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# Anomalous perpendicular magnetic anisotropy in a Co monolayer on Au(111)

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**Abstract**

Fully-relativistic spin-polarized local spin density calculations are performed for an Au covered Co monolayer on Au(111). In good agreement with experiments (i) the enhancement of perpendicular magnetic anisotropy for the capped Co layer, and (ii) a characteristic maximum in the anisotropy energy for one covering Au layer are obtained. The close relationship found between the anisotropies of orbital magnetic moments and the anisotropy energies supplies a partial interpretation of our results in terms of familiar perturbation theory. By using this framework the anomalous behavior of the magnetic anisotropy energies can be well explained due to changes in the interfacial hybridization between Co and Au.

*Keywords:* Thin films; Multilayers – metallic; Anisotropy – perpendicular; Surface

## 1. Introduction

Since many overlayer and superlattice systems have been predicted as candidates for high-density magneto-optical storage, theoretical investigations of the magnetic anisotropy of such systems became of particular interest. Despite extensive experimental and theoretical efforts, a complete, material-specific understanding of the surface magnetic anisotropy, as yet, has not been achieved. Recently several authors have discussed the anomalous perpendicular anisotropy in ultrathin Co films on Au and Pt (111) surfaces. As inferred from in situ polar magneto-optical Kerr effect (PMOKE) measurements, deposition of additional Ag, Cu, Au or Pd layers – forming a trilayer – increases the perpendicular anisotropy [1–4]. Furthermore, it has been revealed that the anisotropy energy has a maximum for coverages between 1 and 2 monolayers (ML). Each experimental paper discusses several possibilities as the theoretical reason of the observed behavior. One reason could be the magneto-elastic anisotropy, but no abrupt change in the structure is observed in the experiments. Although, in principle, a strong lattice mismatch can lead to such a contribution, it has been pointed out by Beauvillain et al. [4] that this can explain neither the measured peak nor the amplitude of the anomalous anisotropy.

In this paper we report results for a single monolayer of Co on Au(111) with different number of additional covering layers of Au. Our aim is to show that the experimental results can be reproduced (both qualitatively and quantitatively) by such calculations, confirming in turn that the anomalous dependence of the magnetic anisotropy energy on the overlayer coverage is caused by the corresponding changes in the electronic band structure.

## 2. Computational details

By treating relativity, spin-polarized scattering and the semi-infinite geometry on the same footing, the fully relativistic spin-polarized screened KKR method [5] has been used to carry out self-consistent calculations for each multilayer $Au_n/Co/Au(111)$ within the local spin density approximation (LSDA) [6], the atomic sphere approximation (ASA) and with a magnetic field pointing perpendicular to the surface. For the number of cap layers we chose $n = 0, 1, 2, 3, 4$ and $\infty$, where $\infty$ refers to the case of a Co interlayer in fcc Au. In all of the calculations a parent lattice of bulk fcc Au was considered, i.e., all nearest neighbor Au–Au, Au–Co and Co–Co distances were chosen to be equal (5.43 a.u.) and no attempt was made to take lattice strain into account.

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For each particular $n$, two subsequent calculations were performed using the previously determined self-consistent potentials, namely, with a magnetic field aligned perpendicular ($\perp$) and parallel ($\parallel$) to the surface. Within the framework of the force theorem [7,8] the magnetic anisotropy energy (MAE), $\Delta E$ is then given as a sum of the band-energy and the magnetostatic dipole–dipole energy contributions, $\Delta E_b = E_b^{\parallel} - E_b$ and $\Delta E_{dd} = E_{dd}^{\parallel} - E_{dd}$, respectively. Further details of the calculations can be found in Ref. [9].

