Overlayer-dependent magnetic moment and anisotropy of a Co monolayer on Cu(100)

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(Received 29 September 1997)

An extensive study of the magnetic moment and of the magnetic anisotropy of a Co monolayer on a Cu(100) substrate capped by an additional $3d$, $4d$, or $5d$ monolayer is presented in terms of first-principles calculations. While for the magnetic moment of the Co and the cap layer systematic trends can be traced that seem to be consistent with the Stoner model, the dependence of the magnetic anisotropy energy on the type of overlayer is found to be less straightforward. However, in some selected cases a correlation of the magnetic anisotropy with the specific features in the electronic structure of the Co and the cap layer can be pinpointed. [S0163-1829(98)00915-1]

Due to its theoretical challenge to solid state physics and technological importance in magneto-optical recording magnetic anisotropies in thin films and multilayers of transition metals have attracted considerable effort in research.[1] In particular, the anomalous perpendicular magnetic anisotropy (PMA) observed in ultrathin Co films on Au and Pt (111) surfaces as coated by additional Ag, Cu, Au, or Pd layers seemed to indicate a close relationship between the material specific electronic structure and the surface magnetic anisotropy.[2] By using *ab initio* computational techniques, it then was shown that indeed the anomalous PMA is a consequence of the change in the interfacial hybridization between the Co and the first cap layer as the thickness of the cap varies.[3,4]

So far experimental and theoretical studies have been mainly confined to caps of noble transition metals (Cu, Ag, Au) or of Pd and Pt. In these studies the magnetic moment of the Co monolayer (ultrathin film) was found to be robust and the actual value of the magnetic anisotropy energy (MAE) resulted from a ''fine tuning'' of the electronic states of Co near the Fermi level. In the case of Pd and Pt the spin polarization of the cap layers turned out to be also important. It is the aim of the present paper to extend systematically the investigations of cap layers to all $3d$, $4d$, and $5d$ transition metal elements on top of a Co monolayer on a Cu(100) substrate.

For each system under consideration, self-consistent calculations have been performed within the local-spin-density approximation (LSDA) and the atomic-sphere approximation (ASA) by solving the Pauli-Schrödinger equation (spin-orbit coupling neglected) by means of the screened Korringa-Kohn-Rostoker (SKKR) method, which has been shown to be extremely suitable to calculate the electronic structure of surfaces and interfaces.[5–7] As in the calculations only spd angular-momentum scattering channels were considered, La, where $4f$ states contribute to the valence band, was excluded from the present study. In order to obtain self-consistency, energy integrations were performed along a semicircular contour in the complex plane using 16 points according to an asymmetric Gaussian sampling, and for the two-dimensional Brillouin-zone integrations 45 $\mathbf{k}_\parallel$ points in the corresponding irreducible wedge (IBZ) were used.

No attempt was made to account for surface reconstruction, relaxation, or strain, i.e., we assumed a perfect parent fcc lattice[8] corresponding to the experimental lattice constant of bulk copper ($a=6.83$ a.u.). Since, as is well known, such lattice distortions do frequently occur at surfaces, the results presented in this paper only refer to a model study. This is most obvious for $4d$ and $5d$ overlayers where large lattice mismatch has to be expected when comparing the corresponding bulk lattice constants to that of bulk Cu. However, as far as trends are concerned, we believe that the present study represents a reliable theoretical description of the electronic structure of capped Co monolayers on Cu(100).

According to the force theorem (see, e.g., Ref. 9) the obtained self-consistent potentials were then used to calculate the band-energy difference $\Delta E_b = E_b^{\parallel} - E_b^{\perp}$ in terms of the fully relativistic spin-polarized SKKR method,[10] where the superscripts $\perp$ and $\parallel$ refer to a normal and a parallel to surface orientation of the effective exchange field, respectively. Here we generally used 325 $\mathbf{k}_\parallel$ points in the IBZ, ensuring a relative accuracy of less than 10% for ΔE_b as was checked in

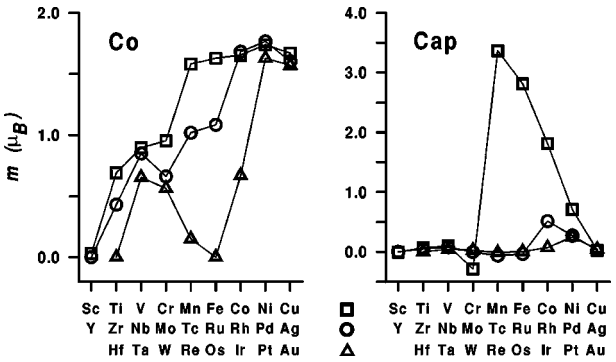

FIG. 1. Calculated magnetic moments of the Co and the cap layer in M/Co/Cu(100) systems, where M denotes a $3d$ (squares), $4d$ (circles), or $5d$ (triangles) transition metal. Solid lines serve as a guide for the eye.

some cases by increasing the number of $\mathbf{k}_{\|}$ points up to 990. The MAE is then given by $\Delta E=\Delta E_{\mathrm{b}}+\Delta E_{\mathrm{dd}}$, where ΔE_{dd} denotes to the magnetostatic dipole-dipole interaction.

