Cap layer influence on the spin reorientation transition in Au/Co/Au

J. Langer,[1] J. Hunter Dunn,[2] A. Hahlin,[3] O. Karis,[3] R. Sellmann,[1] D. Arvanitis,[3] and H. Maletta[1,*]
[1]*Hahn-Meitner-Institut Berlin, Glienicker Str. 100, D-14109 Berlin, Germany*
[2]*Stanford Synchrotron Radiation Laboratory, Stanford, California 94309*
[3]*University of Uppsala, Box 530, S-75121 Uppsala, Sweden*
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X-ray magnetic circular dichroism vector magnetometry provides evidence that the spin reorientation transition in epitaxially grown Au/Co/Au films depends on the thickness of the Au capping layer. Whereas a 1.9-nm-thick Co film with a 2-nm Au cap exhibits the expected pure in-plane orientation of the magnetization in the remanent state at room temperature, the same film with thinner Au cap shows a significant out-of-plane component. These findings are linked with the observation of a cap-induced enhancement of the orbital moment of about $0.05\mu_B$ per Co atom.

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An important topic in thin-film magnetism research is the influence of the reduced dimensionality on the magnetic properties. This is of interest both from a fundamental as well as from a technological point of view. With decreasing layer thickness interesting magnetic effects are often observed. One prominent example is the spin reorientation transition (SRT) in ultrathin Co films of a few monolayers, sandwiched between transition metal films like Au.[1–4] Here the easy axis of Co magnetization changes from in plane to out of plane when the thickness of the Co layer is decreased at a fixed temperature or when the temperature is decreased at a fixed thickness.[5,6] This can be attributed to the different dependence in the magnetic anisotropy constants for Co atoms in the bulk and at the interface. From a microscopic point of view it is suggested that the symmetry breaking of the local environment of magnetic atoms at interfaces should be responsible for the observed effects. In particular an anisotropy in the magnetic orbital moments is expected.[7]

With x-ray magnetic circular dichroism (XMCD) spectroscopy it is possible to separate and determine the spin and orbital magnetic moments in $3d$ transition metals by applying the magneto-optical sum rules.[8] By means of XMCD measurements at the Co $L_{2,3}$ edges of Au/Co/Au layers, it was shown in Refs. 9–11 that an asymmetry of the in-plane and out-of-plane orbital moments develops with decreasing Co thickness. In Ref. 9, the Au cap thickness was held constant around 1.8 nm and the Co thickness varied. However, it is also known from *in situ* polar magneto-optical Kerr effect (MOKE),[12–14] *in situ* torsion magnetometry,[15] and relativistic spin-polarized local spin density calculations[16] that the thickness of the cap also influences the magnetic surface anisotropy in Au/Co/Au sandwiches. Here we report on the influence of the thickness of the Au cap on the Co magnetization in epitaxial Au/Co/Au sandwiches and present, in particular, results obtained by XMCD magnetometry.

The thickness of the Co layer was chosen so that the temperature-driven reorientation of the magnetization from in plane to out of plane occurs as the temperature is lowered below room temperature.[5,6] A staircase sample with four different thicknesses of the capping layer was prepared by *e*-beam evaporation in an ultrahigh-vacuum chamber (base pressure $<3\times10^{-10}$ mbar) at the Hahn-Meitner Institute. Using a $(11\bar{2}0)$ sapphire single-crystal substrate (16 mm $\times$ 39 mm), a 15-nm W(110) bcc layer was first evaporated, followed by a 5.0-nm Au(111) fcc layer. The subsequently evaporated Co layer was 1.9 nm thick, with a (0001) hcp structure. On top, a staircase of 1.0 nm, 1.3 nm, 1.7 nm, and 2.0 nm Au was evaporated in terraces with a width of 9.75 mm. All deposition rates were controlled during growth using a calibrated quartz microbalance. The chemical composition, cleanliness, and occurrence of epitaxial growth were controlled, *in situ*, by means of Auger electron spectroscopy (AES) and low-electron energy diffraction (LEED), respectively. For more details, see Refs. 5 and 6.

