# Spin polarization of Mn layers on Fe(001)

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The spin polarization of Mn layers for a body-centered-tetragonal structure on Fe(001) is studied using a self-consistent tight-binding real-space model within the unrestricted Hartree-Fock approximation to the Hubbard Hamiltonian. We investigate all the possible magnetic structures of Mn overlayers with respect to the Fe majority-spin direction as a function of the exchange integral J of Mn. For an antiparallel alignment of a Mn monolayer with the ferromagnetic Fe substrate, the Mn magnetic moment exhibits a second-order transition from low spin to high spin in a large interval of the exchange integral J  $(0.35 \le J \le 0.85 \text{ eV})$ . The parallel Fe-Mn arrangement exists only for the high spin (J > 0.49 eV), whereas for  $J \le 0.49 \text{ eV}$  this solution undergoes a first-order transition towards the antiparallel Fe-Mn. Interesting cases are obtained for thicker Mn slabs, which show different solutions for all possible interlayer magnetic arrangements of Mn as a function of the interfacial Fe-Mn spin alignment. The Mn shows a tendency to form a layer-by-layer antiferromagnetic structure coupled ferromagnetically with the ferromagnetic Fe substrate.

#### I. INTRODUCTION

Manganese and its alloys manifest a wide variety of structural and magnetic properties. There are complex crystal modifications with many atoms per unit cell, but the simple body-centered cubic (bcc) and face-centered cubic (fcc) structures can only be stabilized at room temperature either by alloying or by epitaxial growth on suitable substrates. Manganese exists in four allotropic modifications exhibiting a complex phase diagram.<sup>1</sup> Up to about 1000 K, crystallographically  $\alpha$ -Mn is bcc with 29 atoms per unit cell and four nonequivalent atomic sites. Between 1000 and 1370 K,  $\beta$ -Mn is simple cubic with two types of atomic sites per unit cell containing 20 atoms. For the next 40 K,  $\gamma$ -Mn is fcc and from there up to the melting point at 1517 K the  $\delta$  phase has the bcc structure. The magnetic and other physical properties are quite different for the various phases and thus strongly depend on the crystal structure. Shull and Wilkinson<sup>2</sup> first established the existence of antiferromagnetism of  $\alpha$ -Mn with Neel temperature ( $T_N = 100$  K) by means of neutron diffraction method. An analysis of the magnetic structure was also made by Kasper and Roberts<sup>3</sup> with the powder sample below  $T_N$ . They showed that the atoms in inequivalent sites have different moments. The actual values of the respective magnetic moments, which were deduced from neutron diffraction data, strongly depend on the form factor chosen in the analysis.<sup>4</sup> A nuclear magnetic resonance study<sup>5</sup> supported these values and derived magnetic moments that are slightly larger than those obtained from neutron data.

The bcc and fcc phases of Mn exist naturally at temperatures that are too high for any type of magnetic order to occur.  $\gamma$ -Mn, for example, is a high temperature

phase, which may also be obtained by rapid quenching and/or alloying with Cu, Ni, Pd, or Fe metals as an antiferromagnet with a magnetic moment of  $2.3\mu_B$ .<sup>6-8</sup> Below the Neel temperature ( $T_N = 540$  K), a large tetragonal distortion occurs with about a 6% contraction. The stability of the antiferromagnetic state in  $\gamma$ -Mn has been confirmed through band-structure calculations.9,10 The microscopic origin of the distortion is explained by Oguchi and Freeman<sup>11</sup> in terms of a directional property of the *d*-band bonding introduced by the antiferromagnetic ordering. At low temperatures, both bcc and fcc phases are predicted to order magnetically if the lattice spacing exceeds some minimum value. Kübler<sup>12</sup> who used the augmented spherical wave (ASW) method<sup>13</sup> has shown that bulk bcc Mn prefers an antiferromagnetic state at lattice parameter a = 3.01 Å with a large magnetic moment. Moruzzi *et al.*<sup>14</sup> have performed total-energy band calculations based on the ASW method for paramagnetic and ferromagnetic states in bcc Mn. The ferromagnetic state undergoes a second-order transition from low spin to nonmagnetic under compression and first-order transition from a low-spin state to high-spin state under expansion. Using a linear combination of the Gaussian orbital method,<sup>15</sup> Fuster et al.<sup>16</sup> obtain similar behavior concerning the ferromagnetic transition in bcc and fcc Mn. The ground state is deduced through the susceptibility calculations as being ferromagnetic for bcc structures in the range 2.75 < a < 2.86 Å and antiferromagnetic for the fcc Mn when a > 3.58 Å. Fujii et al.<sup>17</sup> have performed total-energy band calculation of bcc Mn as a function of the lattice constant from 2.5 to 3.5 Å for paramagnetic, ferromagnetic, and antiferromagnetic states. The calculations were carried out by the linearmuffin-tin-orbital method in the atomic sphere approximation.<sup>18</sup> They have found that bcc Mn undergoes a

