Magnetic anisotropy of Fe_xCo_{1-x} multilayers on Cu(001): Reorientation transition of magnetic moments due to different interlayer coupling

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The magnetic anisotropy energies of Fe_xCo_{1-x} multilayers on Cu(001) have been determined by means of *ab initio* calculations using the fully relativistic, spin-polarized screened Korringa-Kohn-Rostoker method within the local spin density approximation. By utilizing the coherent potential approximation the Fe/Co system was treated within a mean-field approach as a (uniform) randomly disordered alloy. The type of magnetic interlayer coupling—either ferromagnetic or antiferromagnetic—that is energetically more favorable is found to depend on both the film thickness and alloy composition. It seems therefore that the type of magnetic interlayer coupling is also responsible for the reorientation transition mainly because of the strong enhancement of the band energy contribution to the magnetocrystalline anisotropy energy in the case of antiferromagnetic coupling. [S0163-1829(98)01934-1]

I. INTRODUCTION

The magnetic anisotropy of thin films of transition metals has been the subject of quite a few theoretical works in recent years.[1–9] So far, in these studies exclusively pure metallic films on a noble metal substrate have been investigated. The present paper focuses on the much tougher problem of the magnetocrystalline anisotropy of a magnetic alloy film, namely, of thin Fe/Co films on a Cu(001) substrate.

Thin films of these two constituents have different orientations of the easy axis on Cu(001): Fe films up to a thickness of at least 10 monolayers (ML's) show out-of-plane magnetization,[10] while thin Co films are magnetized in plane.[11] These experimental facts have been confirmed also by first-principles calculations (see Refs. 12, 13 and Refs. 14, 15, respectively). Thus, an interesting behavior is to be expected when alloying Fe with Co. The aim of this paper is therefore to study how the easy axis of the magnetization in such an alloy system varies with film thickness and composition, i.e., to address the question of whether regions of different preferred orientations of the moments can be identified as a function of the number of magnetic layers and of the alloy composition and, hence, if there are such regions, to pinpoint also the driving force behind such a reorientation.

It has been shown previously[12,13] that antiferromagnetically coupled Fe layers on Cu(001) exhibit perpendicular

magnetism up to a film thickness of at least seven atomic layers, whereas if the layers are arranged ferromagnetically, the moments are oriented in plane.[16] This indicates that also in the case of Fe-rich Fe/Co thin films the type of magnetic interlayer coupling might be of crucial importance for a correct theoretical description of the magnetic anisotropy energy (MAE). Consequently, we performed self-consistent calculations for each alloy composition and film thickness under consideration by varying the type of magnetic interlayer coupling in order to find the magnetic configuration that corresponds to the lowest total energy. This configuration was then used for the determination of the MAE.

The paper is organized as follows: Section II summarizes the computational details of the self-consistent calculations and the type of magnetic interlayer coupling in the respective ground states; in Sec. III we describe the approach applied to calculate the MAE and analyze the results obtained. Finally, in a conclusion we summarize our results and relate the present work to available experiments.

II. GROUND STATE CONFIGURATIONS

The self-consistent calculations were carried out by using a spin-polarized, *scalar-relativistic* version of the screened Korringa-Kohn-Rostoker (KKR) method[17,18] because of its numerical efficiency. For some cases it has been checked that

a fully relativistic, self-consistent treatment[5] yields results not significantly different from the present ones as the magnetic coupling between layers is to first order determined by the (nonrelativistic) exchange interaction.

