Magnetic behavior of thin Cr layers sandwiched by Fe

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The magnetic behavior of thin layers of Cr in Fe/Cr/Fe(001) trilayers and superlattices is studied using the first principles self-consistent RS-LMTO-ASA (real space – linear muffin-tin orbital – atomic sphere approximation) method. The effects of lattice compression and interface mixing are investigated, and it is shown that they can cause large reductions of the Cr magnetic moments.
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I. INTRODUCTION

Magnetic multilayers exhibiting giant magnetoresistance have stimulated a considerable amount of research on magnetic thin films. The magnetoresistance effect, which is currently being used by the information processing industry to develop magnetoresistive read/write heads, occurs in a variety of systems, but it was originally observed in Fe/Cr multilayers.[1] Adjacent Fe layers in Fe/Cr multilayers couple, mediated by the Cr layer, and the coupling oscillates as a function of the Cr spacer thickness, between ferromagnetic and antiferromagnetic. The Fe/Cr interface quality can significantly affect both the magnetoresistance and the exchange coupling between the Fe layers.[2,3] It influences the ''magnetic contrast'' which is related to the spin dependent transmission and reflection of carriers across the interface. It is easier from a theoretical point of view to treat systems with perfect interfaces having translational symmetry parallel to the layers and for a long time first-principles calculations were restricted to these systems.[4–6] However, experimentally, it is very difficult to completely avoid interface mixing and the formation of islands in the deposition of Cr on Fe.[7,8] Therefore, it is important to consider deviations from perfect interfaces and investigate their influence on the properties of Fe/Cr multilayers. With the advances in the theoretical approaches and better computer facilities, first-principles calculation of multilayers in the presence of interface mixing is now possible.[9,10] However, values for the local moments of Cr sites in the bcc structure are extremely sensitive to small changes in the lattice parameter[11] and, as we show here, a lot of care must be taken when comparing different theoretical results with experiments.

The magnetic properties of Fe/Cr systems have been extensively studied and a considerable amount of information about this system is currently available. The ground state of bulk bcc Cr exhibits a spin density wave (SDW) antiferromagnetism which is not commensurate with the lattice. The wave vector of the SDW lies along one of the [001] cubic directions and its wavelength is ≈ 21 lattice spacings. Theoretically the incommensurate state of Cr is difficult to treat. Recent LSDF-KKR calculations have shown that for the experimental lattice constant a SDW state with a wave vector close to the observed one is energetically more favorable than the antiferromagnetic state, but for the Cr lattice constant which minimize the total energy, the calculated ground state is nonmagnetic.[12] Bulk Cr is in the verge of a transition between magnetic and nonmagnetic states and slight differences in lattice parameters or theoretical procedures can drive the system nonmagnetic. If one neglects incommensurability, bulk Cr is an antiferromagnet with each site having a magnetic moment of $\approx 0.6\ \mu_B$; the magnetic moments of the atoms occupying the body-centered positions align in one direction, and opposite to those in the bcc vertices. The commensurate antiferromagnetism in bulk Cr can be stabilized by the presence of impurities, such as Mn and Fe among others.[13] In Fe/Cr/Fe systems, the presence of the Fe layers affects the magnetic properties of the Cr layer. Since the Cr moments couple antiferromagnetically with the Fe moments, one would expect in-plane ferromagnetic order in Fe/Cr/Fe(001) sandwiches. For sufficiently large Cr thicknesses, the incommensurate SDW may form in the Cr spacer layer.[12] However, below a critical thickness, the Cr layer is unable to sustain a stable incommensurate SDW, even at very low temperatures.[14,15] Previous works have shown that the magnetic moments in the Cr layer depend on the relative orientation of the magnetizations of the adjacent Fe layers as well as on the growth orientation of the multilayer.[16–19] The magnetic ordering of the Cr layer is also strongly influenced by inhomogeneities at the Fe/Cr interfaces. The presence of steps at the interface can lead to noncollinear spin ordering in the Cr layers.[20–22] It is also recognized that in addition to steps, mixing in the Fe/Cr interfaces always occurs, even in the best grown samples.[7,8] The formation of a disordered Fe/Cr alloy at the interface generates frustrations which may substantially suppress the moment of some Cr neighboring sites.[5] Such frustrations may also induce noncollinear arrangements of the local magnetic moments which can be rather intricate, depending on the degree of interface mixing.

A relevant aspect that has not been sufficiently explored is related to the fact that the magnetism of Cr is rather sensitive to the application of pressure.[11,12] The Fe lattice parameter is $\approx 0.6\%$ smaller than that of Cr. Thus, when sandwiched by Fe, the Cr layer is slightly compressed, and this effect is more pronounced for very thin Cr spacers. It is noteworthy that earlier perturbed angular correlation spectroscopy and transport measurements in Fe/Cr(001) multilayers have found no antiferromagnetic order in the Cr layers with thicknesses $t_{\mathrm{Cr}} < 42$ Å.[23,24] Neutron scattering results, however, exhibited magnetic scattering, consistent with a commensurate antiferromagnetic order in Cr on samples with thinner Cr layers.[9,14] It is, therefore, important to determine the electronic structure of Cr layers sandwiched by Fe, taking into account interface mixing and lattice relaxation effects.

