Magnetic structure and anisotropy in Fe/Cu(001) over- and interlayers with antiferromagnetic interlayer coupling

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A first-principles study of the ground state and the magnetic anisotropy of antiferromagnetic fcc Fe/Cu(001) over- and interlayers is presented using the fully relativistic spin-polarized screened Korringa-Kohn-Rostoker method. It is shown that the formation of the antiferromagnetic ground state is highly sensitive to the atomic volume (lattice spacing). Contrary to a previous study of the ferromagnetic state, it is found that for all considered cases, namely up to seven layers of Fe, the magnetization is oriented along the surface normal. [S0163-1829(97)05221-1]

I. INTRODUCTION

It has been shown by several authors[1–7] that ''fcc'' iron films exhibit a rich magnetic structure. Different kinds of ferromagnetic and antiferromagnetic phases can be distinguished, the stability of which depends very sensitively on the atomic volume. Fe films grown on Cu(001) are of particular interest, because the lattice constant (atomic volume) of Cu is close to the lattice spacing in all these phases, therefore small differences of strain can stabilize either one. In a previous paper[8] we discussed in detail the magnetocrystalline anisotropy of the ferromagnetic phase, while the present paper is devoted to the antiferromagnetic one. Here, and throughout the whole paper, ''antiferromagnetic'' refers to a collinearly ordered magnetic thin film system, where the intralayer coupling between the moments is ferromagnetic (see also the discussion in Ref. 9), but some of the different atomic layers couple antiferromagnetically. This kind of antiferromagnetic coupling was found also by Lorenz and Hafner[10] using the noncollinear tight-binding linear muffin-tin orbital method. In the present paper we study Fe over- and interlayers, denoted by Fe_n/Cu(001) and Cu/Fe_n/Cu(001), respectively. In the next section first the magnetic ground states are determined and characterized, and then the magnetic anisotropy energies (MAE) with respect to orientations normal and parallel to the surface (interface) are compared to the ferromagnetic case.[8] Finally, we draw some conclusions concerning the relationship between experimental observations and theoretical investigations.

II. RESULTS AND DISCUSSION

A. Ground-state properties

First, we determined the ground-state configuration for each system, namely, the type of coupling between the layers that corresponds to the lowest total energy. For an overlayer system consisting of n monolayers this, in principle, means considering 2^{n-1} different configurations. In order to reduce the numerical effort, for the overlayer systems we applied the *scalar-relativistic* spin-polarized screened Korringa-Kohn-Rostoker (SKKR) method[11,12] to calculate total energies self-consistently within the local spin-density approximation[13] and the atomic-sphere approximation for the different magnetic configurations, from which in turn the ground state was found. Self-consistent *fully relativistic* calculations were then carried out for the ground-state configurations only. This procedure led safely to the proper ground state, since the type of magnetic coupling between the layers is primarily determined by the (nonrelativistic) exchange interaction, while the calculated physical properties are consistent with a fully relativistic theory. Due to trivial symmetry considerations, for an interlayer film consisting of $n=2m$ or $n=2m+1$ layers the number of possible magnetic configuration is 2^m, i.e., considerably less than in the overlayer case. For these systems, therefore, the fully relativistic SKKR method was used throughout the whole computational scheme. Note, that the self-consistent fully relativistic SKKR calculations were carried out with magnetic moments aligned normal to the surface.

In Table I the calculated (spin-only) magnetic moments for the ground-state configurations are displayed together with a sign indicating a parallel (positive sign) or antiparallel (negative sign) orientation along the surface normal. It is important to mention that for two layers of Fe, both as over- or interlayers, the ferromagnetic configuration is found to be the ground state. For the overlayer systems ($n>2$) the surface and subsurface layers always couple ferromagnetically, carrying an enhanced moment as compared to the antiferromagnetic fcc bulk Fe with the lattice constant of bulk Cu

