overlayer experimentally observed⁶ is favored by the antiferromagnetic polarization of the Cr substrate.

This work was partially supported by DGICYT (Spain) and Junta de Castilla y Leon. We also wish to thank the CNIMAT of Nancy for computing facilities.

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