Stable and variable features of the magnetic structure of fcc Fe/Cu(001) films

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After more than a decade of experimental and theoretical studies of the magnetic structure of the fcc Fe/Cu(001) films, most of the researchers agree that the atomic spins of two surface layers are ferromagnetically ordered whereas the magnetic structure of further layers is not ferromagnetic. The conclusions on the magnetic configuration of the nonferromagnetic part vary broadly from a simple collinear layer-by-layer antiferromagnet to complex incommensurate spin-density wave. We report detailed theoretical study of the fcc Fe/Cu(001) films with 6, 7, and 8 ML coverages. The study is based on the density-functional-theory calculations with the code allowing for the consideration of arbitrary noncollinear magnetic configurations. We suggest a view on the magnetism of the Fe/Cu(001) films that includes the grouping of the layers into blocks with robust collinear magnetic structure whereas the variation in the relative directions of the moments of different blocks is energetically relatively inexpensive. The robust magnetic structure of the three surface layers was found to be ⇓⇑⇑. Further layers form the pairs with robust antiferromagnetic structure. We demonstrate that the formation of the robust magnetic blocks corresponds to a certain hierarchy of the interlayer exchange interactions. We suggest that the individual defect pattern of each fabricated film influences before all the relative orientation of the block magnetizations keeping the intrablock structure intact. The calculated interatomic exchange parameters are used to study magnetic thermodynamics of the films within mean-field approximation. In particular we focus on possible physical reasons of the appearance of the steplike features in the temperature dependence of the magnetization detected experimentally. We emphasize the important consequences of the inequivalence of the Fe atoms belonging to different layers of the film. This inequivalence makes the properties of the films to be essentially different from the properties of the corresponding bulk system. Using a rigid-band model we investigate the dependence of the exchange parameters on the electron number. We critically discuss the possibility of the formation of the incommensurate SDW in the thin Fe/Cu(001) films. The brief formulation of the idea of robust blocking and its experimental confirmation was recently published as a letter [H. L. Meyerheim, J.-M. Tonnerre, L. M. Sandratskii *et al.*, Phys. Rev. Lett. **103**, 267202 (2009)].

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I. INTRODUCTION

Fe films grown on the Cu(001) substrate are among the most intensively studied magnetic films.[1–20] They are characterized by the richness of the structural and magnetic properties that are strongly interconnected with each other. After more than decade of experimental and theoretical research a consensus seems to immerge on a number of important aspects of the structure and magnetism of the Fe/Cu(001) films (see for a detailed discussion, e.g., experimental work by Meyerheim *et al.*[10] or recent review by Vaz *et al.*[21]). Three main coverage regimes are distinguished. For thicknesses below about 4 monolayers (ML) the crystal structure is of distorted fcc type and the magnetic structure of the film is ferromagnetic. For thicknesses above about 10 ML the crystal structure becomes of the bcc type whereas the magnetic structure is also ferromagnetic. The information about the magnetic properties of the films with intermediate coverage is controversial.

In the thickness interval between about 5 and 10 ML the crystal structure is of the fcc type similar to the films with smaller coverage. However, the magnetic structure changes strongly. Most of the researchers agree that the moments of the two surface layers order ferromagnetically whereas the moments of further layers order in a nonferromagnetic configuration. The particular magnetic structure of the nonferromagnetic part remains a matter of debate. Some researchers suggested a type-I antiferromagnetic configuration with alternating directions of the moments of the neighboring layers (see, e.g., Refs. 3 and 16). The first-principles calculations find the minimum of the total energy for more complex magnetic structures, e.g., for the double-layer antiferromagnetic structure (see, e.g., Refs. 13 and 15). Qian *et al.*[6] suggested an incommensurate spin-density wave (SDW) as the magnetic structure of the nonferromagnetic part of the film. Spisak and Hafner[19] disagreed with the conclusion of Qian *et al.* and argued that the formation of this type of structure is energetically unfavorable. Amemiya *et al.*[8,9] reported the study of the magnetic depth profiles with the depth-resolved x-ray magnetic circular dichroism (XMCD) measurements. They confirmed the conclusion of Qian *et al.*[6] about the formation of the incommensurate SDW structure. They agree with previous studies with respect to the ferromagnetic structure of the two surface layers. The moment of the third layer was found to be opposite to the moments of the first two layers. It is worth noting that in the structure suggested by Qian *et al.* the moment of the third layer is parallel to the moments of the two upper layers. Therefore in this important respect two experimental works came to opposite conclusions.

Another important discussion point closely related to the distinguishing of the magnetic behavior of the surface and inner parts of the Fe/Cu(001) films of intermediate coverage

concerns the magnetic critical temperature. A number of experimental groups argue to detect higher temperature of the magnetic phase transition for the surface layers compared to the ordering temperature of the inner layers.[2,3,6] Here there is, however, an important conceptual question: can different parts of a thin-film order at different temperatures? Earlier, a similar problem was discussed for semi-infinite three-dimensional (3D) systems.[22–26] In the latter case, most of the researchers agree that different magnetic transition temperatures are possible for the surface region and for the deep inner part of the system. On a qualitative level this conclusion is very plausible. Indeed the information about enhanced exchange interactions at the surface leading to the enhanced magnetic transition temperature decays when going to deeper layers and becomes negligible at large enough depth. This conclusion cannot, however, be extended to the thin films with few atomic layers since the magnetic moment appeared at the surface layer will influence through interlayer exchange interaction the rest of the film and lead to a noninfinitesimal induced moment in all layers. As a result the magnetic order parameter of an inner layer of the thin film becomes nonzero at the same temperature as the net magnetization of the surface layer. According to the definition of the disorder-order magnetic phase transition as the transformation from the state with zero net magnetization to the state with nonzero net magnetization the phase transition should be considered to have taken place simultaneously in the whole film. This formally exact statement can, however, have small importance from experimental and practical points of view since the character of the temperature dependence of the magnetization of an inner layer can be very different from the corresponding temperature dependence of the surface layer. If in the cooling process the magnetization of the inner layer remains small up to a certain temperature T_i below the ordering temperature of the upper layers T_C and experiences a fast increase at temperatures below T_i it is physically appealing to associate the magnetic ordering in the corresponding layer with temperature T_i. Therefore to understand the temperature-driven processes in thin magnetic films it is very important to access layer-resolved temperature dependence of the magnetization.

The layer-resolved information on the magnetization of the thin films is important for both experimental and theoretical studies of the magnetic films. In this respect there is, however, an asymmetry between theory and experiment. In the theoretical studies the layer-resolved information appears as a necessary component of solving the problem. A critical point in the theoretical studies is the formulation of a realistic mathematical model of the exchange interactions in the system. On the other hand, an experimental extraction of the layer-resolved information on the temperature dependence of the magnetization is a very complex and by far not solved problem. The total net magnetization of the films accessible by the magneto-optical Kerr effect provides information on the sum of the contributions coming from different layers. A unique separation of these contributions is not possible and obtaining of the layer-resolved information necessary relies on assumptions. For example, Qian *et al.* assumed that an additional layer added between substrate and film does not change the magnetic structure of the upper layers and therefore the comparison of the films with different numbers of layers leads to the layer-resolved information about the films. The assumption about identical fragments of the magnetic structure of different films is, however, not well founded because of both mutual influence of the layers of the thin films and different defect patterns of different films.

Qian *et al.* measured the temperature dependence of the magnetization for 6, 7, and 8 ML films. For 6 ML and 8 ML films they detected a steplike increase in the magnetization at a temperature T_i that is about 50 K below T_C of the film, whereas for 7 ML film this additional increase was absent. The authors interpreted the temperature of the additional steplike increase as the ordering temperature of the inner layers. The magnetic structure of the inner layers was suggested to be an incommensurate SDW. The absence of the step in the magnetization curve of the 7 ML film was explained by an accidental compensation of the magnetic moments of the inner part of the film reflecting the properties of the incommensurate spin-density wave.

An important experimental information on temperature dependence of the magnetization is reported by Amemiya *et al.*[8] They compared the measurements for two temperatures 130 and 200 K and noticed that the magnetic signal is stronger at 200 K. Since the signal is expected to be higher for higher net magnetization of the film this decrease in the net magnetization with decreasing temperature reveals an antiparallel contribution of the net moment of the inner layers with respect to the magnetization of the surface layers. In this case a strong increase in the magnetization of the inner layers at temperature T_i diminishes the magnetization of the film as a whole. This feature is again opposite to the feature observed by Qian *et al.* for the 6 and 8 ML films who registered a steplike increase in the magnetization. Obviously the two nominally identical films measured by the two groups differ in the relative orientation of the moments of the inner layers with respect to the surface magnetization.

Further experimental and theoretical efforts are needed to advance the understanding of the magnetic properties of the Fe/Cu(001) films. Apparently the films with nominally identical compositions have different defect patterns that can influence both the ground-state magnetic structure and the temperature dependence of the magnetization. Therefore it is important to distinguish between magnetic properties robust to the inevitable differences in the fabricated films and the features that are expected to vary from film to film. This aspect is one of the important focuses of the present paper.