For disordered layered systems the inhomogeneous coherent potential approximation (CPA) has been discussed in details in Ref. 19. Since our self-consistent calculations rely on standard applications of the local spin-density approximation (LSDA) in the context of random alloys treated within the CPA,[20] no further theoretical details have to be repeated. The same computational parameters as in our previous investigations[13,16] were used. In particular, no attempt was made to account for a relatively small tetragonal expansion of the surface layers along the surface normal.[21]

We thus carried out self-consistent calculations for the $(Fe_xCo_{1-x})_n/Cu(001)$ system for $x = 0.5, 0.6, 0.7, 0.8, 0.85, 0.9,$ and 1.0 and $n = 1,...,7$. For a proper determination of the respective ground state a comparison of the total energies of all possible configurations with respect to different magnetic intra- and interlayer couplings would have been necessary, which, however, would have been untractable to perform. Instead, as previously assumed,[13,16] we exclusively supposed ferromagnetic intralayer coupling (see also Ref. 22). Furthermore, since antiferromagnetic coupling is to be expected only for Fe-rich samples, for each film thickness n beside the ferromagnetic configuration we considered only that configuration which has been identified as the state of lowest energy for the corresponding $Fe_n/Cu(001)$ system (see Table I of Ref. 13). With respect to these two configurations, in the following the one of lower total energy is referred to as the *ground state*. For three Fe_xCo_{1-x} layers we verified that this indeed pertains over the full range of compositions considered. Note furthermore that no concentration gradient with respect to the surface normal was assumed; i.e., the alloy composition was the same in each magnetic layer on Cu(001).

In Fig. 1 the difference of the total energy ΔE_{tot} of the antiferromagnetically coupled system $E_{tot}(AF)$ and that of the ferromagnetic system $E_{tot}(F)$ is displayed as a function of the concentration x for $n = 2, 3, 6,$ and 7. As for $x = 1.0,$[13] for two layers of Fe_xCo_{1-x} the ferromagnetic alignment is energetically favorable over the whole range of concentrations. Remarkably, increasing the Co content hardly changes the energetical difference between the ferromagnetic and antiferromagnetic alignments. For three layers, at $x = 1.0,$ the ($\downarrow\uparrow\uparrow$) configuration (substrate on the left side) turned out to be the ground state as discussed in some detail in Ref. 13. When increasing the Co concentration, ΔE_{tot} is linearly increasing such that for $x = 0.9$ it becomes positive; that is, the ferromagnetic configuration becomes favorable. This tendency can basically be attributed to an increasing ferromagnetic coupling between Co sites as well as between the Fe and Co sites.

Clearly, also for thicker films this tendency holds true. Since, however, for pure Fe films ΔE_{tot} is increasing with film thickness,[13] the Co concentration at which the ground state turns out to be ferromagnetic is expected to increase, too. Although not shown in Fig. 1, for four and five layers the transition is found to be at approximately 85% and 80% of Fe concentration, respectively. As can be deduced from Fig. 1, for six layers the transition occurs at about $x = 0.75$;
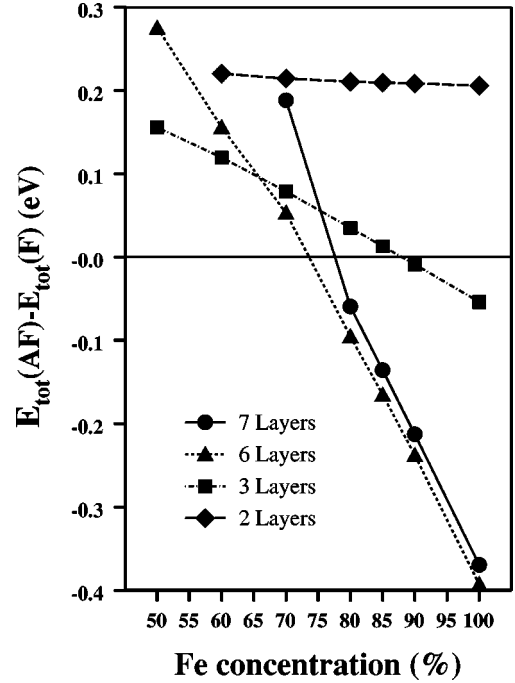

FIG. 1. Total energy differences between the antiferromagnetic and ferromagnetic arrangements of $(Fe_xCo_{1-x})_n/Cu(001)$ for $n = 2$ (solid diamonds, dashed line), $n = 3$ (solid squares, dash-dotted line), $n = 6$ (solid triangles, dotted line), and $n = 7$ (solid circles, solid line) as a function of the Fe concentration.