Figure 1 displays the calculated magnetic moments of the Co and cap layers. At the end of each d row the magnetic moment of Co is quite robust ($1.6\mu_{\mathrm{B}}<m_{\mathrm{Co}}<1.8\mu_{\mathrm{B}}$), dropping, however, rapidly for Cr, Ru, and Ir in the $3d$, $4d$, and $5d$ series, respectively. Near the beginning of the $3d$ row m_{Co} decreases monotonously, while for $4d$ and $5d$ caps it reaches a local maximum for Nb and Ta, respectively. In the case of an Os overlayer and at the very beginning of each of the d rows (Sc, Y, and Hf) the Co layer is found to be even nonmagnetic.

As to be expected, $3d$ elements with a well-localized, open d shell (Mn, Fe, Co, and Ni) have large magnetic moments as overlayers, reaching a maximum of $3.5\mu_{\mathrm{B}}$ for Mn. Concomitantly to the abrupt decrease of m_{Co} for Cr, in the $3d$ row the magnetic moments of the corresponding caps are in turn strongly reduced below Mn. It should be noted that Cr shows a tendency for antiferromagnetic coupling with Co, a possible formation of an in-plane antiferromagnetism within the Cr layer, however, was not investigated. In the $4d$ and $5d$ rows much smaller maxima for the moment of the cap layer are present for Rh and Pt, respectively. Most of the tendencies observed for the magnetic moments of the cap layers can be related to those of $3d$, $4d$, and $5d$ monolayers on Ag or Au substrates,[11] which in turn fit fairly well the simple Stoner criterion.

For the cases of the $3d$ and $5d$ overlayer systems the local densities of states (LDOS) of the Co and the corresponding cap layer as derived from self-consistent paramagnetic calculations are shown in Fig. 2. In both cases, the d band of the overlayer moves gradually up in the energy as its filling increases, and since concomitantly the atomic number decreases, the overlayer's d band gets more and more delocalized. Since $5d$ bands are obviously more delocalized than the $3d$ bands, especially around the Fermi level E_F, they give rise to a different kind of hybridization with the Co d states. The variation of the LDOS of Co at E_F can well be correlated with the variation of m_{Co} in Fig. 1. In particular, for $5d$ caps, the minimum of m_{Co} for Os and the subsequent maximum for W and Ta are clearly mapped in a corresponding minimum and maximum in the LDOS of Co at E_F.

Turning now to the results for the MAE, we first note that

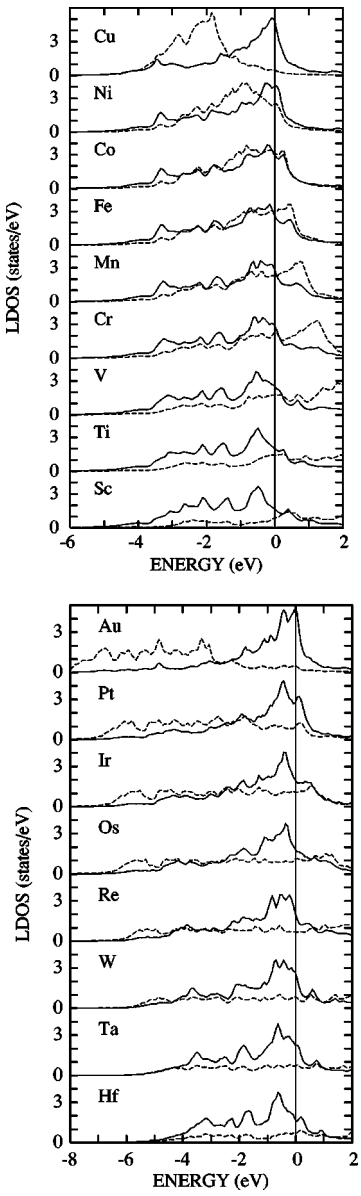

FIG. 2. Calculated paramagnetic local densities of states (LDOS's) for the Co (solid lines) and the cap (dashed lines) layer in M/Co/Cu(100) systems. M: $3d$ (upper panel) and $5d$ (lower panel) transition metals. The zero of energy corresponds to the Fermi level. In each entry the label denotes the corresponding cap.

