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Antiferromagnetic polarization at Mn/V(001) interfaces

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The magnetic map of a few Mn monolayers on V(001) substrates is investigated by *ab initio* density-functional theory within a generalized gradient approximation. We have found that a perfect layer-by-layer growth of the Mn film on V(001) is unstable against interdiffusion between Mn and V at the interface. The magnetic ground states found in the range of 1–3 monolayers (ML) of Mn present layered antiparallel couplings with high magnetic moments at the surface Mn atoms ($\sim 3\mu_B$). A buried Mn ML is shown to be more stable than the Mn monolayer on the V(001) surface and the 2-ML MnV/V(001) surface ordered alloy. A buried alloy, i.e., V/Mn-V/V(001), is also found more stable as compared to clustering Mn buried ML in V(001), and to a clean V(001) surface, whereas the surface ordered alloy is instable against the Mn overlayer and clean V(001) surface. As for the effect of magnetism on the stability of a monolayer with respect to a bilayer, we show that the formation energy sign changes from the paramagnetic to the spin-polarized case.

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I. INTRODUCTION

The formation of local magnetic moments plays a crucial role in the structure of metals and stability of alloys. With the increase of local moments in low-dimensional metallic systems, the importance of magnetism for the structure and stability of these nanosystems becomes more important. From the experimental point of view, it is very difficult to control and to characterize the morphology and growth modes of ultrathin films on different substrates. Two effects can occur: (i) diffusion of atoms across the surface, which could lead to cluster formation; (ii) interdiffusion of atoms into the substrate, leading to bulk alloys or a film of atom clusters covered by substrate atoms. These effects are related to the activation energy needed for diffusion. In this paper we present theoretical results of the interplay between the magnetic behavior and the structural state of thin Mn films deposited on V(001). The Mn and V elements are subject to many controversies about either their structural phases or magnetic properties in nanosystems. A large number of theoretical studies have been devoted to the magnetic properties of the various crystalline phases of Mn.[1–3] It is clearly pointed out that the local-density approximation (LDA) may not be suitable so that generalized gradient approximation (GGA) has to be used.

Mn is a particularly interesting case because, according to Hund's rule, the magnetic moment of the free atom is as large as $5\mu_B$. Thus, it tends to be one of the most studied element.[4] Actually, Mn is incorporated to dope semiconductors for spin injection. Besides, recent *ab initio* calculations[5] have tried to link the Curie temperature of these diluted magnetic semiconductors to the calculated local density of states. In the case of metallic systems, Mn deposited on Co and Fe was particularly studied because it was expected that Mn, being a nonferromagnetic element in its bulk metallic form,[6] can present an induced ferromagnetic behavior[4] in contact with a strong ferromagnet. However, both experimentally and theoretically, it was found that (i) indeed a ferromagnetic polarization appears at the interface *but* this induced polarization is "short-ranged;"[7,8] (ii) intermixing of atoms between the Mn adatoms and the Co (or Fe) substrate takes place, leading to surface ordered alloys.[7,9–12] This exchange of atoms stabilizes the system. Theoretically it has been shown that magnetism can act against interdiffusion,[13] i.e., the formation of an interfacial alloy is stabilized partially or entirely when spin polarization is included.

