Multiple reorientation transition of the magnetization of free surfaces of Fe on Ag(100)

C. Sommers
Laboratoire de Physique des Solides, Campus d'Orsay, Orsay, France

J. Zabloudil
Center for Computational Materials Science, Vienna, Austria

C. Uiberacker
Institut für Technische Elektrochemie, Technical University Vienna, Austria

P. Weinberger
Institut für Technische Elektrochemie, Technical University Vienna, Austria

L. Szunyogh
Department of Theoretical Physics, Technical University of Budapest, Budapest, Hungary
and Center for Computational Materials Science, Vienna, Austria
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The magnetic properties of Ag(100)/Fe$_n$, $n \leqslant 16$, are investigated using the fully relativistic spin-polarized screened Korringa-Kohn-Rostocker method. It is found that (a) ferromagnetic interlayer coupling is the most stable magnetic configuration of free surfaces of Fe on Ag(100) and (b) that in the vicinity of $n=5$ a multiple reorientation transition of the magnetization occurs. The results obtained are in excellent agreement with available experimental data and give an interesting explanation for the occurrence of temperature-dependent effects seen experimentally. [S0163-1829(98)09133-4]

I. INTRODUCTION

In contrast to the Cu(100)/Fe$_n$ system, free surfaces of Fe on Ag(100) attracted much less attention,[1–5] partially because the experimental results for the Cu(100)/Fe$_n$ system seemed and still seem to diverge considerably, giving rise to new measurements and investigations. Another reason for this lesser curiosity in exploring the magnetic properties of free surfaces of Fe on Ag(100) is quite likely the occurrence of a strange regime of reorientation transitions,[4] which did not fit in the usual (traditional) description of surface magnetic anisotropies and which made the design of new experiments unclear. In order to see whether a theoretical study can shed some light on this strange behavior, in the present paper the magnetic properties of Ag(100)/Fe$_n$, $n \leqslant 16$, are investigated as well as the type of magnetic interlayer coupling, which for some time seemed to obscure theoretical descriptions of the magnetic anisotropy energy in the Cu(100)/Fe$_n$ systems.[14,15] Quite clearly, as compared to the Cu(100)/Fe$_n$ system, part of the different structural and magnetic behavior of Ag(100)/Fe$_n$ is correlated to the very different lattice spacings of the bulk substrates, namely 6.8309 and 7.7895 a.u., for fcc Cu and fcc Ag, respectively.

II. COMPUTATIONAL DETAILS

The fully relativistic spin-polarized version[6] of the screened Korringa-Kohn-Rostoker (KKR) method[7] for layered systems[8] is applied to calculate self-consistently the electronic structure and the magnetic properties of free surfaces of Fe$_n$ on Ag(100), $n \leqslant 16$, whereby all interlayer distances refer to an fcc ''parent lattice''[9] corresponding to the experimental lattice spacing of Ag (no surface or interface relaxations). For each system, i.e., for each n, the electronic and magnetic structure of the magnetic configuration corresponding to a *uniform* perpendicular-to-plane orientation of the magnetization in the Fe layers is calculated self-consistently using 45 $k_\parallel$ points in the irreducible part of the surface Brillouin zone (ISBZ) and the local-density functional described by Vosko *et al.*[10]

The obtained self-consistent layer-resolved effective potentials and layer-resolved effective magnetization fields in the spin-polarized Kohn-Sham-Dirac Hamiltonian (see, e.g., Ref. 11) are then used to evaluate differences in the band energies with respect to the following magnetic configurations:

$$\Delta E(\mathcal{C}) = E(\mathcal{C}) - E(\mathcal{C}_0), \tag{1}$$

$$\mathcal{C} = \mathcal{C}_a, \mathcal{C}_i, \mathcal{C}_{ij}, \ldots, \tag{2}$$

where

$$\mathcal{C}_0 = \{\hat{\mathbf{z}}_1, \hat{\mathbf{z}}_2, \ldots, \hat{\mathbf{z}}_n\}, \tag{3}$$