There is a number of earlier studies of the thermodynamics of the fcc Fe/Cu(001) films. Spisak and Hafner[15] used a real-space tight-binding linear-muffin-tin-orbital (TB-LMTO) technique to calculate the ground state and Heisenberg exchange parameters for fcc Fe/Cu(001) films with coverage from one to 6 ML. The exchange parameters were calculated using the torque-force approach by Small and Heine[27] resulting in a formula similar to the formula derived by Lichtenstein *et al.*[28] on the basis of magnetic force theorem. The thermodynamics was studied within an Ising model using Monte Carlo simulations. Camley and Li[16] considered a Heisenberg-type model. Only the coupling between the nearest layers has been included. The values of the exchange parameters were chosen on the basis of experimental infor-

mation on the magnetic transition temperatures. The ground-state magnetic structure was assumed to be ferromagnetic for upper two layers and simple antiferromagnetic for further layers. Correspondingly, two different exchange parameters were chosen, a ferromagnetic one for the upper two layers and an antiferromagnetic one for further layers. The statistical mechanics problem was solved for films up to 11 ML coverage using a self-consistent local mean-field theory. Pajda *et al.*[18] used TB-LMTO method and Lichtenstein's expression for Heisenberg exchange parameters to calculate the Curie temperature of 1 ML film within both mean-field approximation (MFA) and random-phase approximation (RPA). Razee *et al.*[17] used disordered local moment (DLM) approach implemented within Korringa-Kohn-Rostoker (KKR) method in coherent-potential approximation (CPA) to calculate thickness dependence of the Curie temperature for the film coverages from 2 to 8 ML. Summarizing, to our knowledge the temperature dependence of the magnetization has been studied in two works. Spisak and Hafner used calculated exchange parameters and Ising-type model, whereas the calculations by Camley and Li were based on a simplified Heisenberg-type model with parameters chosen on the basis of the experimental information on the magnetic transition temperatures of the films. In works by Pajda *et al.* and Razee *et al.* the estimations of the Curie temperature of the films have been given.

The purpose of the present work is a detailed study of the exchange interactions in the 6–8 ML Fe films on Cu(001). The evaluated parameters of the exchange interactions are used to calculate the layer-resolved temperature dependence of the magnetization. We distinguish between robust features in the properties of the films and the features sensitive to the imperfections of the films. We discuss the formation of step-like features in the temperature dependence of the total film magnetization and comment on the possibility of the incommensurate SDW as the magnetic structure of the inner layers.

Some of the conclusions of the given work were recently briefly formulated in the paper (see Ref. 29) where we emphasize the importance of the hierarchy of the exchange interactions in the Fe/Cu(001) films leading to the formation of the robust magnetic blocks in contrast to a relatively weak exchange connection between blocks. This physical picture was confirmed by the x-ray resonant magnetic scattering measurements allowing for the layer-resolved experimental determination of the film magnetization.[29]

II. CALCULATIONAL TECHNIQUE

The density-functional-theory calculations are performed with the augmented spherical wave (ASW) method.[30] The calculations have been performed in so-called slab geometry where the fcc Fe film is sandwiched between 7–8 Cu layers and 4–5 layers of empty spheres. To simplify calculations this system of layers was assumed to be periodically repeated. The lattice parameter of fcc Fe was chosen equal to the lattice parameter of Cu $a=1.805$ Å.[31]

A most straightforward and widely accepted way to quantitatively describe the exchange interactions in an itinerant-electron system is the mapping of the system on a Heisenberg Hamiltonian. The parameters of the Heisenberg Hamiltonian are found on the basis of the first-principles evaluation of the total-energy differences between various magnetic configurations. A very convenient and efficient method to evaluate the exchange parameters is suggested by the formula derived by Lichtenstein *et al.*[28] that gives the exchange parameters in the form of the second derivatives of the band energy with respect to the infinitesimal deviations of the atomic moments. The derivation of the formula is based on the so-called magnetic force theorem that allows to approximately replace the differences in the total energies of two magnetic configurations by the difference in the sum of the single-particle Kohn-Sham energies. In an ideal Heisenberg system the values of the exchange parameters do not depend on the magnetic configurations used in the calculations. For a complex itinerant-electron system such as fcc Fe films this independence is by far not obvious and must be verified. Also the error introduced by the replacement of the total-energy difference by the band energy difference needs consideration. In the given work we do not use the Lichtenstein's expression for the interatomic exchange parameters. Instead we perform estimations of the energies of selected magnetic configurations as described below.

The basis for the quantitative characterization of the interatomic exchange parameters is the formula of the Heisenberg model

$$H_{eff} = -\sum_{\mu,\nu} \sum_{\substack{\mathbf{R},\mathbf{R}' \\ (\mu\mathbf{R}\neq\nu\mathbf{R}')}} J^{\mu\nu}_{\mathbf{R}\mathbf{R}'} \mathbf{e}^{\mu}_{\mathbf{R}} \mathbf{e}^{\nu}_{\mathbf{R}'}, \tag{1}$$

where the indices μ and ν number different Fe layers and $\mathbf{R}$ and $\mathbf{R}'$ are the two-dimensional (2D) lattice vectors specifying the atoms within layers and $\mathbf{e}^{\mu}_{\mathbf{R}}$ is the unit vector pointing in the direction of the atomic magnetic moment at site $(\mu,\mathbf{R})$.

The interlayer exchange parameters are calculated according the following scheme. Starting with the collinear ground state the moments of layers μ and ν were, first, both rotated clockwise by angle θ and, second, in opposite directions by the same angle. The energy difference of two states takes the form

$$\Delta E^{\mu\nu}(\theta) = 2a[1-\cos(2\theta)]J^{\mu\nu}_0, \tag{2}$$

where $a=1$ if the moments of the layers μ and ν are parallel to each other in the ground state and $a=-1$ if they are antiparallel; the parameter

$$J^{\mu\nu}_0 = \sum_{\mathbf{R}} J^{\mu\nu}_{\mathbf{0R}} \tag{3}$$

characterizes the exchange interaction between layers μ and ν. The knowledge of $\Delta E^{\mu\nu}(\theta)$ for a given θ immediately gives an estimation of $J^{\mu\nu}_0$ [see Eq. (2)].

This relatively simple method does not allow to calculate the intralayer exchange parameters. Here we use so-called frozen magnon approach (see, e.g., Ref. 32 for more details). The DFT calculations are performed for the spiral magnetic configurations for a given layer ν

TABLE I. Layer resolved atomic moments (in μ_B) for fcc Fe/Cu(001) films with 6–8 ML coverage.

	m_1	m_2	m_3	m_4	m_5	m_6	m_7	m_8
6 ML	2.59	2.24	2.20	2.19	2.19	2.83		
7 ML	2.25	2.23	2.15	2.17	2.17	2.18	2.82	
8 ML	2.59	2.22	2.17	2.14	2.17	2.17	2.17	2.81

$$\mathbf{e}_{\mathbf{R}}^{\nu} = [\sin\,\theta^{\nu}\cos(\mathbf{qR}), \sin\,\theta^{\nu}\sin(\mathbf{qR}), \cos\,\theta^{\nu}], \tag{4}$$

whereas the moments of other layers keep the ground state direction. Using the notation $E^{\nu}(\theta,\mathbf{q})$ for the energies of these configurations the exchange parameters between atoms of the layer ν are obtained by means of the 2D back Fourier transformation

$$J_{\mathbf{0R}}^{\nu\nu} = -\frac{1}{N}\sum_{\mathbf{q}} [E^{\nu}(\theta,\mathbf{q})/\sin^2\,\theta]\exp(-i\mathbf{qR}). \tag{5}$$

For the mean-field calculation of the temperature dependencies it is convenient to introduce a cumulative intralayer parameter

$$J_0^{\nu\nu} = \sum_{\mathbf{R}} J_{\mathbf{0R}}^{\nu\nu}. \tag{6}$$

To simplify the notations we will use J_0^{ν} instead of $J_0^{\nu\nu}$ in all cases where this does not lead to confusion.

The mean-field approximation for the calculation of the temperature dependence of the layer magnetizations of the thin film takes the form of a system of equations that must be solved self-consistently for each temperature with respect to the average layer magnetizations $\langle \mathbf{e}_{\nu} \rangle$

$$\langle \mathbf{e}_{\mu} \rangle = L\left(\frac{1}{kT}\sum_{\nu} J_0^{\mu\nu}\langle \mathbf{e}_{\nu} \rangle\right). \tag{7}$$

Here the Langevin function is defined as

$$L(x) = \coth(x) - 1/x. \tag{8}$$

The following comment on the applicability of the MFA to the study of the thermodynamics of the thin films is useful. According to the Mermin-Wagner[33] theorem the 2D system described by isotropic Heisenberg Hamiltonian does not order at any nonzero temperature. The reason for the absence of the long-range magnetic ordering is the Goldstone mode of the gapless long-wavelength magnetic excitations. The destructive role of such excitations increases with decreasing dimension of the system. On the other hand, the magnetic anisotropy present in the 2D systems results in a gap in the spectrum of long-wavelength magnetic excitations. Even a relatively small gap is sufficient to cardinally diminish the role of the long-wavelength excitations.[18,34] This diminishing of the role of the long-wavelength magnetic excitations in the real materials takes place through the magnetic anisotropy leading to the energy gap in the magnon spectrum at zero wave vector. Another factor is the presence of the substrate that effectively increases the dimensionality of the magnetic system. The Curie temperature of several hundreds Kelvins and the bulklike form of the temperature dependence of the magnetization observed for the fcc Fe/Cu(001) films[3,6] show that the magnetic ordering is governed by strong exchange interactions between neighboring magnetic atoms. Since these interactions are adequately taken into account within the MFA it can be expected that the MFA captures important features of the thermodynamics of the fcc Fe/Cu(001) films at least on a qualitative level. On the other hand, it is well known that the MFA overestimates the Curie temperature of the Heisenberg systems. It also does not give correct estimation of the critical exponents describing the approaching by the magnetization of the zero value at the point of the phase transition. The application of more precise but also more time consuming methods such as random phase approximation and Monte Carlo simulation to the study of the thermodynamics of fcc Fe/Cu(001) films is desirable. As mentioned in the Introduction Spisak and Hafner[15] performed the Monte Carlo simulation for an Ising type of the Hamiltonian. However, the use of the Heisenberg-type models for the description of the fcc Fe/Cu(001) films seems to be more appropriate. Actually as we show below even Heisenberg-type models are not sufficient to describe exchange interactions in the fcc Fe/Cu(001) films. The account for higher-order spin interactions and longitudinal fluctuations can be necessary to reach quantitative theoretical description of the magnetic properties of the films. These aspects of the problem should, however, be the subject of separate study.