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first-order transition from a low spin to a high spin for both the ferromagnetic and the antiferromagnetic states. The total-energy calculations indicate that the bcc Mn prefers a paramagnetic state for a lattice constant less than 2.68 Å (under compression), a low-spin ferromagnetic state between 2.68 and 2.95 Å around the minimum of total energy and a high-spin antiferromagnetic state for a lattice parameter above 2.95 Å (under expansion). It is only recently that Sliwko et al.<sup>19</sup> have reported band theoretical results for the electronic and magnetic structure of  $\alpha$  and  $\beta$  phases, whose complicated structures are unique among all elements. Within the ASW method and collinear spin model, they obtained results which agree fairly well with the experimental interpretation based on canted spins. This suggests that in  $\alpha$ -Mn, the canting angle has a minor influence on the absolute value of the magnetic moment as was already discussed by Stoeffler and Gautier.<sup>20</sup> Density functional theory of noncollinear antiferromagnet  $\gamma$ -FeMn shows that its total energy is minimized<sup>21</sup> in the tetrahedral magnetic structure first proposed by Kouvel and Kasper.<sup>22</sup> Mn atoms can occupy a wide range of atomic volumes and be in a number of different structural configurations (see Table II of Cable and Tsunoda<sup>23</sup>).

Because of its exotic structural and magnetic properties, Mn is an interesting candidate for thin-film growth, as it is expected to accept different local configurations. Experimentally, one may attempt to stabilize normally high-temperature phases of a material by epitaxial growth on a suitable substrate. Arrott and co-workers<sup>24</sup> have studied Mn layers on Ru(0001), Fe(001) and Ni(001) in an effort to expand Mn lattice to produce magnetic moments approaching the Hund's rule limit of  $5\mu_B$ . However, no significant magnetic moment on Mn was depicted through ferromagnetic resonance. Epitaxial Mn layers on Ag(001),<sup>25,26</sup> as well as on Cu(001) (Ref. 26) and Pd(111),<sup>27</sup> grow in distorted face-centered-tetragonal (fct) structure. In these cases, the Mn layers take on the inplane spacing of the square surface net of the substrate and distort out of plane yielding the fct structure. On Ag(001) which has approximately the same square surface net as Fe(001), Jonker et al.<sup>25</sup> have grown singlecrystal Mn films and coherent Mn/Ag superlattices and characterized the magnetic behavior with temperature dependent ferromagnetic resonance and magnetic susceptibility measurements. They detect no significant ferromagnetic contribution at temperature down to 5 K, and assumed that Mn is nonmagnetic or antiferromagnetic. Ounadjela et al.<sup>28</sup> and O'Brien and Tonner<sup>29</sup> have discussed, respectively, the structural changes in metastable epitaxial Co/Mn superlattices and the surfaceenhanced magnetic moment and ferromagnetic ordering of Mn ultrathin films on fcc Co(001). Antiferromagnetic ordering of the Mn spacer with a Neel temperature close to 320 K has been evidenced by Henry and Ounadjela.<sup>30</sup> The electronic structure of ultrathin films of Mn grown on GaAs(001) is found to be very different from bulk  $\alpha$ -Mn<sup>31</sup> and the possibility that magnetic ordering exists in the ultrathin overlayer of Mn is proposed. Fisher et al.<sup>32</sup> have recently reported on Mn/Ir(111) superlattices and thin overlayers of Mn and FeMn on Ir(100). Lowtemperature superconducting quantum interference device (SQUID) measurements display different type of magnetization depending on the crystallographic face of Ir substrate and on the concentration of Mn in FeMn. Blügel *et al.*<sup>33</sup> and Blügel and Dederichs<sup>34</sup> have reported on a systematic study, based on the full-potential linearized augmented plane wave (FLAPW) method, for 3*d* transition metals as monolayers on the Pd and Ag surfaces, respectively. Magnetic moments of about  $4\mu_B$  have been obtained in the case of Mn overlayer. Both ferromagnetic  $p(1\times1)$  and antiferromagnetic  $c(2\times2)$  configurations have been explained and similar magnetic moments obtained. However, the antiferromagnetic configuration appears to be stable in agreement with re-

