

Structure and magnetism of self-organized Co islands

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Received: 2 September 1998 / Received in final form: 15 October 1998

Abstract. This investigation turns its attention to magnetic effects of cobalt islands on a non-magnetic substrate. The preparation technique of MBE enables the creation of flat and well-ordered surfaces of hcp-Co(0001) if the evaporation onto a W(110) crystal is carried out at ambient temperatures. Subsequent heating to about 1000 K causes a phase transition from thin films to islands. The investigations are carried out in VUV photoemission with polarized synchrotron radiation, LEED and Scanning Tunneling Microscopy (STM). We employ the technique of magnetic linear dichroism in the angular distribution of photoemission (MLDAD) from valence bands. The MLDAD effect shows significant differences between a flat cobalt layer and cobalt islands. Variations due to the excitation energies are discussed with respect to a fully-relativistic bulk-band-structure calculation.

PACS. 61.46.+w Clusters, nanoparticles, and nanocrystalline materials – 75.25.+z Spin arrangements in magnetically ordered materials – 75.30.Pd Surface magnetism

1 Introduction

Transition metal films on single crystal surfaces have been the subject of an intensive research using a couple of techniques [1]. Nowadays, magnetic nanoparticles open a completely new field of research for both fundamental research as well as technical applications such as magneto-electronics. In this contribution we will concentrate on the structure and magnetic phenomena of cobalt islands on W(110). In our experiments, the islands are created by heating a flat and well-ordered Co film to above 1000 K. *Magnetic Linear Dichroism in photoemission* (MLDAD) serves to study the electronic and magnetic properties with linearly polarized VUV synchrotron radiation. Both magnetic circular and linear dichroism in photoemission have already been established as a powerful technique to study the magnetic properties of ferromagnetic solids and thin films [2–4]. MCDAD and MLDAD in core level photoemission can be theoretically described in the atomic model picture [5–7], where the splitting of the $2p$ level into sublevels with orbital momentum m_l is caused by the electrostatic interaction of the core level with the magnetically polarized valence electrons. In contrast, these techniques are more difficult to be explained when using valence band photoemission, since one has to solve the Dirac equation in order to calculate the band structure or photoemission spectra. First results are now available [8, 9].

Thin cobalt films on a clean tungsten(110) surface have been studied in detail using Low Energy Electron Diffraction (LEED) and Auger Electron Spectroscopy [10,

11], angle resolved ultraviolet photoelectron spectroscopy (UPS) [8, 12, 13], ac- and dc-Magneto-Optical Kerr-effect (MOKE) [14], epitaxial strain and magnetic anisotropy is described by Fritzsche et al. [15]. The first layers have different lattice constants due to strain relaxation, details are shown later. Increasing the substrate temperature during the cobalt evaporation on W(110) to about 790 K leads to a *Stranski-Krastanov* mode with islands of a typical size of 500 nm and a height of about 10 nm [10, 16]. On top of an interfacial monolayer, the adsorbed cobalt atoms form 3D islands.

Cobalt islands with a typical size of some hundred nanometer can also be created on a W(110) single crystal, when annealing an epitaxially grown hcp(0001) cobalt film of less than 10 monolayers to about 1000 K. Here, we present LEED and first STM results on the structure of Co islands on W(110). The MLD ultraviolet photoelectron spectroscopy data are compared to similar measurements on thin cobalt films and to a fully-relativistic bulk-band-structure calculation [8].

2 Experimental details

Magnetic linear dichroism in angle resolved valence band photoemission (MLDAD) has been used in order to study the magnetic properties of thin cobalt films and islands. In our case the investigations have been carried out at the storage ring BESSY in Berlin using linearly polarized VUV-synchrotron radiation. A detailed description of the 6.5 mNIM beamline is given in [17].