since $\Delta E_{\mathrm{dd}}\simeq -0.022$ meV$\times(m_{\mathrm{Co}}^2+m_{\mathrm{cap}}^2)$ (see, e.g., the Appendix of Ref. 10), ΔE_{dd} is only larger in magnitude than 0.1 meV for Mn, Fe, and Co overlayers. Therefore, in what follows we focus on the analysis of ΔE_{b}. The calculated ΔE_{b}'s for the Co and cap layers as well as the total MAE (including also ΔE_{dd}) are shown in Fig. 3. It is important to mention that for an uncovered Co monolayer we obtained a value of -0.38 meV for ΔE_{b}, which is in excellent agreement with previous theoretical results.[12]

Apparently, the variation of the MAE upon changing the element forming the cap is much more complicated than that of the magnetic moments. As can be seen from Fig. 3, in most cases a cap enhances the MAE as compared to Co/Cu(100), although within a particular row of transition-metal elements the MAE changes sign several times. The anomalous PMA mentioned in the Introduction shows up as

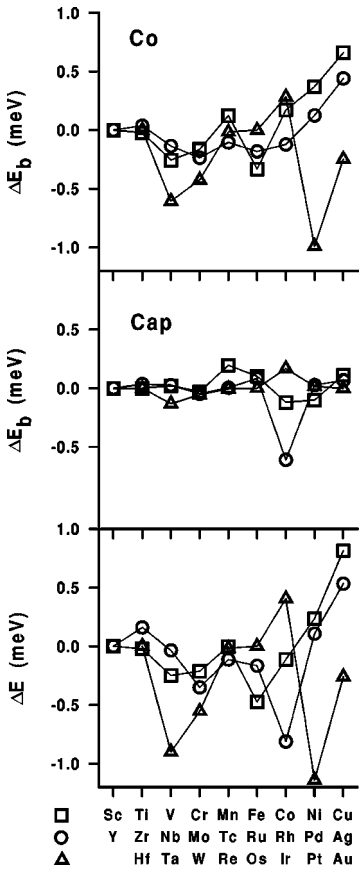

FIG. 3. Calculated MAE for M/Co/Cu(100) systems. M: $3d$ (squares), $4d$ (circles), and $5d$ (triangles) transition metals. Upper and middle panel: Respective band energy contributions, ΔE_{b}, of the Co and the cap layer. Lower panel: total MAE, $\Delta E=\Delta E_{\mathrm{b}}+\Delta E_{\mathrm{dd}}$. Solid lines serve as guide for the eye.

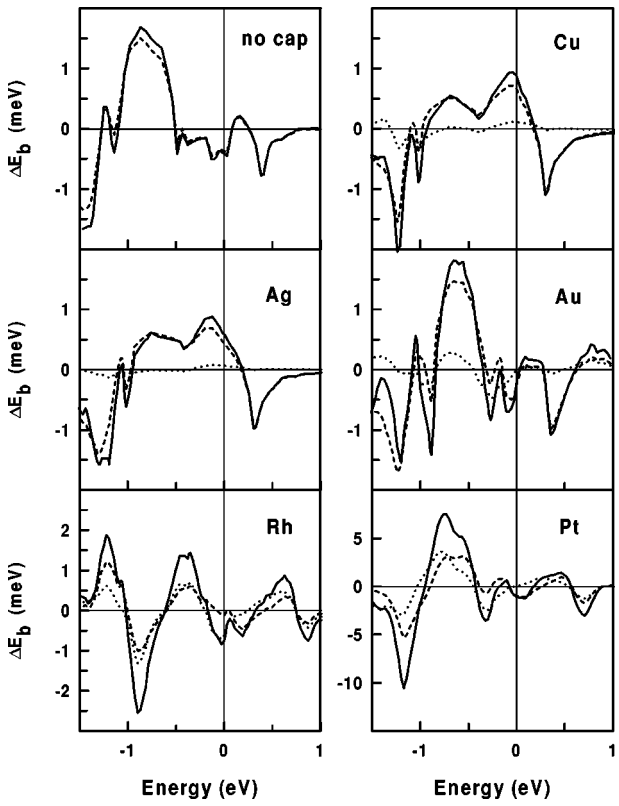

FIG. 4. Band energy anisotropies ΔE_{b} as a function of the highest occupied energy for Co/Cu(100) and M/Co/Cu(100) (M=Cu, Ag, Au, Rh, and Pt). Solid line, total; dashed line, contribution of Co; dotted line, contribution of cap. The energy zero refers to the Fermi level.

a large positive value for a Cu and Ag cap. Surprisingly enough, in the case of a Au cap the MAE is negative. In the $3d$ and $4d$ rows, when going from the end (Cu and Ag) to roughly the middle (Fe and Ru) the contribution of Co to ΔE_{b} is gradually decreasing and then starts to oscillate. The biggest negative value of ΔE_{b} for Co is found for a Pt overlayer, while the only noticeable contribution from a cap layer refers to Rh.