At low temperature, in its crystal form, Mn assumes a complicated crystal structure, but at high temperature it presents a fcc phase called γ-Mn, and a bcc phase called δ-Mn.[14] The two phases can be stabilized at low temperature by epitaxial growth of Mn on a substrate having fcc or bcc crystallographic structure. Indeed, different experimental studies[15–21] have shown that these simple structures could be stabilized at room temperature by alloying (γ-fcc) or by pseudomorphic epitaxy (δ-bcc) on appropriate substrates. Meanwhile, Tian *et al.*[22] concluded that one cannot affirm which among δ-Mn or γ-Mn is the equilibrium phase of Mn films grown on V(001), Pd(001), or Fe(001) as it is reported in many papers.[15–21] In addition, studies of $3d$-metal overlayers on noble metals have shown that Mn adopts in-plane antiferromagnetic (AF) structure on (001) surfaces of Cu, Ag, Au, Pt, and Pd.[23–26] The magnetic properties of Mn are very important for the stabilization of a new class of materials, i.e., ordered two-dimensional magnetic alloys where no ordered bulk alloys exist. The case of surface alloy is very interesting, because Mn displays a high magnetic moment as well as an outward relaxation that reduces the atomic coordination, and thus enhances the magnetic moment. As a result, there is a gain in magnetic energy which stabilizes the structures: $Mn_{0.5}Cu_{0.5}/Cu(001)$, $Mn_{0.5}Cu_{0.5}/Cu(110)$, $Mn_{0.5}Ni_{0.5}/Ni(001)$, $Mn_{0.5}Pd_{0.5}/Pd(001)$, and $Mn_{0.5}Ag_{0.5}/Ag(001)$.[27–31] These systems were already investigated theoretically as well as experimentally. One can cite also the spin-polarization studies of various surface alloys of Mn on Co(001) which were studied by Meza-Aguilar *et al.*[32]

Mohammed *et al.*[33] have investigated the magnetic properties of Cr on Mn using a real-space tight-binding approach for three crystallographic orientations and found that magnetism is more favored for the (111) direction than for less open (001) and (011) ones. In previous work, Khalifeh *et al.*[34] have reported calculations on a Mn/Cr system using a real-space tight-binding approach in the different crystallographic orientations. They found ferromagnetic sheets of both metals for the planes (001) and (111), and magnetic $c(2\times2)$ configuration for the (011) plane.

Vanadium bulk is known to be nonmagnetic but, theoretically, it presents a magnetic moment when its lattice parameter is sufficiently expanded. Morruzi and Marcus[35] have found an AF state in bcc vanadium at expanded volumes using the augmented-spherical-wave method. However, the magnetism of the V(001) surface is still subject to controversy from both experiment and theory. Rau and co-workers,[36,37] using electron-capture spectroscopy found that the topmost layer V(001) is ferromagnetic with a Curie temperature of 540 K. Using the same experimental technique, the same authors found that 1 ML of V on Ag(001) is ferromagnetic whereas with magnetooptic Kerr measurements, Fink *et al.*[38] found no magnetization where calculations predict antiferromagnetism.[39] Bihlmayer *et al.*[40] investigated the V(001) surface using the full potential linear augmented plane-wave method and concluded that in the case of very thin V films, a surface magnetic moment can be stabilized while for thicker and relaxed films no surface magnetism can be found. Robles *et al.*[41] studied the V(001) surface using two *ab initio* methods: Tight-binding linear muffin-tin orbital atomic sphere approximation (TB-LMTO-ASA) within the GGA of Langreth-Mehl-Hu[42] (LMH) and GGA of Perdew-Wang[43] (PW91) and the pseudopotential linear combination of atomic orbitals code SIESTA (spanish initiative for electronic simulations with thousands of atoms) within the GGA of the Perdew-Burke-Ernzerhof[44] (PBE) approach. They found that calculations within TB-LMTO lead to the same conclusions as those of Bihlmayer *et al.*,[40] whereas pseudopotential calculations lead to a high magnetic moment even for a thicker V slab. By using tight-binding method, M'passi-Mabiala *et al.*[45] investigated the polarization at the Cr/V interface in different crystallographic orientations and V thicknesses. The Cr polarization depends drastically on the crystallographic face considered. The (001) and (111) faces show layered antiferromagnetic (LAF) couplings whereas the (011) face displays in-plane AF order.