configuration appears to be stable in agreement with recent photoemission experiments.<sup>35</sup> Ultrathin films of Mn on Cu(001) and Ni(001) have received much attention recently, due to the existence of stable alloys which form at 1/2 ML coverage.<sup>36-41</sup> These surface alloys form at room temperature and display a high-spin moment.<sup>40,41</sup> Epitaxial Mn layers have been deposited by Arrott and

Epitaxial Mn layers have been deposited by Arrott and co-workers on Fe(001).<sup>24,42</sup> A careful search by Squid magnetometry and ferromagnetic resonance revealed no detectable magnetic moment and Mn was assumed to be either nonmagnetic or antiferromagnetic. Later on, the growth and structure of the Mn films were monitored by reflection high energy electron diffraction and low energy electron diffraction.<sup>43</sup> A Mn wedge shape was formed by slowly moving a shutter across the Fe(001) whisker during evaporation. Electron diffusion showed that Mn grows monolayer by monolayer in a bct structure (a=2.87 Å, c=3.27 Å). The measured atomic volume is 13.44 Å<sup>3</sup>, which is significantly larger than for all phases of bulk Mn. The ability of Mn to form tetragonal structures may be the reason for the relative stability of the bct overlayers.<sup>11</sup> The interlayer exchange coupling between Fe layers separated by Mn is shown to be antiferromagnetic (AF) for interlayer thicknesses above 4 ML. For thickness above 7 ML a 2-ML oscillation in the AF exchange coupling is depicted. To investigate the origin of this oscillating coupling, ASW calculations were performed with the experimental values of the lattice parameters<sup>43</sup> for a Fe<sub>5</sub>/Mn<sub>7</sub>(001) multilayer. Parallel (P) and antiparallel (AP) configurations have been considered at the Fe/Mn interface. The P configuration was found to be more stable in contrast to the Fe/Cr situation where the arrangement at the interface is found to be AP.<sup>20,44</sup> For both solutions, the Mn spacer displays an AF ground state with large moments. More recently, Walker and Hopster<sup>45</sup> have performed measurements on the magnetic state of epitaxial overlayers of Mn grown on Fe(001) through spin-polarized electron energy loss spectroscopy (SPEELS). Nonzero exchange asymmetries were found, demonstrating that the surface layer of Mn overlayers has a net magnetic moment. For thicknesses above 5 ML, the exchange asymmetry oscillates with a period of about two atomic layers proving that the Mn forms ferromagnetic (100) sheets and that these sheets align antiferromagnetically. Their conclusion appears in agreement with the recent calculations.<sup>20,43,46</sup> Thus, there appears a clear contradiction with the experimental studies of Purcell et al.,43 who observed only AF interlayer exchange coupling above 7 ML thickness, with oscillatory modulations in the coupling strength.

In order to shed some light on this contradiction, we have studied the magnetic order and the critical behavior of bct Mn on Fe(001) with the measured parameters of Purcell et al.<sup>43</sup> Both P and AP arrangements at the Fe/Mn interfaces, together with F or AF configurations of the Mn film were considered. The magnetic order is derived within a self-consistent tight-binding real-space model within the unrestricted Hartree-Fock approximation to the Hubbard Hamiltonian. This method gives a good description of the itinerant d electrons of transition metals elements of the middle of the first row.<sup>47</sup> For the elements like Fe and Mn, the effect of the sp electron in the magnetic moments do not exceed 10%.<sup>48</sup> This model has been extensively used to study the electronic struc-tures of multilayers,<sup>49</sup> overlayers,<sup>50,51</sup> vicinal surface<sup>51</sup> and small clusters<sup>52</sup> on Ag(001). The rest of the paper is organized as follows. In Sec. II, we present briefly the theoretical model used for the calculation of both bcc Mn and bct Mn overlayers on Fe(001). The calculation is performed versus J, the many-body Hubbard-like parameter representing the intrasite exchange interactions. Results are discussed in detail in Sec. III. We start with a calculation of spin polarization in bcc Mn versus J in order to compare our results with those of Fujii et al.<sup>17</sup> who studied the effect of volume change on the stability of the magnetic ground state. Then we study the bct bulk Mn with the parameters measured by Purcell et  $al.^{43}$  We continue with 1-3 ML of Mn on Fe(001). We investigate all available solutions (stable and metastable) in order to find out if a possible discrepancy between Purcell et al. and Walker and Hopster may be related to a stabilization of a metastable solution by specific growth conditions. In the conclusion reported in the Sec. IV, we discuss the extension to rough interfaces as done previously at the Fe/Cr interface.<sup>53,54</sup>