however, for seven layers this critical Fe concentration seems to rise slightly. Obviously, at any concentration ΔE_{tot} for seven layers is above that for six layers. It is thus difficult to estimate what happens for even thicker films. Since the hierarchy of metastable antiferromagnetic configurations gets quite complicated for thicker films (see also Fig. 2 of Ref. 13), a determination of the ground state in terms of total energies for such films seems to be a rather difficult numerical task. On the other hand, as—depending on the experimental situation, preparation, etc.—for larger film thicknesses and Fe concentrations a fcc-to-bcc-type structural phase transformation occurs,[10] the present restriction to seven layers coincides in about with the range of the fcc-type growth of Fe-rich Fe_xCo_{1-x} overlayers on Cu(001).

III. MAGNETOCRYSTALLINE ANISOTROPY ENERGIES

A. Calculation of the MAE

In principle the MAE is defined as the difference in energy between a uniform in-plane ($\parallel$) and a uniform normal-to-plane ($\perp$) orientation of the magnetization in the system of consideration,

$$\Delta E = E(\parallel) - E(\perp). \tag{1}$$

Relying on the force theorem (see, e.g., Refs. 2 and 23) the MAE contains two contributions, namely, the band energy ΔE_b and the magnetostatic dipole-dipole energy ΔE_{dd},

$$\Delta E = \Delta E_b + \Delta E_{dd}. \tag{2}$$

Details of how to calculate these two contributions to the MAE for layered systems within a fully relativistic, spin-

polarized approach as used in here are discussed in Ref. 5. Since, however, in the present case one has to deal with a two-component alloy, short attention will be given in the following to the definitions of these two quantities in the case of statistical disorder.

For layered systems the band energy $E_b(\delta)$ for a given orientation of the magnetization ($\delta=\parallel$ or $\perp$) is defined within the CPA as the concentration-weighted sum over all relevant layers in the surface region (including a few vacuum and Cu layers):

$$E_b(\delta) = \sum_p \sum_\alpha c_{p\alpha} E_b^{p,\alpha}(\delta), \qquad (3)$$

where p specifies a certain layer, and α refers to a particular component of concentration $c_{p\alpha}$. Note that in a homogeneously alloyed multilayer $c_{p\alpha}$ is the same in all layers p. The quantities $E_b^{p,\alpha}$ are given by

$$E_b^{p,\alpha}(\delta) = \int_{-\infty}^{\epsilon_F} (\epsilon - \epsilon_F) n_\alpha^p(\epsilon;\delta) d\epsilon, \qquad (4)$$

where $n_\alpha^p(\epsilon;\delta)$ is the layer and component projected local density of states (see also Ref. 19). It should be noted that since the Fermi level ϵ_F is determined by the nonmagnetic substrate, one and the same ϵ_F applies for all magnetic orientations of the overlayer system.

In Ref. 5 we also discussed a method in order to calculate the dipole-dipole interaction energy of two-dimensional translational invariant systems. Since in terms of a general complex medium the CPA has to recover this kind of translational invariance in a multicomponent alloy, the moments arising from Fe and from Co have to be weighted with their respective concentrations such that to each site in a given layer p a uniform magnetic moment applies:

$$\langle \mathbf{m}_p \rangle = \sum_\alpha c_{p\alpha} \mathbf{m}_{p\alpha}, \qquad (5)$$

where $\langle\rangle$ denotes an average over statistical configurations and $\mathbf{m}_{p\alpha}$ refers to the magnetic moment of component α in layer p. It should be noted that by using the above averaged magnetic moments in Eq. (A2) of Ref. 5, one in fact neglects vertex corrections of the kind $\langle \mathbf{m_R m_{R'}} \rangle - \langle \mathbf{m_R} \rangle \langle \mathbf{m_{R'}} \rangle$, where $\mathbf{R}$ and $\mathbf{R'}$ refer to two different sites.