The difficulty in controlling and characterizing the morphology and growth modes at the interface is intimately related to the difficulties in the understanding of the interfacial magnetism of these systems. So the growth mode depends on the energetic barriers between the different process. For this purpose, we investigate the magnetic polarization of Mn on V(001) in the range of 1–3 ML. To begin with the case of 1 ML, we have examined the possible stabilization of the surface ordered alloy and the buried one against phase separation. We have considered the following structural configurations: (i) The Mn overlayer on V(001), (ii) the buried Mn monolayer, i.e., V/Mn/V(001), (iii) the ordered Mn-V surface ordered alloy, i.e., $(\mathrm{Mn}_{0.5}\mathrm{V}_{0.5})/\mathrm{V}(001)$, (iv) the buried alloy $\mathrm{V}/(\mathrm{Mn}_{0.5}\mathrm{V}_{0.5})/\mathrm{V}(001)$ (v) the 2-ML surface ordered alloy $(\mathrm{Mn}_{0.5}\mathrm{V}_{0.5})_2/\mathrm{V}(001)$. For systems with an equal numbers of V and Mn atoms in the unit cell the computed energies are directly comparable. Difficulties arise when the numbers of V and Mn atoms are inequivalent. This happens, for example, in the presence of 1-ML-thick ordered alloy, in either the surface or buried. To determine the formation energy in these specific cases, we follow Blügel formula.[13] For example, for the stability of the surface ordered alloy against phase separation this formula gives

$$\Delta E=E_{\mathrm{Mn\text{-}V/V(001)}}-(0.5)(E_{\mathrm{Mn}/V(001)}+E_{\mathrm{V/V(001)}}), \qquad (1)$$

where $E_{\mathrm{Mn\text{-}V/V(001)}}$, $E_{\mathrm{Mn/V(001)}}$, and $E_{\mathrm{V/V(001)}}$ are the total energies of the Mn-V surface alloy on the V(001), Mn monolayer on the V(001) substrate, and V on V(001), respectively.

For the stability of the buried alloy the formula is

$$\Delta E=E_{\mathrm{V/Mn\text{-}V/V(001)}}-(0.5)(E_{\mathrm{V/Mn/V(001)}}+E_{\mathrm{V/V(001)}}), \qquad (2)$$

where $E_{\mathrm{V/Mn\text{-}V/V(001)}}$, $E_{\mathrm{V/Mn/V(001)}}$, and $E_{\mathrm{V/V(001)}}$ are total energies of the V/Mn-V/V(001) buried alloy in V(001), Mn buried monolayer in the V(001) substrate, and the clean V in (001) orientation, respectively.

We have also studied the possibility of bilayer formation against 1 ML of Mn comparing the total energy of Mn ML on V(001) with a bilayer covering 50% of the V(001) surface and leaving 50% uncovered:

$$\Delta E=E_{\mathrm{Mn/V(001)}}-(0.5)(E_{\mathrm{Mn/Mn/V(001)}}+E_{\mathrm{V(001)}}), \qquad (3)$$

where $E_{\mathrm{Mn/Mn/V(001)}}$ is the total energy of the bilayer Mn on V(001).

Our paper is organized as follows: Sec. II provides a brief account of the theoretical model used for calculations; Sec. III discusses the nonmagnetic case; Sec. IV details the spin-polarized calculations; Sec. V sums up the conclusions reached in the analysis.

II. A BRIEF OUTLINE OF THE METHOD

Our results were obtained with a scalar-relativistic version of the TB-LMTO-ASA method. First of all, we determined the lattice parameter for bcc bulk V by total-energy minimization using different functionals: the local spin-density approximation LSDA of von Barth–Hedin (LSDA-vBH),[46] the LSDA of Vosko-Wilk-Nusair (LSDA-VWN),[47] the GGA-LMH,[42] and the GGA-PW91.[43] The results of Table I show clearly that GGA-LMH fits well the experimental lattice parameter[48] because it is only 0.35% lower, whereas the GGA-PW91 is 1.75% higher than the experimental lattice parameter.

Even if the GGA-LMH functional fits better the lattice parameter we have used the GGA-PW91 in our calculations because it leads to a very improved description of the structural and magnetic properties of Mn.[49] In our attempt to study the system we have considered the lattice parameter obtained with the two functionals. We have found that the magnetic properties are not affected significantly in this interval of lattice parameters, so we have carried out our calculations by using the experimental parameter.[48]

TABLE I. Lattice parameter (in a.u.) of V bulk calculated with different functionals of the exchange correlation energy compared to the experimental one.

	a_{exp} (Ref. 48)	LDA (VBH)	LDA (VWN)	GGA (LMH)	GGA (PW91)
a_0 (u.a)	5.7	5.61	5.62	5.68	5.80

In the present calculations, we modeled the structure of the samples by using seven layers of V(001) as a substrate. This approximation is appropriate because the results do not change when considering thicker V slabs in our calculations. We also assumed an epitaxial growth of 1–3 ML of Mn by considering the vanadium lattice parameter. We added to the supercell 5 ML of empty spheres to assure a cutoff interaction between supercells. The calculations were performed by using 400 k points in the irreducible Brillouin zone.