In this article we have investigated the effect of pressure and interface mixing on the local magnetic moments of very thin Cr layers sandwiched between Fe slabs in Fe/Cr(001) multilayers. We have performed first-principles calculations of the electronic structure of $\mathrm{Fe}/\mathrm{Cr}_n/\mathrm{Fe}(001)$ trilayers and $\mathrm{Fe}_3/\mathrm{Cr}_n(001)$ superlattices (with $n=1$, 3, 5, and 7 atomic planes) for different values of the lattice parameter and degrees of interface mixing, using a self-consistent and real space linear-muffin-tin method within the atomic sphere approximation (RS-LMTO-ASA method). The magnetic moments of all inequivalent sites are calculated in each case. The different roles played by interface mixing and lattice relaxation in the reduction of the local magnetic moment of sites in the Cr layers are discussed. The plan of the paper is as follows: in the next section, a brief account of the theory employed in our calculations is given. In Sec. III, we discuss our results for $\mathrm{Fe}/\mathrm{Cr}_n/\mathrm{Fe}(001)$ sandwiches with perfect Fe-Cr interfaces. Calculations for $\mathrm{Fe}_m/\mathrm{Cr}_n(001)$ superlattices considering interface mixing are presented and discussed in Sec. IV. Finally, in Sec. V, we compare our results with previous works and draw our main conclusions.

II. THE RS-LMTO-ASA SCHEME

In this section we give a brief outline of the RS-LMTO-ASA method used in our calculations. A detailed description of this procedure can be found elsewhere.[25–27] The RS-LMTO-ASA is a first-principles and self-consistent scheme which is based on the LMTO-ASA formalism.[28,29] It is similar to the reciprocal-space-LMTO-ASA method; however, in contrast, the local density of states is determined in real space, using the recursion method,[30] rather than by solving the eigenvalue problem in k space. The RS-LMTO-ASA is an order N method which can be used to treat complex systems with large number of inequivalent atoms in the unit cell. It has been successfully applied to study crystalline systems,[25] substitutional and interstitial impurities in metallic hosts,[26,31] as well as other local defects in metals,[32] and metallic surfaces,[27,33] yielding results which agree well with those obtained by other first-principles approaches. The RS-LMTO-ASA is a linear method, and its solutions are more accurate around a given energy E_ν, which is usually chosen to be the center of gravity of the occupied part of the band. It also uses the atomic sphere approximation (ASA), where the Wigner Seitz (WS) cell around each site is substituted by a WS sphere with the same volume. In the RS-LMTO-ASA method, the solutions of the Schrödinger-like equations within the WS spheres are obtained by the same procedures as those used in the k space-LMTO-ASA method, including the approximations which are usually made in treating the exchange and correlation terms.

In the RS-LMTO-ASA scheme we work in the orthogonal representation of the LMTO-ASA formalism,[29] for which the overlap is approximately given by the unit matrix. Using the first-order approximation, where only linear terms in $(E - E_\nu)$ are retained, the Hamiltonian H takes the usual tight-binding form, and the corresponding eigenvalue problem can be written as[25,26]

$$(H-E)u=0,$$

where

$$H=\overline{C}+\overline{\Delta}^{1/2}\overline{S}\,\overline{\Delta}^{1/2}.$$

$\overline{C}$ and $\overline{\Delta}$ are the potential parameters and $\overline{S}$ is the structure constant in the tight-binding LMTO-ASA representation. $\overline{S}$ is entirely determined by the lattice structure, i.e., by the position of the lattice sites. The potential parameters ($\overline{C}$ and $\overline{\Delta}$) are related to the solutions of the Schrödinger-like equation inside the WS spheres around each site, and are determined self-consistently. The corresponding wave functions are given by

$$\Psi_E=\sum_{RL}\,[\varphi_{l\nu}(r_R)+(E-E_\nu)\dot{\varphi}_{l\nu}(r_R)]Y_L(\hat{r}_R)u_{RL}(E),$$

where $\varphi_{l\nu}(r)$ and $\dot{\varphi}_{l\nu}(r)$ are the radial part and its first energy derivative (at $E=E_\nu$) of the solution of the Schrödinger equation in the WS sphere at site R. $Y_L(\hat{r}_R)$ are spherical harmonics with quantum numbers $L=(l,m)$, and $u_{RL}(E)$ are the solutions of the eigenvalue problem given above.

