Magnetic anisotropy of thin films of Co on Cu(111)

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The magnetic anisotropy of epitaxial Co_N/Cu(111),$1\leqslant N \leqslant 7$, films is investigated in terms of the relativistic spin-polarized screened Korringa-Kohn-Rostoker method by taking into account uniform relaxations of the Co interlayer distance between -4% and $+3\%$ with respect to the Cu parent lattice. While the spin-orbit coupling induced (band energy) part of the magnetic anisotropy is found to favor a perpendicular magnetization for $N \geqslant 2$, because of the dominating contribution of the magnetic dipole-dipole interaction to the magnetic anisotropy energy, an in-plane magnetization is energetically preferred for essentially all relaxations and layer thicknesses. Only for $N=2,3$ the anisotropy between an in-plane and a perpendicular orientation of the magnetization is not significantly different. The theoretical results are in good agreement with recent experiments based on pulsed layer deposition.

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I. INTRODUCTION

Growth, morphology, and magnetic structure of ultrathin films of Co on Cu(111) have been a matter of intensive experimental investigations in the recent past. Prepared by molecular-beam epitaxy (thermal deposition, TD) Co films (ML) grow in a face-centered-cubic (fcc) structure below a thickness of 2 ML by forming mostly 2 or 3 ML high islands,[1,2] above this thickness they undergo a gradual fcc $\rightarrow$hcp transformation[3,4] aided by hexagonal-close-packed (hcp) stacking faults. By using Pb as a surfactant on Cu(111) the quality of growth of Co films has been considerably improved, however, this leads to a substantial change in the magnetic properties of the system, namely induced by a Pb overlayer.[5] An experimental method using pulsed layer deposition[6,7] (PLD) made it possible to reduce significantly the number of stacking faults during the initial growth of Co films on Cu(111) and thus to delay the fcc$\rightarrow$hcp structural transition to about 6 ML of Co:[8,9] in contrast to perpendicularly magnetized thermally deposited hcp films,[9] these films show an overall in-plane magnetization.

As the practically perfect layer-by-layer growth of the PLD films represents an ideal situation for theoretical investigations, the purpose of the present paper is to calculate and discuss the magnetic anisotropy properties of epitaxial Co_N/Cu(111) ($1\leqslant N\leqslant 7$) films. To our knowledge, *ab initio* calculations of the magnetic anisotropy energy (MAE) have been reported so far only for Co_1/Cu(111) (in fact, also as capped by additional Cu overlayers) by Zhong *et al.*[10] There a MAE of -0.31 meV per unit cell (in-plane) was found for a Co ML occupying positions of a perfect fcc Cu parent lattice, while for a self-consistently relaxed Co monolayer ($\sim$7.6% inward relaxation) a MAE of -0.30 meV per unit cell was reported. In many cases the effect of layer relaxations is known to be decisive for the MAE, most prominently for Ni/Cu(100) film systems, where it gives rise to an inverse reorientation transition with increasing film thickness.[11]

II. THEORETICAL APPROACH

The following theoretical study of the MAE of thin films of Co on Cu(111) is performed by using the fully relativistic spin-polarized screened Korringa-Kohn-Rostoker (KKR) method[12] in the context of the spin-polarized local-density functional as parametrized by Vosko *et al.*[13] Various uniform relaxations R with respect to the interlayer distance in the substrate (fcc Cu), $-4\%\leqslant R\leqslant +3\%$, have been considered by extending this method to systems of layers sharing only the same in-plane translational symmetry but otherwise can differ in respective interlayer distances.[11,14] It should be noted that the same in-plane translational symmetry is a necessary requirement for making use of two-dimensional lattice Fourier transformations. For each system, i.e., for each uniform relaxation R and number of Co layers N, the electronic and magnetic structure is calculated self-consistently for a ferromagnetic configuration corresponding to an orientation of the magnetization perpendicular to the planes of atoms. In all systems investigated, an additional buffer of three substrate Cu layers is treated self-consistently. It was found that within the atomic sphere approximation 30 $k_\parallel$ points in the hexagonal irreducible surface Brillouin zone guarantee the necessary numerical accuracy for the effective potentials and effective exchange fields treated.