III. DENSITY-FUNCTIONAL-THEORY STUDY OF MAGNETIC STATES OF THE FE/CU(001) FILMS

First, we performed calculations for various collinear magnetic configurations of the films. The lowest-energy states obtained for the 6–8 ML films are ⇑⇑⇓⇓⇑⇑, ⇓⇑⇑⇓⇓⇑⇑, and ⇓⇓⇑⇑⇓⇓⇑⇑. Throughout the paper we number the layers starting from the interface layer. Correspondingly in the notation ⇑⇑⇓⇓⇑⇑ the most left arrow corresponds to the interface layer. The ground-state values of the atomic moments are collected in Table I. The calculations performed for films with smaller coverage show that we get good agreement with the results by Asada and Bluegel[13] who systematically studied the energetics of the collinear magnetic structures in the fcc Fe/Cu(001) films.

To verify the possibility of a noncollinear ground state we carried out self-consistent calculations of the magnetic structure starting from different arbitrary chosen noncollinear magnetic configurations. A number of interesting and unexpected results have been obtained in these calculations. The resulting numerically self-consistent magnetic structures were noncollinear and different for different initial configu-

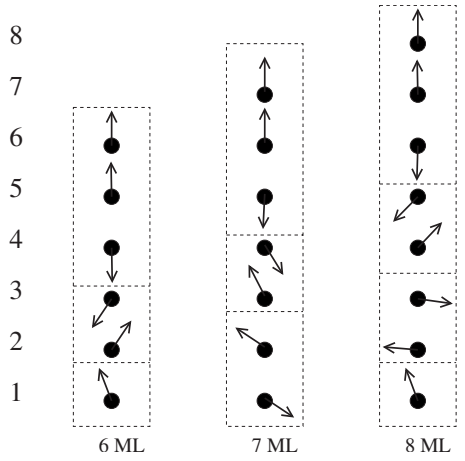

FIG. 1. Typical numerically self-consistent magnetic configurations of the 6–8 ML fcc-Fe/Cu(001) films. Broken-line boxes show the blocks of layers with robust magnetic structure. The layers are numbered from interface to surface.

rations. In Fig. 1 we show typical numerically self-consistent states obtained in such calculations for the films of different thickness. In agreement with previous DFT studies none of the noncollinear magnetic structures had the total energy lower than the lowest-energy collinear structure. The energy difference was, however, rather small, typically of the order of 0.1 mRy per Fe atom.

The analysis of the quasistable structures of the type shown in Fig. 1 has shown that they have the following common features: (i) the magnetic moments of the three surface layers are almost collinear to each other and form a magnetic configuration of the type ⇓⇑⇑. (ii) Deeper layers form pairs with the directions of the moments in the pairs being close to collinear antiparallel. (iii) The relative directions of the moments of different groups vary strongly from calculation to calculation.

The results of these calculations agree with the main body of previous studies concerning the parallel orientation of the moments of the two surface layers. They also support the conclusion of the experimental work by Amemiya *et al.*[8] about an antiparallel direction of the moment of the third layer relative to the moments of the first two layers.

Simultaneously these results suggest a modification of the picture proposed in previous studies. Thus instead of the considering the film as the combination of two parts: two upper ferromagnetic layers and a nonferromagnetic rest of the film, our calculations suggest a different physically relevant grouping of the layers. The surface part of the film is formed not by two ferromagnetic layers but by three layers ordered as ⇓⇑⇑. The further layers form pairs. The relative directions of the moments of these pairs are close to collinear antiparallel. The dividing of the 6–8 ML films into groups of layers according to this scheme takes the form [1][23][456], [12][34][567], and [1][23][45][678]. Here the square brackets contain the numbers of the layers belonging to one group. The calculations suggest that the relative directions of the moments within the groups are substantially robuster features of the magnetic structure of the films than the relative directions of the moments of different groups.

As already mentioned above the formation of the robust magnetic blocks was recently detected experimentally.[29] The directions of the moments of different blocks were found to be noncollinear.[35]

Interestingly, alone this type of the grouping of the layers allows to give an interpretation of the observation by Qian *et al.*[6] that an additional step appears in the temperature dependence of the total film magnetization for 6 and 8 ML films but not for 7 ML film. Indeed, because of the antiparallel directions of the moments of the layers, the groups containing two layers can be considered as magnetically almost compensated and, therefore, not contributing substantially to the net magnetization of the film. On the other hand, the surface group of all films and the interface group [1] of the 6 and 8 ML films are not magnetically compensated. Therefore if the interface layer of the 6 and 8 ML films orders at lower temperature then the surface part of the film and the directions of the ordered moments of the both parts are parallel to each other, the appearance of the positive step in the magnetization curve of the type reported by Qian *et al.* is expected.

Although useful for the interpretation of the experimental results of Qian *et al.* this qualitative consideration cannot replace a quantitative study of the exchange interactions and temperature dependencies of magnetization that are discussed in the following sections.

IV. EXCHANGE PARAMETERS AND THERMODYNAMICS OF 6 ML FILMS

We begin with the detailed study of the 6 ML film.

A. Interlayer exchange parameters

In Fig. 2, we present the interlayer exchange parameters for pairs of neighboring layers. The parameters are given as a function of the angle between the moments of the layers used in the calculations [see Eq. (3)]. The left panel shows the results of the self-consistent calculations (SLFCs) whereas the right panel contains the results of the force theorem (FT) calculations.

The nearest-neighbor interactions presented in Fig. 2 are the strongest and therefore determining the main features of the magnetic structure of the film and of its thermodynamics. However, the exchange interactions between more distant layers are also sizable and can play significant role in the formation of the relative orientations of the magnetic moments. In Fig. 3 we present as an example the interactions between the interface and surface layers and all other layers. These interactions are also θ dependent. Some of them change sign with increasing θ. Since the weak exchange interactions between distant layers are more sensitive to finer details of the electron structure than the nearest-neighbor (nn) exchange interactions it is to expect that they can strongly depend on both the particular defect pattern of the fabricated film and on the method of solving the DFT problem. Below discussing the statistical mechanics studies we will begin with the calculation where all interlayer exchange interactions are included. Next, intending to distinguish the most robust features of the exchange interactions we neglect

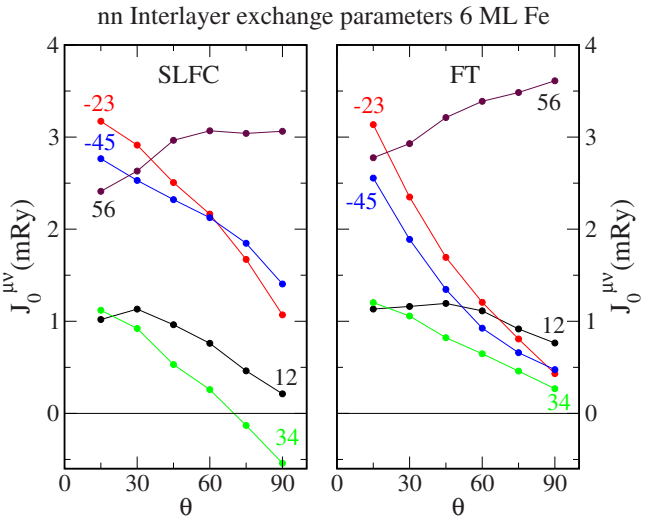

FIG. 2. (Color online) The interlayer exchange parameters for pairs of neighboring layers of 6 ML film. The parameters are given as functions of the angle between the moments of the layers used in the calculations. The left panel shows the results of the self-consistent calculations, whereas the right panel contains the results of the force theorem calculations. The numbers give the pairs of interacting layers. The mines sign before the number means that the sign of the exchange parameter is reversed for better visualization.

the interlayer exchange interactions beyond the nn interactions.

Let us, first, consider the SLFC parameters calculated for the deviation angle θ=15°. (To remind, since in the calculation of the exchange parameters the moments of different planes deviate in different directions the angle between moments is 2θ.) The strongest interlayer interaction is between second and third layers followed by 4–5 and 5–6 interlayer interactions. The interactions 1–2 and 3–4 are substantially weaker. This difference in the values of the interactions

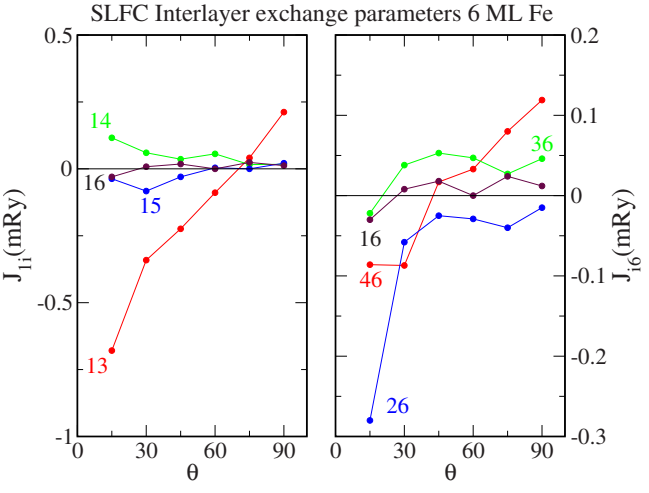

FIG. 3. (Color online) The interlayer exchange parameters for pairs of layers that are not nearest neighbors in the film. The parameters are given as functions of the angle between the moments of the layers used in the calculations. Both panels show the results of the self-consistent calculations. The reader should notice the difference in the scales of the ordinate axes in this figure and in Fig. 2.

agrees with the splitting of the layers into groups [1], [23], and [456] with robust intragroup magnetic structure and much less robust relative orientation of the magnetizations of different groups.

