Multiple collinear magnetic arrangements in thin Mn films supported on Fe(001). Antiferromagnetic versus ferromagnetic behavior

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Abstract

We present a theoretical study of the magnetic properties of thin Mn films of 6 and 7 monolayers supported on Fe(001). The ab-initio Tight Binding Linear Muffin Tin Orbital (TB-LMTO) method was used to investigate the competition between ferromagnetic (F) and antiferromagnetic (AF) couplings within the system. We found several collinear magnetic solutions that may coexist at room temperature. The most stable configurations are characterized by AF coupling between the surface and subsurface Mn layers together with F coupling between Mn and Fe at the interface. The ground state arrangements for the 6 and 7 Mn films display opposite sign of the surface magnetic moment relative to the Fe substrate. The implications of these results in the possible onset of non-collinear magnetism when a step is present at the interface are discussed in comparison with Cr/Fe systems where non-collinear magnetism has been recently reported.

Key words: Magnetism; Transition metal systems; Surfaces and interfaces
PACS: 75.70.-i; 71.20.Be

Although Mn does not stabilise in a bulk bcc phase at room temperature, several results show that body-centered tetragonal (bct) Mn films can grow

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pseudomorphically on the Fe(001) substrates. It has been shown with quantitative low energy electron difraction (LEED) [1], extended X-ray absorption spectroscopy (EXAFS)[2] and scanning tunneling microscopy [3] that Mn deposited at room temperature on the Fe(001) surface grows in a layer-by-layer mode up to more than 10 monolayers (ML). The Mn layers grow pseudomorphically on the Fe(001) surface presenting a bct structure with an in-plane lattice constant of 0.287nm and interlayer spacing of 0.166nm. For the first three Mn layers the interlayer spacing is smaller due to a limited intermixing between Mn and Fe [3].

Mn is exactly in the middle of the 3d transition metal (TM) series, just between Fe, which is a natural ferromagnet in the bulk, and Cr, which is an antiferromagnet. Therefore, Mn stands as one of the more complex 3d TM from the point of view of the magnetic coupling, and it is a clear candidate to exhibit a great variety of magnetic properties. On the one hand, Mn has a large number of d-holes available to be spin-polarized so that high magnetic moments are expected in low-dimensional regimes; on the other hand, Mn is expected to have tendencies to present ferromagnetic (F) or antiferromagnetic (AF) couplings depending on the growth conditions, like those related with the local chemical environment, for instance the hybridization with other TM elements.

The system formed by the deposition of a bct Mn film on Fe(001) is a clear candidate to investigate the above facts, since both the surface effect associated with the loss of neighbors (low-dimensional regime) and the Mn/Fe interface (hybridization effect) occurs at the same time. This system, which is the focus of the present work, has been experimentally investigated by spin-polarized electron energy loss spectroscopy[4] and scanning electron microscopy with polarization analysis[5]. A layered AF (LAF) arrangement was found in the Mn film, that is, each Mn atomic plane is AF coupled with both its upper and lower Mn planes and the coupling within each Mn atomic plane being F. The complexity of the Mn/Fe system from the magnetic point of view was corroborated by the ab initio calculations of Elmouhssine *et al.*[6] for one Mn monolayer on Fe(001), for which multiple magnetic solutions were found, some of them nearly degenerated. Therefore, it is expected that other magnetic arrangements close in energy to the ground state can also exist for the supported Mn film. The aim of the present work is to perform a systematical study of different magnetic solutions (and the relative energy difference) of thin Mn films of 6 and 7 monolayers supported on Fe(001). The in-plane lattice constant (0.287nm) and inter-layer spacings (0.166nm) used in the calculations are those measured in the experiments[1,2] We will investigate the competition between the F and the AF couplings within these systems. We consider F coupling within each (001) plane, as observed also in the experiments for the supported Mn films.

Taking into account the F character of the Fe substrate together with the tendency to AF coupling in the Mn film, different degrees of magnetic frustration are expected for the different magnetic solutions if atomic roughness or steps are present at the interface. In this context, the analysis of the competing magnetic interactions intended in this work will provide valuable information. Besides, the presence of multiple magnetic solutions would provide compelling evidence of the possibility that Mn may adopt non-collinear magnetic arrangements under certain conditions.