The MAE E_a,

$$E_a = E(\parallel) - E(\perp), \tag{1}$$

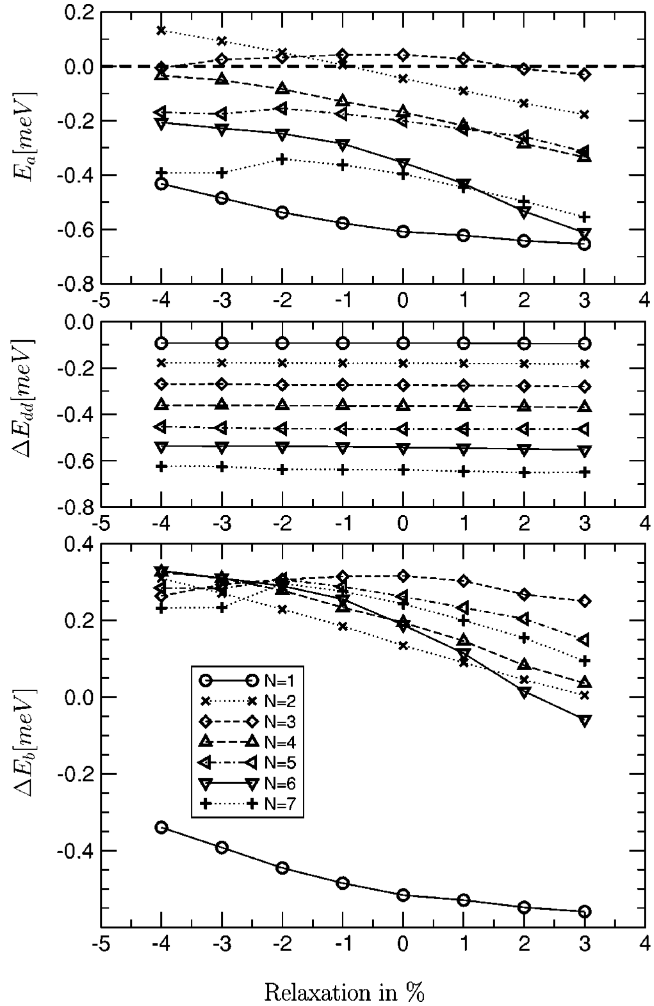

FIG. 1. Calculated total magnetic anisotropy energies E_a, band energy anisotropies ΔE_b, and magnetic dipole-dipole energy differences ΔE_{dd} for Co_N/Cu(111) ($1 \leqslant N \leqslant 7$) as a function of a uniform relaxation R.

defined as the energy difference between a uniform in-plane and a uniform perpendicular orientation of the magnetization, is obtained by making use of the magnetic force theorem,[12,15] namely, as a sum over the respective band energy difference ΔE_b and the magnetic dipole-dipole energy contribution ΔE_{dd},

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

It is worthwhile to mention that ΔE_b, evaluated here with 690 $k_\parallel$ points in the ISBZ in order to guarantee a relative accuracy of below 5%, can be identified as the contribution to the MAE induced by the spin-orbit interaction, while ΔE_{dd} is a purely classical term denoted usually as the shape anisotropy.