TABLE I. Calculated spin-only magnetic moments [μ_B] of Fe in Cu_mFe_n/Cu(001) multilayers, $n=2,3,\ldots,7$, corresponding to the antiferromagnetic ground state. The numbering of the Fe layers increases from the vacuum (or capped) side towards the bulk. For comparison the case $n=2$ is included here although the ground state refers to the ferromagnetic configuration. A parallel or antiparallel orientation of the magnetization along the surface normal is indicated by a plus and minus sign, respectively.

n	m	Fe(7)	Fe(6)	Fe(5)	Fe(4)	Fe(3)	Fe(2)	Fe(1)
2	0						-2.340	2.355
	∞						-2.207	2.207
3	0					-2.121	2.297	2.820
	∞					2.232	-1.428	2.232
4	0				2.262	-1.430	2.259	2.788
	∞				-2.130	-2.094	2.094	2.130
5	0			-2.224	1.453	-1.506	2.245	2.793
	∞			2.507	2.147	-1.609	2.147	2.507
6	0		2.523	2.089	-2.029	-2.022	2.162	2.789
	∞		2.509	2.069	-2.029	-2.029	2.069	2.509
7	0	-2.232	1.534	-1.450	1.779	-1.457	2.264	2.802
	∞	2.520	2.264	-1.939	-1.753	-1.939	2.264	2.520

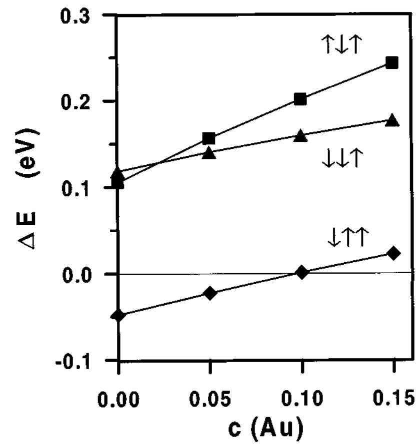

FIG. 1. Total-energy differences of the possible antiferromagnetic configurations with respect to the ferromagnetic configuration of three Fe overlayers on $Cu_{1-c}Au_c$(001) as a function of the Au concentration. '' ↑'' and ''↓'' refer to a parallel or antiparallel orientation of the magnetization in a layer with respect to the surface normal, respectively.

[$\sim 1.65\mu_B$ (Ref. 14)]. This feature has been found also by other authors both experimentally[15] and theoretically.[16,17,10]

Returning to Table I, one can see that as the subsequent layers couple mostly antiferromagnetically, buried Fe layers with a layer index ranging between 3 and $n-1$ carry even smaller magnetic moments. In particular, for the overlayer system with $n=5$ and 7, where the buried Fe layers couple antiferromagnetically, we find the magnetic moments to be very close to that of fcc bulk Fe. The Fe layer at the interface with the substrate, however, systematically carries a higher magnetic moment. A similar type of effect was already seen for the ferromagnetic Fe_n/Cu(001) (Ref. 8) and also Fe_n/Au(001) over- (Ref. 18) and interlayers,[19] attributed mainly to the weak hybridization between Fe and the substrate.

As studied extensively in the past,[14,20–22] the magnetic ground state of bulk fcc Fe depends very sensitively on the volume. In order to demonstrate that this feature survives even for ultrathin films of ''fcc'' iron, we performed as a case study a calculation of a Fe_3 overlayer on a statistically disordered $Cu_{1-c}Au_c$(001) substrate using the coherent potential approximation (for a detailed discussion in the case of multilayers, see Ref. 23). It should be noted, that for Au concentrations of less than about 15% the equilibrium lattice constant of $Cu_{1-c}Au_c$ follows Vegard's law fairly well.[24] In Fig. 1 the energy difference between the possible antiferromagnetic states and the ferromagnetic state is shown. From this figure one can see that the antiferromagnetic configuration (↓↑↑) is indeed very close in energy to the ferromagnetic one and that for concentrations of Au greater than 10% the ferromagnetic configuration becomes the ground state. Since this concentration refers only to a 3.6% increase in the atomic volume with respect to the pure Cu substrate, layer relaxation close to the surface, which was not included into the present study, may easily mimic this effect. This is in accordance with the experiments in Ref. 25 revealing that for thin films of Fe below four monolayers the tetragonal distortion due to interlayer relaxation is around 5% and that the ground state is ferromagnetic.