The exchange parameters strongly depend on the angle θ revealing serious restrictions in the applicability of the Heisenberg model to the description of the energetics of the fcc Fe/Cu(001) films. A more complex model Hamiltonian must be used to fully describe the energetics of the system in terms of the parameters independent of the characteristics of magnetic configurations. The higher-order pair exchange interactions such as a biquadratic exchange and/or the interactions between three and more atomic moments have to be taken into account. In the present paper we do not consider these more complex types of interactions. Instead we calculate the temperature behavior of the magnetization using the Heisenberg parameters corresponding to different θ and analyze the θ dependence obtained.

Figure 2 shows that the ferromagnetic exchange interaction between two upper layers increases with increasing θ, whereas all other interactions are strongly decreasing. The 3–4 interaction even changes the sign at large θ demonstrating that this partial interaction can favor the reorientation of the relative directions of the moments of the third and fourth layers. The increase in the 5–6 interaction reveals the robustness of the ferromagnetism of the upper two layers and explains the consensus of the experimental and theoretical studies concerning this feature despite strong scattering in other properties. The separation of the interlayer exchange interactions into stronger 2–3, 4–5, and 5–6 interactions and weaker 1–2 and 3–4 interactions remains valid up to largest θ.

The physical consequences of the θ dependence of the exchange parameters can be viewed from two different points. First, this complex behavior of the exchange interactions helps to understand the origin of the numerous quasistable states obtained in the DFT calculations with different starting configurations. Note that these calculations correspond to zero temperature and reflect the properties of the energy as a function of the magnetic configuration. Second, in the study of the thermodynamics the angle dependence of the exchange parameters can be related to the temperature dependence of the effective interatomic exchange parameters. Indeed, the process of thermal magnetic disordering results in increasing deviations of the atomic moments from the ground-state directions. If to associate a certain extent of local magnetic disorder with each temperature and express this disorder in terms of characteristic angle θ, the θ dependence of the exchange parameters can be considered as determining the temperature dependence of these parameters. The assignment of a certain angle to a certain temperature is not, however, straightforward since it must rely on the knowledge of the temperature dependence of the short-range magnetic order (SRMO). Although it is obvious that the SRMO decreases with increasing temperature the detailed character of the temperature dependence of this characteristic remains the matter of debates even for the elemental bulk ferromagnets.[36,37] The calculations of the temperature dependence of the SRMO is beyond the scope of the given paper. We investigate the sensitivity of the Curie temperature and

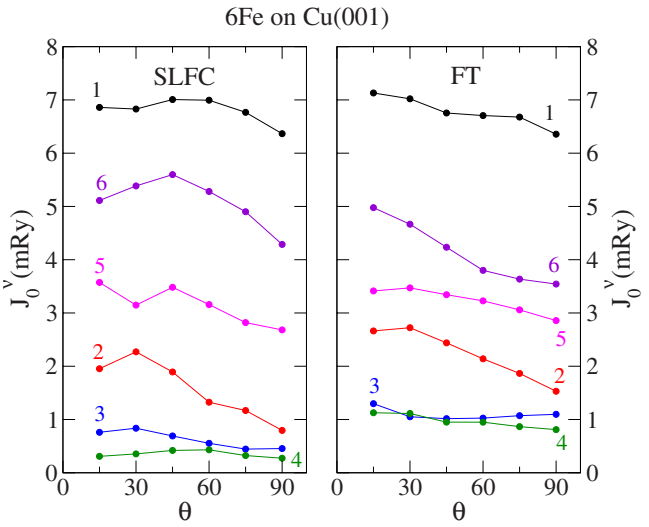

FIG. 4. (Color online) Intralayer exchange parameters J_0^ν calculated for the 6 ML film both self-consistently (left panel) and with the use of the force theorem (right panel). The parameters are given as functions of the angle between the moments of the layers used in the calculations. The labels at the curves give the corresponding layer number ν.

temperature dependence of the magnetization to the variation in the exchange parameters taking into account that the application of low-θ parameters corresponds to the assumption of a strong SRMO at the Curie temperature whereas the use of the exchange parameters obtained for $\theta=45°$ corresponds to the neglect of the SRMO at T_C.

The comparison of the exchange parameters obtained in the self-consistent calculations with the parameters resulting from the FT calculations (Fig. 2) shows that on a qualitative level both provide a similar picture. For example the nearest-neighbor parameters at $\theta=15°$ are rather close to those obtained self-consistently and preserve the separation into stronger and weaker interacting pairs. Also here the 5–6 interaction increases with increasing θ, whereas other interactions have the trend to decrease. However, quantitatively the difference is substantial. For instance 2–3 and 4–5 interactions decrease stronger with increasing θ in contrast to 1–2 and 3–4 interactions that decrease weaker. The error of the use of the FT instead of the self-consistent calculations is expected to increase with increasing θ.

B. Intralayer exchange interactions

Above we discussed the calculated parameters of the interlayer exchange interactions. To study the thermodynamics of the films the knowledge of the interlayer interactions is not sufficient. The process of the thermal magnetic disorder depends on both intralayer and interlayer exchange interactions.

In Fig. 4 we show intralayer parameters J_0^ν calculated for the 6 ML film both self-consistently and with the use of the force theorem. The parameters are strongly layer dependent. The θ dependence is in general weaker than for the interlayer interactions (Fig. 2).

Two patterns are again qualitatively similar. The strongest interaction is obtained for the interface layer followed by the first and second surface layers and second interface layer. The weakest intralayer exchange interactions are in the two middle layers. On the quantitative level there are substantial differences in the self-consistent and FT patterns. To these differences belong the θ dependence of the J_0^6 and the values of J_0^2, J_0^3, and J_0^4. For the inner layers 2–4 the J_0^ν parameter from the SLFC calculations are always smaller than from the FT calculations. The difference is especially remarkable for the fourth layer where J_0^4 calculated, for example, for $\theta=15°$ drops from 1.13 mRy for FT calculation to 0.31 mRy for SLFC calculation.

It is worth noting that the layer dependence of the exchange parameters is much stronger than the layer dependence of the magnetic moments (see Table I). For example, if we compare the nn interlayer exchange parameters calculated for $\theta=15°$ the strongest exchange interaction 2–3 is more than three times larger than the weakest exchange interaction 1–2. For the intralayer exchange parameter we get a huge value of 22 for the relation between strongest J_0^1 and weakest J_0^4. These results demonstrate that the values of interatomic exchange interactions are very sensitive to the details of the electronic structure. Since the interatomic exchange interactions govern the thermal magnetic disordering the strong variation in the exchange interactions in the film can result in a strong difference in the temperature dependence of the magnetization of different layers. This means that the magnetization depth profile at an elevated temperature can be strongly different from the magnetization profile in the ground state. Not only the relative values but also the relative orientations of the atomic moments can be temperature dependent. Below we will discuss this issue in some detail.

C. Thermodynamics of 6 ML fcc Fe/Cu(001) film

We begin with the discussion of the temperature dependence of the magnetization calculated with the SLFC exchange parameters obtained for $\theta=15°$ (Fig. 5). The calculations are performed in the heating regime with temperature increasing from zero to values corresponding to the paramagnetic state. At each temperature the layer magnetizations obtained on the previous temperature step are used as starting values for the MFA self-consistent procedure. The figure shows that the magnetizations of the second and third layers drop at substantially lower temperature than the magnetization of other layers. If to associate the inflection point separating low-temperature concave behavior from high-temperature convex behavior with an effective ordering temperature of the corresponding layer, we find that for the second and third layers this temperature is about 500 K. It is substantially lower than the Curie temperature of the film $T_C^{MFA}=770$ K. At about 700 K the system experiences the first-order phase transition that is caused by the flipping of the direction of the moment of the interface layer. The discontinuity of the magnetization of the interface layer is about $0.54\mu_B$, the discontinuity in the second layer magnetization is $0.12\mu_B$. The magnetization of the third layer remains close

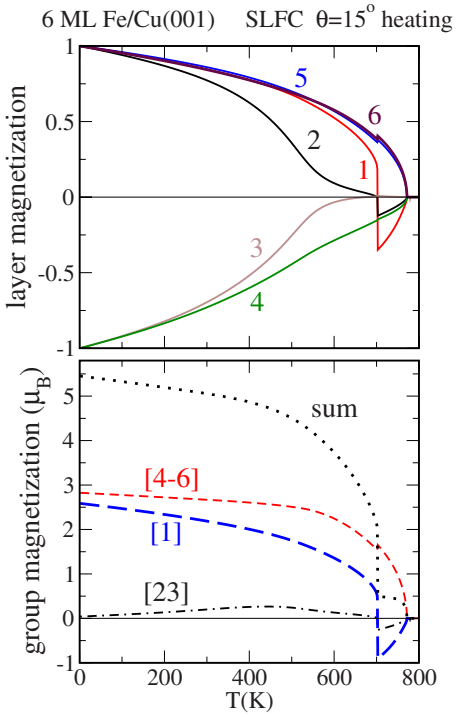

FIG. 5. (Color online) The temperature dependence of the magnetization. The calculations are performed with the SLFC exchange parameters in the heating regime. The exchange parameters are calculated for $\theta=15°$. The exchange interactions between all layers are taken into account. Upper panel: layer-resolved temperature dependence of the magnetization. All curves are normalized to unity at $T=0$. Lower panel: group magnetizations for the groups of layers [1], [23], and [456]. The total magnetization of the film.