In our calculations for Fe/Cr/Fe(001) trilayers we have fixed the Fermi energy E_F of the system to be that of bulk Fe. Charge transfers in each inequivalent site have been determined by integrating the corresponding local density of states (LDOS) up to E_F. These charge transfers were then used in a two-dimensional Ewald summation[34] to obtain the Madelung potential and the value of the electrostatic potential (VES) at each site. When treating the semi-infinite Fe layers, only the Fe sites occupying the first two atomic planes close to the Fe-Cr interface have been treated self-consistently in our calculations. For those further away we have taken the bulk Fe parameters $\overline{C}$ and $\overline{\Delta}$. The use of bulk parameters does not render the procedure less accurate, provided a sufficiently large number of atomic planes close to the interface is treated self-consistently.[26,33]

We have performed our RS-LMTO-ASA calculations for superlattices using a supercell approach in which translational symmetry, with the supercell periodicity, is assumed in the direction perpendicular to the layers. In this case, the Fermi level of the system is determined, at each iteration, by filling the bands with the correct number of electrons, and the electrostatic potential VES is obtained, in the usual way, by Ewald sums.

III. RESULTS FOR Fe/Cr/Fe(001) SANDWICHES

Our aim is to investigate the effect of the Fe layers on the magnetic state of the Cr spacer layer in Fe/Cr/Fe(001) sandwiches. The natural reference system with which one should compare the Cr layer results is bulk Cr. One may ask, for instance, whether the presence of the Fe layers (or the compression of the Cr lattice caused by them) enhances or diminishes the magnitude of the Cr local magnetic moments with respect to its bulk value. As mentioned earlier, the ground state of bcc Cr shows an incommensurate SDW antiferromagnetism, where the maximum observed local moment is $0.59\mu_B$, for an equilibrium WS radius of 2.684 a.u.[13,35] However, the commensurate antiferromagnetic state of Cr is close to the ground state, and most theoretical calculations of the electronic structure of bulk Cr assume it as an approximation for the ground state of Cr. Nevertheless, even in this case, the theoretical determination of the correct local moment in bulk Cr is rather subtle. The calculated moment of bulk bcc Cr is extremely sensitive to the value of the lattice parameter used, as well as to details of the calculations.[11,12,36] It has been reported, in the context of LMTO-ASA calculations, that it is necessary to consider a slightly larger lattice parameter than the one experimentally observed to obtain a reasonable description of the magnetic properties of bulk bcc Cr.[11] Moruzzi and Marcus,[36] using total-energy band calculations within the local spin density approximation (LSDA), have shown that bcc Cr has a first-order transition from non-magnetic to antiferromagnetic behavior at lattice constants expanded by approximately 1.5%. Therefore, in studying the changes of the Cr moments due to the presence of Fe layers in Fe/Cr systems, it is important to calculate the magnetic moment of pure Cr with the same method, to set our reference state on an equal footing. This is necessary in order to be able to infer the correct trends. For example, if an LSD calculation for an Fe/Cr system assumes a lattice constant at which pure bulk Cr is nonmagnetic, any finite local magnetic moment found in the Cr layer of the Fe/Cr system would represent an enhancement of the Cr moment. On the other hand, the same results for the Fe/Cr systems would be interpreted differently if the magnetic moment found for pure Cr is $m_{\mathrm{Cr}}=0.6\ \mu_B$. Therefore, before proceeding to investigate Fe/Cr systems, we use the RS-LMTO-ASA scheme to calculate the electronic structure and the magnetic moment of pure bulk bcc Cr.

In all our calculations we have used the LSDA with the exchange and correlation potential of von Barth and Hedin.[37] We have considered a basis set with nine orbitals per site representing the valence *s*, *p* and *d* electrons, and have used the Beer and Pettifor terminator,[38] with a cutoff parameter $LL=20$ in the recursion chain. For bcc Cr, we consider two sublattices, allowing the occurrence of antiferromagnetic alignment. A cluster of ≈ 3600 atoms was used in the calculations. We found that the local moments of the Cr sites are extremely sensitive to the lattice parameter. The system does not develop a local magnetic moment if the experimental lattice constant of Fe is used. However, it is found to be antiferromagnetic, with $m_{\mathrm{Cr}}=0.54\ \mu_B$, for a Cr lattice parameter $a_{\mathrm{Cr}}=2.935$ Å, which is $\approx 1.75\%$ larger than the experimentally observed bulk Cr value. This is in good agreement with other LSD calculations,[11,36] and variations of the lattice parameter in this range will be used in our investigations of Fe/Cr systems.