B. Results on the MAE

Figure 2 shows the calculated values of ΔE_b, ΔE_{dd}, and ΔE for film thicknesses of two to seven layers of Fe_xCo_{1-x} on Cu(001) as a function of the Fe concentration x. Note that in all cases these values correspond to the so-called ground state configuration (see Sec. II). For two layers and—although not shown in Fig. 2—for one layer of Fe/Co the MAE is negative for all concentrations under consideration; i.e., the magnetization is always in plane. This occurs despite the fact that for an increasing amount of Fe, ΔE_b clearly gets positive, since the magnitude of the negative ΔE_{dd} increases, too. For three layers and for concentrations up to $x=0.85$, ΔE_b is quite stable around 0.4 eV, but because of the large negative value of ΔE_{dd}, the MAE is still negative. At $x=0.9$, i.e., at the concentration at which the ground state
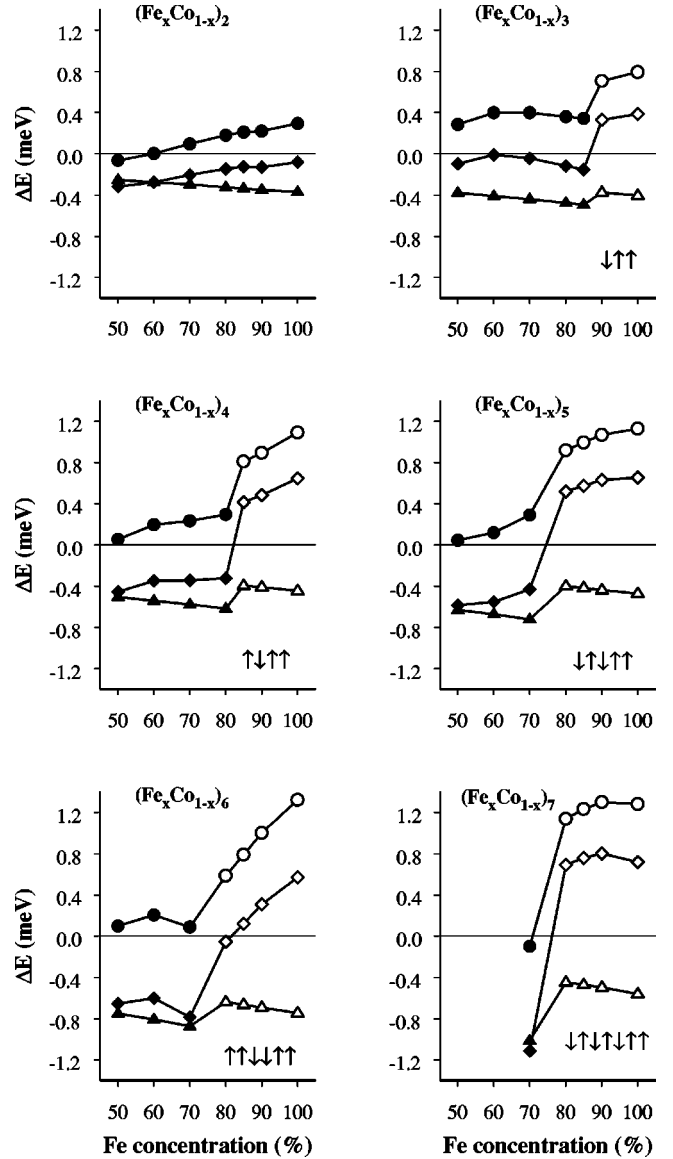

FIG. 2. Calculated magnetic anisotropy energies as a function of the Fe concentration for $(Fe_xCo_{1-x})_n/Cu(001)$ for $n=2,3,4,5,6,7$. The type of antiferromagnetic arrangement is indicated by arrows, whereby the Cu substrate is on the left hand side. Solid circles, ΔE_b for the ferromagnetic configuration; open circles, ΔE_b for the antiferromagnetic configuration; solid triangles, ΔE_{dd} for the ferromagnetic configuration; open triangles, ΔE_{dd} for the antiferromagnetic configuration; solid diamonds, ΔE for the ferromagnetic configuration; open diamonds, ΔE for the antiferromagnetic configuration.

refers to antiferromagnetic interlayer coupling (see Fig. 1), a sudden increase can be seen in the MAE. This shows up in both contributions of the MAE. As discussed in Ref. 13, antiferromagnetic coupling reduces the magnetic moment of Fe; therefore, ΔE_{dd} clearly drops in magnitude. On the other hand, a strong enhancement of ΔE_b can be observed. A layer and composition resolved analysis shows that similarly to the case of pure Fe films this enhancement is entirely due to Fe sites.

