Noncollinear magnetism in rough ultrathin γ-Fe films

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(Received 24 April 1998)

In ultrathin face-centered cubic Fe films on Cu(100) substrates only the surface and subsurface layers are ferromagnetically coupled, while the interior of the films shows various antiferromagnetic configurations, depending on the thickness of the films. We show using *ab initio* local-spin-density calculations that for films with more than four monolayers this leads to a frustration of the magnetic interactions in the vicinity of a step and hence to a noncollinear magnetic structure. As a consequence rough ultrathin γ-Fe films cannot be characterized by a simple uniaxial magnetic anisotropy.
[S0163-1829(98)05530-1]

Recently the complex structural and magnetic properties of ultrathin fcc Fe films grown on Cu(100) have attracted much interest.[1–11] Two distinct preparation conditions resulting in films with different morphologies and different magnetic properties have been identified.[2] In the first procedure, the Fe film is deposited at low temperature (LT) ($T \lesssim 120$ K) and afterwards annealed at room temperature. LT-deposited films grow in a Stranski-Krastanov mode, resulting in a pronounced surface roughness increasing with the average thickness of the film. In the second procedure the film is deposited at or above room temperature (RT). The RT-deposited films grow in a layer-by-layer mode, their surface roughness decreases with increasing thickness of the film.

In the limit of very low film thickness LT- and RT-grown films share two common characteristics: (a) After the onset of ferromagnetism at 1.3 to 2 monolayers (ML) follows region I with up to 3 to 4 ML characterized by an almost homogeneous magnetization of the entire film confirmed by the investigation of the linear magnetooptical Kerr effect (MOKE)[3,4] and perpendicular magnetic anisotropy. In this region the structure of the films is slightly tetragonally distorted fcc with characteristic buckling reconstructions.[5,6] (b) Films with more than 3 to 4 ML (region II) show a surface magnetization nearly equal to that of region I while the interior of the film has been described as either in a low-moment state (which could be antiferromagnetic, see below)[7,8] or as paramagnetic at room temperature.[9,10] These films show perpendicular magnetic anisotropy, their structure is described as fcc with a slight relaxation of the top layer.[5,6] It is remarkable that both the surface magnetization and the spin asymmetry are almost unaffected by the structural and magnetic phase transition at 4 ML.[4] For the RT-grown films region II extends up to 10 to 12 ML with almost constant magnetization and still perpendicular anisotropy.[3,4] In contrast, LT-deposited films show a transition to in-plane anisotropy much earlier, between 5 and 6 ML (Ref. 1) while conserving their fcc structure and reduced magnetization for thicknesses up to 10 to 12 ML. In the transition range from perpendicular to in-plane anisotropy the magnetic properties of the films show an interesting temperature dependence.[11] (c) Above 10 to 12 ML both RT and LT films become predominantly bcc, with a homogeneous magnetization of the entire film and in-plane anisotropy. It has been speculated that the magnetization reversal in LT films at $\sim$5 ML is likely to be driven by a roughness-induced modification of the magnetic properties.[2,4]

First-principles calculations[12–16] of the electronic and magnetic structures of fcc Fe/Cu(100) films agree on the following. (a) A ferromagnetic coupling exists between the surface (S) and the first subsurface (S-1) layer, with a surface moment that is strongly enhanced over the moment in bulk bcc Fe. (b) Films with up to three ML are entirely ferromagnetic, but in films with four and more ML antiferromagnetic coupling is found in the interior of the film. There is evidence for the coexistence of stable and metastable high- and low-spin configurations.[15,16] (c) The magnetic moments show only a small variation under a tetragonal distortion of the film,[14] in contrast to studies on bulk fcc and fct iron.[17] (d) The magnetic anisotropy remains perpendicular even for the thickest layers (7 ML) covered in these studies.[15] Altogether these results represent a rather satisfactory explanation of the experimental observations on RT-deposited films with atomically flat surfaces.

In this paper we address the question of a possible influence of the surface-roughness of the LT-grown films on their magnetic properties via *ab initio* calculations in the local-spin-density approximation.[18] It is clear that for the thinnest films with a homogeneous ferromagnetic polarization a possible roughness of the surface will affect the magnetization only marginally. On the other hand, steps in partially antiferromagnetic films will unavoidably lead to a mismatch between layers with different spin orientations. We show that this leads to the formation of a noncollinear magnetic structure in the vicinity of the step. The immediate consequence is that rough films cannot be characterized by a simple uniaxial surface anisotropy, in contrast to smooth films.