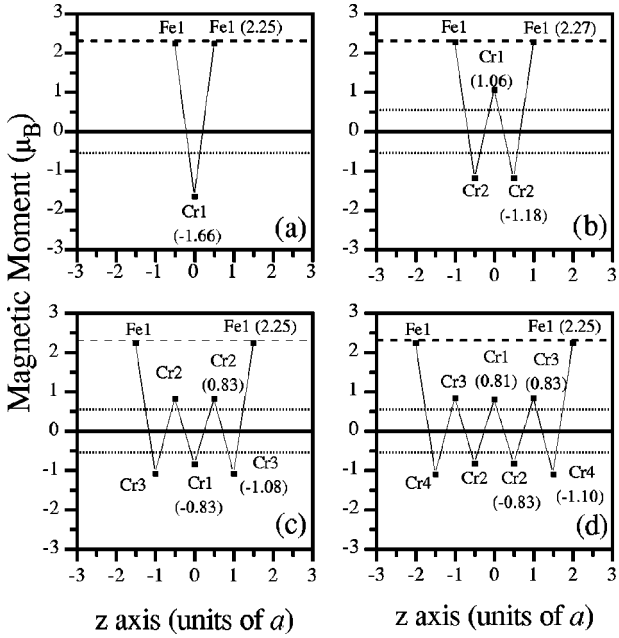

FIG. 1. Magnetic moment distribution for $\mathrm{Fe/Cr}_n\mathrm{/Fe(001)}$ sandwiches with $a=2.935$ Å [$n=1$ (a), $n=3$ (b), $n=5$ (c), and $n=7$ (d)]. Dashed (dotted) line shows the bulk Fe (Cr) moment for the lattice parameter used in the calculations.

Having established the range of lattice parameters which we will consider, we now present our results for $\mathrm{Fe/Cr}_n\mathrm{/Fe(001)}$ sandwiches. Initially, we take a common lattice constant $a=2.935$ Å for the whole Fe/Cr system, and investigate the magnetic moments of the Cr sites for different thicknesses of the Cr spacer layer. This is a reasonable choice of *a* to study the tendencies obeyed by the local magnetic moments in unstrained Cr layers sandwiched by Fe, because the calculated magnetic moment of pure Cr, with this lattice constant, is rather close to the observed value. We have performed calculations for Cr layers with thicknesses $n=1$, 3, 5, and 7 atomic planes.

Our RS-LMTO-ASA calculations for Fe/Cr/Fe(001) sandwiches require the determination of the Fermi level of the system and the bulk potential parameters associated with the Fe sites far from the Fe/Cr interface. In order to obtain these quantities we have performed a calculation for bulk Fe with the chosen lattice constant $a=2.935$ Å. For this value of *a* we found a magnetic moment $m_{\mathrm{Fe}}=2.31\ \mu_B$, which is slightly larger than the observed value of $2.21\ \mu_B$ obtained with the Fe lattice constant $a_{\mathrm{Fe}}=2.861$ Å. Having obtained the parameters for bulk Fe we can now use the procedure described in Sec. II to calculate the electronic structure and magnetic moments in the Fe/Cr/Fe(001) sandwiches.

In Fig. 1 we show the RS-LMTO-ASA results for the magnetic moment distribution of (a) $\mathrm{/Fe/Cr_1/Fe(001)}$, (b) $\mathrm{/Fe/Cr_3/Fe(001)}$, (c) $\mathrm{/Fe/Cr_5/Fe(001)}$, and (d) $\mathrm{/Fe/Cr_7/Fe(001)}$ sandwiches. In all cases we have labeled the central Cr atomic plane as Cr1, and the Fe plane at the Fe/Cr interface as Fe1. The other Cr planes are numbered in increasing order from Cr1 to the interface, as shown in the corresponding figures. For comparison we also show the local magnetic moments of bcc Cr (dotted line) and Fe (dashed line), both

TABLE I. Magnetic moment profiles in $Fe_2/Cr_3(001)$ superlattices, calculated with different lattice parameters $a_1=2.935$ Å and $a_2=2.872$ Å (arithmetic average of bulk Fe and Cr observed lattice constants). For comparison, we also show earlier first-principles LMTO-ASA results. All magnetic moments in units of μ_B.

	RS-LMTO-ASA		LMTO-ASA (Ref. 4)
	a_1	a_2	a_2
Fe1	2.43	2.23	2.1
Cr2	−0.96	−0.59	−0.5
Cr1	0.79	0.43	0.4

TABLE III. Local magnetic moments (in μ_B) of the Fe at the interface (Fe1), of the Cr at the interface (Cr4), of the other adjacent Cr layers (Cr2) and (Cr3), and of the Cr in the central layer (Cr1), for the system $Fe/Cr_7/Fe(001)$ sandwiches.

a(Å)	Cr1	Cr2	Cr3	Cr4	Fe1
2.935	0.81	−0.83	0.83	−1.10	2.25
2.905	0.54	−0.56	0.58	−0.84	2.16
2.884	0.43	−0.44	0.46	−0.71	2.10
2.872	0.33	−0.34	0.36	−0.60	2.06
2.861	0.25	−0.27	0.29	−0.53	2.02

calculated with the same lattice constant $a=2.935$ Å. We have found that the Fe-Cr interface planes couple antiferromagnetically, and that the magnetic moment of the Cr sites at the Cr/Fe interface, is substantially enhanced in comparison with the bulk Cr value. The magnetic moment of the interfacial Cr sites is larger for $n=1$ ($m_{Cr}=1.66\mu_B$) when they are surrounded by Fe layers on both sides, and decreases to $m_{Cr}=1.10\mu_B$ when the Cr layer has seven atomic planes. The moment of the central-plane sites (Cr1) is also enhanced, but it decreases as the distance to the Fe-Cr interface increases, approaching the bulk-Cr value for sufficiently large Cr layer thicknesses.