III. NONMAGNETIC CASE

In this section we present non-spin-polarized calculations performed for Mn layers on the V(001) substrate in order to check the stability of the Mn/V interfaces in a nonmagnetic approach. We consider different structural configurations of alloys in the range of 1 ML of Mn on V(001) in order to determine their relative stabilities. Using GGA-PW91, we find that when we go from a Mn ML towards the buried Mn ML and towards the 2-ML surface alloy we gain, respectively, 10.05 mRy/atom and 3.96 mRy/atom. The formation energies of the 1-ML surface alloy and the buried alloy are, respectively, 0.27 mRy/atom and -0.84 mRy/atom (Table II, column 2). This means that the latter is more stable than the buried Mn ML, whereas the former is instable with respect to the Mn overlayer.

We also found that Mn ML is instable compared to a bilayer formation. Indeed, the formation energy according to the formula cited above is 1.16 mRy/atom (Table II, column 2). As a main result of the present energetic study, we conclude that the 1-ML surface alloy is instable against interdiffusion.

TABLE II. Formation energies in nonmagnetic and magnetic cases (in mRy/atom) for the surface alloy Mn-V/V(001), buried alloy V/Mn-V/V(001) against phase separation and for a monolayer against a bilayer formation ($\Delta E<0$ means energetically favored).

System	Formation energy nonmagnetic	Formation energy magnetic
Mn-V/V(001)	0.27	0.86
V/Mn-V/V(001)	-0.84	-0.15
Mn/V(001)	1.16	-0.46

The idea of this non-spin-polarized calculation is to check the effect of magnetism on the stabilization of the various structural and chemical configurations.

IV. SPIN-POLARIZED CALCULATIONS

In the foregoing section we consider different magnetic phases as input for the chemical configurations. In the range of one Mn monolayer on V(001), we perform the same study as in the nonmagnetic case by considering the energy of the most stable magnetic state of each structure. We studied all the magnetic phases with and without polarization of the interface vanadium in the case of one, two, and three Mn monolayers on V(001). All the chemical structures under investigation are found to be magnetic.

To simplify the notations, we note the polarization of the ith plane by $[\uparrow]_i$ or $[\downarrow]_i$ with $i=S,\ S-1,\ S-2,\ I,\ I-1$, where S represents the surface atoms and I is the V interface layer.

The LAF solution stands for the layered antiferromagnetic coupling between the ferromagnetic layers going from the surface to the interface regardless of the atoms chemical type ($[\uparrow]_S[\downarrow]_{S-1}\cdots[\uparrow]_{S-3}/[\downarrow]_I[\uparrow]_{I-1}$), whereas $c(2\times2)$ corresponds to an in-plane AF coupling between atoms of the surface layer S.

A. 1 Mn ML on V(001)

For one Mn ML on V(001), the only converged solutions that we obtained are $c(2\times2)$ and the layered antiferromag-

TABLE III. The table shows the magnetic configurations for 1, 2, 3 ML's of Mn on V(001) obtained (second column). The third to the sixth column indicate the magnetic moments (in μ_B) on Mn atoms and V at the interface (I). The difference in energies (in mRy/atom) of the different solutions vs the ground state (noted 0.00) are reported in the eighth column. (Mn$_1$) and (Mn$_2$) represent the two inequivalent atoms in a plane.