### **II. CALCULATION MODEL**

Basically, the calculation model deals with *d*-band tight-binding scheme in the unrestricted Hartree-Fock approximation of the Hubbard Hamiltonian.<sup>47</sup> We assume spin-independent canonical hopping integrals up to the second neighbors varying as the inverse of the fifth power of the interatomic distance  $R_{ij}$  between the *i*th and the *j*th neighbor atoms:<sup>55</sup>

$$dd (\sigma, \pi, \delta)_{ij} = (6, -4, 1) dd \delta_b (R_b / R_{ij})^5 , \qquad (1)$$

where  $R_b$  is the corresponding distance in the equilibrium bulk. The factor (6, -4, 1) indicates that  $dd\sigma(dd\pi)$  is 6 (-4) times  $dd\delta$ . The lattice parameters of Mn are chosen according to the experimental results of Purcell,<sup>43</sup> who found that Mn grew in bct structure on Fe(001) with in-plane spacing a=2.87 Å and the out-of-plane spacing c=3.27 Å (c/a=1.14). At the Fe/Mn interface, the two-center hopping integrals  $ddk_{\text{FeMn}}$  [ $k = (\sigma, \pi, \delta)$ ], are deduced from the bandwidths of the bulk transition metals for FeFe and MnMn pairs and from the Shiba<sup>56</sup> prescription:

$$ddk_{\rm FeMn} = [ddk_{\rm FeFe} ddk_{\rm MnMn}]^{1/2}$$
<sup>(2)</sup>

for FeMn pairs. The Fe-Mn distance is chosen as the mean value between a and c: this leads to an increase of 7% of the distance between Mn and Fe planes at the interface as compared to the lattice spacing of bulk Fe. This value is in reasonable agreement with recent FLAPW calculations.<sup>61</sup> The diagonal terms of the Hamiltonian are spin dependent and expressed, within local charge neutrality on each site *i*, by

$$E_{i\sigma} = E_i^0 + V_i - \sigma \frac{J_i}{2} M_i , \qquad (3)$$

where  $\sigma$  is the spin index and  $E_i^0$  the *d*-energy level in the paramagnetic state of the bulk corresponding to the element at site *i*. The site-dependent term  $V_i$  is the potential that assures the local charge neutrality. The potential is adjusted self-consistently with the requirement that the electronic occupation  $q_i$  of each atom is the same as in the corresponding bulk. The local magnetic moment is obtained from the difference between the number of electrons of spin up and spin down:  $M_i = N_{i\uparrow} - N_{i\downarrow}$ , where  $N_{i\sigma}$  is given by

$$N_{i\sigma} = \int_{-\infty}^{E_F} n_{i\sigma}(E) dE \quad . \tag{4}$$

The spin-polarized density of state  $n_{i\sigma}$  is calculated by means of the recursion method applied in the real space.<sup>57</sup> The term  $J_i$  stands for the shifts due to intraatomic exchange interactions. In the present work, the calculations are performed with a fixed exchange integral J of Fe fitted in order to reproduce its bulk magnetic moment  $(2.21\mu_B)$ . Such procedure cannot be applied for Mn, because of its particular bct structure on Fe(001),<sup>43</sup> which does not exist in the bulk form. *Ab initio* calculations for bcc Mn<sup>58</sup> give a value of J=0.75 eV, but with some uncertainties due to the correlation effects in the spin-polarized local-spin-density-approximation (SP-LSDA) calculations, as shown by Stollhoff.<sup>59</sup> Therefore, we study the magnetic moments as function of  $J_{Mn}$  as we have done in our previous calculations for V slabs,<sup>50</sup> Voverlayers on Fe,<sup>52</sup> Rh islands on Ag,<sup>52</sup> and for bulk Mn by Süss and Krey.<sup>60</sup>