It is interesting to compare our results with others found in the literature[4,39] for Fe/Cr(001) multilayers with similar Cr spacer layer thicknesses. Our calculation for the $Fe/Cr_3/Fe(001)$ sandwich [see Fig. 1(b)] give values of m_{Cr} which are greater than the experimentally observed bulk Cr value ($m_{Cr}\approx 0.6\ \mu_B$) for all Cr sites. However, previous calculations for $Fe_2/Cr_3(001)$[4] and $Fe_3/Cr_3(001)$ (Ref. 39) supercells have found smaller values of m_{Cr}, apparently indicating that the presence of the Fe layers reduces the Cr magnetic moments in these systems. Sandwiches and superlattices have different geometries, but the tendencies obtained for the Cr moments in the presence of Fe at an ideal Fe-Cr interface should be similar, and the origin of this apparent discrepancy must be investigated. In Tables I and II, we present RS-LMTO-ASA results for the $Fe_2/Cr_3(001)$ and $Fe_3/Cr_3(001)$ systems, considering different values of the lattice constant. It is clear that the RS-LMTO-ASA calculations agree with those of Refs. 4 and 39, provide the same lattice constant is used in both ccalculations. On the other hand, due to limitations of the LSDA, calculations performed using the lattice constant listed in columns two and three of Tables I and II yield magnetic moments for bulk bcc Cr which are close to zero. Therefore, as in the case of the trilayer, the presence of Fe in the $Fe_2/Cr_3(001)$ and $Fe_3/Cr_3(001)$ superlattices leads, in all cases, to an enhancement of the Cr moments relative to its pure bulk value (obtained with the same lattice parameter). We have seen that the moment of Cr is extremely sensitive to changes in the lattice parameter and it is interesting to investigate the effect of these changes on the local moments at the Cr sites in Fe/Cr/Fe(001) sandwiches. Since the lattice parameter of Fe is somewhat smaller than that of Cr, one would expect the Cr layer lattice spacing to be slightly reduced in $Fe/Cr_n/Fe(001)$ sandwiches, particularly for thin Cr layers. To investigate the effect of such compression, we have considered the $Fe/Cr_7/Fe(001)$ system and calculated the magnetic moments at all inequivalent Cr sites, for different values of the lattice parameter. The calculated results are shown in Table III for the central (Cr1) and subsequent Cr atomic planes (Cr2 and Cr3), as well as for the interfacial Fe-Cr planes (Fe1 and Cr4), as a function of lattice parameter. The local moment of the Fe sites (Fe1) does not change much when the lattice parameter is varied, but the Cr local moments are substantially reduced as the lattice parameter decreases. The Cr1 magnetic moment is reduced from $m_{Cr}=0.81\ \mu_B$ for $a=2.935$ Å to $m_{Cr}=0.25\ \mu_B$ when the experimental lattice constant of bcc Fe ($a_{Fe}=2.861$ Å) is used. As in the case of the superlattices considered in Tables I and II, the Cr moments in $Fe/Cr_n/Fe(001)$ sandwiches can be

TABLE II. Magnetic moment profiles in $Fe_3/Cr_3(001)$ superlattices, calculated with different lattice parameters: column one with $a_1=2.935$ Å; columns 2 and 3 with an unit cell whose lengths along the [100] and [001] directions are $a_2=2.829$ Å and $a_3=2.875$ Å, respectively. For comparison, we also show earlier ASW (augmented spherical waves) results. All magnetic moments in units of μ_B.

	RS-LMTO-ASA		ASW (Ref. 39)
	a_1	a_2,a_3	a_2,a_3
Fe1	2.22	1.94	1.80
Fe2	2.57	2.48	2.45
Cr2	−0.89	−0.48	−0.35
Cr1	0.64	0.30	0.25

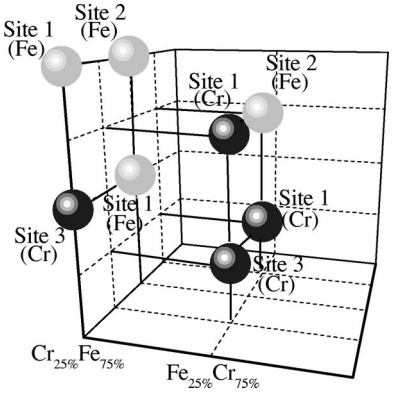

FIG. 2. Schematic representation of the two layers $(Cr_{25\%}Fe_{75\%})(Cr_{75\%}Fe_{25\%})$ rough Fe-Cr interfaces, used in the system $Fe_m/Cr_n(001)$. Each site is identified by numbers.