The x-ray absorption and XMCD spectroscopy measurements were performed at the undulator beamline 5.2 at the Stanford Synchrotron Radiation Laboratory (SSRL), using a specially tailored UHV chamber.[17] This setup is optimally suited for vector magnetometry since both the sample and magnetizing coils rotate independently. The helicity of the x rays was fixed during the experiments. All spectra were recorded in the remanent state. The XMCD signal was obtained by switching the magnetization direction either parallel or antiparallel to the photon spin. This was done in the in-plane or out-of-plane direction of the sample using pulses of 170 Oe. This applied field is an order of magnitude larger than the in-plane coercive field of the sample. As in earlier studies, the in-plane remanent magnetization lies along the substrate W [1–10] directions.[5] The spectra presented here were recorded using the photocurrent from the sample. Simultaneously, total electron yield measurements were done, using a channel plate detector. This latter method was found to produce identical results. The undulator supplies nearly 100% circularly polarized light. The beamline optics delivers a spot size of about 1 mm on the sample, well below the size of the terraces defined by the regions of different cap thicknesses. For the evaluation of the spectra a well-established procedure described elsewhere was used, yielding results on a per atom basis.[18] The spectra have been corrected for saturation effects in the photocurrent, which become pronounced at small incidence angles according to the procedure described in Ref. 19. An effective electron escape depth of 1.7 nm was assumed as determined from these investigations.[19]

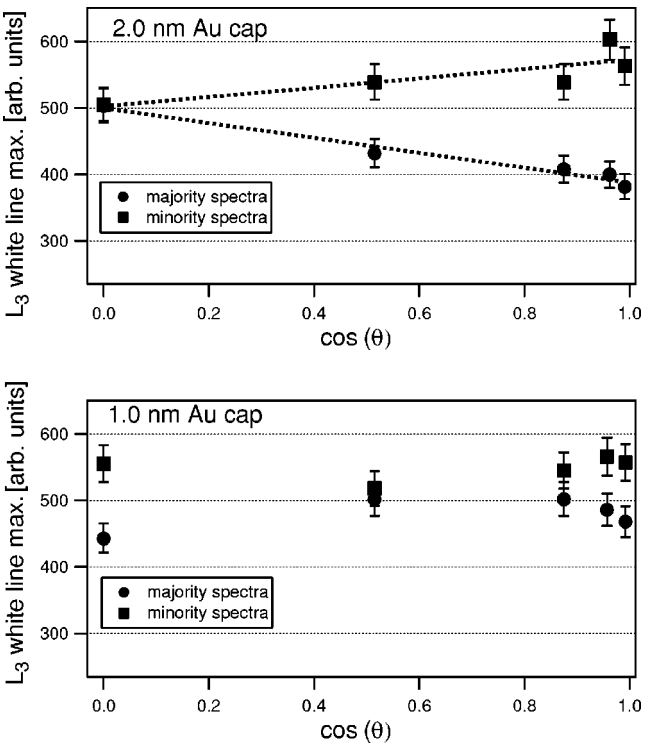

FIG. 1. Angular dependence of the Co L_3 peak heights for two different cap layer thicknesses on a 1.9-nm Co film grown on Au. Values are plotted vs the cosine of the x-ray angle of incidence Θ. The data are corrected for saturation effects in the secondary channel at small x-ray incidence angles.

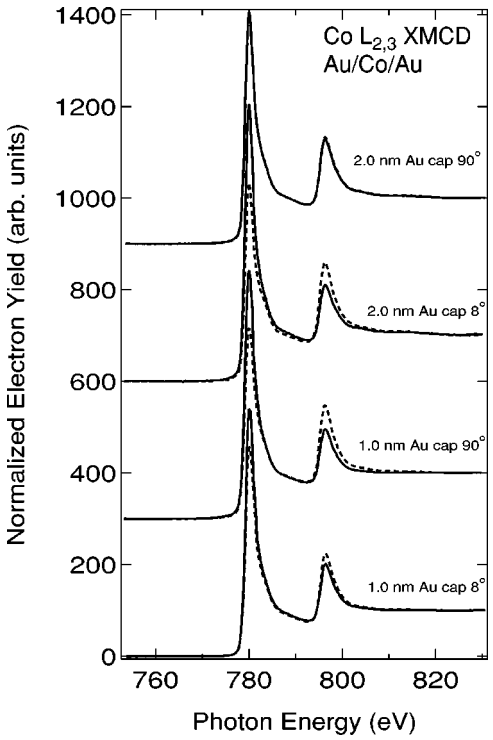

FIG. 2. Measured x-ray absorption spectra for the Co layer capped with 1.0 nm and 2.0 nm Au at normal and grazing x-ray incidence. The spectra have been corrected for saturation effects.