## 3. Results and discussion

The calculated MAEs are shown in Fig. 1 together with the experimental results as deduced from the room temperature measurements of Ref. [4] for thicker Co films by extrapolation to the monolayer limit. Similar to the case of a Cu(001) substrate [10], for the uncovered monolayer the theory predicts in-plane orientation with a small anisotropy energy. In contrary, the experimental curve in Fig. 1 exhibits out-of-plane direction of the magnetization even in absence of Au coverage ($n = 0$). This might be due to the approximate extrapolation scheme used when deriving this curve from the experimental data, and/or to the fact that perfect pseudomorphism of Co on Au (as was assumed in the calculation) is never obtained experimentally.

By depositing an additional layer of Au onto the Co layer, both in theory and in experiment, there is an abrupt increase in the MAE implying a strong tendency to perpendicular magnetization. For more than 1 ML coverage the MAE slightly drops and rapidly converges with increasing coverage to its interface value. It should be noted that the theoretical values for the noninteger coverages were obtained in terms of the Coherent Potential Approximation [11,12] by supposing random distribution of the Au atoms and vacancies at the lattice positions of the incomplete

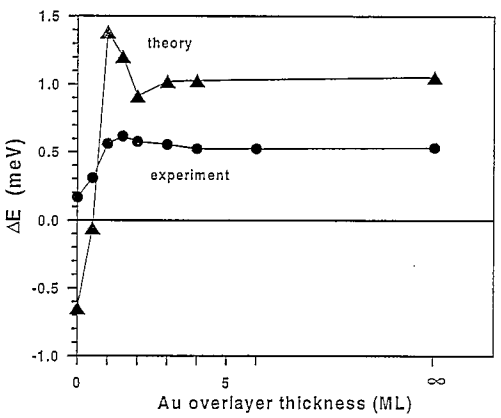


Fig. 1. Total (band + dipolar) magnetic anisotropy energies $\Delta E$ for a Co monolayer on Au(111) with different Au coverages. Triangles: calculated results; circles: experimental results from Ref. [4]. Solid lines serve as a guide to the eye.

Table 1
Calculated layer resolved band energy differences $\Delta E_b = E_b^{\parallel} - E_b^{\perp}$ (in meV) for $Au_n/Co/Au(111)$ films. In the first row, integer arguments with a prime label layers on the substrate side and those without a prime label layers on the substrate side and those without a prime label the cap layers. For $n = 0.5$ and 1.5 only the Au contributions are shown for the surface layers Au(1) and Au(2), respectively

| $n$ | | ... | Au(2) | Au(1) | Co | Au(1′) | Au(2′) |
|---|---|---|---|---|---|---|---|
| 0 | | | | | −6.626 | 0.045 | 0.001 |
| 0.5 | | | | 0.121 | −0.087 | 0.058 | 0.005 |
| 1 | | | | 0.197 | 1.207 | 0.157 | 0.018 |
| 1.5 | | | −0.007 | 0.183 | 0.952 | 0.142 | 0.014 |
| 2 | | | 0.012 | 0.161 | 0.711 | 0.144 | 0.011 |
| 3 | | −0.015 | 0.002 | 0.156 | 0.834 | 0.153 | 0.014 |
| 4 | 0.001 | 0.000 | 0.014 | 0.145 | 0.793 | 0.147 | 0.013 |
| ∞ | 0.001 | −0.002 | 0.012 | 0.149 | 0.821 | 0.149 | 0.012 |

surface layer. As can be seen from Fig. 1, all qualitative features of the experimental data are well reproduced, in particular the maximum at about 1 ML coverage. The theoretical values, however, are bigger than the experimental values by a factor of two. This factor can be accounted for intermixing at the Au/Co interface(s) as shown for $Fe_nCu(001)$ multilayers [13] and/or for surface roughness effects.

In order to investigate to what extent the different layers contribute to the MAE, in Table 1 the layer resolved band energies are shown. The main contribution to $\Delta E_b$ comes from the cobalt layer, however, for the covered samples a remarkable increase of $\Delta E_b$ arises also due to the Au layers closest to the Co layer. Obviously, the contributions from both the Co layer and the neighboring Au layers reach their maximum at 1 ML Au coverage giving rise to the characteristic peak seen in Fig. 1. This indicates that the changes in the hybridization between the Co and Au layers introduced by the cap layers is the main course of the anomalous magnetic anisotropy.