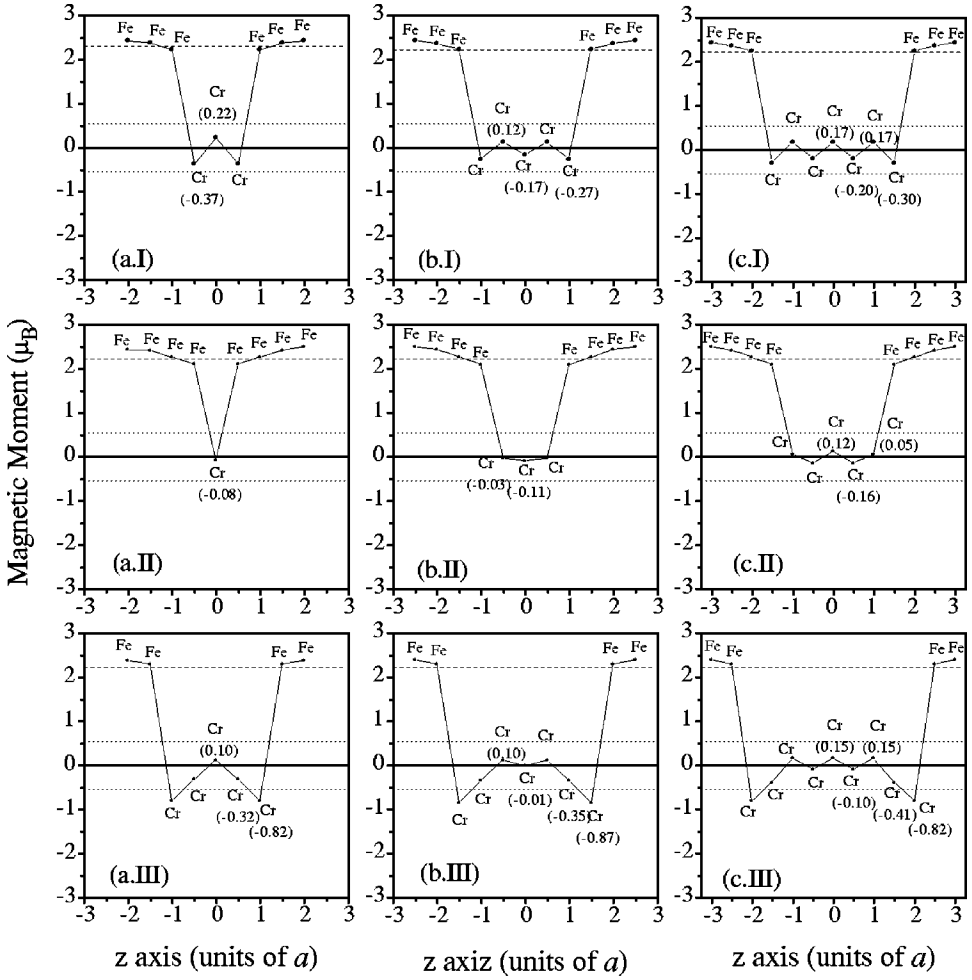

FIG. 3. Magnetic moment distribution for the (a) $Fe_3/Cr_1(001)$, (b) $Fe_3/Cr_3(001)$, and (c) $Fe_3/Cr_5(001)$ superlattices with $a = 2.935$ Å, along the three lines representative of the inequivalent sites as explained in the text. The dashed (dotted) line shows bulk Fe (Cr) moments for the lattice parameter used in the calculations.

smaller than $0.59\mu_B$. However, for perfect interfaces, and when the Fe layers are ferromagnetically aligned, they are always enhanced relative to the Cr bulk value which, in turn, can be drastically reduced under pressure. For the smaller lattice constant considered in Table III, the moment of the Cr1 sites in $Fe/Cr_n/Fe(001)$ sandwiches tends to zero as n increases, because this is the bulk-Cr value for $a_{Cr} = 2.861$ Å.

IV. Fe/Cr(001) SUPERLATTICES IN THE PRESENCE OF INTERFACE MIXING

Ideal interfaces are difficult to obtain experimentally and some degree of mixing between adjacent Fe-Cr planes at the interface is usually present in real systems. Therefore it is important to consider deviations from perfect interfaces. Fe/Cr(001) superlattices with several degrees of interface mixing have been studied by Stoeffler *et al.*[16] using a parametrized electronic structure approach. It was found that the introduction of interfacial ordered compounds to simulating a mixing of 25% at the interface can lead to strong reductions of local moments in the Cr layers. We have seen that a compression of the Cr lattice can also cause a reduction of the Cr moments in Fe/Cr(001) superlattices and sandwiches. In this section we use the RS-LMTO-ASA method to investigate the combined effects of compression and interface mixing on the local moments at the Cr sites.