The Co signal-to-background or high-energy continuum edge jump ratio was found to be 0.19 in the case of the 2-nm cap and 0.38 in the case of the 1-nm Au cap. These values are sufficiently large to allow for an analysis of the dichroic signal as presented earlier.[18–20]

Magnetometry was performed at room temperature for all four sample regions by taking XMCD spectra at five different x-ray incidence angles Θ. Here Θ denotes the angle between incident beam and sample surface. The magnetic field was in all cases applied in plane except for the 90° measurements, where it was applied parallel to the sample normal, i.e., out of plane. The XMCD intensity is proportional to $\boldsymbol{k}\cdot\boldsymbol{M}$ where $\boldsymbol{k}$ is the propagation vector of the light and $\boldsymbol{M}$ the direction of the magnetization in the sample. We take the XMCD difference at the L_3 peak maximum as a measure of the projection of the Co layer magnetization along the $\boldsymbol{k}$ vector. For example, at normal x-ray incidence, one can evaluate whether any remanent component is pointing out of plane. At grazing incidence one measures the in-plane components of the remanence. For a magnetic monodomain, a cosine angular dependence is expected, as indicated by the scalar product given above.

We examined the magnitude of the XMCD difference at the L_3 peak maximum as a function of the incident x-ray angle. In Fig. 1 we show the recorded intensity at the majority and minority L_3 peaks, respectively, as a function of the cosine of the x-ray incidence angle Θ. Data for both 1-nm and 2-nm Au caps are presented. The corresponding spectra, normalized to yield a signal on a per-atom basis, are shown in Fig. 2.

The data of both Figs. 1 and 2 indicate a qualitatively different magnetic behavior of the Co layer depending on the thickness of the Au cap. In the region with the 2-nm Au cap there is no visible XMCD signal at the L_3 peaks at normal x-ray incidence and maximum XMCD difference at grazing incidence. For angles in between, the intensities varies linearly (Fig. 1). This dependence is expected in case of a complete in-plane magnetization,[19] having no out-of-plane component. On the other hand, in the region capped with 1 nm Au a XMCD difference was detected at grazing (8°) as well as at normal incidence (90°) (Figs. 1 and 2). However, the difference at grazing and normal incidence was not as big as for the 2-nm Au cap at grazing incidence (Fig. 2), indicating a reduced remanent magnetic contribution at each of these directions. Interestingly a nearly vanishing splitting at intermediate angles is found in the case of the remanent magnetization for the 1-nm Au cap when the field is applied in plane. A possible interpretation can be based on the splitup into magnetic domains. A model based on a state of coexisting phases has also been proposed recently for the system Co/Au(111).[21] A detailed discussion of these models in connection to a detailed analysis of the XMCD angular dependence goes beyond the scope of the present communication. The important finding is that in the thin Au cap case, both in-plane and out-of-plane magnetization components do exist in the remanent state. An intermediate situation is observed for the regions with 1.3-nm and 1.7-nm Au caps. For both these regions a nonvanishing magnetization component was observed also at normal x-ray incidence, indicating that in

the remanent state the resulting magnetization vector has an out-of-plane component.