Denoting the magnetic moment in the cell centered around the atomic position $\mathbf{R}$ by $m_{\mathbf{R}}$ the (classical) magnetic dipole-dipole interaction energy is given (in atomic Rydberg units) by

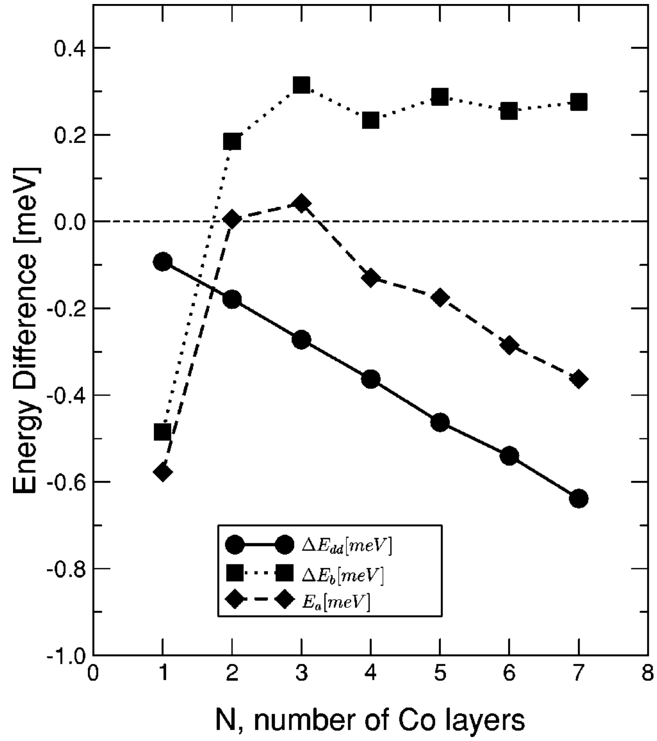

FIG. 2. Band energy anisotropies ΔE_b (squares), dipole-dipole energy differences ΔE_{dd} (circles), and magnetic anisotropy energies E_a (diamonds) as a function of the number of Co layers for the experimentally given relaxation of $R = -1\%$.

$$E_{dd} = \frac{1}{c^2} \sum_{\mathbf{R},\mathbf{R}'}{}' \left\{ \frac{m_{\mathbf{R}} \cdot m_{\mathbf{R}'}}{|\mathbf{R}-\mathbf{R}'|^3} - 3\frac{[m_{\mathbf{R}} \cdot (\mathbf{R}-\mathbf{R}')][m_{\mathbf{R}'} \cdot (\mathbf{R}-\mathbf{R}')]}{|\mathbf{R}-\mathbf{R}'|^5} \right\}. \tag{3}$$

This expression can be evaluated very efficiently by making use of the underlying two-dimensional translational symmetry; for further theoretical and computational details, see Refs. 12 and 16. Note that due to the definition in Eq. (1), positive/negative values of E_a imply a perpendicular/in-plane orientation of the magnetization.

III. RESULTS

In Fig. 1 the MAE is displayed together with the corresponding band energy and magnetic dipole-dipole energy contribution for Co_N films ($1 \leqslant N \leqslant 7$) on Cu(111) as a function of the uniform relaxation rate R of the cobalt interlayer distance. The results show that for $N=1$ an in-plane orientation is clearly favored, while for $N=2, 3$ the MAE is around zero; for increasing contractions, a small perpendicular magnetic anisotropy occurs. For $N \geqslant 4$, however, an in-plane anisotropy develops with increasing film thickness. The two contributions to the MAE, namely, the band energy and the magnetic dipole-dipole energy, have significantly different properties: ΔE_{dd}, favoring always an in-plane magnetization, is essentially independent of relaxations, at least in the regime investigated, and increases in good approximation