The interfacial Cr-Fe mixing produces a disordered alloy which can extend over a few atomic planes beyond the Fe/Cr interface. Such random mixing generates frustrations of magnetic character, which may induce intricate non-collinear spin alignments. Presently, however, it would be computationally prohibitive to include all these degrees of freedom in our first-principles calculation. Often simpler models can be used to understand the behavior of more complex systems, and here, to illustrate the effect of compression in the presence of interface mixing, we restrict our calculations to the interfacially ordered compounds and collinear spin arrangements.

To simulate the Fe-Cr superlattices, we have used supercells consisting of $(Cr_{25\%}Fe_{75\%})$ $(Cr_{75\%}Fe_{25\%})/Cr_n/(Cr_{75\%}Fe_{25\%})$ $(Cr_{25\%}Fe_{75\%})/Fe_3$ with $n=1$, 3, 5, and 7, to generate large clusters of up to 6000 atoms which were used in the calculations. The potential parameters for all inequivalent atoms in the supercell were obtained self-consistently and the Fermi level was determined by filling the available states with the correct number of valence electrons in the system. Due to interface mixing there are three inequivalent atoms in each of the (001) planes of the bcc structure and our

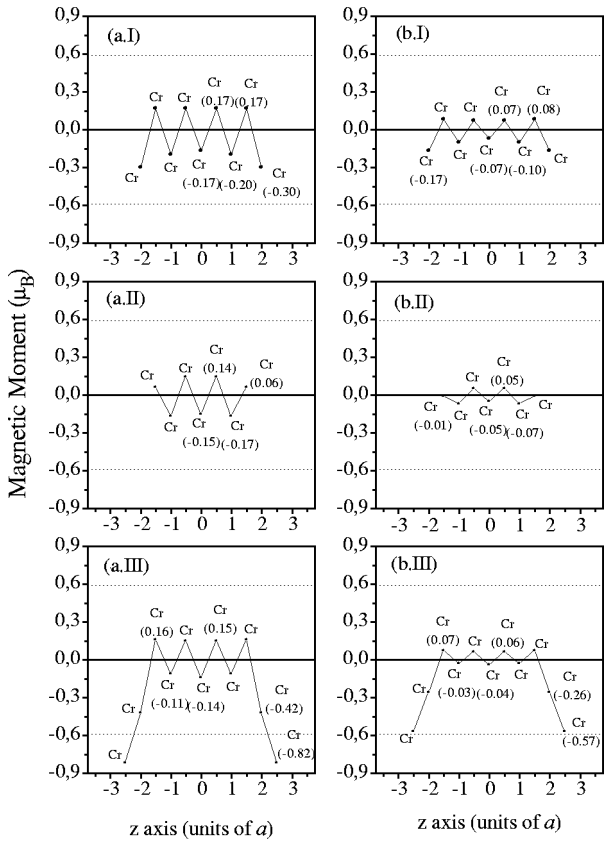

FIG. 4. Magnetic moment distribution for the $Fe_3/Cr_7(001)$ superlattices with (a) $a=2.935$ Å and (b) $a=2.861$ Å, along the three lines (I, II, and III) representative of the inequivalent sites, as specified in Fig. 3. The dotted line ($0.59\mu_B$) indicates the experimentally observed value in bcc Cr.

results will be plotted following the lines of sites designated by 1 (two degenerate branches), 2, or 3 in the schematic representation of the mixed interface planes of Fig. 2.

In Fig. 3 we show results for Fe/Cr(001) superlattices with $n=1$ (a), 3 (b) and 5 (c), obtained using a lattice constant $a=2.935$ Å, which should represent well the magnetic behavior of unstrained Cr layers. Values for the Cr moments in the case of $n=7$ are shown in Fig. 4(a). Three sets of boxes (I, II, and III) are shown in each case. In the first box (I) we show the behavior of the moments along the line of sites designated by 1 in Fig. 2. The two degenerate paths of this type go through majority sites at the interface: Fe atoms in mixed ($Cr_{25\%}Fe_{75\%}$) planes and Cr atoms in mixed ($Cr_{75\%}Fe_{25\%}$) planes. In the second box (II), we show moments along line 2, which pass through Fe atoms in all mixed planes, while in the third box (III), we show results for the line 3 which passes through Cr atoms in these same planes. We see that the local moments at the central plane of the thin Fe_3 layer are slightly increased when compared to bulk values, while the moment at the minority Fe sites in the mixed planes (see box II) is slightly decreased. These variations are rather small, and our results agree with the experimental observations,[19] which indicates that the magnetism of the Fe is not significantly affected by the presence of Cr neighbors. The local moments of Cr atoms in the mixed planes (box III), with the same number of Fe neighbors, have similar values of magnetic moments in the different $Fe/Cr_n(001)$ systems: the Cr atoms in the mixed ($Cr_{25\%}Fe_{75\%}$) planes have moments around $-0.8\mu_B$, enhanced relative to that of bcc Cr due to the presence of the Fe neighbors, while those in ($Cr_{75\%}Fe_{25\%}$) planes show moments of around $0.4\mu_B$. As expected,[10] the local moments at the unmixed Cr planes are drastically reduced by interface mixing. The moments at Cr sites of the central plane in the Cr_n layer increase as n is increased, and should tend to the value for bcc Cr in the asymptotic limit of n large. Our results confirm the general behavior first described by the parametrized tight-binding calculations of Stoeffler *et al.*[16] The qualitative agreement between the first-principles calculations and the parametrized results of the literature is not surprising. One can show that a sound parametrization and the use of approximate charge neutrality are expected to give a good description of the electronic structure of transition metal systems.[25,40]