By cooling the sample close to liquid nitrogen temperature and probing the XMCD response in an identical manner to what has been discussed above for room temperature, it was checked that all sample regions showed a out-of-plane magnetization with no significant in-plane XMCD signal. The XMCD difference at the L_3 lines for the out-of-plane magnetization was within experimental error of the same size for all regions. Thus, all regions showed the expected temperature-driven SRT. Indeed, a smooth and reversible temperature-driven SRT is found for the region with the 2-nm Au cap, as we reported elsewhere.[20] These last observations confirm the intrinsic character of our findings and the high quality of the samples. During the measurements we also checked, using x-ray absorption (oxygen and carbon *K* edges), that no sizable amounts of oxygen and carbon impurities could be detected in the sample. In addition, we checked that the Co signal-to-background ratio at the high-energy continuum (Co edge jump ratio) decreased in an exponential manner as a function of the Au cap thickness, with an effective electron escape depth of about 1.7 nm. As mentioned above, this value is in agreement with the saturation correction procedure.[19] All these findings are compatible with epitaxial, high-quality, smooth layers of Au and Co of high purity. They indicate that the observed cap dependence of the magnetic properties is of an intrinsic character for the SRT in the Au/Co/Au system.

In a second step in the analysis of the XMCD spectra, the spin moments M_S and orbital moments M_L of the different regions were quantified by using the magneto-optical sum rules[8] as presented earlier in the case of thin Co films.[18,19] The number of occupied *d* states was set to 7.5. Since we probe the remanent state, we also correct the data for the fact that the magnetization is reduced compared to the magnetically saturated state, as determined from the element specific hysteresis loops. For the latter experiment experiment the beam energy was fixed at the energy of the Co L_3 peak and the intensity of the reflected specular beam (angle of incident light equals angle of reflected light) was recorded with a diode while changing the magnitude of the in-plane applied magnetic field. Given the fact that we do not systematically measure the XMCD response in both the easy and hard magnetic directions, we use data from the literature for the dipolar correction in the sum rules.[9,11] This approach is justified as the above-mentioned data where obtained from measurements performed for the same system (Au/Co/Au) for a thickness of the Au cap corresponding to our 2.0-nm cap. The transferability of the dipolar correction term for the case of the thick Au cap is justified given (i) the magnetic response reported in the literature for similar systems and (ii) the corresponding orientation of the magnetization and its temperature dependence.[9,11] An analysis of the present set of data in terms of spin and orbital moments on a per-atom basis allows us to gain further insight into the orbital moment anisotropy related with the local environment of the Co atoms. The orbital moment anisotropy as is now discussed appears directly linked with the occurrence of the SRT observed here. As we observe experimentally, a change in the

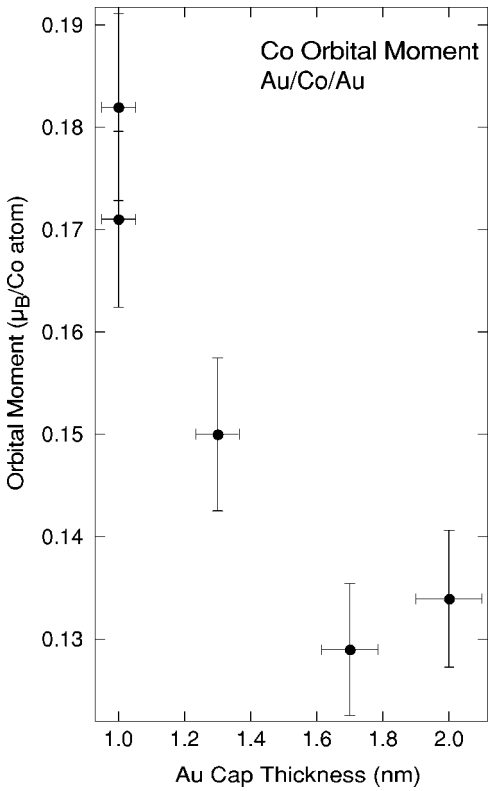

FIG. 3. The orbital moment per Co atom is given as derived by applying the XMCD sum rules. The spin moment was found to be constant, 1.7(2) Bohr magnetons (μ_B) per Co atom, for all sample regions. The given error bars are relative. They include uncertainties in the relative area determination, but do not include limitations due to the fact that the sum rules are based on a simplified atomic model. An absolute error in the order of 20% is appropriate.