Turning now to the case of interlayers, it is interesting to observe from Table I that — at least for $n\geqslant 4$ — the two layers next to the Fe/Cu interfaces couple ferromagnetically. In Fig. 2 the magnetic moments and the total-energy differences with respect to the ferromagnetic configuration of possible antiferromagnetic configurations are shown for two systems, namely, for interlayers with five and six Fe layers, respectively. These two cases serve also as an illustration for different distributions of magnetic moments with respect to an even and an odd number of Fe layers. As can be seen, all of the different antiferromagnetic configurations shown are energetically more favorable than the ferromagnetic one. Remarkably, the difference between the ground-state energy and the total energy of the subsequent metastable state is fairly small ($<$ 0.05 eV). In general, there seems to be a tendency that the moments corresponding to layers with antiferromagnetically coupled nearest layers are much smaller.

B. Magnetic anisotropy energies

As was discussed in detail in Ref. 18, by using the force theorem the magnetic anisotropy energy (MAE) consists of a sum of two contributions, namely the band energy and the magnetostatic dipole-dipole interaction energy, ΔE_b and ΔE_{dd}, respectively. Here we use the notation $\Delta E_\alpha = E_\alpha^\parallel - E_\alpha^\perp$ ($\alpha = b$ or dd), where $\parallel$ and $\perp$ refer to the cases that in each layer the magnetic field points uniformly parallel or perpendicular to the surface (interface). Technically the parallel orientation was obtained from the perpendicular one by a simultaneous rotation of $\pi/2$ around the y axis in all layers. Again, the numerical details were the same as used previously to study the ferromagnetic case.[8]

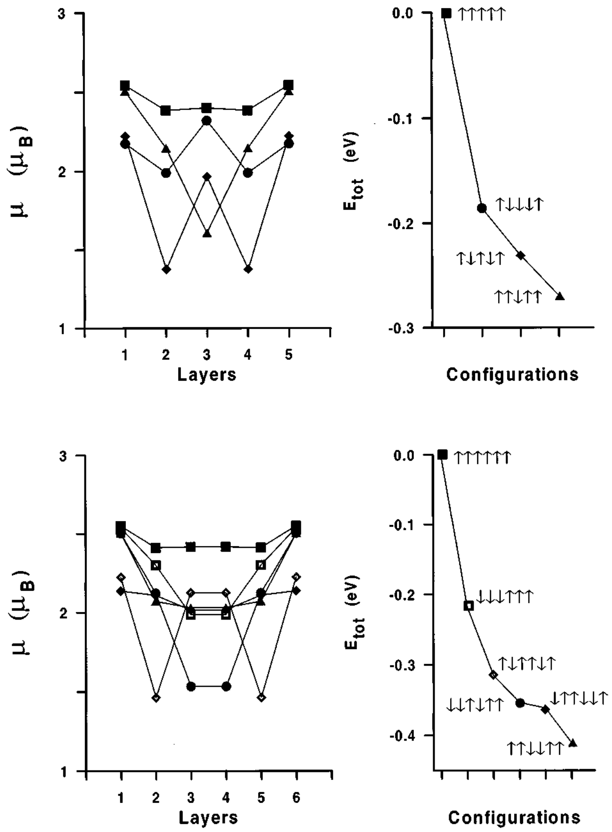

FIG. 2. Left-hand side: layer resolved magnetic moments for the possible magnetic configurations in the Cu/Fe_5/Cu(001) (upper sheet) and Cu/Fe_6 /Cu(001) (lower sheet) interlayer systems. Right-hand side: total-energy differences with respect to the ferromagnetic configuration for the corresponding interlayer systems and magnetic configurations on the left-hand side.