Mn coverage	Magnetic configuration	μ_s	μ_{s-1}	μ_{s-2}	μ_I	μ_{I-1}	Energy
1 ML	$[\uparrow]_S/[\downarrow]_I[\uparrow]_{I-1}$	3.33			1.02	0.04	0.00
	$c(2\times2)$	3.26			0.00	-0.05	5.94
2 ML	$[\uparrow]_S[\downarrow]_{S-1}/[\uparrow]_I[\downarrow]_{I-1}$	3.89	-1.10		0.41	-0.13	0.00
	$c(2\times2)$	3.84	0.00		0.00	0.00	1.29
3 ML	$[\uparrow]_S[\downarrow]_{S-1}[\uparrow]_{S-2}/[\downarrow]_I[\uparrow]_{I-1}$	3.96	-2.59	2.00	-0.74	0.09	0.00
	$[\uparrow\downarrow]_S[\uparrow]_{S-1}[\downarrow]_{S-2}/[\uparrow]_I[\downarrow]_{I-1}$	-4.11 3.84 (Mn$_1$) (Mn$_2$)	2.09	-1.83 -1.30 (Mn$_1$) (Mn$_2$)	0.62	-0.05	0.24
	$[\uparrow]_S[\uparrow]_{S-1}[\downarrow]_{S-2}/[\uparrow]_I[\downarrow]_{I-1}$	3.86	2.49	-1.70	0.62	0.03	0.9
	$c(2\times2)$	$\pm$ 3.89	0.00	$\pm$ 0.69	0.00	0.00	1.59

TABLE IV. Magnetic moments (in μ_B) for the different alloy configurations. In parentheses (Mn), (V) indicate one of the two inequivalent atoms in the plane.

Magnetic moment	$Mn_{0.5}V_{0.5}/V(001)$ LAF	$V/Mn_{0.5}V_{0.5}/V(001)$ LAF	V/Mn/V(001) LAF	2-ML $Mn_{0.5}V_{0.5}/V(001)$ LAF
μ_s	1.41(V) 3.25(Mn)	−1.39(V)	2.15(V)	1.6(V) 3.61(Mn)
μ_I	−0.52(V)	0.86(Mn) 0.45(V)	−1.39(Mn)	−0.82(V) −0.91(Mn)
μ_{I-1}	$-0.21(V_1)$ $-0.03(V_2)$	0.00	0.41(V)	$0.02(V_1)$ $0.09(V_2)$

netic structure LAF. The results are shown in Table III. The LAF is found to be the ground state. The magnetic moment of the Mn atom at the surface is $3.33\mu_B$; the reduction of the coordination number at the surface favors this relatively high magnetic moment. We note also a sizable induced magnetic moment on V atoms at the interface ($1.02\mu_B$). The metastable $c(2\times 2)$ solution is found at 5.94 mRy/atom higher in energy.

Then we investigated the $Mn_{0.5}V_{0.5}$ surface ordered alloy one layer thick on V(001). The converged magnetic solution is LAF. The results are reported in Table IV. The magnetic moment of Mn surface atoms is equal to $3.25\mu_B$, which represents a reduction of 2.4% compared to the overlayer case. This decrease can be explained by an interaction with V second-nearest-neighbors. The induced magnetic moment on V surface atoms is slightly increased up to $1.41\mu_B$. Hamad *et al.*[50] studied recently the magnetic properties of $Cr_x(Mn_x)V_{1-x}$ and Cr_xMn_{1-x} alloys on V(001) with different concentrations ($x=0.25$, 0.50, and 0.75) using a self-consistent real-space tight-binding method in the Hartree-Fock approximation of the Hubbard Hamiltonian. These authors found an AF coupling between the Mn surface atoms and the interface V atoms. The net surface magnetization increases with Cr(Mn) concentration in the $Cr_x(Mn_x)V_{1-x}/V(001)$ system, whereas no variations arises in the $Cr_xMn_{1-x}/V(001)$ system. In the case of an ordered surface alloy $Mn_{0.5}V_{0.5}$ on the V(001) substrate, they have found LAF behavior with a moment of $3.33\mu_B$ for Mn surface atoms and $1.44\mu_B$ induced magnetic moment on V surface atoms. Our results confirm their findings.