$$\mathcal{C}_a = \{\hat{\mathbf{x}}_1, \hat{\mathbf{x}}_2, \ldots, \hat{\mathbf{x}}_n\}, \tag{4}$$

$$\mathcal{C}_i = \{\hat{\mathbf{z}}_1, \hat{\mathbf{z}}_2, \ldots, \hat{\mathbf{z}}_{i-1}, -\hat{\mathbf{z}}_i, \hat{\mathbf{z}}_{i+1}, \ldots, \hat{\mathbf{z}}_n\}, \tag{5}$$

and n is the number of Fe layers. Configuration $\mathcal{C}_0$ refers therefore to a reference configuration, in which in all n layers of the Fe film on Ag(100) the orientation of the magnetization points along the surface normal, denoted by $\hat{\mathbf{z}}$, which in

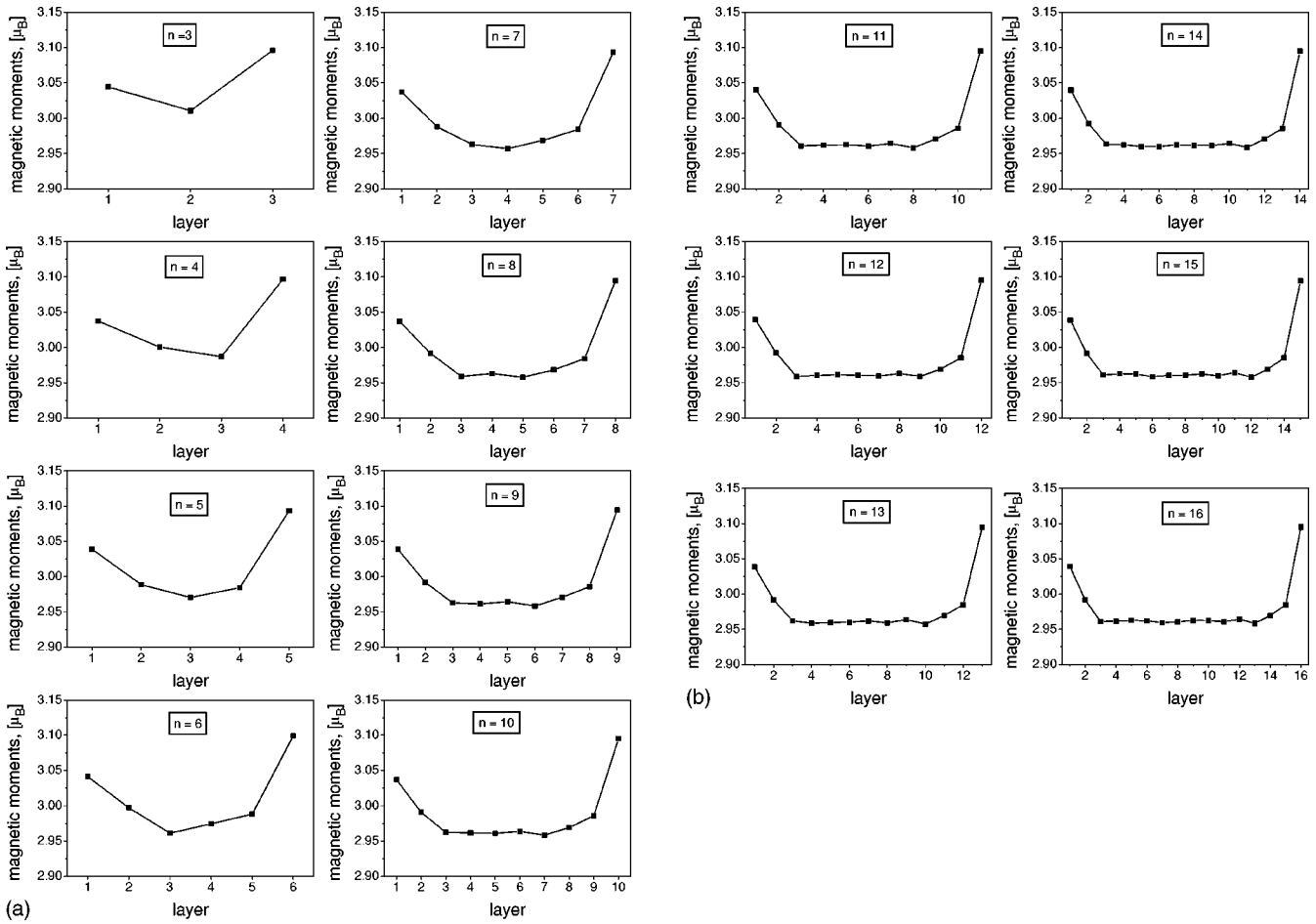

FIG. 1. (a) Layer-resolved magnetic moments of free surfaces of Ag(100)/Fe$_n$ for $n \leqslant 10$. The total number of Fe layers is marked explicitly, the labeling of layers starts at the Ag/Fe interface. (b) Layer-resolved magnetic moments of free surfaces of Ag(100)/Fe$_n$ for $n > 10$. The total number of Fe layers is marked explicitly, the labeling of layers starts at the Ag/Fe interface.

turn corresponds to the one used in the self-consistent calculations. Configuration $\mathcal{C}_a$ comprises the case that in all n Fe layers the orientation of the magnetization is in-plane, while configurations $\mathcal{C}_i$ specify cases in which, with respect to the reference configuration only in the ith Fe layer, the orientation of the magnetization points along a different direction, namely along $-\hat{\mathbf{z}}$. In Eq. (1) $\Delta E(\mathcal{C}_a)$ refers then to the band energy contribution to the magnetic anisotropy energy,[6,12] while the $\Delta E(\mathcal{C}_i)$ reflect single ''spin-flip'' energies.[13] Consequently a double spin-flip energy $\Delta E(\mathcal{C}_{ij})$ is then defined by Eq. (1) if in this equation $\mathcal{C}$ refers to