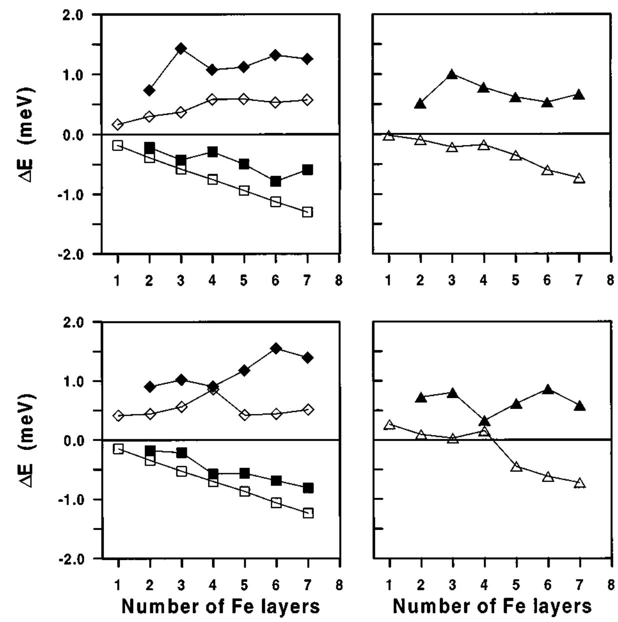

FIG. 3. Calculated magnetic anisotropy energies for the Fe_n/Cu(001) overlayer (upper panels) and for the Cu/Fe_n/Cu(001) interlayer (lower panels) systems. Diamonds: ΔE_b, squares: ΔE_{dd}, triangles: $\Delta E = \Delta E_b + \Delta E_{dd}$. Full symbols: antiferromagnetic ground states (but for $n=2$), open symbols: ferromagnetic state (Ref. 8). The solid lines serve as a guide for the eye.

In our previous paper[8] we found that ferromagnetic overlayers of Fe on Cu(001) have in-plane orientations of the magnetization for all thicknesses. However, experimentally it was shown that the magnetization is perpendicular to the surface even for rather thick overlayers of Fe. As mentioned earlier, it seems to be an emerging consensus that these films are antiferromagnetically ordered, show perpendicular magnetism up to a quite large overlayer thickness (10–14 monolayers), and that the reorientational transition is accompanied by a structural phase transition[6] from a fcc to a bcc type lattice (for a discussion of the meaning of such lattices in context with a two-dimensional translational symmetry see Ref. 9).

Indeed, as is shown in Fig. 3, we found that in the antiferromagnetic ground state, for both the overlayer and interlayer cases, the magnetization is perpendicular for all thicknesses, and that no tendency for a switching to an in-plane orientation at higher thickness can be read off. The reason for this behavior is twofold. First, with the exception of $n=4$ for the interlayer case, ΔE_b is much higher for the antiferromagnetic ground state than in the ferromagnetic configuration. Second, the reduced moments characterizing the antiferromagnetic ground state give rise to magnetic dipole-dipole contributions smaller in magnitude than in the ferromagnetic case. As a result, in the antiferromagnetic multilayers the positive band energy anisotropy outweighs the negative dipole-dipole term leading thus to perpendicular magnetism.

In previous studies[8,18] the layer-resolved analysis of ΔE_b provided a very useful tool to analyze and interpret the MAE. In Figs. 4 and 5 the resolution of ΔE_b with respect to layers is shown for the over- and interlayers, respectively. For comparison, the corresponding entries for the ferromagnetic systems are also displayed. From these figures it is apparent that ΔE_b has a fundamentally different spatial origin in the antiferromagnetic ground states as compared to the ferromagnetic ones. As can be seen in Fig. 5, this difference is most obvious for thicker interlayer systems ($n \geqslant 4$), where in the ferromagnetic case the buried layers contribute only negligibly to the MAE and the main contributions come from the Fe layers at the two interfaces, while in the antiferromagnetic case all layers contribute significantly. A particular exception is the middle layer in the Cu/Fe_7/Cu system, which has a very low contribution to the MAE. Although not presented here, a similar distribution characterizes the MAE of the Cu/Fe_9/Cu system with a (↑↑↑↓↓↓↑↑↑) configuration, as buried Fe layers with only ferromagnetic nearest layer coupling contribute remarkably less to the MAE than those with antiferromagnetic nearest layer coupling. Therefore, it seems that the MAE for the buried layers is governed by the *local* configuration.