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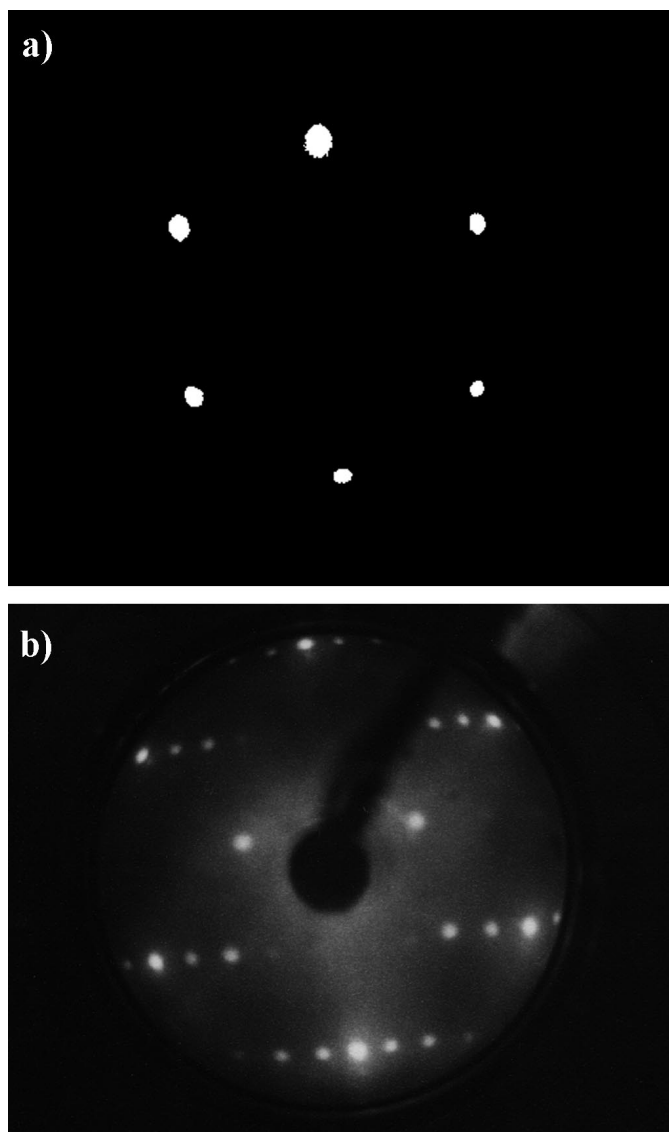


Fig. 1. LEED patterns of thin epitaxial cobalt film (upper part) and 3D cobalt islands (lower part) on clean W(110).

The MLDAD experiment is performed in a ultrahigh vacuum chamber with a base pressure of 2×10^{-10} mbar. It is equipped with a combined LEED-Auger system for surface characterization and a rotatable hemispherical photoelectron analyzer with a resolution of about 200 meV. The setup of the experiment (see [8]) exhibits a “chirality” being essential for dichroic effects. For observing magnetic linear dichroic effects, the magnetization vector \mathbf{M} has to point perpendicular with respect to the plane of incidence. The angle of incidence is about 45° with respect to the surface normal.

A W(110) crystal serves as substrate for the growth of epitaxial cobalt films. The crystal is cleaned by heating in oxygen and flashing to 2600 K. Cobalt films are evaporated by an electron beam evaporator on W(110) at room temperature. An integrated ionization gauge-like flux monitor facilitates a reproducible growth rate; it is typically set to one layer per minute. In contrast to bulk hcp Co with its

easy axis being perpendicular to the basal plane, the surface anisotropy causes the easy magnetization axis to lie *in-plane* [18]. In our experiment the films are magnetized by a current pulse through a coil close to the sample along the $[1\bar{1}0]$ direction of the tungsten substrate.

3 Results and discussion

Cobalt overlayers on W(110) grow layer-by-layer and form an hcp(0001) structure above two monolayers. A careful annealing of these films to about 400 K leads to LEED patterns corresponding to flat and well-defined cobalt(0001) surfaces, as can be seen in upper part of Fig. 1. Cobalt films on tungsten show the Nishiyama–Wassermann orientation, i.e., the hcp Co $[11\bar{2}0]$ direction is parallel to the W[001] axis. Up to about 0.7 ML, a pseudomorphic growth is observed. In this region the strain is large along the W[001] direction (20.8%) and small (3%) parallel to the W $[1\bar{1}0]$ axis. With increasing coverage the lattice begins to compress. Now the LEED-patterns show a (4×1) structure which is oriented along the [001] direction of the substrate. Every fifth cobalt atom corresponds to every fourth W-atom. The values for the misfit have changed to 1.5% in the W[001] direction and to 3% in the $[1\bar{1}0]$ -direction for the compressed film [15].

