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

R. Hammerling, C. Uiberacker, J. Zabloudil, and P. Weinberger

Center for Computational Materials Science, Technische Universität Wien, Getreidemarkt 9/134, 1060 Vienna, Austria

L. Szunyogh

Center for Computational Materials Science, Technische Universität Wien, Getreidemarkt 9/134, 1060 Vienna, Austria

and Department of Theoretical Physics, Budapest University of Technology and Economics, Budafoki út. 8, 1521 Budapest, Hungary

J. Kirschner

Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle/Saale, Germany

(Received 26 March 2002; revised manuscript received 6 June 2002; published 1 August 2002)

The magnetic anisotropy of epitaxial Co/N/Cu(111), \( 1 \leq N \leq 7 \), 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 \( N \geq 2 \), 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.

DOI: 10.1103/PhysRevB.66.052402

PACS number(s): 75.30.Gw, 75.70.Ak, 75.70.Cn

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 (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, 6, 7 in contrast to perpendicularly magnetized thermally deposited hcp films, 8 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) \( (1 \leq N \leq 7) \) films. To our knowledge, \textit{ab initio} calculations of the magnetic anisotropy energy (MAE) have been reported so far only for Co/N/Cu(111) \( (1 \leq N \leq 7) \) films, 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 relax-

\[ E_a = E(\parallel) - E(\perp), \]

\[ E_a = E(\parallel) - E(\perp), \]
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, namely, as a sum over the respective band energy difference $D_{E_b}$ and the magnetic dipole-dipole energy contribution $D_{E_{dd}}$.

\[
E_a = D_{E_b} + D_{E_{dd}}. \tag{2}
\]

It is worthwhile to mention that $D_{E_b}$, evaluated here with 690 $k$ 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 $D_{E_{dd}}$ 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

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

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$_N$/Cu(111) ($1 \leq N \leq 7$) as a function of the uniform relaxation rate $R$. 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 $N>4$, 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: $D_{E_{dd}}$, favoring always an in-plane magnetization, is essentially independent of relaxations, at least in the regime investigated, and increases in good approximation.
linearly with the number of film layers. This simple behavior of $\Delta E_{dd}$ 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 $\Delta E_{dd}$ the band energy difference $\Delta E_b$ does depend on both the thickness of the film and the relaxation. In agreement with the theoretical investigations of Zhong et al.\textsuperscript{10} $\Delta E_b$ 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 $\Delta E_b$ favors a perpendicular magnetization for $N>2$.

For the specific case of $R=\sim -1\%$ (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 $N=2$, $\Delta E_b$ oscillates for $N \geq 3$ around about $0.3$ meV with an amplitude that reduces by increasing the number of cobalt layers. The fact that for $N \geq 3$ the band energy difference is not significantly changing with the film thickness can be deduced from Fig. 3, showing the layer-resolved contributions of $\Delta E_b$ for the thickest system under consideration, Co$_7$/Cu(111). As can be seen $\Delta E_b$ 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 $\Delta E_b$. It should be noted that a similar oscillating behavior of the MAE was found for Co films on Cu(100).\textsuperscript{17} Obviously, however, $\Delta E_{dd}$ increases in magnitude with $N$, and thus results in an in-plane magnetization for $N \geq 4$.

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.

PLD grown Cu(111)/Co$_N$ films show an in-plane easy axis of magnetization for all thicknesses investigated\textsuperscript{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. 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.

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.

ACKNOWLEDGMENTS

Financial support was provided by the Fonds zur Förderung der wissenschaftlichen Forschung (Project No. W004), the Austrian Ministry of Science (Project No. GZ 45.504), and the Hungarian National Science Foundation (Grants No. OTKA T030240 and T037856). The collaboration between L.S. and the Austrian partners was partially sponsored also by the Research and Technological Cooperation Project between Austria and Hungary (Contract No. A-23/01).