In order to pursue this point we carried out two model calculations for an interlayer containing 26 identical Fe layers, namely, one with only ferromagnetic interlayer coupling and the other one with alternating antiferromagnetic coupling. It turned out that as suggested in Fig. 5 in the ferromagnetic case exclusively the Fe layers at the interfaces contributed sizably to the MAE, whereas in the layered

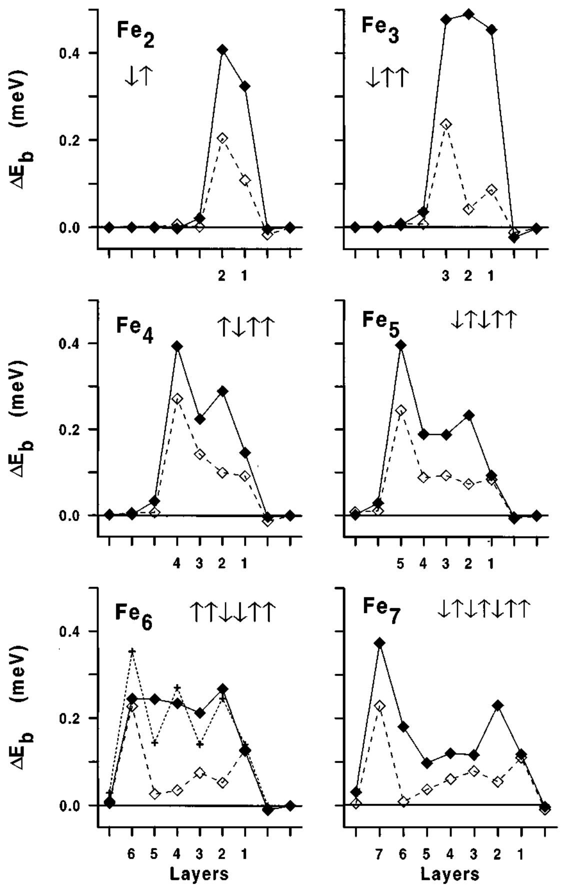

FIG. 4. Layer-resolved band energy contributions to the MAE of the antiferromagnetic ground state of Fe_n/Cu(001) overlayers (full symbols). In each entry the corresponding spin configuration is indicated by arrows. Open symbols refer to the results for the ferromagnetic configuration in Ref. 8. For Fe_6/Cu(100) the data for the ↑↓↑↓↑↑ state is also shown with crosses (see text). Only the Fe layers are numbered in the sequence as in Table I. The lines serve as a guide for the eye.