In our experiments cobalt islands are generated by heating thin epitaxial cobalt films to about 1000 K. The phase transition from the epitaxial films to the 3D island structure can easily be seen in LEED investigations where the hcp-structure changes to a satellite pattern around the tungsten spots, cf. lower part of Fig. 1. In contrast to the well ordered and flat bcc(110) iron islands [19], cobalt islands show a preferential ordering along the W $[1\bar{1}0]$ axis. The transition from a thin cobalt film (atomically flat terraces with a width of some hundred nm) to three dimensional island growth has been studied in-situ with scanning tunneling microscopy, see Fig. 2. The growth of the Co islands strongly depends on carbon contamination of the W(110) surface, which can easily be created by segregation of carbon from the tungsten bulk when cleaning the crystal by flashing.

Typically, a carbon contamination on W(110) forms a $c(15 \times 3)$ superstructure [20] which can be observed in LEED studies especially at low kinetic energies (down to $E_{\text{kin}} = 20$ eV). The carbon atoms arrange in two different domains on the surface, cf. Fig. 2c–d. Thermal evaporation of cobalt onto this carbon-induced superstructure leads to a generation of fcc(100) cobalt films [18] oriented in two domains (Fig. 2e). When annealing these films to temperatures above 700 K only islands with a height of 1–2 ML are created which are usually oriented along the domains of the carbon structure, see Fig. 2f. Similar results have been found by other groups using Scanning Auger Microscopy under ultrahigh vacuum condition [19]. Up to now no element-specific analysis in between two islands has been carried out.

Figure 3 displays photoelectron spectra taken from a 7 ML thick Co film and an cobalt island system (3.5 ML