To investigate the combined effects of compression and interface mixing in Fe/Cr(001) superlattices, we have performed RS-LMTO-ASA calculations for the system with $n=7$, taking $a=2.861$ Å, the experimentally observed lattice constant of bcc Fe. In Fig. 4 we show results for the local moments at Cr sites for two different values of the lattice parameter: $a=2.935$ Å [Fig. 4(a)] and $a=2.861$ Å [Fig. 4(b)]. The notation is the same as in Fig. 3, but to better represent the small Cr moments, the larger Fe moments were not included. The calculated moment for Cr in the commensurate antiferromagnetic state is close to this value for $a=2.935$ Å, but goes to zero for $a=2.861$ Å. Comparing the values in Figs. 4(a) and 4(b), we see that a compression of the Cr lattice can further suppress the moments at the Cr sites. The Cr atoms at the mixed Fe-Cr planes sustain considerable local moments [see box 4(b) III] even under compression, but the moments of Cr located in the Cr_7 layer are already extremely small, below $0.1\mu_B$. Furthermore, the Cr moment in the central Cr plane should tend asymptotically to zero (the bulk value for the compressed lattice) as the number of planes in the Cr layer is increased; as long as the lattice remains compressed, the Cr moments in the Cr_n layer should remain under $0.1\mu_B$ for n greater than seven. Such small moments may be difficult to detect experimentally. Based on our results, we suggest that the combined effect of interface mixing and compression of the Cr lattice due to the presence of Fe layers could be responsible for the experimentally observed[19,23] paramagnetic behavior of the thin Cr layers in Fe-Cr sandwiches.

V. CONCLUSIONS

We have used the first-principles RS-LMTO-ASA scheme to study the magnetic behavior of extremely thin Cr layers in Fe-Cr sandwiches and superlattices. The effect of compression of the Cr layer in these systems was also investigated. The calculations, some performed for large supercells of 76 atoms, illustrate the application of this order-N real-space approach to the study of the electronic structure and magnetic behavior of complex metallic systems.

We have considered $Fe/Cr_n/Fe(001)$ sandwiches, consisting of n atomic planes of Cr embedded in Fe along the (001) direction, and have obtained results for very thin Cr layers with $n=1$, 3, 5, and 7. In all cases, the local moments

at the Cr sites were larger than $0.59\mu_B$, the experimentally observed value in bcc Cr, being enhanced due to the presence of the Fe layers. A similar tendency was obtained for Cr moments in $Fe_3/Cr_3(001)$ and $Fe_2/Cr_3(001)$ superlattices. By comparing the superlattice results with those of other first principle approaches in the literature, we have shown that, due to the sensibility of the Cr moment to small variations of the lattice parameter, when investigating the behavior of bcc Cr layers in the presence of other elements, extra care must be taken to isolate the effect of compression from other effects. When performing first-principle calculations in these systems it is important, as a consistency check, to calculate bcc Cr under the same conditions and with the same method.

Since the lattice parameter of bcc Fe is slightly smaller than that of bcc Cr, it is interesting to investigate the magnetic behavior of Fe-Cr sandwiches under compression. Here we have performed RS-LMTO-ASA calculations for $Fe/Cr_7/Fe(001)$ sandwiches, as a function of lattice parameter. The results indicate that the moments of Fe sites are not very sensitive to small variations of the lattice parameter, but the local moments at the Cr sites are significantly reduced by compression.

Finally, we have presented RS-LMTO-ASA calculations for $Fe/Cr_n(001)$ superlattices, with $n=1$, 3, 5, and 7, in the presence of interface mixing. Our results for the unstrained Cr lattice are in qualitative agreement with those of Ref. 16, showing that interface mixing can substantially reduce the moments of the Cr layers. We have also investigated the role of compression of the Cr lattice in the presence of interface mixing in Fe/Cr(001) superlattices. Our results suggest that the combined effects of interface mixing and compression of the Cr lattice could be responsible for the paramagnetic behavior observed for thin Cr layers[19,23] in Fe/Cr(001) systems.

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