Our calculations are based on a real-space tight-binding linear-muffin-tin-orbital (TB-LMTO) technique[19] based on a canonical transformation from the standard LMTO Hamiltonian to the most localized tight-binding basis. The screened structure-constants of the TB-LMTO have been calculated for the nearest- and next-nearest-neighbor environments, the two-center TB Hamiltonian has been calculated to second order in the Löwdin expansion.[19,20] We use a scalar relativistic local-spin-density (LSD) Hamiltonian with variable spin-quantization axes on each site, augmented by a spin-

orbit interaction term. The calculations are performed self-consistently using the real-space recursion technique.[21] Self-consistency is achieved in a three-step procedure. First we perform a paramagnetic calculation until the charge density is converged. In the second step the spin-density is converged for fixed collinear directions of the spins. In the third step we add spin-orbit coupling and we allow for local fluctuations of the spin-quantization axes. Dipole-dipole interactions accounting for the shape anisotropy are also included. The calculation is iterated until magnitudes and directions of the local moments are converged. For charge and spin densities as well as for the directions of the moments we use the optimized Broyden mixing proposed by Kresse and Furthmüller.[22] For further technical details of the noncollinear calculations we refer to our earlier papers[23,15] which are based, however, on a local-density-plus Hubbard (LDA-Hubbard) Hamiltonian.

Our main objective is the calculation of the magnetic structure in the vicinity of a step in a Fe/Cu(100) film. Figure 1 shows a side view of our model for a 4 ML/5 ML step. The local spin-densities have been calculated self-consistently for the 32 Fe atoms in the vicinity of the step, the 7 Cu atoms in the first layer at the interface and the first layer of vacuum spheres (again seven spheres). The sites in the second and third Cu layer and the second vacuum layer have been treated as equivalent, counting each as one type of atomic sites. This makes altogether 49 inequivalent atomic sites. For the inner core of 23 Fe atoms (shown with a lighter shading in Fig. 1) the directions of the moments are allowed to deviate from the easy axis perpendicular to the surface. The more distant environment of the step has been modelled by a large cluster with free boundary conditions in the plane perpendicular to the step edge and periodicity along this direction, with the potential and TB parameters fixed at their values determined from self-consistent calculations for 4 ML and 5 ML films. Using 9, 15, and 37 recursion levels for *s*, *p*, and *d* orbitals on Fe sites in the noncollinear calculation leads to a cluster with altogether 7840 atoms (20 layers with 392 atoms) and about 2300 neighbors to each iron site.

Table I summarizes the results for flat Fe/Cu(100) films with up to 7 ML Fe, aligned at the Fe/Cu interface. Only the magnetic ground state in each layer has been considered, possible metastable states[15] are ignored. The calculations admit in principle a canted spin structure, but we find that in the ground state the moments are aligned along the surface normal. The confrontation of our self-consistent LSD results with the earlier calculations[15] based on a Hubbard-type exchange Hamiltonian with a constant Stoner parameter confirms the validity of this approximation. Within each film one has ferromagnetic coupling between the surface and subsurface Fe layer (the ''magnetically live'' surface observed by Li *et al.*[8]), and layered antiferromagnetism (but not necessarily in a one-by-one alternating sequence) in the interior of the film. It is immediately evident that for four and more monolayers the layered antiferromagnetism is strongly disturbed at a monolayer step (as marked in Table I). The frustrations marked in Table I refer to the case where the directions of the global magnetization are parallel on both sides of the step (i.e., the step occurs within a single magnetic domain). If we assume an antiparallel orientation of the global magnetic moments (i.e., if we assume that the step coincides with the boundary of two magnetic domains), the role of frustrated and matching layers is just inverted. If we allow the directions of the magnetic moments to relax from the direction perpendicular to the film fixed by the anisotropy of the flat films, a noncollinear structure will appear in the vicinity of the step.