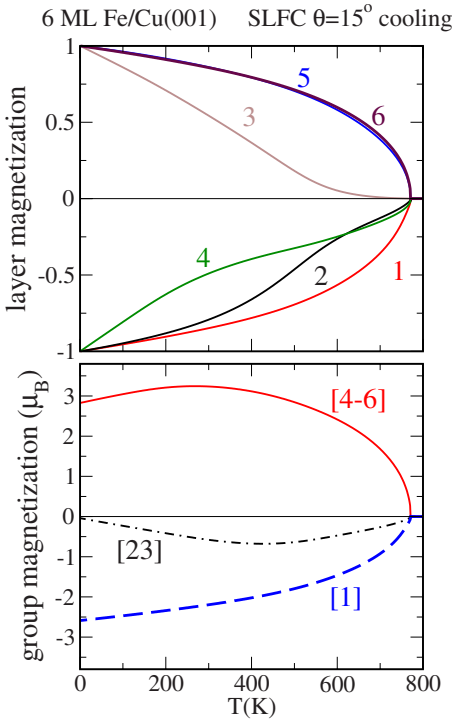

FIG. 6. (Color online) The same as in Fig. 5 but in cooling regime.

to zero. The discontinuity of the magnetizations of other layers is weak.

The analysis of the calculations shows that the origin of this first-order phase transition is in weak antiferromagnetic interactions between two surface and two interface layers. As long as the magnetic moments of the third and fourth layers are comparable in value to the moments of other layers these weak antiferromagnetic interactions do not play important role since the relative directions of the moments are governed by strong nn interactions. When, however, the magnetizations of the third and fourth layers drop strongly as a result of the thermal disordering, the interaction between the distant layers with large net moments becomes important and leads to the change in the relative orientation of the layer magnetizations. In the lower panel of Fig. 5 we show the temperature dependence of the magnetization of the three groups of layers and the total magnetization. As the result of the order-order phase transition the total magnetization has a steplike feature at about 700 K.

The occurrence of the first-order phase transition in the heating process makes interesting to perform the MFA simulations for the cooling process starting at the temperature above the Curie temperature and decreasing it to 0 K. The corresponding magnetization curves are shown in Fig. 6. For temperatures above 700 K the curves are identical to those corresponding to the heating process. No additional phase transition is, however, obtained. The high-temperature magnetic structure ⇓⇓⇑⇓⇑⇑ is preserved up to 0 K. This irreversibility of the thermal processes in the MFA simulations can be explained as follows. The configuration ⇓⇓⇑⇓⇑⇑ corresponds at each temperature to a self-consistent solution of the MFA equations and therefore to a minimum of the free energy. This minimum can be either local or global. It is characteristic for the first-order phase transitions that the same state corresponds to a global minimum for some temperatures and to a local minimum for other temperatures. The temperature at which the energies of two minima become equal corresponds to the temperature of the phase transition. In the nature, the system transforms from the state at a local minimum to the global-minimum state by means of the fluctuations that overcome the barrier between local and global minima. These transforming fluctuations are, however, not present in the MFA simulations. This explains why in the cooling process the system remains in the ⇓⇓⇑⇓⇑⇑ state up to 0 K although the ground state of the system is ⇑⇑⇓⇓⇑⇑ and therefore, in the nature, the transition from ⇓⇓⇑⇓⇑⇑ to ⇑⇑⇓⇓⇑⇑ must take place.

This explanation rises the question: why system does not remain in the ⇑⇑⇓⇓⇑⇑ state during the heating process. The reason for this is the absence of a self-consistent MFA solution for the ⇑⇑⇓⇓⇑⇑ state for $T>T_i$, where T_i is the temperature of the first-order phase transition in Fig. 5. This means that the ⇑⇑⇓⇓⇑⇑ configuration does not correspond to a minimum of the free energy for $T>T_i$ and the system transforms discontinuously to ⇓⇓⇑⇓⇑⇑ structure.

For comparison, we repeated MFA simulations using the exchange parameters obtained with the FT calculations for $\theta=15°$. These simulations (not shown) also give the first-order phase transition in the heating process. The tempera-

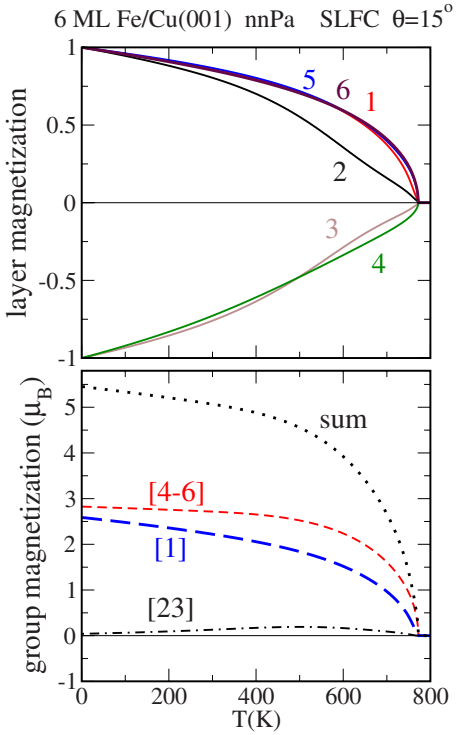

FIG. 7. (Color online) The same as in Fig. 5 but with only nearest-neighbor interlayer interactions taken into account.

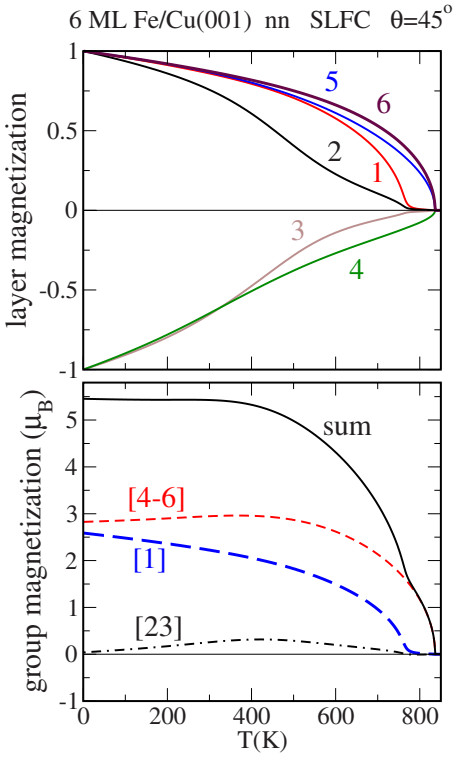

FIG. 8. (Color online) The same as in Fig. 7 but with exchange parameters calculated for θ=45°.

ture of the transition and the details of the temperature dependence of the layer magnetizations differ, however, considerably.

For us, an important aspect of the consideration of the first-order phase transition is the demonstration that it can lead to a steplike feature in the temperature dependence of the total magnetization. Although the possibility of such a phase transition in thin films is plausible, a reliable prediction of its occurrence and its temperature for a concrete film is presently hardly possible since, as stated above, the weak exchange interactions between distant layers are expected to be sensitive to both the defect pattern in the film and the details of the technique for the DFT calculations.[38] Therefore after having emphasized the importance of the results presented in Fig. 5, in the rest of the paper we will consider simpler models where only nn interlayer exchange interactions are taken into account. This simplification agrees with our strategy to separate the most robust features in the exchange interactions and thermodynamics of the films.

If we neglect the interlayer exchange interactions beyond nn the temperature dependence of the magnetization takes the form shown in Fig. 7. In this case no first-order phase transition is obtained and the heating and cooling processes are identical. On the other hand, the Curie temperature of the film and the temperature dependence of the magnetization of the surface group changes rather weakly. The $m(T)$ dependence for the interface layers 1–3 experienced stronger changes. The inflection point at about 500 K seen on $m_2(T)$ and $m_3(T)$ curves in Fig. 5 does not appear in the nn calculations. The absence of the first-order phase transition changes strongly the behavior of the interface magnetization $m_1(T)$ at temperatures above 700 K.

To study the influence on the thermodynamic properties of the θ dependence of the exchange parameters we plot in Fig. 8 the temperature dependence of layer magnetizations calculated for θ=45°. The comparison of Figs. 7 and 8 shows that the use of the exchange parameters calculated for θ=45° leads to a change in the Curie temperature from 770 to 840 K. There are substantial changes in the form of the magnetization curves. Now there is clear difference in the character of approaching zero between the magnetizations of 1–3 and 4–6 layers. The magnetizations of the 4–6 layers approach zero with an infinite slope at the Curie temperature of the film whereas the magnetizations of the 1–3 layers become very small already at temperature of about 770 K and approach the Curie temperature of 840 K in the form of a convex curve close to the abscissa axis. Effectively we get in this case two different Curie temperatures, correspondingly, for the 4–6 layers and for the 1–3 layers. The Curie temperature of the 4–6 layers is at the same time the Curie temperature of the film as a whole in a strict definition of the order parameter. The reason for the separation of the Curie temperatures of the two groups of layers obtained for the exchange parameters corresponding to θ=45° is in strongly decreased 3–4 exchange interaction compared to the θ=15° case (see Fig. 2).