The relative stability of the different magnetic configurations investigated here are deduced from the total-energy expression  $E_T$  given by

$$E_T = \sum_{i} \left[ \int_{-\infty}^{E_f} E\left\{ \sum_{\sigma} n_{i\sigma}(E) \right\} dE - q_i V_i + \frac{1}{4} J_i M_i^2 \right].$$
(5)

# **III. RESULTS**

Before computing the spin polarization of Mn layers on Fe(001) versus exchange integral J, we have determined the band structure of bcc Mn. This has been done in order to compare with total-energy band calculations of bcc Mn as a function of lattice constant of Fujii *et al.*<sup>17</sup> They have carried out the total-energy band calculations as a function of lattice parameter for paramagnetic, ferromagnetic, and antiferromagnetic states. For a = 2.68 Å, a first-order transition—from a paramagnetic to a low-spin ferromagnetic—is obtained. This F state is stable until a=2.95 Å, where a first-order transition—from low-spin ferromagnetic to high-spin antiferromagnetic—appears. We have reported on Fig. 1(a) the F and AF behavior occurring in bcc Mn as a function of the exchange integral J. The F magnetic state appears for a value of J slightly smaller than one at which the AF state appears. At J=0.54 eV, a second-order transition from a P to F (with a low spin) takes place followed by a first-order transition from F to AF at J=0.60 eV. If we consider only the ground state [Fig. 1(b)], it can be noticed that our model gives the same transitions

(versus J) as those obtained by Fujii *et al.*<sup>17</sup> (versus *a*). For  $J_{Mn} = 0.75 \text{ eV}$ ,<sup>58</sup> the difference between F and AF states is 0.15 eV per atom.

For the bct configuration with the parameter measured by Purcell *et al.*,<sup>43</sup> the results are drastically different [Figs. 1(c) and 1(d)]. A first-order transition between *P* and AF appears for a *J* value of 0.48 eV, whereas the *F* state appears for J = 0.53 eV [Fig. 1(c)]. The AF state is now the ground state above J = 0.48 eV, in agreement with similar findings by Oguchi and Freeman.<sup>11</sup> We have also investigated the effect of different lattice parameters. For c/a = 1.11 [Fig. 1(e)], both AF and *F* phases appear



FIG. 1. Magnetic moment per atom (a) and total energy (b) in terms of the exchange integral J for the ferromagnetic (F) and the antiferromagnetic (AF) states in bulk bcc Mn. (c) and (d) represent, respectively, the same quantities for bct Mn with c/a = 1.14. Magnetic moments per atom versus J for F and AF phases in bulk bct Mn, when c/a = 1.11 (e) and c/a = 1.18 (f).

for J=0.5 eV, the ground state being clearly AF for J>0.55 eV. For c/a=1.18 [Fig. 1(f)], the ground state is definitively AF for any value of J>0.46 eV. From this study, we can conclude that for bulk Mn, tetragonal distortion stabilizes the AF phase.

The magnetic polarization map obtained in the case of a Mn monolayer on a Fe(001) semi-infinite substrate for the antiparallel (AP) and the parallel (P) Fe-Mn configurations are given, respectively, on Figs. 2 and 3. We report local magnetic moments as function of the exchange integral J of Mn on the Mn layer and on the two nearest neighbors Fe layers. In the case of an antiparallel Fe-Mn configuration (Fig. 2), the calculated magnetic moments on Mn layer undergo a second-order transition from low spin to high spin in the range  $0.35 \le J \le 0.85$ eV. The polarization of the Mn layer with P Fe-Mn configuration (Fig. 3) exists only in the high-spin region (J > 0.49 eV). We observe a spin inversion on the Mn atom towards the AP solution for the values of the exchange integral J < 0.49 eV. In the high-spin region, the calculated magnetic moments on the Mn layer are slightly higher for the P than for the AP Fe-Mn.