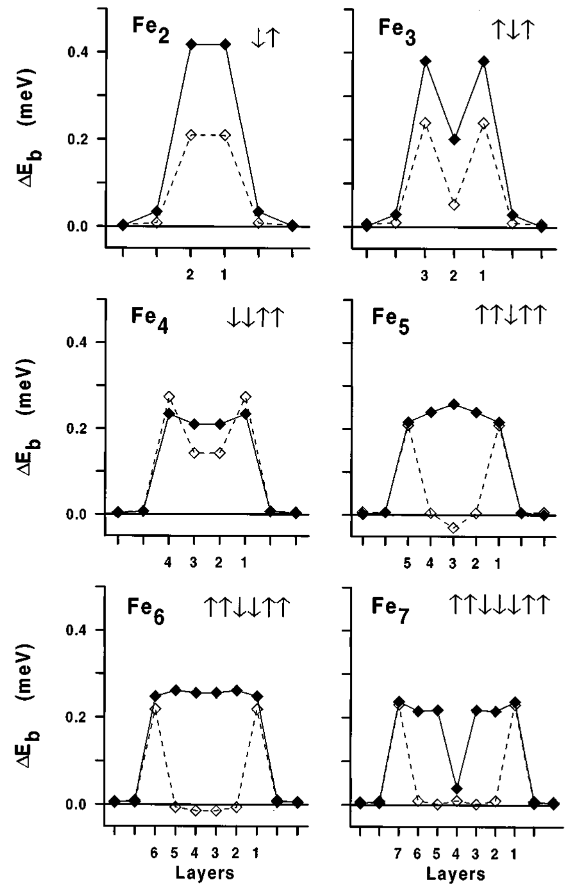

FIG. 5. Layer-resolved band energy contributions to the MAE of the antiferromagnetic ground state of Cu/Fe_n/Cu(001) interlayers (full symbols). In each entry the corresponding spin configuration is indicated by arrows. Open symbols refer to the results for the ferromagnetic configuration in Ref. 8. Only the Fe layers are numbered in the sequence as in Table I. The lines serve as a guide for the eye.

antiferromagnetic case all the Fe layers had nearly the same contributions (~ 0.2 meV/layer). This indicates that in a fcc bulk Fe system with antiferromagnetic coupling the same order of magnitude for the MAE can be expected.

Although the situation in the case of overlayers is much more complicated, a similar behavior seems to apply there. As, with exception of $n=6$, the Fe layer at the interface (labeled by n) couples antiferromagnetically to layer $n-1$, an enhanced contribution to the MAE arises from this layer in comparison to the ferromagnetic case. From Fig. 4 it is also obvious that in the antiferromagnetic ground states buried Fe layers generally have a larger contribution to ΔE_b than their ferromagnetic counterparts. The ferromagnetic coupling at the surface (see Sec. II A) apparently shows up for $n \geqslant 4$, and causes nearly identical contributions to ΔE_b from the surface layers for both the antiferromagnetic and the ferromagnetic cases. However, unlike in the ferromagnetic case, the subsurface Fe layer (labeled by 2), which couples antiferromagnetically to layer 3, contributes systematically more to ΔE_b than the surface layer. Note that for the Fe_3/Cu(001) system the surface layer, coupled ferromagnetically to the subsurface layer, has a surprisingly large contribution to the MAE.

Since the ground-state configurations of magnetic couplings for Fe_5/Cu(001), (↓↑↓↑↑), and for Fe_7/Cu(001), (↓↑↓↑↓↑↑), are in their sequence very different from that obtained for Fe_6/Cu(001), (↑↑↓↓↑↑), it seems to be particularly illuminating to present also the layer-resolved magnetic anisotropy energy of Fe_6/Cu(001) for the configuration (↑↓↑↓↑↑), which is energetically very close to the ground state. As can be seen from the corresponding entry of Fig. 4, if the layer-resolved band energy contributions of this particular configuration are considered then the above-mentioned trends become more transparent. It is quite obvious from this example that the calculated anisotropy energies are rather sensitive to the magnetic configuration, i.e., the type of the magnetic coupling between the layers.

III. CONCLUDING REMARKS

We presented an *ab initio* study of the magnetic states and the magnetic anisotropy of Fe over- and interlayers on Cu(001) based on a fully relativistic spin-polarized band theory. It has been shown that the ground state of these films depends very much on the volume, which can be influenced by changes in interlayer (and intralayer) distances. The actual values of such layer relaxations are likely to depend on the growth conditions and might be different in different experiments. We also showed that the magnetic anisotropy energies are rather different for the ferromagnetic and the various antiferromagnetic configurations. Different magnetic configurations as a consequence of different growth conditions might turn out to be yet another source of experimental disagreements. Despite these possible ambiguities, however, the basic experimental observation for Fe films grown on Cu(001), namely a perpendicular orientation of the magnetization even for rather thick Fe films, was shown in this paper to be the result of antiferromagnetic coupling between the Fe layers, a result that pertains to overlayer systems (free surfaces) as well as to interlayer systems (surface capped with a thick layer of Cu).

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