The calculations were performed using a scalar-relativistic version of the ab-initio k-space TB-LMTO method [7] within the atomic-sphere approximation and with the generalized gradient approximation in the form of Perdew for the exchange and correlation potential[8]. The number of k points were increased until final convergence was obtained for 135 k points in the irreducible Brillouin Zone. In order to simulate the surface the super-cell technique was used, considering enough layers of empty spheres to assure that there was no interaction between the surfaces of adjacent super-cells. The criterion used to assume that two supercells are isolated was that the charge of the central layer of empty space is smaller than 10^{-4} electrons. 5 monolayers of empty spheres were enough.

In tables 1 and 2 we report some of the different magnetic arrangements found, respectively, for the 6 and 7 layers Mn films on Fe(001), together with the total energy difference with respect to the ground state solution. High magnetic moments are obtained, particularly at the surface sites which are the less coordinated sites. This is due to the large number of d-holes available to be spin-polarized in the low-dimensional regime. The ground state magnetic solution (LAF solution) coincides with the one found in the experiments[4,5]. It is interesting to notice that although the magnetic coupling within the Mn film is AF-like, a F coupling is obtained between Mn and Fe at the interface. This can be understood if we consider that the electron states at the interface resulting from Fe-Mn hybridization must have some Fe character, and thus an enhanced tendency towards the F coupling compared with pure Mn states.
In figure 1, we have plotted the local densities of states (LDOS) at some layers of the LAF solution of Mn_6/Fe(001). The large spin-splitting is evident particularly at the surface and subsurface Mn layers. Besides, the change of sign of the splitting of the LDOS between these Mn layers reflects their AF coupling, in contrast to the situation at the interface, where the same sign of the splitting is reflected in the LDOS of Mn and Fe. It is well known that the magnetic properties of TM systems are sensitive to changes in the atomic environment. In order to test the stability of the LAF ground state under surface relaxation, we have performed additional calculations considering an artificial relaxation of $\pm 5\%$ in the surface and subsurface Mn layers. The LAF solution remains the most stable one and the energy difference between the metastable solutions and the LAF do not change appreciably except for the

less stable solutions 4 and 5, being solution 4 degenerated with solution 3 in the expanded geometry and degenerated with solution 5 in the compressed geometry. The local magnetic moments only change in no more than 0.1 μ_B except for the subsurface Mn layer whose moment increases in about 0.2-0.4 μ_B under 5% expansion (compression).

The tendency of pure Mn also towards the F coupling is reflected in most of the magnetic solutions, where some layers of the Mn films are ferromagnetically coupled. Besides, some of those solutions for the Mn film are close in energy to the LAF one, a clear prove of the small energy needed to change the magnetic coupling from AF to F in this material. This can be qualitatively understood taking into account that Mn is located in the TM series between an antiferromagnet (Cr) and a ferromagnet (Fe) so that the Mn d-electrons exhibit tendencies to both magnetic couplings. Between the different magnetic arrangements found, those having the lowest energies (solutions 1, 2 and 3) have two common characteristics: (i) AF coupling between the surface and subsurface Mn layers and (ii) F coupling between Mn and Fe at the interface. This means that in the case that a magnetic configuration other than the LAF one were stabilized (at finite temperature for instance), the magnetic trends described at both the surface layers and the interface layers of the system would persist. The most stable magnetic arrangements for the 6 and 7 Mn films (with thickness differing in one monolayer) display opposite sign of the surface magnetic moment relative to the Fe substrate (compare the Mn on the surface Mn(S) in solution 1 of tables 1 and 2).

The above results have important implications in the magnetic behavior of non-ideal Mn/Fe interfacial systems. For instance, the presence of monoatomic steps at the interface of supported Mn films on Fe would disturb the antiferromagnetic order at the edges, being the origin of magnetic frustration. This fact, together with the existence of multiple magnetic solutions, makes this system a clear candidate to exhibit noncollinear magnetism under interfacial defects like monoatomic steps or other more general structural changes. In this context, we have recently reported a non-collinear magnetic behavior of Cr films supported on a stepped Fe(001) substrate[9]. Due to the computational limitation of the ab-initio methods, that complex system was investigated in the framework of a semiempirical noncollinear tight-binding model parameterized to TB-LMTO results of ideal interfaces. The resulting non-collinear behavior was related to the magnetic frustrations at the interface. However, the magnetic frustration at the Mn/Fe interface is expected to be lower than in the Cr/Fe case in view of the tendency of Mn to exhibit F couplings with a low energy cost. Then, a non-collinear study of such stepped Mn/Fe systems is quite interesting at present. The TBLMTO results reported in the present study for ideal Mn/Fe interfaces will serve us also to parameterize and test our TB model. Noncollinear calculations of Mn films supported on a stepped Fe(001) substrate are in progress and will be reported at due time.