For the AP configuration (Fig. 2) there are two distinct regions on both sides of the critical point  $J_c(AP)=0.52$ eV and  $M_c(AP)=2.28\mu_B$ . On both sides of this particular point, the absolute values of the lower  $[M < M_c(AP)]$  and the higher  $[M > M_c(AP)]$  magnetic moments have similar mathematical behavior:  $|M - M_c(AP)| \propto |J - J_c(AP)|^{\alpha}$ , but with different critical exponents  $\alpha$ , respectively, equal to 0.2 and 0.4. In the case of the P Fe-Mn configuration



FIG. 2. Magnetic moment per atom of Mn surface layer, Fe interface layer Fe(i), and Fe(i-1) layer in terms of the exchange integral J of Mn, in the case of one Mn monolayer aligned antiparallel to a ferromagnetic Fe(001) semi-infinite substrate.



FIG. 3. Magnetic moment per atom of Mn surface layer, Fe interface layer [Fe(i)], and Fe(i-1) layer, in terms of the exchange integral J of Mn, in the case of one Mn monolayer aligned parallel to a ferromagnetic Fe(001) semi-infinite substrate.

(Fig. 3), which appears only for J greater than 0.49 eV, the curvature towards the high-spin follows the same power law as obtained for the AP alignment with a critical exponent  $\alpha = 0.3$ . However, we observe a remarkable difference on the Fe polarization. In the case of AP configuration (Fig. 2), the magnetic moment on the second iron layer [Fe(i-1)] is slightly increased as compared to the bulk value, but remains nearly constant with the variation of J. The atom at the interface (Fe(i))presents a smaller and decreasing magnetic moment as the exchange integral increases from the critical point. The P configuration at the interface Fe-Mn (Fig. 3) shows a totally different effect on the Fe polarization: the Fe moment at the interface increases with J, whereas its nearest neighboring Fe atom has a decreasing magnetic moment.

For J=0.75 eV,<sup>58</sup> the *P* configuration between Fe and Mn is energetically favored (0.04 eV per atom) in agreement with FLAPW calculations.<sup>61</sup> Let us mention also that in Ref. 61, the calculated total energy versus Fe-Mn distance shows that the ground state for the AP configuration is obtained for a Fe-Mn distance equal to the lattice parameter of pure Fe, whereas the ground state for *P* configuration is obtained for an increase of the interfacial Fe-Mn distance of 6%. This is in reasonable agreement with the fact that we have chosen for the Fe-Mn distance a mean value between those in Fe and Mn (in the bct configuration).

Interesting results are obtained when the Mn film thickness is greater than 1 ML. Basically, we expect re-

sults qualitatively similar to those obtained experimentally by many authors<sup>62,63</sup> on Cr/Fe(001). However, if both materials Mn and Cr have a net tendency to antiferromagnetism, the difference comes from the particular structure (bct) of Mn on Fe(001), which permits high magnetic moments on Mn atoms. When Mn is alloyed with small amounts of Fe<sup>8</sup> and quenched to room temperature, it forms a fct lattice with a very large magnetic moment of  $2.3\mu_B$  compared to only  $0.2\mu_B$  for Cr. Let us notice that the local moment of the Mn impurity in Fe has theoretically<sup>64</sup> and experimentally<sup>65</sup> the same alignment as that of Fe, whereas the Cr impurity exhibits an antiparallel alignment.<sup>64</sup> However, no general rule can be derived from the fact that parallel configuration between Fe and Mn is stable in the case of Mn as impurity in Fe and for Mn films on Fe(001). In the particular case of Cr, AP alignment is obtained in the case of Cr impurity in Fe and for Cr overlayer on Fe(001), but P alignment is obtained in the case of Fe-Cr compounds.<sup>66</sup> This leads us to investigate all the possible solutions of the interlayer Mn configuration with respect to Fe majority-spin direction.

Figure 4 shows the calculated magnetic moments on 2Mn/Fe(001) system for antiparallel Fe-Mn configuration and layer-by-layer antiferromagnetic structure in Mn film. For this solution called AP1, the polarization of Fe atoms at the interface is reduced as compared to the bulk value, whereas the nearest Fe(i-1) layer acquires a higher magnetic moment. This effect is reversed in the case where we consider a parallel interfacial Fe-Mn



FIG. 4. Magnetic moment per atom of Mn surface layer [Mn(S)], Mn interface layer [Mn(i)], Fe(i) layer, and Fe(i-1) layer, in terms of the exchange integral J of Mn, in the case of two Mn monolayers having an antiferromagnetic structure and an antiparallel Fe-Mn configuration at the interface with a ferromagnetic Fe(001) semi-infinite substrate (AP1 solution).