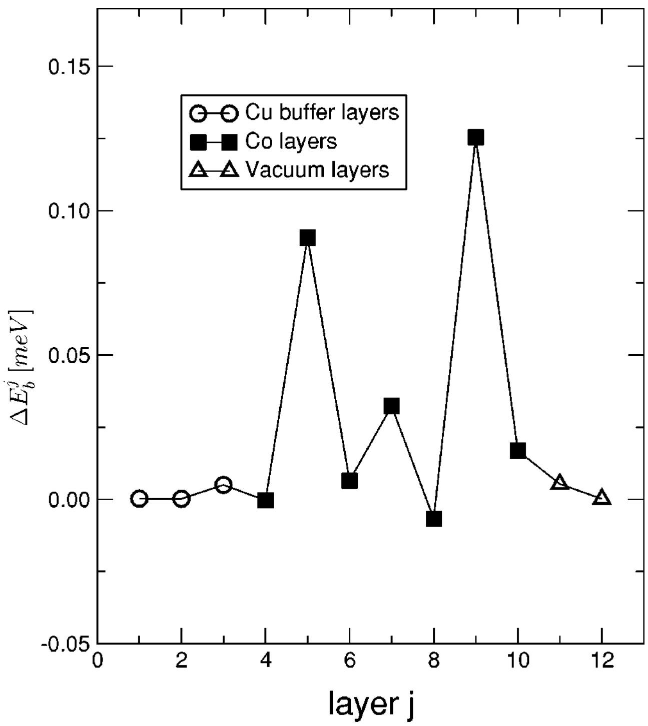

FIG. 3. Layer-resolved band energy anisotropies ΔE_b^j for a film of seven cobalt layers with a uniform relaxation of $R = -1\%$.

linearly with the number of film layers. This simple behavior of ΔE_{dd} results from the dominating spin-only Co magnetic moments, which are fairly insensitive to both relaxations and the thickness of the film. In contrast to ΔE_{dd} the band energy difference ΔE_b does depend on both the thickness of the film and the relaxation. In agreement with the theoretical investigations of Zhong *et al.*[10] ΔE_b is negative for $N=1$; however, it seems to show a more pronounced dependence on R as compared to the one that can be deduced from Ref. 10. For not-too-large values of R the band energy ΔE_b favors a perpendicular magnetization for $N \geqslant 2$.

For the specific case of $R = -1\%$ (closest to the experiment, see Ref. 8), the variation of the MAE and its contributions with respect to N is visualized in Fig. 2. After an abrupt jump from about -0.5 meV at $N=1$ to nearly 0.2 meV at $N=2$, ΔE_b oscillates for $N \geqslant 3$ around about 0.3 meV with an amplitude that reduces by increasing the number of cobalt layers. The fact that for $N \geqslant 3$ the band energy difference is not significantly changing with the film thickness can be deduced from Fig. 3, showing the layer-resolved contributions of ΔE_b for the thickest system under consideration, Co_7/Cu(111). As can be seen ΔE_b is mainly located in the cobalt layers second closest to the interface and to the surface; the corresponding contributions from the three most interior cobalt layers alternate in sign, but are remarkably smaller in magnitude. Therefore, the interior of the Co film does not contribute significantly to ΔE_b. It should be noted that a similar oscillating behavior of the MAE was found for Co films on Cu(100).[17] Obviously, however, ΔE_{dd} increases in magnitude with N, and thus results in an in-plane magnetization for $N \geqslant 4$.

Changes in physical quantities such as charges or magnetic moments with respect to the magnetic orientation are usually very small as compared to their absolute values. In the first row of Fig. 4 the layer resolved total (orbital and spin) magnetic moments for the in-plane and the perpendicular orientation are shown for three (first column) and seven (second column) layers of Cobalt. In the second and third row of this figure the corresponding layer resolved orbital and spin magnetic moment differences are displayed for the same systems. One can see in Fig. 4 that the absolute value of the difference in the orbital magnetic moments is one order of magnitude larger than the corresponding difference in the spin magnetic moments. It should be noted, however, that considering the actual size of the anisotropy energy, see Fig. 2, the minute differences to be read off from Fig. 4 are not surprising at all.