As far as the buried Mn ML, i.e., V/Mn/V(001), is concerned, the only magnetic configuration that converged is the LAF solution (Table IV shows that). The effect of the surface and the Mn atoms polarization enhances the magnetic moments of the V atoms at the surface ($\sim 2\mu_B$). At the same time, the increasing number of the V nearest-neighbor atoms favors the hybridization. The latter decreases strongly the Mn magnetic moment which reaches $1.39\mu_B$. It is important to notice that vanadium tends to kill magnetism.

In the case of a buried ordered alloy, i.e., $V/(Mn_{0.5}V_{0.5})/V(001)$, the only converged solution is LAF as shown in Table IV. We notice here that, on the one hand, the magnetic moment of Mn atoms decreases drastically down to $0.86\mu_B$, which represents a reduction of 74% as compared to the magnetic moment in the case of the surface layer. This can be related to the hybridization with larger number of vanadium first-nearest-neighboring atoms. On the other hand, the V moment at the surface shows an increase up to $1.39\mu_B$ which is probably due to both surface effect and Mn polarization.

Finally, for the 2-ML surface ordered alloy, i.e., $(Mn_{0.5}V_{0.5})_2/V(001)$ all the input magnetic configurations converged to the LAF where the surface and subsurface layers are antiferromagnetically coupled as shown in Table IV. Surprisingly, the magnetic moment of Mn at the surface is higher ($3.61\mu_B$) than in all the other systems that we considered till now. Mn atoms at the surface have two nearest-neighbor Mn atoms (less interaction with V atoms); this may explain the result obtained.

From an energetic point of view, we summarize the spin-polarized calculations by retaining only the stable magnetic configurations. We find that, going from a Mn ML towards the buried ML and towards the 2-ML alloy we, respectively, gain 1.70 mRy/atom and 0.41 mRy/atom. The formation energy of the surface alloy is 0.86 mRy/atom, which means that the latter is instable against the phase separation, i.e., the clean V/V(001) and a ML of Mn on V(001). Contrary to the surface alloy, the formation of the buried alloy is favored (0.15 mRy/atom) as regards the clean V(001) and the buried Mn ML (Table II, column 3).

The conclusions here are the same as those in the nonmagnetic calculations. The formation energy for the surface alloy increases moving from the paramagnetic case to the spin-polarized one. Thus, the main result of this part of the work is that the magnetism in this system plays an important role in the segregation of Mn atoms on V(001). It is probably due to the high magnetic moments observed on Mn atoms when it has more Mn neighbor atoms.

We have compared the LDOS of a homogenous ML [Fig. 1(a)] with that of some different alloyed configurations [Figs. 1(b)–1(d)]. There are two aspects of the LDOS that must be noticed.

First, the presence of a pronounced minimum in the middle of the majority *d* band between the bonding and antibonding states in the case of the perfect Mn overlayer. This minimum is absent for the alloyed configurations. We discuss this result in terms of surface spins taking into account only first-nearest neighbors (NN's). Hence, we attribute this pronounced minimum to the Mn-Mn 3*d* hybridization of the LAF magnetic configuration with spins of the same sign lying at the surface plane. In the in-plane AF configuration $c(2\times 2)$, one Mn atom has some first NN's spins at the surface plane of the opposite sign and next NN's spins having same sign, and thus, the Mn-Mn 3*d* overlap of the same