FIG. 5. Magnetic moment per atom of Mn surface layer [Mn(S)], Mn interface layer [Mn(i)], Fe(i) layer, and Fe(i-1) layer, in terms of the exchange integral J of Mn, in the case of two Mn monolayers having an antiferromagnetic structure and a parallel Fe-Mn configuration at the interface with a ferromagnetic Fe(001) semi-infinite substrate (P1 solution).

configuration and layered antiferromagnetic Mn-Mn structure (Fig. 5). For this solution called P1, the local magnetic moments on both Fe layers are very close to the bulk value near the critical point, whereas for  $J \ge 0.55$ eV, the calculated magnetic moments on Fe(i-1) layer are slightly smaller than those obtained on the Fe interface atoms. For the Mn layers, we obtain nearly the same values of the magnetic moments on the surface as well on the subsurface layer for both solutions (AP1 and P1). Energetically, it appears that the P1 solution with a parallel Fe-Mn configuration at the interface is more stable (by 0.08 eV per atom for J = 0.75 eV). It must be noticed that the two magnetic configurations appear at nearly the same critical point J = 0.49 eV. For completeness, we have also computed the solutions for  $J_{\rm Mn} < 0.49$ eV. For these particular values of J, the polarization of all Mn atoms is parallel to the polarization of Fe substrate.

We have also investigated the layer-by-layer ferromagnetic structure in Mn film coupled parallel (P2) and antiparallel (AP2) with the ferromagnetically polarized Fe substrate. These solutions that are found to be higher in energy exist, because of the high magnetic moments occurring on the Mn atoms. In fact, the AP2 and the P2 configurations reported on Figs. 6 and 7, respectively, show an important polarization of Mn atoms. However, the magnetic moment on Fe is not qualitatively affected by the nature of the Fe-Mn configuration at the interface, contrary to the previous considered cases AP1 and P1. We do not observe a remarkable change in polarization



FIG. 6. Magnetic moment per atom of Mn surface layer [Mn(S)], Mn interface layer [Mn(i)], Fe(i) layer, and Fe(i-1) layer in terms of the exchange integral J of Mn in the case of two Mn monolayers having a ferromagnetic structure and an antiparallel Fe-Mn configuration at the interface with a ferromagnetic Fe(001) semi-infinite substrate (AP2 solution).

on the Fe atoms when the interfacial Fe-Mn configuration is reversed. In the present case, the Fe(i-1) atom has always the highest magnetic moment as compared to its neighbor Fe(i) atom. The polarization of the Fe is less important when we consider a parallel Fe-Mn configuration than for the antiparallel Fe-Mn configuration. In addition, for the same ferromagnetic layered structure in Mn, the parallel configuration Fe-Mn



FIG. 7. Magnetic moment per atom of Mn surface layer [Mn(S)], Mn interface layer [Mn(i)], Fe(i) layer, and Fe(i-1) layer, in terms of the exchange integral J of Mn, in the case of two Mn monolayers having a ferromagnetic structure and a parallel Fe-Mn configuration at the interface with a ferromagnetic Fe(001) semi-infinite substrate (P2 solution).



FIG. 8. Magnetic moment per atom of Mn surface layer [Mn(S)], Mn(S-1) layer, Mn(i) layer, Fe(i) layer, and Fe(i-1) layer, in terms of the exchange integral J of Mn, in the case of three Mn monolayers having antiferromagnetic structure and antiparallel Fe-Mn configuration at the interface with a ferromagnetic Fe(001) semi-infinite substrate (AP3 solution).

is energetically favorable. To be more precise, when P2and AP2 solutions are both present [see Figs. 6 and 7], the P2 configuration is lower in energy as compared to AP2, for any value of J. Let us mention, that the AP2 solution appears more readily with the exchange integral J than the P2 configuration. When we decrease the exchange integral J, we observe a first-order transition from the AP2(P2) to P1(AP1) solution. This can be explained by the fact that Mn prefers an antiferromagnetic structure and thus it is energetically easier to reverse the spin of the interfacial Mn atom than the moment of the surface atom that is higher.