The above effects caused by antiferromagnetic coupling apply also for the MAE of films thicker than three monolayers. In the range of the antiferromagnetic ground state ΔE_{dd} decreases in magnitude with decreasing Fe content. Once the
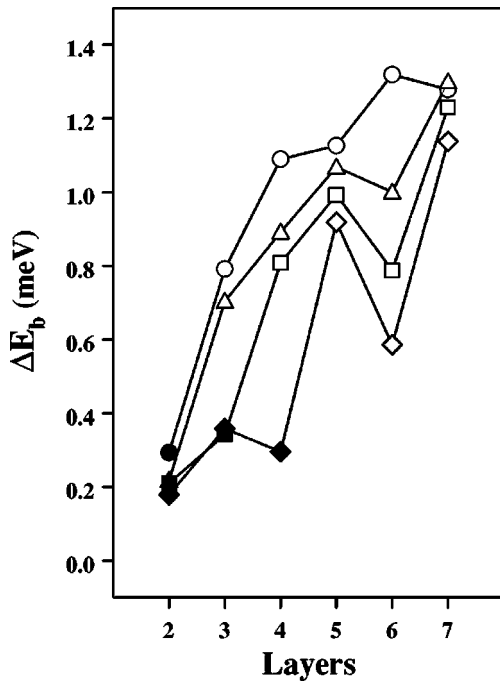
FIG. 3. Comparison of the calculated values of ΔE_b for different alloy compositions as a function of the number of Fe/Co layers. Circles, Fe$_{1.0}$Co$_{0.0}$; triangles, Fe$_{0.9}$Co$_{0.1}$; squares, Fe$_{0.85}$Co$_{0.15}$; diamonds, Fe$_{0.8}$Co$_{0.2}$. Open symbols refer to the antiferromagnetic configurations as displayed in Fig. 2 and solid symbols to the ferromagnetic one.

FIG. 4. ''Phase diagram'' of the reorientation transition in terms regions of perpendicular and in-plane magnetization. Solid circles in the shaded region correspond to a positive MAE and therefore to a perpendicular magnetization. Open circles indicate a negative MAE and an in-plane orientation of the magnetic moments.

system becomes ferromagnetic ΔE_{dd} suddenly increases due to the enhanced Fe moments. Beyond this transition, i.e., by further decreasing the Fe concentration, ΔE_{dd} again decreases. The change of ΔE_b with respect to x is even more dramatic: in each case at the transition from the antiferromagnetic to ferromagnetic state it drops by more than 0.5 eV, in the seven-layer case even by more than 1 eV. Therefore, whenever the ground state is found to be ferromagnetic the MAE turns out to be negative, while for an antiferromagnetic ground state it is positive. The only exception—namely, that an antiferromagnetic ground state does not correspond to an out-of-plane orientation of the magnetization—is the case of six layers at an alloy composition of Fe$_{0.8}$Co$_{0.2}$, which, however, can be attributed to the somewhat different arrangement of antiferromagnetic coupling as compared to the other cases [see also the case of Fe$_6$/Cu(001) (Ref. 13)]. As compared to the ferromagnetic arrangement the enhancement of ΔE_b caused by antiferromagnetic coupling is clearly seen from Fig. 3.