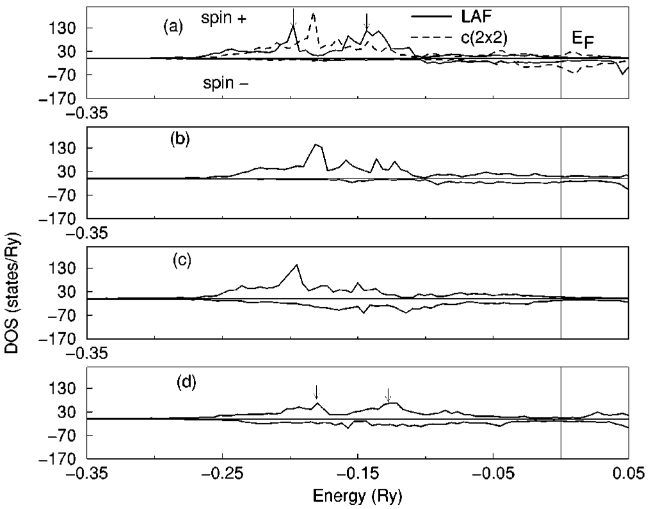

FIG. 1. LDOS of Mn atom for (a) monolayer Mn on V(001), (b) the surface alloy (Mn-V)/V(001), (c) 2-ML surface alloy $(\text{Mn-V})_2/\text{V}(001)$, (d) the buried Mn layer V/Mn/V(001). Full line represents the ground-state solution (LAF) and the metastable solution $c(2\times2)$ is given in dashed line. The arrows indicate the two main peaks of the spin-up band in the ground state.

sign spins is weak. This could explain the disappearance of the minimum in the LDOS in this case [Fig. 1(a)].

Second, we observe in the cases of different alloyed configurations [Fig. 1(b)–1(d)], intensities at the energy position of the main peaks but for the opposite spin direction. We attribute these intensities to the effect of hybridization between orbitals of Mn atoms at the surface and atoms at the subsurface of the opposite sign. These intensities are more pronounced for the case of 2-ML alloy, i.e., $(\text{Mn}_{0.5}\text{V}_{0.5})_2/\text{V}(001)$ [Fig. 1(c)] related to the existence of first NN's Mn atoms at the subsurface. This is contrary to what happens when we have only V nearest neighbors at the subsurface as in the case of $\text{Mn}_{0.5}\text{V}_{0.5}/\text{V}(001)$ [Fig. 1(b)] where these intensities are not significant. The LDOS of Mn atoms in the case of the buried Mn ML displayed in Fig. 1(d) shows two main peaks in majority band due to the effect of Mn atoms in the plane, and the additional main peaks for opposite spin due to the strong hybridization with the 8 V NN's.

B. 2 and 3 ML on V(001)

In this section we consider the magnetic map of 2 and 3 ML's of Mn. For the $(\text{Mn})_2/\text{V}(001)$ system we obtained two converged solutions: the ground-state magnetic configuration consists of an antiparallel coupling between the surface S, subsurface $S-1$, and the subsubsurface $(S-2)$ planes and the metastable solution $c(2\times2)$ which is 1.29 mRy/atom high in energy (Table III). The magnetic moment at the surface is now higher $(3.89\mu_B)$ and it is 17% enhanced as compared to the case of Mn/V(001). This is due to the large number of Mn neighbors. Using Eq. (3), we studied the possible formation of a bilayer against a monolayer, and we found that the magnetism inverts the sign of the formation energy $(-0.46$ mRy/atom$)$. This means that the monolayer is more stable. The same behavior was found by Asada *et al.*[51] in Mn films on Fe(001).

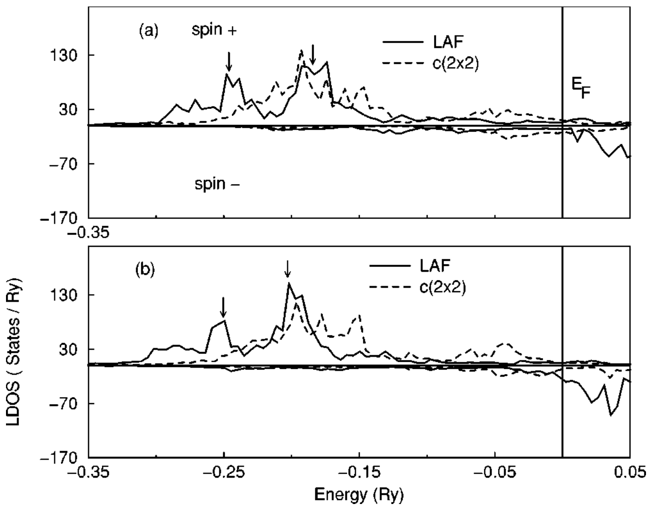

FIG. 2. LDOS at the surface layer for (a) two layers of Mn on V(001), (b) three layers of Mn on V(001). Full line represents the layer AF ground-state solution (LAF) and dashed line the metastable solution $c(2\times2)$. The arrows indicate the two main peaks of the spin-up band.