Taking into account the conclusions deduced from the results of 2Mn/Fe(001), we have investigated the expected stable configurations only by considering a layer-bylayer antiferromagnetic structure on thicker Mn films having a parallel or antiparallel configuration at the Fe-Mn interface. We report on Figs. 8 and 9 the calculated magnetic moments on each atomic layer for an antiparallel (AP3) and for a parallel (P3) arrangement. We find again the same behavior of the Fe layers with respect to the interfacial Fe-Mn configuration already observed in the case of the 2Mn/Fe(001). The AP3 arrangement tends to decrease the magnetic moment of the Fe interface atom in comparison with bulk value, whereas the moment is slightly increased on its neighbor Fe(i-1)atom. The P3 arrangement induce nearly the same and constant magnetic moment  $(2.2\mu_B)$  on the two atomic Fe layers. The magnitude of the Mn polarization follows the



FIG. 9. Magnetic moment per atom of Mn surface layer [Mn(S)], Mn(S-1) layer, Mn(i) layer, Fe(i) layer, and Fe(i-1) layer, in terms of the exchange integral J of Mn, in the case of three Mn monolayers having antiferromagnetic structure and parallel Fe-Mn configuration at the interface with a ferromagnetic Fe(001) semi-infinite substrate (P3 solution).

classical atomic coordination law, since we observe the higher magnetic moment on the less coordinated atoms. The energy calculation shows that the configuration with a layer-by-layer antiferromagnetic structure on Mn coupled parallel with the ferromagnetic Fe(001) substrate (P3) is favored appreciably with a difference in energy of 0.1 eV.

# **IV. CONCLUSION**

We have presented through a self-consistent calculation the magnetic polarization of the Mn layers for a body-centered-tetragonal structure on Fe(001). The ground state electronic structure of this particular system is described within the unrestricted Hartree-Fock approximation of the Hubbard Hamiltonian. We have investigated the antiferromagnetic layer-by-layer structure in Mn, with respect to the majority spin of Fe. The results obtained display a rich variety of solutions. In all cases, it is found that the stable Fe-Mn configuration at the interface is of the parallel type. The antiparallel configuration appears with a higher energy, in agreement with previous calculations.<sup>20,43,61</sup> Layered antiferromagnetic structure appears to be the ground state for the Mn overlayers. Of course, we have also obtained ferromagnetic structure in the Mn overlayers, but this solution is unstable. The highest magnetic moment for Mn atoms is obtained at the surface.

The main difference between Mn and Cr on Fe(001) seems to be about the configuration at the interface with Fe. The Fe-Cr configuration is of antiparallel type at (001) surfaces,<sup>67</sup> whereas the Fe-Mn configuration has been found to be parallel. Antiparallel configuration has also been obtained numerically for Fe-Mn, but its energy is higher. In the case of thin Cr layers on Fe substrate with vicinal surfaces (or steps), some Fe-Cr bonds are frustrated,<sup>68</sup> i.e., they are coupled ferromagnetically so that the magnetic moment on this particular Cr atom is strongly diminished. This is not the case for Mn on Fe vicinal surfaces, because as we have discussed before, P or AP configurations between Fe-Mn bonds do not drastically change the values of the magnetic moments of Mn at the interface.<sup>69</sup>

Concerning the Fe atoms near the interface, it is clear that the nature of Fe-Mn configuration has a dramatic effect on it. As in the case of Cr (Refs. 67 and 53) or V (Ref. 51) on Fe(001) a strong decrease of the magnetic moment on Fe is observed when the Fe-Mn configuration is antiparallel. This is not the case for the parallel alignment where the Fe atoms keep more or less the bulk value.

A detailed comparison with the SPEELS experiments<sup>45</sup> is difficult. However, there is a reasonable agreement between this experiment and the present work, concerning the Mn surface layer with a magnetic moment of about  $3\mu_B$ . Also Fe-Mn parallel configuration and the layered antiferromagnetic structure can explain the two-period oscillation of the exchange asymmetry found by SPEELS.<sup>45</sup> However, these oscillations are only present above 5 ML of Mn. This should be related to some kind of interface roughness that has been taken into account in the case of Fe-Cr interface.<sup>70</sup> This leads to dramatic effects, at least for 3 to 4 ML.<sup>70</sup> This kind of study would be most useful in order to shed some light on the difference between Purcell *et al.*<sup>43</sup> and Walker and Hopster<sup>45</sup> results. Such a study is presently under progress.

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