$$\mathcal{C}_{ij}=\{\hat{\mathbf{z}}_1,\hat{\mathbf{z}}_2,\ldots,\hat{\mathbf{z}}_{i-1},-\hat{\mathbf{z}}_i,\hat{\mathbf{z}}_{i+1},\ldots,\hat{\mathbf{z}}_{j-1},-\hat{\mathbf{z}}_j,\hat{\mathbf{z}}_{j+1},\ldots,\hat{\mathbf{z}}_n\}, \tag{6}$$

i.e., if with respect to the reference configuration the orientation of the magnetization is switched simultaneously in layers i and j from $\hat{\mathbf{z}}$ to $-\hat{\mathbf{z}}$. As was recently[13] shown, the band energy difference $\Delta E(\mathcal{C}')$ for a configuration with $\leqslant n/2$ spin flips with respect to the reference configuration can be approximated in terms of a Heisenberg-like model of the following kind:

$$\Delta E(\mathcal{C}') \sim \frac{1}{2}\sum_{i,j\leqslant n/2}\Delta E(\mathcal{C}_{ij}). \tag{7}$$

Furthermore, by expressing the double spin-flip energies as

$$\Delta E(\mathcal{C}_{ij})=\Delta E(\mathcal{C}_i)+\Delta E(\mathcal{C}_j)+V_{ij}, \tag{8}$$

where the V_{ij} can be viewed as vertex corrections, it turned out[13] that for free and capped surfaces of Fe on Cu(100), which show antiferromagnetic interlayer coupling, even a model with $V_{ij}=0, \forall i,j$, gave the correct configuration of lowest energy.

All band-energy differences presented in this paper were evaluated within the force theorem approximation (see, in particular, Ref. 12) by using 990 $k_\parallel$ points in the ISBZ and by applying the group-theoretical methods described in Ref. 6. In principle, the magnetic anisotropy energy and the spin flip energies also contain a contribution arising from the magnetic dipole-dipole interaction,[6,14–16] which, however, in the present paper is only evaluated and discussed in the case of the magnetic anisotropy energy.