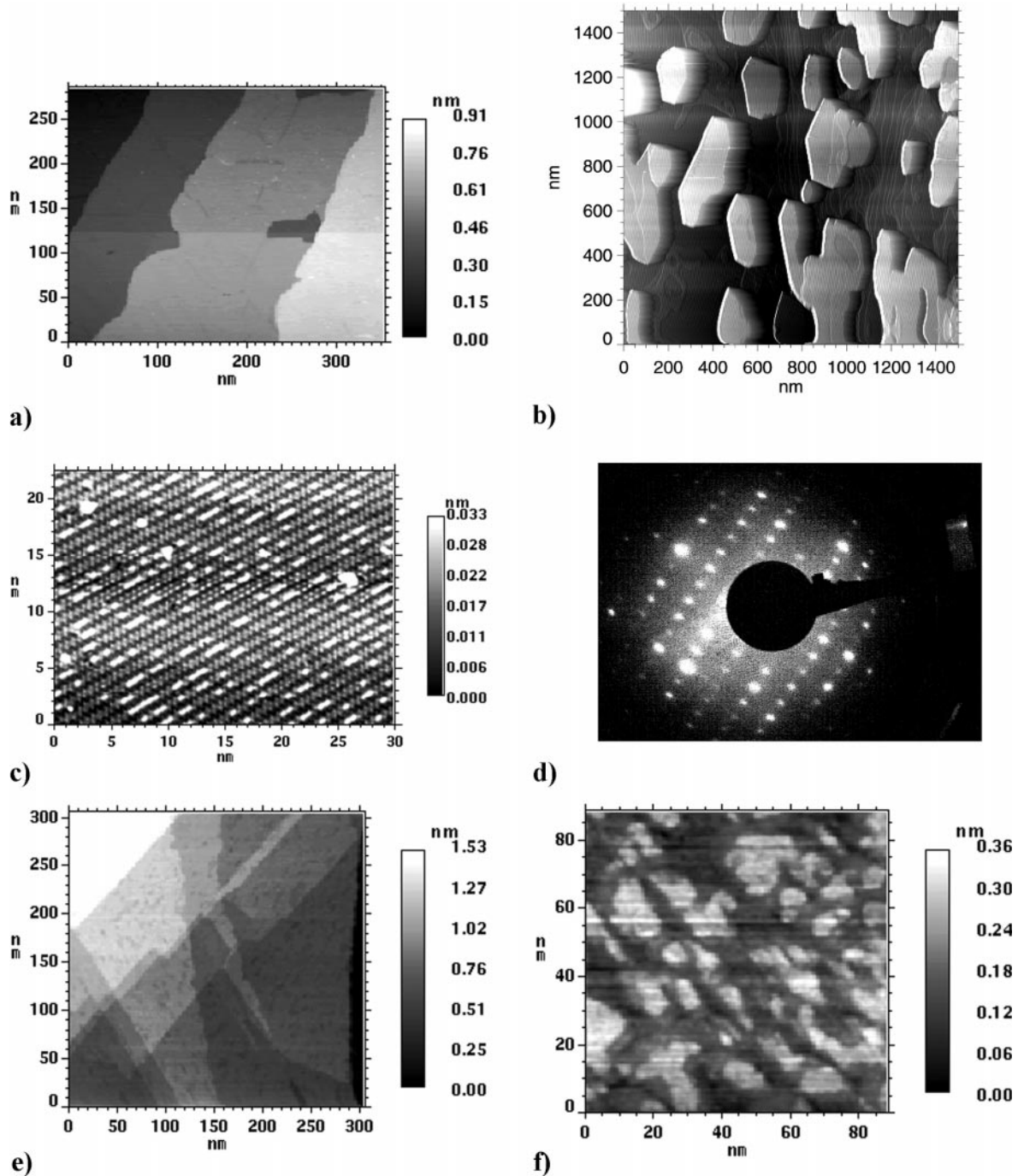


Fig. 2. STM images showing an atomically flat cobalt layer (a) after careful annealing to about 500 K and three dimensional Co islands (b) after annealing to 1000 K. In case of a carbon contamination of the W(110) sample (c) easily to be observed in LEED (d), already thin cobalt films exhibit two domains with different orientations (e). Annealing these films (e) leads to a completely different growth of the Co islands (f).

equivalent coverage). The data have been recorded in remanence after pulse magnetization in opposite directions (M^+ and M^-) using linearly polarized light at $h\nu=16$ eV. The lower panel shows the corresponding asymmetry, i.e., the difference between the two photoemission spectra divided by the sum. Typical values for the asymmetry are less than 10%, and, of course, the asymmetry varies with the photon energy [8]. Both photoemission data clearly

show an MLDAD effect. In the right panel, the intensity difference in the peak close to the Fermi level E_F is much more pronounced. For thin Co films, the MLDAD effect nearly vanishes in this structure, but an asymmetry is visible in the peak located at about 1.2 eV binding energy. The MLDAD effect is caused by a hybridization of bands with different symmetry in the initial state and a non vanishing exchange splitting in the final state to-