Au cap thickness can also induce a reorientation of the Co magnetization. Any deviation from the expected values of the spin and orbital moments, in conjunction to theory, indicates how the electronic state of the Co layer is modified. The application of the sum rules yields a constant spin moment of $1.7(2)\mu_B$ per Co atom, independent of the Au layer thickness. The total error in the spin moment determination is $0.2\mu_B$ per Co atom, while the relative error is only in the order of 5%. For the orbital moment an increase of 20% is observed with decreasing Au cap thickness, as shown in Fig. 3. To our knowledge this is the first time such an observation has been made. Previous results on cap-induced effects for epitaxial films have been reported, but were limited to the overall magnetization and the Curie temperature.[22] Our present observations clearly link the out-of-plane magnetization to the variation in the orbital moment anisotropy, as one would expect based on general arguments.[7] The data of Fig. 3 show that in the case of the 1.0-nm Au cap, two distinct points are obtained corresponding to the in-plane (lower value) and out-of-plane (higher value) responses, respectively. These values are, however, still close to the experimental error. For the intermediate cap thicknesses both an in- and out-of-plane magnetic response was observed in the XMCD data.

The variation of the orbital moment that occurs between 1

nm and 2 nm Au cap thickness is in the same range as the variation reported earlier at constant Au cap thickness but with variable Co layer thickness.[9,11] A variation from about $0.08\mu_B$ to $0.14\mu_B$ per Co atom was reported as the Co thickness is varied for the in-plane orientation (hard axis) for a Au/Co/Au staircase sample between 4 and 11 Co atom layers.[9,11] The corresponding out-of-plane values (easy axis) are 0.21–0.14.[9,11] Here we obtain values from 0.130(7) to 0.182(10) by varying the Au cap thickness. This yields a variation of the orbital moment of about $0.05\mu_B$ per Co atom. All values of the orbital moment reported here are in the same numerical range as the ones reported earlier.[9,11] Following the treatment by Bruno,[7] we obtain a spin-orbit magnetic anisotropy energy of 1.25×10^{-4} eV per Co atom using the same corrective constants as presented for Au/Co/Au earlier.[9,11] This energy is still larger than the spin-spin dipole interaction of about 9×10^{-5} eV per Co atom. The data of Fig. 3 therefore explain, within this simple qualitative model, the observation of out-of-plane components as the Au cap thickness decreases.

A dependence of the orbital moment, in ultrathin Co layers, on the variation of the thickness of the cap layer in the order of a few monolayers has not been reported so far. On the other hand, it is well known that the anisotropy energy—and hence the transition from in-plane to out-of-plane magnetization—can be influenced by evaporating a cap in the submonolayer range. As discussed previously, a nonuniform cap of the Co with Au can be excluded as an explanation to bring both findings in one picture. Uncapped Co exhibits an in-plane orientation of the magnetization, which turns out of plane upon capping. The region with the thick Au cap should therefore show an out-of-plane orientation rather than the layer with the thin cap. Furthermore, the intensity ratio between Au and Co signals in the AES spectra showed an exponential dependence on the cap layer thickness. The same exponential attenuation was also found in the XMCD edge jump ratios. This indicates a uniform growth of the Au cap.

We expect that structural and electronic structure modifications may occur upon capping. Further crystallographic experiments are necessary to link the results in Fig. 3 to electronic structure effects originating in the modified symmetry of the Co layer atoms upon capping. The precise study of the functional dependence of the data in Fig. 3 allows us to discriminate interface from volume effects for the Co film as noticed earlier.[9,11] Here, a nonlinear dilution law would best describe our results, supporting a model where cap-induced changes occur primarily close to the Co/Au interface. A precise analysis goes beyond the scope of the present communication.

In conclusion, XMCD vector magnetometry provided evidence that the spin reorientation transition in epitaxially grown Au/Co/Au thin film layers depends on the thickness of the Au cap. Whereas a 1.9-nm-thick Co film with 2-nm Au cap exhibited the expected pure in-plane orientation of magnetization at room temperature, Co films of the same thickness but with thinner Au caps showed a significant out-of-plane component in the remanent state. These findings were linked to the observation of a cap-induced variation in the orbital moment anisotropy of Co. We observed an enhancement of the Co orbital moment from $0.13\mu_B$ per atom for a 2-nm Au cap to $0.18\mu_B$ for a 1-nm Au cap.

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*Corresponding author. Electronic address: maletta@hmi.de

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