III. RESULTS AND DISCUSSION

In Fig. 1 the layer-resolved magnetic moments of free surfaces of Fe on Ag(100) are shown. It is interesting to note that beyond $n=8$ a characteristic pattern evolves: starting from either interface, Fe/Ag or Fe/vacuum, at about 4–5 layers the magnetic moment falls off to a nearly constant value in the middle of the magnetic film. This implies that in terms

of the moments the influence of interfaces is confined to about 4–5 times the interlayer distance. This characteristic influence of the interfaces will be seen later on in the discussion of the spin-flip energies. Clearly enough, in the middle of thick films the value of the moment in a hypothetical fcc bulk Fe is not reached, simply because in all Ag(100)/Fe$_n$ systems the lattice spacing and the Fermi energy of the substrate, namely of Ag(100) applies.

A. Magnetic anisotropy energy

In Fig. 2 the band energy and magnetic dipole-dipole contributions to the magnetic anisotropy energy are displayed together with the latter quantity. Beyond about eight layers of Fe the band energy part starts to oscillate with a period of two, a phenomenon that was already detected in theoretical calculations for free and capped surfaces of Co on Cu(100), see also the discussion in Ref. 16. As can be seen in this figure, the magnetic dipole-dipole energy, the physical source of the so-called shape anisotropy, grows linearly with the number of Fe layers and for thicker Fe layers determines the sign of the magnetic anisotropy energy:

$$\Delta E(\mathcal{C}_a)=\begin{cases} >0; & \text{out-of-plane,} \\ <0; & \text{in-plane.} \end{cases} \tag{9}$$

Quite clearly, for $n>6$ the orientation of the magnetization is in-plane, while for $n\leqslant 3$ an out-of-plane orientation is found. The interesting cases are of course $n=4,5$, since for $n=5$ the anisotropy energy is marginally positive, suggesting that between four and six layers temperature-dependent effects have to be expected. Furthermore, since for four layers of Fe the magnetic anisotropy energy is negative, a multiple reorientation transition of the magnetization between three and six layers can be deduced. It should be noted that for very thin films this peculiar shape of the anisotropy energy is entirely caused by the strong oscillations of the band energy part with respect to the number of Fe layers.

B. Spin-flip energies

In the case of free and capped surfaces of Fe on Cu(100) it was found that antiferromagnetic coupling between the Fe layers is decisive for the orientation of the magnetization to be either in-plane or out-of-plane, see in particular the discussion in Refs. 14,15. Recently it was also shown that in the case of Fe-rich alloys the layer and concentration-dependent reorientation transition seen experimentally[18] for (Fe_cCo_{1-c}) films on Cu(100) is indeed also a consequence of the setting-in of antiferromagnetic coupling between layers.[17] It is therefore of quite some interest to see whether such antiferromagnetic interlayer coupling would also occur in Fe films on Ag(100).

In Fig. 3 the single spin-flip energies are displayed for all cases of Ag(100)/Fe$_n$ investigated. The most interesting fact of course is that for all n these spin-flip energies are positive. Furthermore, since all double spin flip energies (not shown here) are positive, it can be ruled out that in the systems Ag(100)/Fe$_n$ any kind of configuration with antiferromagnetic interlayer coupling is of lower energy than the one corresponding to ferromagnetic coupling. In view of the properties of free and capped surfaces of Fe on Cu(100), this indeed is an important statement, since the anisotropy energy shown in Fig. 2 does refer to the actual ground state and not just to the case of *assumed* ferromagnetic coupled layers of Fe on Ag(100).