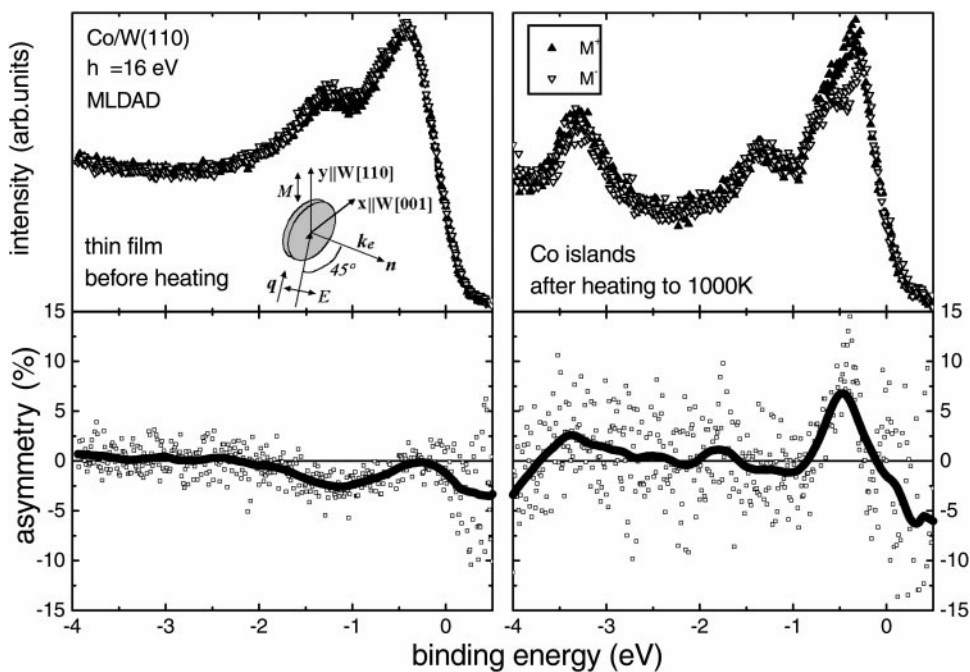


Fig. 3. Photoelectron spectra of Co 3d valence band for a well-ordered hcp(0001)-surface (left) and Co islands (right part) on W(110) taken for opposite in-plane magnetization directions (full triangles: M^+ , open triangles: M^-). In the lower part, the corresponding asymmetry is displayed with the solid curve giving an average. The inset displays the experimental setup with the induced chirality. The magnetization M has been carried out perpendicular to the orbit plane of the storage ring.