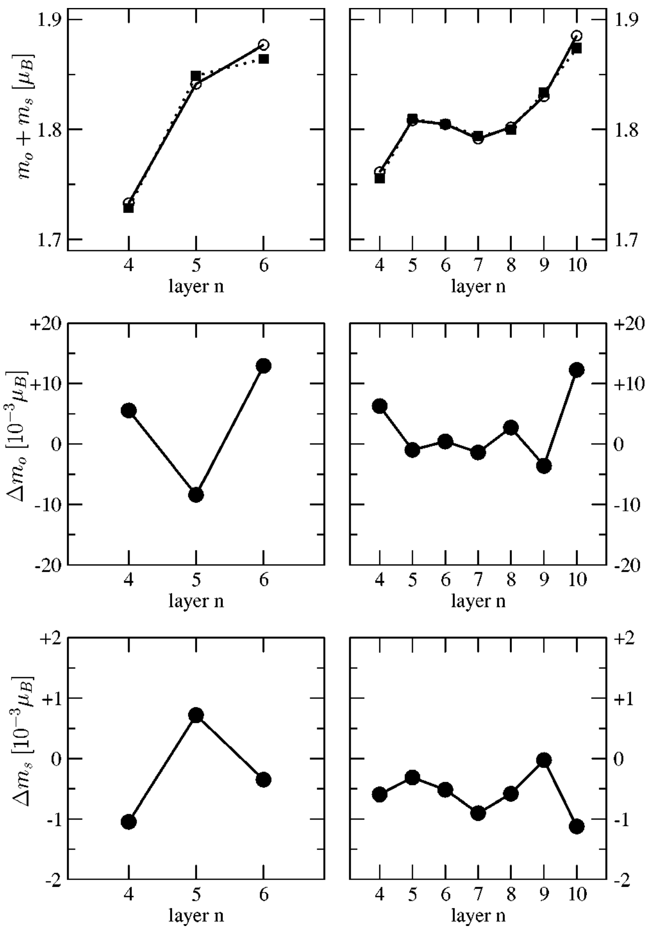

FIG. 4. Layer resolved total magnetic moments (top; circles correspond to the perpendicular orientation of the magnetization and squares correspond to the in-plane orientation) and differences in the orbital (middle) and spin (bottom) magnetic moments with respect to the orientation of magnetization. The left column refers to the three-layer thick Co film, the right column to a film with seven cobalt layers. Only the cobalt layers are shown.

PLD grown Cu(111)/Co_N films show an in-plane easy axis of magnetization for all thicknesses investigated[9] ($N = 2, \ldots ,15$) in good agreement with our theoretical results. When comparing experimental and theoretical results, one should keep in mind, however, that for very thin films the Curie temperature drops significantly. This means that the measurement temperature of 230 K is no longer far below T_c, see Ref. 9. Usually, when approaching T_c from below, the anisotropy falls more rapidly with increasing temperature than the magnetization. Therefore, in the experiment, the magnetic dipole-dipole contribution can overwhelm the tiny perpendicular band energy anisotropy, pulling the magnetization in-plane. In contrast to this behavior, TD films do also show a perpendicular easy axis of magnetization for various film thicknesses, in addition to a more three-dimensional-like growth mode of the films. These facts and the present results prove once again the strong dependence of the magnetic properties of thin films on the experimental growth technique. Unfortunately, in *ab initio*–like descriptions at best the electronic temperature (via the Fermi-Dirac distribution function) for the band energy part can be taken into account. This implies that for a rigorous study of temperature effects in these systems a Heisenberg-like model with *ab initio*–like parameters is needed in order to calculate corresponding free energies and to attempt to evaluate critical temperatures. Evaluation schemes for such *ab initio* parameters, however, are still under discussion and, at present, not available.

IV. CONCLUSION

We have investigated *ab initio* the magnetic anisotropy energy of the system $Co_N/Cu(111)$ using the fully relativistic spin-polarized screened KKR method by taking into account uniform interlayer relaxations in the Co film between $R=-4\%$ and $+3\%$. It was shown that the calculations predict an in-plane easy axis of magnetization for essentially all thicknesses and relaxations. Furthermore, the results prove that the main contributions to the magnetic anisotropy arise from the Cu/Co interface and the Co/Vac surface: relaxations therefore do not influence the anisotropy energy in a very sensitive way. This, in turn, justifies the simplified model of uniform relaxations used here instead of specific relaxation profiles. The obtained results are in good agreement with the experimental findings on PLD ultrathin films of the same system.[9]

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