For ferromagnetic multilayers Bruno [14] has developed a perturbational treatment of the spin–orbit coupling interaction as combined with a tight-binding formalism to calculate the magneto-crystalline anisotropy. Within this theory the MAE is closely related to the anisotropy of the orbital magnetic moment, $\Delta m_{orb} = m_{orb}^{\parallel} - m_{orb}$. For a more than half filled d shell and by neglecting spin–channel coupling a negative proportionality between $\Delta E$ and $\Delta m_{orb}$ can be predicted, as confirmed recently also by means of X-ray magnetic circular dichroism for thicker of Au/Co/Au films [15,16]. In Fig. 2 the calculated $\Delta m_{orb}$'s are shown (with a negative sign) as a function of the thickness of the Au cap. In agreement with the observed in-plane anisotropy, for the uncapped case an enhancement of the orbital magnetic moment with respect to the parallel direction is found. For the capped cases displaying perpendicular anisotropy this enhancement is dramatically re-

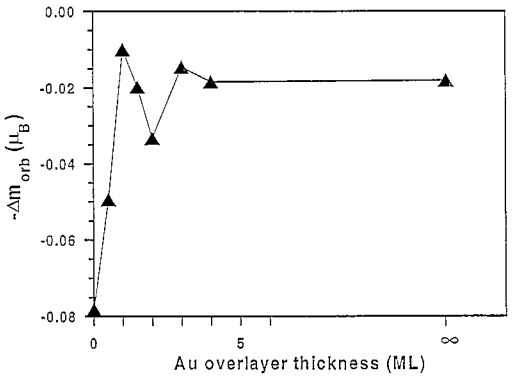


Fig. 2. Calculated values of orbital moment anisotropies, $\Delta m_{orb} = m_{orb}^{\parallel} - m_{orb}^{\perp}$, as shown with a negative sign, for $Au_n$/Co/Au(111) multilayers. The solid line serves as a guide to the eye.

duced, i.e., the perpendicular orbital magnetic moment is increased relative to $n = 0$, however, $\Delta m_{orb}$ does not change sign as would be expected from the simple tight-binding model. Comparing, however, Fig. 2 to Fig. 1 reveals that the shape of $\Delta m_{orb}$ accurately follows that of $\Delta E$, consequently, since $\Delta E_{dd}$ ($\sim -0.09$ meV) was fairly insensitive to the coverage, that of $\Delta E_b$. This at least implies that the changes in $\Delta E_b$ induced by the Au overlayer can possibly be interpreted in terms of perturbation theory.

In Fig. 3 the $d_{z^2}$ and $d_{xz,yz}$-like orbital projected minority DOS's of Co and the $p_z$-like DOS of Au(1) (see the labels in Table 1) are shown. Note that the $z$-axis is perpendicular to the layers, while the $x$ and $y$-axes are in-plane, with $x$ along a nearest neighbor direction. Due to spin–orbit induced $L_x$ and $L_y$ couplings with the $d_{yz}$ and $d_{xz}$ states, respectively, the big portion of $d_{z^2}$ states at the Fermi level ($E_F$) clearly explains the in-plane anisotropy for $n = 0$. For $n = 1$ the $d_{z^2}$ peak at $E_F$ considerably reduces, while a $d_{yz}$ peak at $E_F$ arises, whereas the $d_{xz}$ component of the DOS remains practically unchanged. This is hardly surprising in terms of hybridization because the lattice positions closest to a Co atom in the neighboring Au layers are nearly aligned along the $yz$-direction. Subsequently, a corresponding peak at $E_F$ was observed in the $p_z$-like DOS of the cap Au. So a strong tendency to perpendicular anisotropy is expected due to $L_z$ coupling between $d_{yz}$ and $d_{xz}$ states as indeed obtained in our calculations.