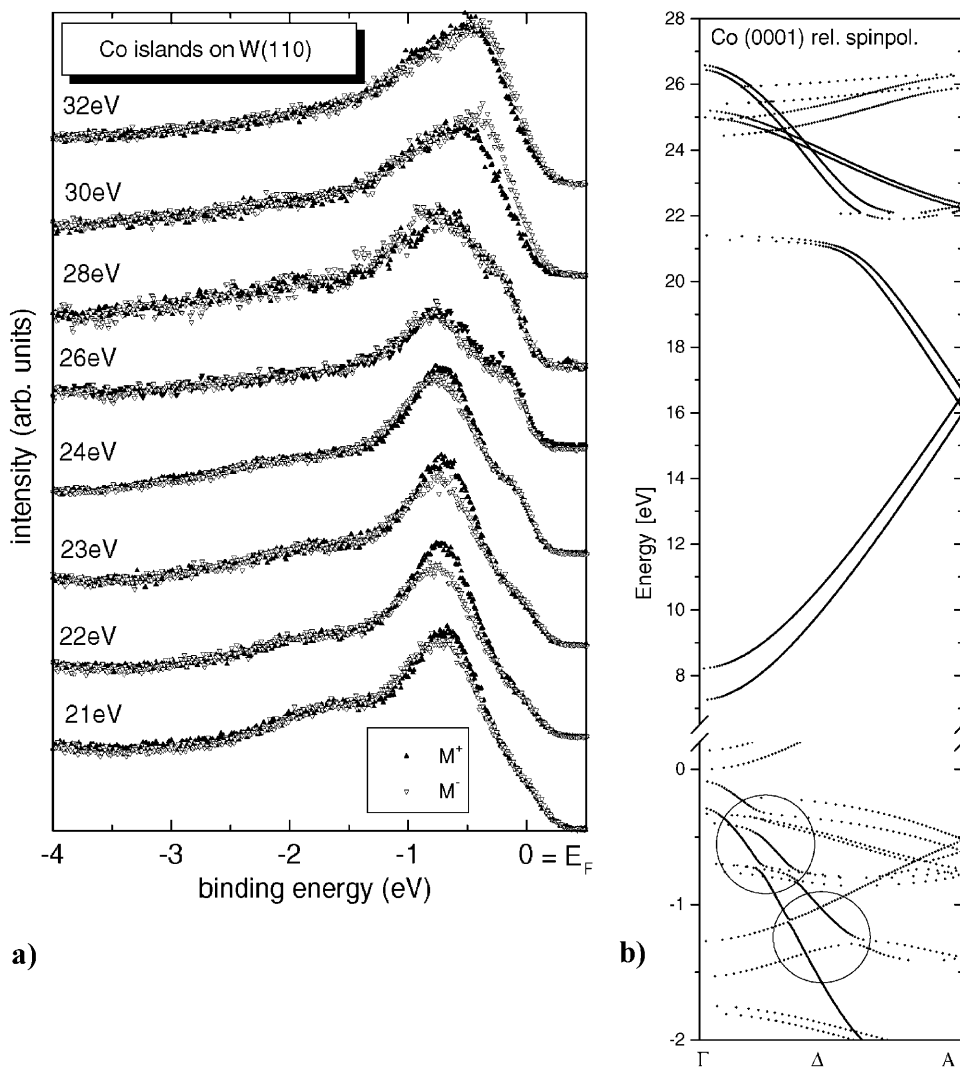


Fig. 4. Left part: Energy dependence of the photoelectron spectra carried out analogously to Fig. 3. The islands were created by heating an epitaxial cobalt film to about 1000 K. Right part: For comparison, a fully-relativistic band structure for hcp(0001) bulk cobalt with in-plane magnetization is displayed. This calculation takes into account both spin-orbit interaction and exchange splitting on the same level of accuracy. The circles in the initial states show two regions where hybridization effects are present.

gether with the chiral experimental setup. Details for thin cobalt films are described in [8] with respect to a fully-relativistic band structure calculation. The photoemission peak in the cobalt island spectra located around a binding energy of 3.5 eV originates from a tungsten resonance and is only visible between 15–17 eV photon energy. For a 7 ML thick Co film, the intensity is completely attenuated due to the high surface sensitivity of ultraviolet photoelectron spectroscopy. The photoemission peak at -3.5 eV binding energy (right part) shows a small asymmetry even though the tungsten bulk is indeed not magnetically ordered. Similar effects have also been observed in spin resolved photoemission (e.g. Co/W(110) [8]) and in MCDAD (thin iron films on Cu(100) [21]). This phenomenon can be explained by the spin-dependent attenuation of electrons (here from the W substrate) passing through a magnetic layer creating a very effective spin filter. The inset in Fig. 3 shows the arrangement of incoming photon beam \mathbf{q} (with the linear polarization vector \mathbf{E} being in the orbit plane), the orientation of the tungsten crystal (surface normal \mathbf{n}) and the magnetization vector \mathbf{M} . The photoelectrons are detected in normal emission for thin films or close to normal emission (in the orbit plane less than 10° with respect to \mathbf{n}) in the case of Co islands.

The energy variation of the photoelectron spectra is displayed in Fig. 4 (left part) in the range between 21 eV and 32 eV, the data have been recorded analogously to Fig. 3. In the right part of Fig. 4, a band structure of bulk hcp(0001) cobalt is shown, which has been fully relativistically calculated for an in-plane magnetization (cf. [8]). Since the MLDAD is caused by spin-orbit as well as exchange-splitting, both effects have been taken into account on the same level of accuracy. The electron distribution curves show an energetic dispersion in the prominent feature near E_F . From 21 eV to about 28 eV, the binding energy is nearly constant (0.75 eV). Above 28 eV, the dispersion starts from 0.75 eV to 0.4 eV. Additionally, a new feature in the spectra is showing up directly at the Fermi level (from 24 eV upwards) and becomes dominant above 30 eV. This peak can be attributed to a direct transition from the initial states near the Γ -point (just below E_F) into the final state band near 25 eV, and disperses to higher binding energies (0.4 eV). Furthermore, the cobalt band structure has two gaps in the final states as displayed here, the first around 22 eV and the second between 27 eV and 29 eV [22]. A possible influence from the underlying tungsten substrate on our experimental data can be excluded, since photoemission spectra of clean tungsten (not shown in this contribution) do not exhibit any significant structure in the interesting region between the Fermi level and a binding energy of $E_{\text{bin}} = 1.5$ eV. Compared to data from thin epitaxial films of about 7 monolayers, the photoelectron intensity in the peak structure the Fermi level (cf. Fig. 4, $h\nu=24-28$ eV, $E_{\text{bin}} \approx 0.25$ eV) is suppressed and below 24 eV the main peak (located at around 0.7 eV) appears at higher binding energies. In principle, photoemission spectra from hcp(0001) Co films with a thickness above 4 ML can be regarded as bulk-like cobalt data [13]. Differences in the photoelectron intensity are mostly discussed with respect to lower coverages, whereas shift in the binding