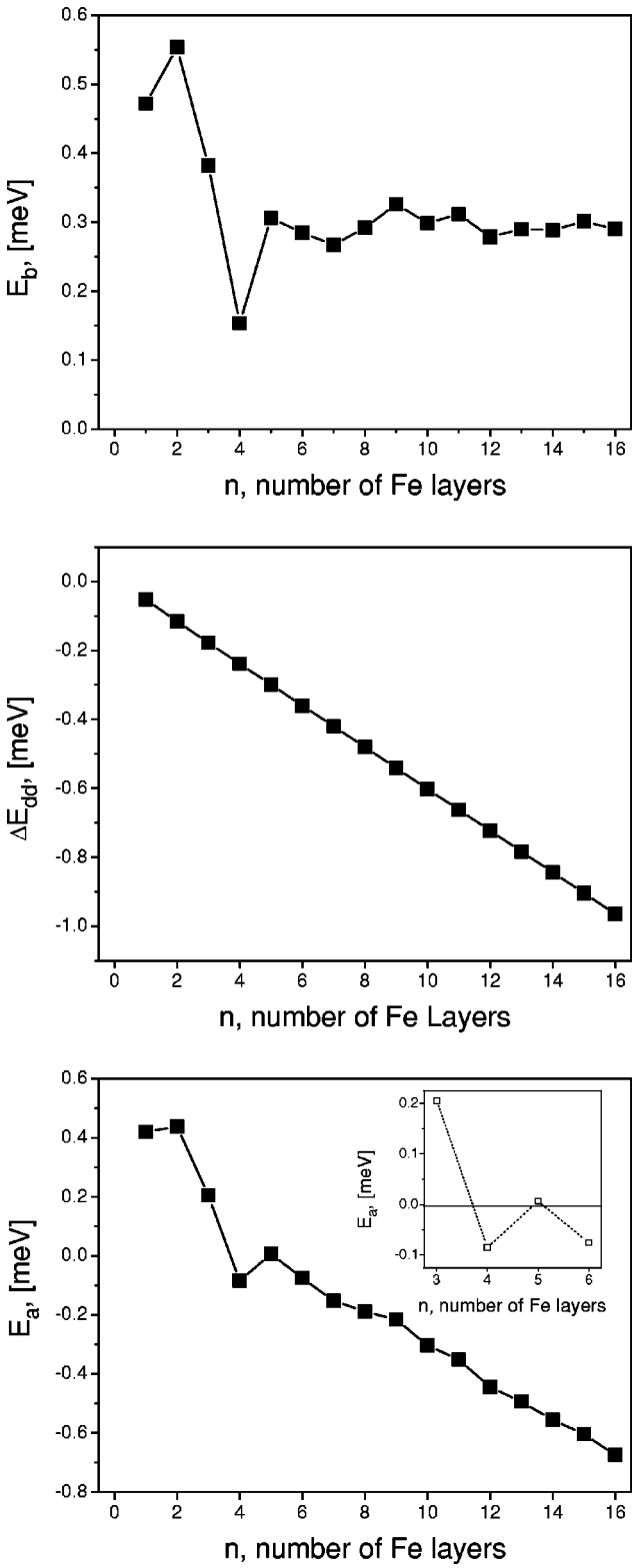

FIG. 2. Band energy (top) and magnetic dipole-dipole contribution (middle) to the magnetic anisotropy energy (bottom) of free surfaces of Fe$_n$ on Ag(100). The inset shows the magnetic anisotropy energy in the vicinity of $n=5$.