As was also stressed in a similar study of the Co/Cu(111) system [17], since the bands of the cap layer at the surface are higher in energy than the corresponding bands in the topmost substrate layer, a more stronger hybridization on the capped side is expected with the minority Co bands. Since for $n = 2$ the Au(1) layer becomes further from the surface and therefore the energetical separation of the Au p bands and the Co minority d bands increases, the hybridization between them obviously reduces, pushing thus the $d_{yz}$ states of Co somewhat away

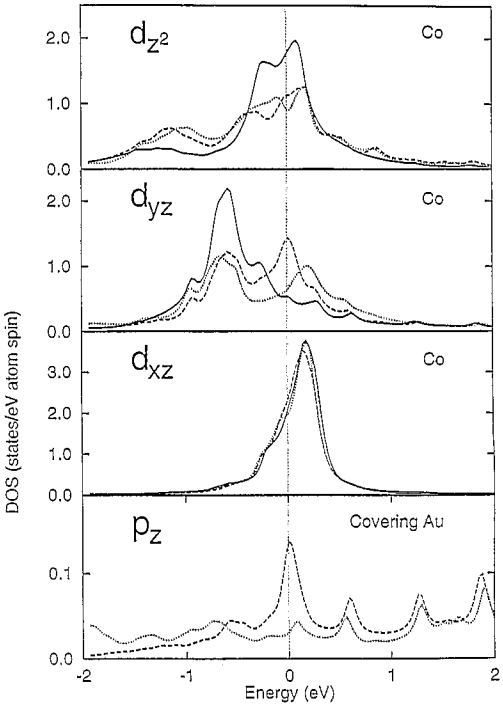


Fig. 3. Orbital resolved $d_{z^2}$, $d_{yz}$ and $d_{xz}$-like minority spin densities of states of Co and $p_z$-like states of Au(1) in $Au_n$/Co/Au(111) multilayers. Solid line: $n = 0$, dashed line: $n = 1$, dotted line: $n = 2$. The Fermi level is chosen to be the origin of the energy scale.

from $E_F$ (see Fig. 3b). Following from the above considerations this results into a decrease of the MAE. Clearly, subsequent deposition of Au layers hardly influences the Au(1) layer any more. In Fig. Fig. 4 we plotted the difference between the calculated Madelung potentials of Au(1) and Co, $\Delta V_{Mad}$. The obvious correlation between $\Delta E$ in Fig. 1 and $\Delta V_{Mad}$ against the thickness of the Au

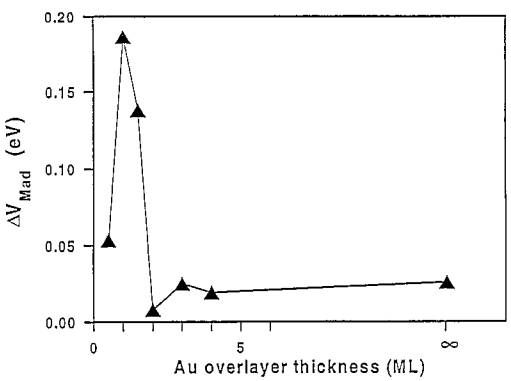


Fig. 4. Differences between the calculated Madelung potentials of the Au(1) and the Co layers in $Au_n$/Co/Au(111) multilayers. The solid line serves as a guide to the eye.

cap also supports the essential role of the relative energetical position of the Co and Au(1) bands in determining the MAE.

In summary, we showed that the anomalous magnetocrystalline anisotropy observed in the $Au_n/Co/Au(111)$ system can be reproduced by electronic structure calculations. As the function of coverage, the orbital moment anisotropy is found to correlate with the MAE in agreement to the prediction of simple perturbation theory. The anomalous behavior of the MAE can be satisfactorily understood in terms of overlayer-induced changes in the electronic structure, governed by the relative energetical positions of the Au and Co bands.

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