This decreased interaction is now not sufficient to make the thermal disordering a common process for the whole film. Taking J_{34} to be exactly zero does not change the general form of the curves (not shown). The complete separation of the thermodynamics of the 1–3 and 4–6 layers leads, however, to changes in details: (i) the magnetizations of the 1–3 layers become now exactly zero at temperature of about 760

K and (ii) they approach zero with infinite slope. Correspondingly the steplike feature in the total magnetization takes the form of a sharp kink with discontinuous change in the slope. For nonzero J_{34} the feature is smoothed but still clearly seen (Fig. 8).

Putting $J_{34}=0$ in the calculation with exchange parameters obtained for $\theta=15$ also leads to the separation of the Curie temperatures of the two parts of the film (not shown). However, the difference of the two Curie temperatures is in this case rather small: 753 K for layers 1–3 and 770 K for layers 4–6. The closeness of the Curie temperatures is an accidental result of the balance of the intralayer and interlayer exchange interactions in the two parts of the film. A stronger intralayer exchange interaction in the interface layer is compensated by stronger interlayer exchange interaction of the surface layers. With J_{34} taken into account the exchange interaction between different parts of the film is sufficient to result in a common concave approaching of zero by the magnetizations of all layers (Fig. 7).

The temperature dependences of the layer magnetizations shown in Figs. 5–8 were obtained with the exchange parameters determined on the basis of the self-consistent calculations of magnetic configurations. The application of the FT exchange parameters calculated for $\theta=15°$ gives in the case of the nn approximation the physical picture that is qualitatively similar to the picture obtained with the SLFC parameters. Since the FT calculations of the exchange parameters are much less expensive, in the studies of the 7 and 8 ML films reported below we restrict ourselves to the use of the FT exchange parameters. In the statistical mechanics simulations for the 7 and 8 ML films only nn interlayer parameter are taken into account.

V. 8 ML FCC FE/CU(001) FILMS

Next we discuss the films with 8 ML thickness. The number of layers in such a film has the same parity as for the 6 ML films discussed above. As we will see this brings certain similarities in the properties of the films. The study of the 7 ML film will be presented in the next section.

The nn interlayer exchange parameters are plotted in Fig. 9. Note that in the MFA simulations reported in this and following sections we use the exchange parameters obtained for $\theta=15°$. This is logical since the FT calculations give the most reliable results for small deviation angles. We, however, decided to show the θ dependences of the exchange parameters in Fig. 9 and Figs. 12, 14, and 15 to allow for a better comparison of different physical situations studied and to visualize the scale of the angle dependence in various cases that can be stimulating for further studies with more complex model Hamiltonians.

There are clear similarities in the behavior of the exchange parameters of the 6 and 8 ML films. Considering the parameters corresponding to $\theta=15°$ we get stronger interactions for pairs 7–8, 6–7, 4–5, and 2–3 and weaker interactions for pairs 5–6 and 1–2. This hierarchy of interactions corresponds to the grouping of layers [1][23][45][678] following from the self-consistent DFT calculations discussed above. The layers within the same group interact stronger

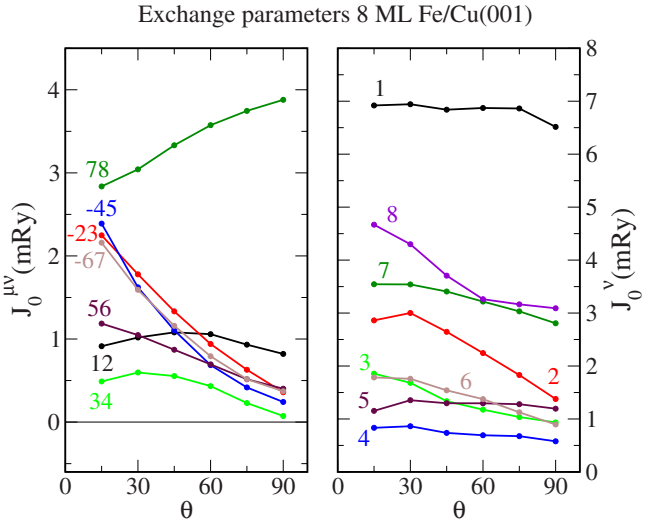

FIG. 9. (Color online) Nearest-neighbor interlayer (left panel) and intralayer (right panel) exchange parameters of the 8 ML fcc Fe/Cu(001) film as a function of angle θ used in the calculations. Calculations are performed with the use of the force theorem. The labeling of the curves corresponds to the labeling used in Figs. 2 and 4.

than the layers of different groups. We again obtained strong θ dependence of the exchange parameters. The ferromagnetic J_{78} parameter increases with increasing θ whereas the main part of other parameters decrease.

The comparison of intralayer exchange parameters for 6 and 8 ML films [Figs. 4 and 9] also shows many similar features. The strongest intralayer exchange interaction of about 7 mRy is obtained for the interface layer. The next in value interactions correspond to the two surface layers followed by the second interface layer. The inner layers have the smallest intralayer exchange parameters.

In the MFA simulation of the thermodynamics of 8 ML film with exchange parameters obtained for $\theta=15°$ (see Fig. 10) the interface magnetization approaches zero at 760 K that is only by 20 K smaller than the corresponding temperature for the surface magnetization determining the Curie temperature of the film. The magnetizations of the [23] and [45] blocks are very weak. In the $\Downarrow\Downarrow\Uparrow\Uparrow\Downarrow\Downarrow\Uparrow\Uparrow$ configuration the magnetizations of the [678] and [1] blocks strongly compensate each other. Resulting in a small peak of the total magnetization just below the Curie temperature and monotonous decrease with decreasing temperature. This form of the m(T) dependence differs from those obtained in the measurements of Qian *et al.*[6] The difference $m[678]-m[1]$ is close to a typical bulk magnetization and is in good correlation with the bulklike dependence measured by Qian *et al.* In the theoretical curve there is a very weak feature at about 760 K that hardly can be detected experimentally. Thus the steplike feature in the temperature magnetization of the 8 ML film is not obtained in these simulations.

In the context of this difference between experiment and theory it is interesting to address the question if the Fe-Cu intermixing present at the Fe-Cu interface can be the reason for the appearance of the steplike feature in the temperature dependence of the total magnetization. The intermixing is

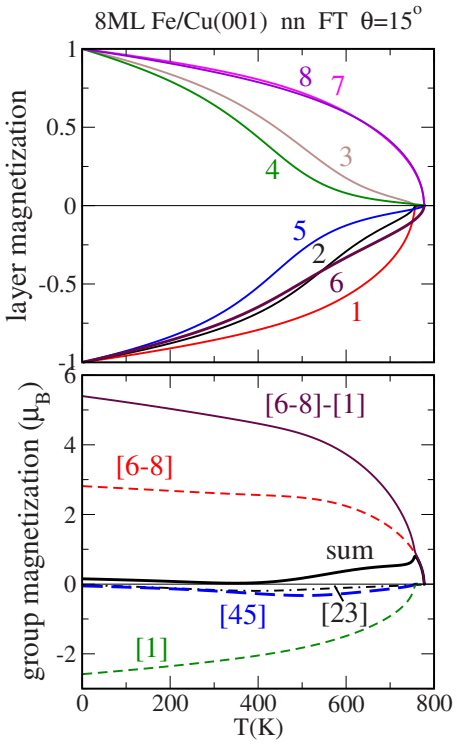

FIG. 10. (Color online) The temperature dependence of the magnetization for 8 ML film. The simulations are performed with the FT exchange parameters calculated for $\theta=15°$. The exchange interactions between nearest layers only are taken into account. Upper panel: layer-resolved temperature dependence of the magnetization. All curves are normalized to unity at $T=0$. Lower panel: group magnetizations for the groups of layers [123], [45], [67], and [8]. The total magnetization of the film.

known to be strong in particular for the films produced by pulse laser deposition.[7,39]

To simulate a 30% intermixing in the MFA calculation the following changes have been made in the model: (i) the atomic moment of the interface layer is decreased by 30%, (ii) the intralayer exchange interactions for the interface layer are multiplied by factor $0.7^2=0.49$, and (iii) interlayer exchange parameter J_{12} is multiplied by factor 0.7. The calculations with these parameters result in substantially changed form of the magnetization $m_1(T)$ of the interface layer (Fig. 11). The interface magnetization approaches zero at about 500 K that is at substantially lower temperature compared to the surface layers. The sum of the surface and interface magnetizations results now in a negative-step feature at 500 K, whereas the difference $m[678]-m[1]$ gives a positive step similar to the step reported by Qian *et al.*

This numerical experiment leads us to the conclusion that the steplike features in the temperature dependence of the magnetization can be the consequence of the intermixing at the interface of the film and substrate. Obviously the intermixing can substantially vary from film to film resulting in different appearance of the steplike features for different films.

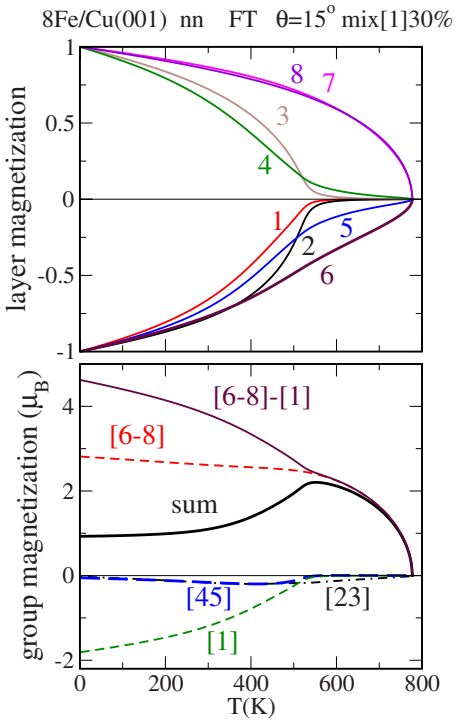

FIG. 11. (Color online) The same as in Fig. 10 but with the Fe-Cu intermixing in the interface layer.