For $(\text{Mn})_3/\text{V}(001)$ we obtained four solutions summarized in Table III.

(i) The ground state (LAF) corresponds to an antiparallel coupling between Mn atoms in successive planes $([\uparrow]_s[\downarrow]_{s-1}[\uparrow]_{s-2}/[\downarrow]_I[\uparrow]_{I-1})$.

(ii) The configuration $([\uparrow\downarrow]_s[\uparrow]_{s-1}[\downarrow]_{s-2}/[\uparrow]_I[\downarrow]_{I-1})$ is 0.24 mRy/atom higher in energy than the ground state.

(iii) The configuration $([\uparrow]_s[\uparrow]_{s-1}[\downarrow]_{s-2}/[\uparrow]_I[\downarrow]_{I-1})$ is 0.9 mRy/atom higher in energy than the ground state.

(iv) The $c(2\times2)$ is 1.59 mRy/atom higher in energy than the ground state.

It is worth mentioning that the interfacial coupling is always of antiferromagnetic-type between nearest-neighboring atoms irrespective of their chemical type. This holds true for all the chemical structures studied in this work.

Compared to the magnetic moment obtained in the case of one Mn $(3.33\mu_B)$ and two Mn $(3.89\mu_B)$ ML's on V(001), the magnetic moment of an Mn atom at the surface layer for the ground state is, respectively, 19% and 2% higher.

Figures 2(a) and 2(b) display the LDOS of 2 and 3 ML's of Mn on V(001) for, respectively, LAF configuration and the in-plane AF configuration. As in the case of 1 ML, the hybridization between Mn atoms belonging to the surface plane presents a minimum. Besides, for the metastable configuration $c(2\times2)$ as it was pointed out for the overlayer, the weak overlap between the Mn atoms with the same moment's sign are next NN's at the surface and thus the minimum disappears.

V. CONCLUSION

We have presented *ab initio* calculation of magnetic properties of 1–3 ML of Mn on a V(001) substrate. We also studied the possible stabilization of ordered alloys in the case of 1 ML of Mn on V(001). The main results of our work could be summarized as follows.

(i) The sign of the formation energy does not change when going from nonmagnetic to spin-polarized calculations. The magnetic energy gained is not sufficient to stabilize the surface ordered alloy despite the high magnetic moment obtained. However, the formation energy increases going from the paramagnetic to the magnetic case, which favors the segregation of Mn atoms onto the vanadium substrates. This is mainly due to the increase of the spin polarization of the Mn atom when it is surrounded by more Mn atoms. We also found that a ML formation is favored over a bilayer formation, where magnetism changes the sign of the formation energy.

(ii) The Mn atoms induce sizable magnetic moments in V and lead to layered AF coupling in all the structural configurations with a high magnetic moment of Mn of the order $\sim 3\mu_B$. The interfacial Mn/V coupling is antiferromagnetic in all cases. The $c(2\times 2)$ configuration is found instable as in the case of Mn-Cr systems,[34,33] in contrast with different studies of Mn on noble metals. Indeed, the general trend is that the deposition of $3d$ metals on (001) noble metals prefers a $c(2\times 2)$ magnetic structure for V, Cr, and Mn and a ferromagnetic structure for Fe, Co, and Ni.[26] On the other hand, the deposition on (001) magnetic metals shows a layered antiferromagnetic coupling for V, Cr, Mn and a ferromagnetic coupling for Fe, Co, and Ni as found by Asada *et al.*[51] for $3d$ metals on Fe(001) and by Khalifeh *et al.*[34] for Mn on Cr(001).

(iii) The magnetic moment of Mn at the surface, at least up to 3 Mn ML's, increases with the number of Mn overlayers. The Mn atom with many V atoms in its neighborhood has strong tendency to decrease its magnetic moment. When we go away from the V film, it is clear therefore that Mn can increase its magnetic moment. This trend can be explained through the LDOS of the different chemical and magnetic structures.

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