energy usually arise due to the development from a two-dimensional to a three-dimensional band structure. Here the energetic shift observed between the Co island data and photoemission data from a 7 ML Co film can, of course, originate due to the confinement of the nanostructured Co islands.

The MLDAD effect shows a maximum (i.e., highest intensity difference) at 22 eV which may be connected to the band gap in this region. Since this gap is quite small and the exchange splitting in the final state above the gap is clearly present, the intensity differences can occur from a preferential excitation for one magnetization direction into these empty states. Furthermore, the peak maxima (for opposite magnetization direction M^+ and M^-) clearly have different energetic positions which are caused by hybridization effects where the energetic bands are not allowed to cross each other (avoided crossover). In the energetic region from 26 eV to 28 eV no final states of the free electron parabola are available and hence, no significant MLDAD can be observed. A possible reason could be the absence of an exchange splitting in the final state. A more detailed discussion of the photoemission curves could give further information of the electronic band structure of cobalt in reduced symmetry.

Nevertheless, the MLDAD photoemission data shown here, clearly demonstrate that 3D cobalt islands with a coverage equivalent to 3.5 ML are ferromagnetically ordered with a remanent magnetization along the W[1 $\bar{1}$ 0] direction. The magnetic effect for cobalt islands cannot simply result from a possibly existing cobalt interface layer. Spin resolved photoemission data of Co/W(110) [12] as well as MOKE measurements [14] prove that at room temperature one monolayer of cobalt has no remanent magnetization, the Curie temperature lies around 260 K. The critical thickness for the onset of the ferromagnetism is about 1.6 layers at room temperature, for thinner films, the Curie temperature T_C is reduced. For one cobalt monolayer, T_C lies in the region of 260 K.

Furthermore, Garreau *et al.* [14] observed that the Curie temperature of thin cobalt films below 3 monolayers decreases when annealing the films above 440 K, even though the stress in the adsorbed layer is released. Above a thickness of 3 ML, on the other hand, T_C increases after annealing. One possible argument for explaining this behavior is that the Co islands created by tempering are large enough to enable a ferromagnetic coupling. The lateral finite size effect should play no important role, the Curie temperature is determined by the heights of the islands. Below this coverage, the Co structure acts as a superparamagnetic system with most likely small and disconnected islands. Therefore, the magnetic phenomenon observed here is connected with the Co island structure.

4 Summary

The STM data clearly display a completely different geometric order for thin epitaxial hcp cobalt films (atomically flat terraces with a width of some hundred nm)

and three dimensional cobalt islands after annealing these films to about 1000 K. The growth conditions of both thin films as well as 3D islands are strongly dependent on the cleanliness of the tungsten substrate. Up to now it has not been proven that a monolayer of cobalt is located between the island structure and the tungsten substrate. Our LEED data, showing both a (4×1) superstructure and a sixfold symmetry belonging to the hcp structure, are strongly pointing to a complete cobalt overlayer on tungsten.

By the technique of magnetic linear dichroism in photoemission we have investigated magnetic and electronic properties of cobalt islands on W(110). MLDAD arises from an energetic splitting of the final states in combination with a hybridization of bands with different spins and symmetry in the initial state provided the setup exhibits a chirality. Both structures, i.e., epitaxial Co films and cobalt islands on W(110), have a remanent magnetization along the axis of the $[1\bar{1}0]$ tungsten substrate.

We gratefully acknowledge support by BESSY (Berlin) and helpful discussion with J. Braun, Univ. Osnabrück. This work has been supported by the Deutsche Forschungsgemeinschaft DFG via contract DFG ME 835/14.

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