VI. 7 ML FCC FE/CU(001) FILMS

The calculated parameters of the exchange interactions between nn layers are given in Fig. 12. We obtain strong 6–7, 3–4, and 5–6 interactions. The 1–2 interaction is now in an intermediate region. 4–5 and 2–3 interactions are substantially weaker. Again the grouping of the layers [12][34][567] is supported on a qualitative level.

In the pattern of intralayer exchange interactions for the 7 ML film we get substantial difference compared to 6 and 8 ML films. The most important difference is the decreased

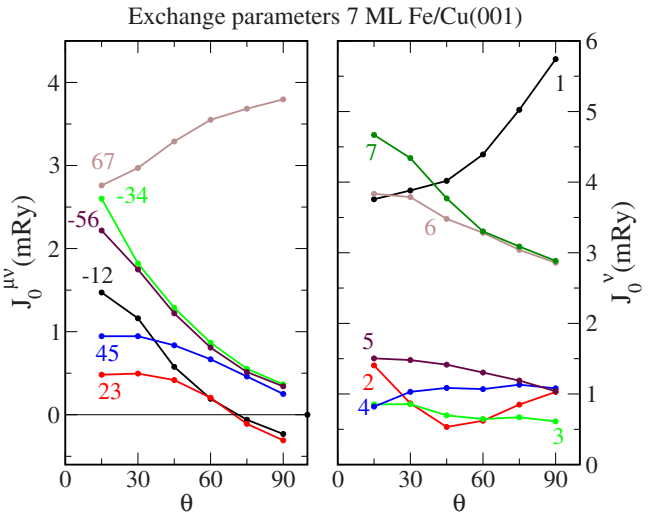

FIG. 12. (Color online) Nearest-neighbor interlayer (left panel) and intralayer (right panel) exchange parameters of the 7 ML fcc Fe/Cu(001) film as a function of angle θ used in the calculations. Calculations are performed with the use of force theorem.

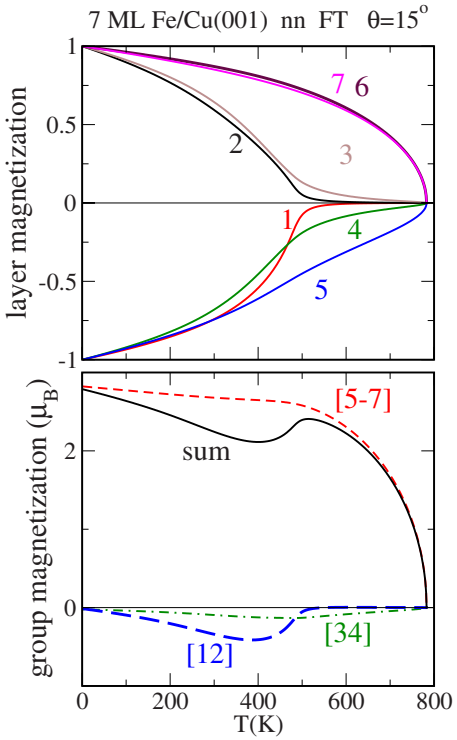

FIG. 13. (Color online) The temperature dependence of the magnetization for 7 ML Fe/Cu(001) film. The calculations are performed with the FT exchange parameters. The exchange parameters are calculated for $\theta=15°$. The exchange interactions between nearest layers only are taken into account. Upper panel: layer-resolved temperature dependence of the magnetization. All curves are normalized to unity at $T=0$. Lower panel: group magnetizations for the groups of layers [12], [34], and [567]. The total magnetization of the film.

value of the intralayer exchange interaction for the interface layer. For $\theta=15°$, parameter J_0^1 has value of 3.76 mRy compared to 7.13 mRy for 6 ML film and to 6.92 mRy for 8 ML film. J_0^1 increases with increasing θ. However, at $\theta=45°$ it still close to 4 mRy.

The results of the MFA simulation with exchange parameters corresponding to $\theta=15°$ are presented in Fig. 13. The behavior of the magnetizations of different layers is very different. The effective Curie temperature of the surface 5–7 layers is close to 780 K and determines the Curie temperature of the film as a whole. The effective Curie temperature of other layers is substantially smaller. For layers 1–3 it is about 500 K. In the group-magnetization plot we see that the surface three layers give a bulk-type contribution whereas the pairs of layers within the groups [34] and [12] compensate each other strongly. This compensation is almost perfect for the [34] group. For the [12] group the difference in the interface layer 1 and inner layer 2 leads to less complete mutual compensation. As a result the temperature dependence of the [12]-group magnetization is strongly nonmonotonous and has a maximum at about 400 K. The contribution of the [12] group to the total magnetization leads to a nonmonotonous feature also in the total magnetization. Such a feature is in principle observable although it is less pronounced than the step arising as a sum of the magnetizations of noncompensated groups.

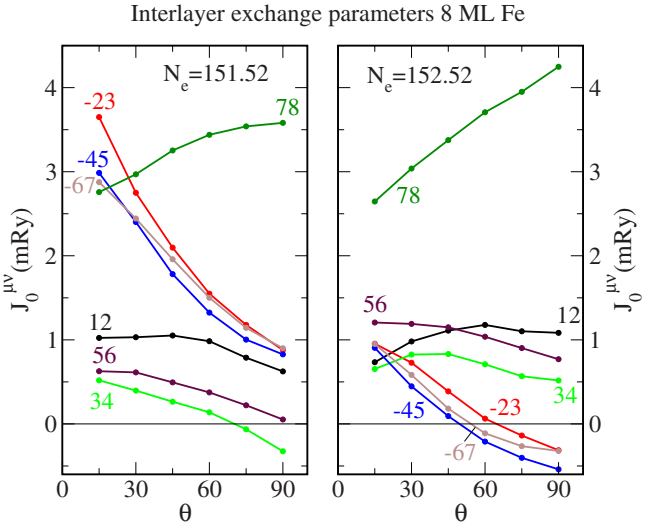

FIG. 14. (Color online) Nearest neighbor interlayer exchange parameters for 8 ML film for the number of valence electrons decreased (left panel) and increased (right panel) by half an electron per unit cell. The numbers give the pairs of interacting layers. The mines sign before the number means that the sign of the exchange parameter is reversed for better visualization. Calculations are performed with the use of the force theorem.

VII. SENSITIVITY OF THE EXCHANGE INTERACTIONS TO THE VARIATION OF THE ELECTRON NUMBER

The presence of defects in the films makes it interesting to study the dependence of the exchange parameters on the small variation in the number of valence electrons. This study is performed with the use of the force theorem and in the rigid-band approximation. This approximation means that the electron structure was calculated for the nominal number of electrons, whereas the occupation of the bands and the calculation of the band energy was performed for the number of electrons per unit cell decreased and increased by half an electron. We present calculations for the 8 ML film. Since the total number of valence electrons per unit cell N_e is 152 the relative variation in the electron number is rather small. Still the effect of this variation on the exchange parameters is remarkably strong.

The calculated exchange parameters are presented in Figs. 14 and 15. We first consider interlayer exchange parameters (Fig. 14). The influence of the variation in the number of electrons is very different for different pairs of the layers. This influence is relatively weak for the surface and interface layers. On the other hand it is very strong for the pairs 2–3, 4–5, and 6–7. These interactions decrease strongly with increasing electron number. Two comments should be made here. First, the ferromagnetic exchange interaction between two surface layers is very robust with respect to the variation in N_e. This result is again in very good correlation with the common consensus on the ferromagnetism of the upper two layers for the films studied in different groups. Second, the hierarchy of the exchange interactions discussed above and considered as the basis for the grouping of the layers into the blocks with robust relative orientations of the layer moments becomes even more pronounced for smaller N_e. On the other

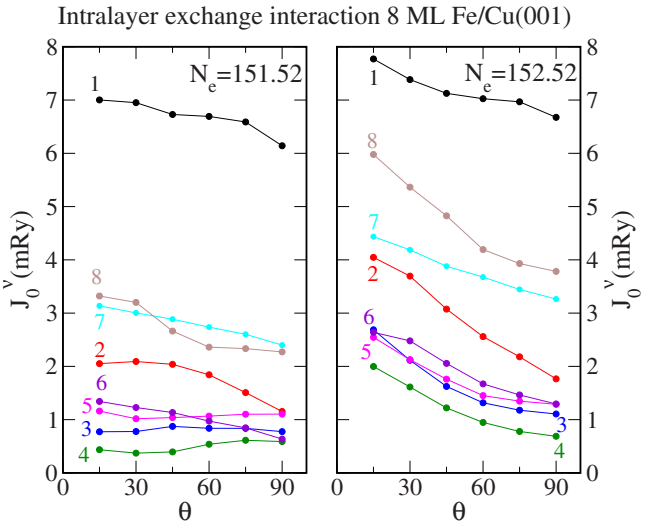

FIG. 15. (Color online) Intralayer exchange parameters for 8 ML film for the number of valence electrons decreased (left panel) and increased (right panel) by half an electron per unit cell. Calculations are performed with the use of the force theorem.

hand, for the increased N_e the blocking of the inner layers is expected to be less distinct.

The consideration of the intralayer exchange parameters (Fig. 15) shows that the trend in their variation with the change in the electron number is opposite to the trend in the variation in the interlayer parameters. There is clear tendency to the increase in intralayer exchange interactions with increasing N_e. The increase is weaker for the interface layer and stronger for inner and surface layers.