In the following, ΔE_{b} is analyzed by considering the following quantity:

$$\Delta E_{\mathrm{b}}(\varepsilon)=\int_{\varepsilon_b}^{\varepsilon} d\varepsilon' \Delta n(\varepsilon')\cdot(\varepsilon'-\varepsilon), \qquad (1)$$

where ε_b and $n(\varepsilon)$ denote the bottom of the valence band and the LDOS, respectively, and which has frequently been used to show the band filling properties of the MAE.[3,12] In Fig. 4, for some selected cases $\Delta E_{\mathrm{b}}(\varepsilon)$ is plotted together with its Co and cap contributions roughly in the regime of the Co minority band.

In the uncovered case, $\Delta E_{\mathrm{b}}(\varepsilon)$ reaches a maximum of more than 1.5 meV at about -0.9 eV and then drops rapidly to negative values, being negative also at E_F. Above E_F, a maximum and a minimum in $\Delta E_{\mathrm{b}}(\varepsilon)$ are found at about 0.2 eV and 0.4 eV, respectively. In terms of perturbation theory,[13] this characteristic curve of $\Delta E_{\mathrm{b}}(\varepsilon)$ can mainly be attributed to the distribution of minority $d_{xz,yz}$ and d_{z^2} states: $d_{xz,yz}$ states have a pronounced minimum just below E_F, while d_{z^2} states are located well around E_F.[14]

As seen in Fig. 4, the situation is remarkably different in the presence of a Cu or Ag cap. While the previously mentioned maximum at -0.9 eV moves up in energy and is considerably smaller than in the uncapped case, the subsequent maximum located at 0.2 eV above E_F for the Co/Cu(100) system is enhanced and shifted below E_F, causing thus a strong positive MAE in these two cases. Recalling again the arguments of perturbation theory, this enhancement of the MAE is due to the hybridization of Co d states with sp states of the cap layer, which results in a lowering of the Co d_{z^2} states around E_F, and also the minimum in the $d_{xz,yz}$ states below E_F disappears.

The case of a Au cap, however, is much more similar to that of the uncovered system, since a minimum of the $d_{xz,yz}$ states is located just below E_F and therefore $\Delta E_{\mathrm{b}}(\varepsilon)$ drops rapidly for -0.5 eV $<\varepsilon<$ -0.3 eV. Although eventually vanishing at E_F, the contribution of the Au layer to $\Delta E_{\mathrm{b}}(\varepsilon)$ is generally larger than that of Cu or Ag, which in turn can be related to the larger spin-orbit coupling. In Fig. 4 we also show the cases of a Rh and a Pt cap. Apparently, in these cases the cap layers contribute as much to $\Delta E_{\mathrm{b}}(\varepsilon)$ as the Co layer. This might be a consequence of the enhanced spin polarization in the cap layers which, especially for Pt, is accompanied by a large spin-orbit coupling, causing thus the giant values of $\Delta E_{\mathrm{b}}(\varepsilon)$ seen in the corresponding entry of Fig. 4.

Finally, it is interesting to note that the variation of m_{Co} with respect to the cap layer as seen in Fig. 2 can hardly be

correlated with the corresponding variation of ΔE_b of Co in Fig. 4. This is, in particular, obvious for the series of Cu, Ni, Co, and Fe as well as for that of Ag, Pd, and Rh, where m_{Co} first slightly increases and then decreases, remaining, however, roughly of the same size, while ΔE_b of Co remarkably drops. Although the main qualitative reasoning for these variations can be deduced from the overlap of the Co and the cap states, a detailed investigation of such an argumentation in terms of perturbation theory and of spin-orbit induced coupling for each of the different cases shown, lies beyond the purpose of the present paper.

The trends found in the size of the magnetic moments follow closely the Stoner model of ferromagnetism. According to our results the MAE depends in a rather complicated manner on the material of the cap, indicating that ultimately only studies based on *ab initio* techniques are able to predict correctly the MAE of technologically important materials. This is even more compulsory if, as in fact is necessary, one aims to include also structural and/or compositional anomalies related to surfaces and interfaces, which may affect considerably the value of the MAE in these systems.[3] Progress in research to include these effects is currently under way.

This paper resulted from a collaboration within the TMR Network on ''Ab initio calculations of magnetic properties of surfaces, interfaces and multilayers'' (Contract No. ERB4061PL951423). In addition, financial support by the Austrian Ministry of Science (GZ 308.941/2-IV/3/95) and the Hungarian National Scientific Research Foundation (OTKA Nos. T022609 and T021228) is kindly acknowledged.

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[14] Note that the d_{xx} and d_{yz} states are coupled through the L_z angular momentum component, whereby the d_{z^2} and $d_{xz,yz}$ states are coupled through the $L_{x,y}$ angular momentum components giving rise to positive or negative MAE, respectively. [See, e.g., J. Stöhr and H. König, Phys. Rev. Lett. **75**, 3748 (1995).]