Finally, in Fig. 4 a schematic representation of regions of in plane or perpendicular magnetization can be given in terms of a ''phase diagram'' of the reorientation transition. In this figure points corresponding to an in-plane magnetization are displayed as open circles, while those for which perpendicular magnetization pertains are indicated as solid circles. In this plot also the data for a monolayer of Fe/Co are shown. By comparing this ''phase diagram'' with what has been said previously about the transition from ferromagnetic to antiferromagnetic coupling, it is now apparent that in regions where the layers are coupled antiferromagnetically, perpendicular magnetization prevails [except for
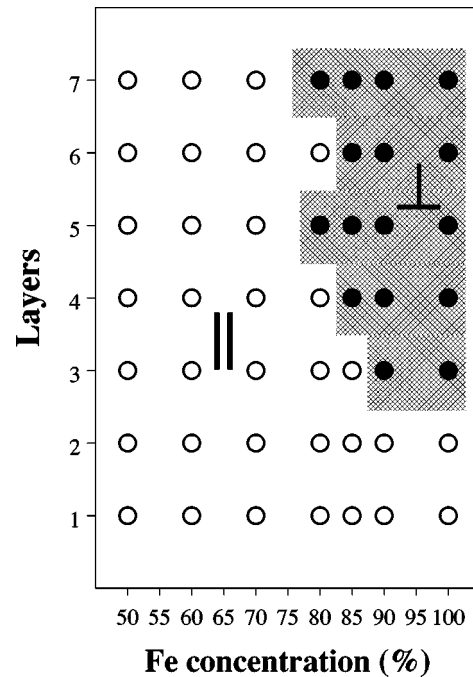
(Fe$_{0.8}$Co$_{0.2}$)$_6$], while in regions of ferromagnetic coupling the films show in-plane magnetization.

IV. CONCLUSIONS

In summary, reorientations between an in-plane and a perpendicular orientation of the magnetization are predicted in films thicker than two monolayers of Fe/Co and at alloy compositions with Fe concentrations above approximately 70%. As one moves from three to five layers a reorientation occurs at decreasing Fe concentrations. Above this film thickness, however, the range of compositions for the reorientation seems to remain approximately constant. The mechanism responsible for the different orientations of the moments can be reduced to different types of coupling between layers, which in turn depends on the thickness of the film and on the composition of the alloy.

The calculated concentration range of only in-plane magnetization fits rather well to a recent experimental study by Dittschar et al.[24] There is also good agreement between the calculated anisotropy energies at the reorientation (e.g., for $n=3$, $\Delta E_b \simeq 0.5$ meV, which with respect to the in-plane spacing of fcc Cu, namely, $a=4.83$ a.u., corresponds to a value of about 1.2 mJ/m^2) and what has been fitted in Ref. 24 ($E_{SOC} \simeq 1.1$ mJ/m^2). Furthermore, as can be deduced from Fig. 2, in agreement with experiment this anisotropy energy gradually increases when the film thickness increases.

The experimental data,[24] however, do suggest that the reorientation transition occurs already at a film thickness of two monolayers, and that above four monolayers, only in-plane magnetization is present (except for Fe concentrations close to 1, which, however, was not investigated in the cited

experimental study). In order to comment on this discrepancy, it should be recalled that the present results correspond to $T = 0$ K, while the measurements were carried out in a temperature range between 120 and 300 K. Since at a given composition finite temperatures most likely decrease the critical thickness of the film (or for a given film thickness decreases the critical Fe concentration) at which the reorientation occurs, the ''phase field'' of perpendicular orientation in the phase diagram of Ref. 24 is expected to move to larger film thicknesses as the temperature approaches $T = 0$ K.

Furthermore, as mentioned already in Sec. II., the present calculations do not take into account the experimentally recorded[24] tetragonal expansion of the Fe/Co film ($c/a \sim 1.04$) for film thicknesses of $n < 4$ and for Fe concentrations $x > 0.7$. Since like in the case of Ni overlayers on Cu(100) (Refs. 7 and 25) such lattice modulations can be of

quite some importance for the MAE of ultrathin films, within this particular range of layers n and concentrations x an inclusion of these modulations might improve the agreement between the present results and the available experimental data.

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