The opposite trends in the variation in the interlayer and intralayer exchange interactions lead to the expectation that they will tend to compensate each other in the thermodynamic properties. Indeed, the change in the temperature dependence of the magnetization with variation in the electron number is not dramatic. In Fig. 16 we show the magnetization curves obtained for $\Delta N_e=-0.5$. The Curie temperature of the film varies from 771 K for $\Delta N_e=-0.5$ to 845 K for $\Delta N_e=0.5$. There are interesting changes in details. For example, in the case of $\Delta N_e=-0.5$ we obtain the effective Curie temperature of the surface layer about 720 K to be lower than the corresponding value of the interface layer (720 versus 770 K). In the algebraic difference of the surface and interface blocks there is a step feature that is rather close to the T_C of the film because of the closeness of the effective Curie temperatures of the surface and interface.

VIII. COMMENTS ON INCOMMENSURATE SDW

Since there are two experimental works arguing to detect an incommensurate magnetic structure in the fcc Fe/Cu(001) films it is worth to comment on this issue. The suggestion of an incommensurate magnetic structure was first made by Qian *et al.*[6] to describe the total magnetization of the films of different thicknesses. Note that the formation of a spin-density wave with the periodicity incommensurate with the periodicity of the crystal lattice needs, first, the periodic lattice of atoms that are equivalent to one another in the absence of the magnetic structure and, second, a certain physical mechanism that leads to the formation of a periodic magnetic structure with periodicity incommensurate with the atomic lattice. Two possible mechanisms of this type are the competition between ferromagnetic and antiferromagnetic interatomic exchange interactions and the Fermi-surface nesting.

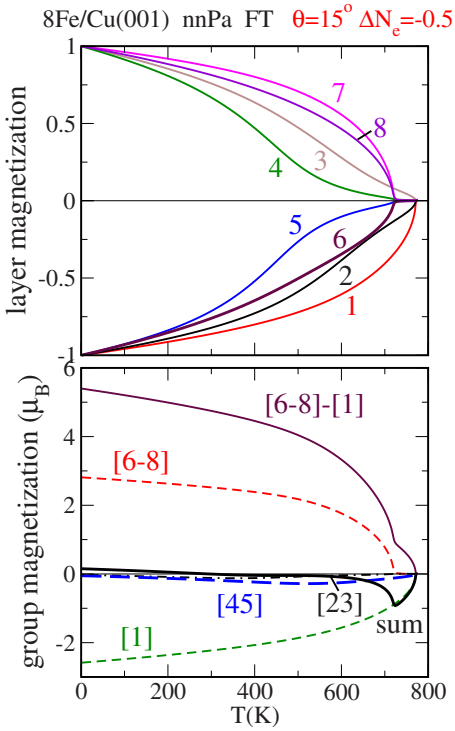

FIG. 16. (Color online) The same as in Fig. 10 but for the decreased number of the valence electrons $\Delta N_e=-0.5$.

Considering the 6 ML film we notice, first, that the surface and interface layers have different atomic environments compared to each other and to the inner layers. Even if to accept a rather unrealistic assumption that four inner layers of the 6 ML film are approximately equivalent to each other these four atomic layers are not sufficient for the establishing a magnetic structure that can be considered as periodic with period of about 2.7 ML. The conclusions by Qian *et al.* are essentially based on the comparison of the magnetizations measured for different films. This comparison assumes that all films have ideal atomic structure and that the same incommensurate magnetic structure propagates coherently through all layers available starting from the surface part of the film. Obviously the assumption of the identical structure of different films is rather unrealistic. Our study reported above shows that there are features of the magnetic structure of the films that are sensitive to the defects in the films. Therefore two fabricated films of nominally the same thickness can be rather different in the magnetic properties because of the different defect patterns. As a consequence, the comparison of different films must be done with caution. On the basis of these arguments we argue that the conclusion about the formation of the incommensurate magnetic structure in the thin Fe/Cu(001) films is not well founded.

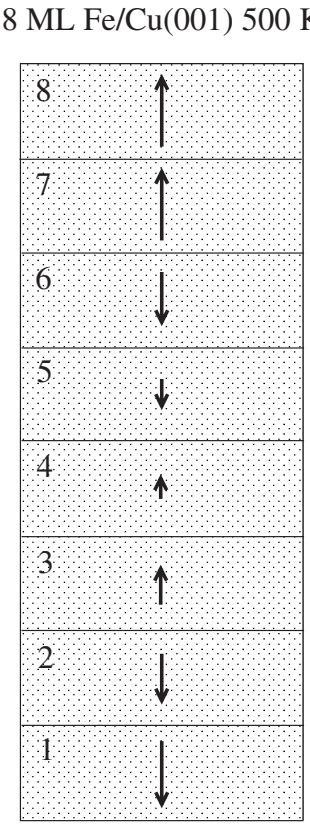

FIG. 17. The magnetization profile of 8 ML film at $T=500$ K.

In contrast to Qian *et al.* Amemiya *et al.*[8,9] come to the conclusion about the formation of an incommensurate magnetic structure by analyzing the depth profile of the same film. It is, however, very important to notice that the measurement of the magnetization profile is performed at an elevated temperature. As we have shown the magnetizations of different layers can have very different temperature dependence. Because of the different temperature dependences the magnetization profile measured at an elevated temperature can remind a spin-density wave since the magnetizations of different layers decrease differently. As an example we show in Fig. 17 the calculated magnetization profile for 8 ML film at $T=500$ K. We see very strong variation in the magnetization from layer to layer that might be attempted to be interpreted in terms of the formation of SDW. But as we have shown such an interpretation is not correct. Instead we deal with the temperature-dependent properties of the magnetic structure of a thin film that is not characterized by an incommensurate periodicity. In the ground state the values of the magnetizations of different layers do not vary strongly.

IX. CONCLUSIONS

We studied the magnetic properties of fcc Fe/Cu(001) films with 6, 7, and 8 ML coverages. Several purposes have been pursued. Among them, first, to distinguish between robust features in the exchange interactions and magnetic structure of the films and the features that are expected to vary strongly from film to film as a result of uncontrolled pattern of defects, second, to consider different physical scenarios of the formation of steplike features in the temperature dependence of the total magnetization, third, to analyze the possibility of the formation of the incommensurate spin-density waves in thin Fe/Cu(001) films reported by two experimental groups.

We suggest a grouping of the layers into blocks with robust collinear magnetic structure whereas the variation in the relative directions of the moments of different blocks is energetically relatively inexpensive. We demonstrate that this blocking corresponds to a certain hierarchy of the interlayer exchange interactions. The blocking has the following form: the magnetic structure of the three surface layers has the form $\Uparrow\Uparrow\Downarrow$. Further layers form the pairs with robust antiferromagnetic structure. This picture differs from the usually considered grouping into two ferromagnetic surface layers and a nonferromagnetic rest of the film. The magnetic structure suggested by us is in agreement with previous works concerning ferromagnetism of the two surface layers and with conclusion of Amemiya *et al.* about opposite directions of the magnetizations of the third layer and upper two layers. This grouping allows to give a qualitative interpretation of the formation of the step features in the temperature dependences of the magnetizations of 6 and 8 ML films in contrast to the 7 ML film where the step feature has not been observed.

We performed MFA calculation of the temperature dependence of the magnetization focusing in particular on the possible physical mechanisms of the formation of the step features in the temperature dependence of the magnetization of the film. We distinguish between first-order phase transition governed by the exchange interactions between distant layers and the difference in the effective magnetic transition temperature in different blocks of layers. The steplike features in the total magnetization can be both positive and negative depending on the detailed pattern of the exchange interaction in the films.

Using rigid-band model we investigated the dependence of the exchange parameters on the electron number. We demonstrate strong sensitivity of the exchange parameters to the variation in the number of electrons. Since the directions of the variation in the interlayer and intralayer exchange parameters are opposite they partially compensate each other in the thermodynamic properties.

We critically discuss the possibility of the formation of the incommensurate SDW in the thin Fe/Cu(001) films. We come to the conclusion that the formation of such structures is improbable and suggest the strong difference in the temperature dependence of the magnetization of different layers as the reason for the observed magnetic depth profile reported by Amemiya *et al.*[8,9]

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[31] It is worth noting that the inequivalence of the atomic layers in the film leads to the difference of the distances between different pairs of the neighboring layers. This difference was experimentally detected [see, e.g., earlier experiment (Ref. 4) on the low energy electron diffraction (LEED) or recent experiment (Ref. 10) on x-ray diffraction]. A typical value of the variation in the interlayer distance in the internal part of the film is about 1%, whereas for the upper most layer it can be substantially larger. For example, for the 8 ML film the distance of the upper most layer to the next one was detected to increase to 1.88 ± 0.015 Å that is by more than 4% with respect to the Cu interlayer distance. Test calculations with varied interlayer distances have shown that all qualitative conclusions formulated in the paper remain valid. In particular the calculations show that the strong ferromagnetic exchange interactions between two upper Fe layers are determined not by the increased interlayer distance but by the reduced coordination of the Fe atoms of the surface layer.

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[38] The restriction of our statistical-mechanics calculations to the mean-field approximation leaves open the question if the first-order phase transition is an intrinsic property of the model. The application of a more elaborated statistical-mechanics technique should be carried out to verify if the phase transition predicted by the MFA calculations takes place in this form.

[39] The small values of the moments of the interface layers of the fcc-Fe/Cu(001) films detected in the recent experiment (Ref. 29) can be explained by strong Fe-Cu intermixing.