This work was supported by Junta de Castilla y León, Spain (Grant VA 073/02), NATO (Project PST.CLG 979708) and INTAS (Project 03-51-4778).

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Table 1
Some of the different magnetic configurations (one per column), obtained with the TBLMTO method for 6 Mn layers supported on Fe(001). The local magnetic moments (in units of μ_B) at each Mn plane and at the Fe interface are given. The total energy difference with respect to the most stable solution is also provided.

layer	1	2	3	4	5
Mn(S)	-3.80	3.76	3.73	-3.74	3.82
Mn(S-1)	1.74	-1.93	-2.09	2.50	-1.74
Mn(S-2)	-3.05	2.85	2.92	2.99	3.05
Mn(S-3)	2.81	2.85	-2.87	-2.83	-2.80
Mn(S-4)	-2.91	-2.78	-2.86	-2.82	3.04
Mn(Int)	2.82	2.89	2.88	2.89	-2.21
Fe(Int)	2.45	2.46	2.44	2.44	2.44
$\Delta E(eV)$	0.00	0.07	0.16	0.26	0.37

Table 2
As in Table 1 for 7 Mn layers supported on Fe(001).

layer	1	2	3	4	5
Mn(S)	3.80	3.76	3.76	-3.78	-3.74
Mn(S-1)	-1.72	-2.42	-1.95	-2.72	2.47
Mn(S-2)	3.00	-2.97	2.78	2.75	2.99
Mn(S-3)	-2.85	2.89	2.79	2.75	-2.81
Mn(S-4)	2.71	2.83	-2.78	-2.79	-2.82
Mn(S-5)	-2.85	-2.79	-2.79	-2.81	2.61
Mn(Int)	2.82	2.87	2.88	2.90	2.02
Fe(Int)	2.44	2.45	2.43	2.44	2.01
$\Delta E(eV)$	0.00	0.24	0.31	0.53	0.95

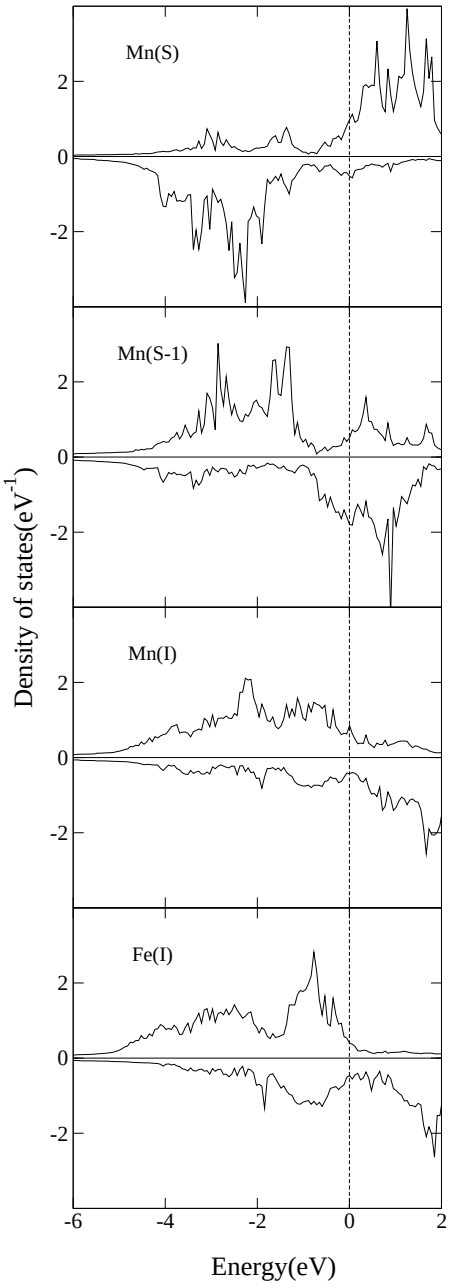

Fig. 1. Local density of states (LDOS) at some layers of the Mn_6/Fe(001) system: surface (S), subsurface (S-1) and interface (I). The vertical dashed line at 0 eV represents the Fermi level.