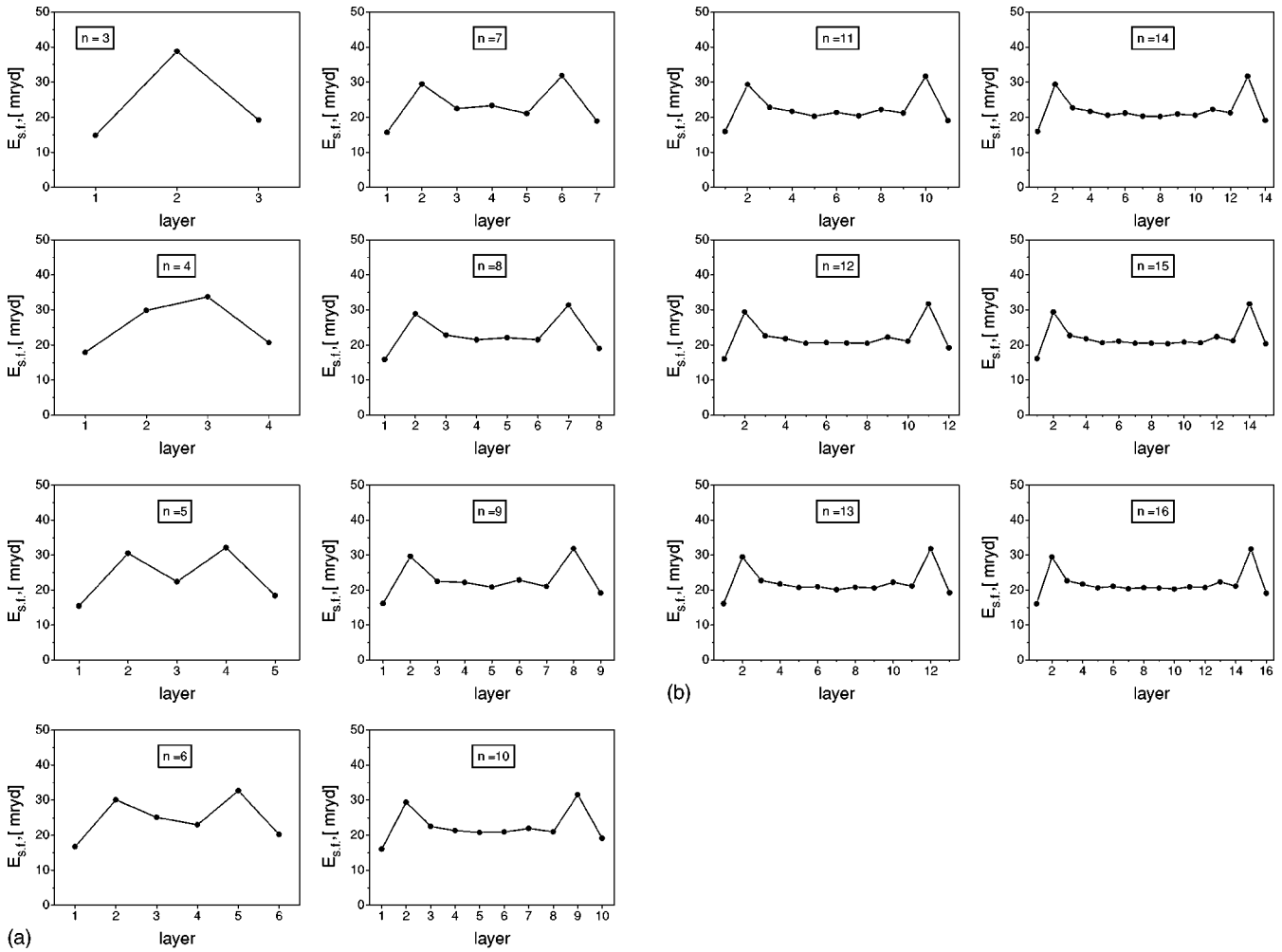

FIG. 3. (a) Spin-flip energies E_{sf} for $n \leqslant 10$. The total number of Fe layers is marked explicitly, the labeling of layers starts at the Ag/Fe interface. (b) Spin-flip energies E_{sf} for $n > 10$. The total number of Fe layers is marked explicitly, the labeling of layers starts at the Ag/Fe interface.

Inspecting Fig. 3 in more detail, one easily can see that for $n \geqslant 9$ the shapes of the single spin-flip energy curves are very similar, namely variations over four layers of Fe next to an interface and a nearly constant value in the middle of the film. Although it is not important in the present case, Fig. 3 shows very nicely that when using a Heisenberg-like model as defined in Eqs. (7) and (8), it is sufficient to consider only a surprisingly small number of such Heisenberg-like parameters in order to describe the energetics of very thick magnetic films. It should be recalled from the discussion of the layer-resolved moments that the same kind of influence of the interfaces was also seen there.