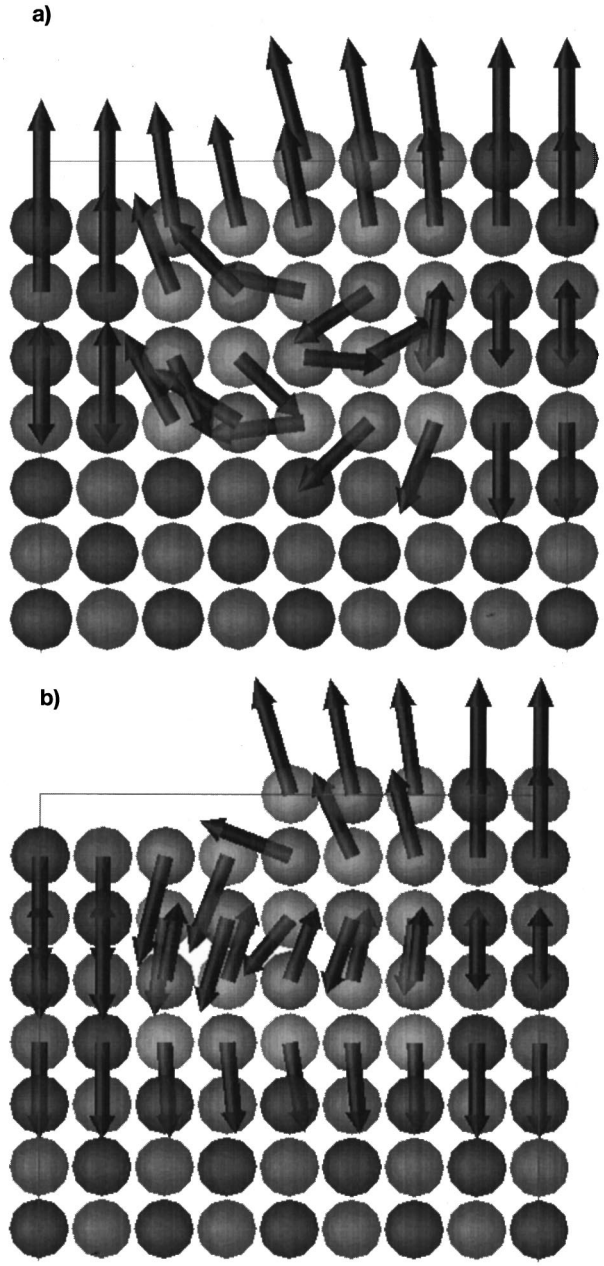

FIG. 1. Noncollinear magnetic structure in the vicinity of a 4 ML/5 ML step in a fcc Fe film on a Cu(100) substrate (side view in the direction of the step edge, showing the atoms in two neighboring planes). Charge and spin densities and the directions of the magnetic moments have been calculated self-consistently on the sites in the immediate vicinity of the step (shown as light grey spheres). The arrows represent the magnitudes and directions of the magnetic moments on the Fe sites. (a) illustrates the solution for a parallel orientation of the magnetization on both sides of the step, (b) for a case of antiparallel orientation. See the text.

Here we concentrate on the results obtained for a 4 ML/5 ML step. Figure 1(a) shows a pictorial representation of the

TABLE I. Magnetic moments in smooth fcc Fe/Cu(100) films with up to seven monolayers of Fe. A dark bullet marks the points where frustrated intralayer exchange coupling will appear at a monolayer step. The present LSD results are given in parentheses and compared with the results from the earlier TB-Hubbard calculations (Ref. 15).

Number of ML in film	1	2	3	4	5	6	7
				Surface			
Layer							
Fe7							2.77
Fe6						2.77	2.25
Fe5					2.75(2.77)	2.20 •	−1.90
Fe4				2.81(2.84)	2.27(2.21) •	−2.36 •	1.63
Fe3			2.82(2.82)	2.38(2.41) •	−1.70(−1.71)	−2.31	−1.45
Fe2		2.76	2.34(2.43) •	−1.95(−1.96) •	1.86(1.72)	2.11	2.15
Fe1	2.71	2.22	2.20(2.38)	2.41(2.30) •	−2.27(−2.24) •	2.51 •	−2.21
				Interface			

magnetic structure resulting from a self-consistent noncollinear calculation for the case of parallel global moments on both sides of the step. Table II lists the magnitudes of the perpendicular and in-plane components of the magnetic moments in the vicinity of the step. We find that in the first three layers from the Fe/Cu interface where layers with opposite moment meet at the interface a gradual rotation of the moment takes place in the transition region, accompanied by

TABLE II. Perpendicular and in-plane (in parentheses) components $\mu_i^{\perp}$ ($\mu_i^{\parallel}$) (in μ_B) of the magnetic moments in the vicinity of a 4 ML/5 ML step in an fcc Fe/Cu(100) film. The lines labeled (a) refer to a configuration with a parallel, those labeled (b) to an antiparallel orientation of the global magnetization on both sides of the step. See the text.