This vicinity range of about 3–5 layers next to an interface was also seen in theoretical calculations of other systems such as Au(100)/Fe$_n$ (Ref. 6) and Cu(100)/Fe$_n$.[14,15] If the vicinity ranges of the two interfaces, namely with respect to the substrate and vacuum (cap), respectively, overlap then most likely hybridization effects between interface states are present, which partially are responsible for the physical properties of very thin films. It should be noted that viewed in terms of a one-dimensional band structure corresponding to a potential with a *finite* range periodicity such interface states refer to so-called band-edge states.[19]

C. Comparison to experiment

Berger and Hopster[1] measured very thin films of Fe on Ag(100). For a film of 4.3 monolayer (ML) thickness they find at low temperatures an out-of-plane orientation of the magnetization, increasing the temperature, however, a reorientation transition is observed at about 220 K, above which the magnetization is orientated in-plane. They also argue that at low temperature the system is in a single-domain state that does not seem to be too sensitive to an elevation of temperature. Looking again at Fig. 2, their data fit very well to the present calculations, which show between four (in-plane) and five (out-of-plane) layers of Fe (a) a reorientation transition and (b) a nearly vanishing (positive) magnetic anisotropy energy at five layers that facilitates a strong temperature dependence of this quantity. In a second paper[2] the same authors investigated the magnetic properties of a 3.8 ML thick film of Fe on Ag(100). They find a perpendicular orientation of the magnetization at low temperatures and a reorientation transition at about 370 K. This again is in excellent agreement with the present calculations, see Fig. 2, since from 3 to 4 ML's of Fe the orientation of the magnetization switches from out-of-plane to in-plane, respectively. As for 3

ML of Fe the anisotropy energy is considerably positive, the crossover occurs close to 4 ML, which is in fact what the experiment shows.

Hicken *et al.*[3] recorded the so-called K_1 anisotropy constant for Ag(100)/Fe$_n$ versus the Fe film thickness by investigating different growth runs. Their data show that depending on the growth run K_1 changes sign at about 3–6 ML's of Fe, being positive (in-plane orientation) for all thicknesses above 6 ML's of Fe. Since they go out to almost 50 ML, one easily can guess from Fig. 2 that as the magnetic dipole-dipole contribution dominates the anisotropy energy for increasingly thicker layers of Fe, beyond 16 ML of Fe an in-plane orientation of the magnetization has to be expected from theoretical calculations.

From magneto-optical measurements Cownburn *et al.*[4] concluded that with respect to the film thickness t their data seem to fall into four classes of behavior: (1) out-of-plane easy direction, full remanence ($t \leqslant 4.3$ ML); (2) out-of-plane easy direction, reduced or zero remanence; (3) in-plane easy direction, zero remanence, and (4) in-plane easy direction, full remanence ($t \geqslant 6.6$ ML), whereby cases 2 and 3 arise from $t \sim 5$ ML. The data discussed in Ref. 4 not only confirm the data given by Berger and Hopster,[1,2] but fit very well the theoretical results shown in Fig. 2, since at exactly $n=5$ a break in the magnetic anisotropy energy as a function of Fe-layer thickness occurs.

Quite clearly for very thin films the growth conditions and growth mode become very important for any surface specific physical quantity. In a recent study using helium scattering Canepa *et al.*[5] showed that growing Fe on Ag(100) follows a kind of Stranski-Krastanov mode, whereby for $t>1$ some degree of three-dimensional island formation also occurs. A different growth mode is most likely therefore responsible for the small differences in the critical film thickness for the two types of reorientation transitions traced in the experiments reported in Refs. 1, 2, and 3, respectively.

IV. CONCLUSION

In this paper we presented a calculation of the magnetic anisotropy energy for Fe films on Ag(100) by using the fully relativistic spin-polarized screened KKR method. We not only proved that a ferromagnetic coupling of layers is the most stable one for the given lattice spacing, but also called attention to the vicinity regimes of the interfaces, Ag/Fe and Fe/vacuum, in terms of layer-resolved magnetic moments and single spin-flip energies. The calculated anisotropy energy as a function of Fe film thickness shows multiple reorientation transitions, which not only is in excellent agreement with available experimental data, but also partially explains the curious behavior of the experimentally recorded data of the magnetization at about 5 ML of Fe.

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