Layer	$\mu_i^{\perp}$ ($\mu_i^{\parallel}$)								
					Surface				
5(a)					2.79(-0.75)	2.76(-0.49)	2.75(-0.34)	2.76	2.77
5(b)					2.76(-0.85)	2.72(-0.55)	2.74(-0.41)	2.77	2.77
4(a)	2.84	2.86	2.79(-0.39)	2.50(-0.53)	2.31(-0.45)	2.29(-0.32)	2.24(-0.16)	2.24	2.21
4(b)	-2.84	-2.84	-2.60(-0.64)	-2.28(-0.89)	0.83(-2.13)	1.99(-0.92)	2.12(-0.56)	2.21	2.21
3(a)	2.41	2.41	2.22(-0.85)	1.65(-1.60)	0.44(-2.09)	-1.15(-1.71)	-1.82(-0.26)	-1.71	-1.71
3(b)	-2.41	-2.41	-2.31(-0.24)	-2.23(-0.63)	-1.34(-1.09)	-1.76(-0.61)	-1.72(-0.26)	-1.71	-1.71
2(a)	-1.96	-1.98	-1.82(0.85)	-1.31(1.40)	-0.16(1.79)	0.85(1.53)	1.71(0.15)	1.72	1.72
2(b)	1.96	-1.96	1.84(0.31)	1.83(0.60)	1.72(0.62)	1.75(0.38)	1.75(0.17)	1.72	1.72
1(a)	2.30	2.30	2.01(-1.07)	1.23(-1.90)	-0.23(-1.61)	-1.56(-1.61)	-2.06(-0.79)	-2.26	-2.24
1(b)	-2.30	-2.30	-2.26(0.13)	-2.17(0.20)	-2.11(0.36)	-2.14(0.18)	-2.20(0.08)	-2.24	-2.24
					Interface				
Average(a)	1.40	1.40	1.30(-0.37)	0.71(-0.66)	1.03(-0.75)	0.64(-0.52)	0.56(-0.28)	0.56	0.55
Average(b)	-1.40	-1.40	-1.33(-0.11)	-1.21(-0.18)	0.37(-0.62)	0.51(-0.30)	0.54(-0.19)	0.55	0.55
					0.85(-0.52)				
					-0.11(-0.37)				

a slow increase of the moments from the 5 ML to the 4 ML side. During the rotation the antiferromagnetic coupling between the first, second, and third layers from the interface is preserved. In the ferromagnetically coupled surface layers we find an increased moment at the upper, and a reduced moment at the lower edge of the step as expected from the increased, respectively, decreased coordination number with respect to the flat surface. In addition we observe a characteristic canting of the moments close to the edge with respect to the surface normal. This effect is independent of the frustration of the magnetic interactions in the deeper layers of the film and is expected to appear also at steps in homogeneously ferromagnetic films.

The case where the step edge coincides with the boundary between two magnetic domains is shown in Fig. 1(b). Here the important point is that the frustration appearing in the ferromagnetic bilayer at the surface is released by a nearly perpendicular orientation of the moments at the frustrated sites. The strong canting at the step edge also induces a canting of the spins in the deeper layers. In the nonrelaxed collinear configuration, the energy of the configuration (a) with parallel moments is 37.9 meV lower than that of the antiparallel configuration (b). The noncollinear relaxation reduces to 21.4 meV, but configuration (a) remains favored.

Similar noncollinear structures appear also at other steps. As can be seen from Table I, in 4 ML/6 ML or 5 ML/7 ML steps the frustration and hence the noncollinearity reaches the surface at the lower step edge and will hence have a particularly large effect on the magnetic surface anisotropy.

Due to the canting of the magnetic moments in the vicinity of the step the magnetization acquires an in-plane component, for both parallel and antiparallel orientation of the global moments on both sides of the step (see Table II). In a rough film, however, up and down steps will occur in equal number so that the in-plane component of the magnetization will average out over a larger area of the film. The perpendicular component of the magnetization varies essentially continuously across the step, with a small enhancement [configuration (a)], respectively, reduction [configuration (b)] precisely at the step where except for the ferromagnetic top bilayer the moments have a predominant in-plane orientation. Hence the noncollinearity induced by the steps will hardly manifest itself in measurements of the magnetization. On the other hand, we expect a substantial influence on the magnetic anisotropy of the film. Investigations of the growth and morphology[2] of Fe/Cu(100) films have shown that the surface roughness of LT-grown films increases dramatically at average thicknesses between 3 and 5 ML. For a film with a coverage of 5 to 6 ML it can be estimated that the density of steps is very high, up to a step every 10 Å. Hence the magnetic structure of such rough films will be essentially noncollinear. The effect on the magnetic anisotropy is more difficult to estimate. Our recent work on the anisotropy of ideally flat films has demonstrated[15] that the main contribution to a perpendicular anisotropy comes from the ferromagnetically coupled surface layers, with a further contribution from the interface layer. Here we have shown that due to the frustration of the exchange interactions, a noncollinear magnetic ordering appears in the interior of the film and at the interface (if the step occurs within a single magnetic domain) or at the surface (if the step coincides with a domain-boundary). Hence the either the interface or the surface contribution to the anisotropy will be strongly reduced, and the dipole contribution favoring in-plane anisotropy will eventually dominate. It appears that the roughness-induced noncollinearity is the clue to the spin-reorientation transition at 5 to 6 ML in rough films instead at 10 to 12 ML as in flat films.

This work was supported by the Austrian Ministry for Science, Research, and Art through the Center for Computational Materials Science within the project ''Magnetism on the Nanometer Scale'' (Grant No. GZ. 45.378/2-IV).

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