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# Pseudomorphic Co/Cr(1 1 0) films investigated by magnetic dichroism in photoemission and MOKE

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## Abstract

The electronic and magnetic properties of heteroepitaxial Co/Cr(1 1 0) films are investigated by magnetic dichroism in UV photoemission and the magneto-optical Kerr effect (MOKE). The Cr(1 1 0) host supports pseudomorphic growth of single crystalline and uniform Co films several nanometers in thickness. Pronounced magnetic dichroism occurs in the valence band region close to the Fermi edge. The observed spin-averaged valence band spectra and the asymmetry in photoemission intensity upon magnetization reversal are discussed in conjunction with the well-established spin-dependent band structure of body-centered cubic Co. The azimuthal dependence of the magnetic dichroic effect suggests two-fold easy axis behavior with the spontaneous in-plane magnetization aligned along [0 0 1], which is corroborated by complementary MOKE measurements. © 1999 Elsevier Science B.V. All rights reserved.

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## 1. Introduction

Heteroepitaxial growth of ultrathin metal films opens up the possibility to generate artificial crystal structures which do not exist in the bulk. This approach is especially relevant for the study of magnetic materials since structural features have significant impact on inherent magnetic properties in the thin film regime [1,2]. One such material which exists in different crystal structures is the 3d metal cobalt: while the equilibrium bulk state of Co corresponds to HCP structure ($a = 2.51$ Å, $c/a = 1.62$), it may adopt face-centered or body-centered cubic arrangement in epitaxial films. Various low-indexed metal substrate surfaces are known to support the formation of FCC Co [3,25–27]. Less work, however, has been reported which succeeded in stabilizing pseudomorphic BCC structure: apart from the prominent case of Co films grown on GaAs substrates [4], tetragonally distorted BCC structure has been observed on Fe(1 0 0) [5,28] and FeAl(1 0 0) [6] below $\sim 10$ ML coverage. We recently showed that pseudomorphic growth of Co in BCC arrangement is also achieved on the Cr(1 1 0) surface [7]: epitaxial films with a high degree of uniformity and

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long-range order are formed up to several nanometers in thickness. Here, we report on the electronic and magnetic properties of these Co/Cr(1 1 0) films investigated by magnetic dichroism in photoemission, in conjunction with complementary magneto-optical Kerr effect (MOKE) measurements. It is shown that the spin-averaged valence band emission close to the Fermi edge and the easy axis behavior of the spontaneous magnetization are in accord with characteristic features previously reported for BCC Co on GaAs(1 1 0) [4,8,9].

## 2. Experimental

All experiments were performed under UHV conditions. A hemispherical electron energy analyzer (VG CLAM-2) and conventional laboratory light sources generating Al $K_\alpha$ and HeI radiation were used to measure photoelectron spectra and magnetic dichroism in the valence band region. For dichroic measurements the sample magnetization could be reversed by an external field of ±160 Oersted (Oe) generated by a pair of Helmholtz coils. The field orientation with respect to the specimen was adjustable by polar rotation of the coils and azimuthal rotation of the sample. Complementary magnetic information was obtained from in situ measurements of the magneto-optical Kerr effect (MOKE) [10,29]. For this purpose, an intensity stabilized 5 mW He–Ne laser was used as a polarized light source.

Similar to preparation procedures given in the literature [11] a chemo-mechanical polished Cr(1 1 0) crystal was cleaned by simultaneous $Ne^+$ sputtering (800 eV, $I_0 \sim 3\ \mu A$) and annealing at 900 K. After repeated preparation cycles a sharp (1 × 1) LEED pattern was obtained indicating that this cleaning procedure leads to a well-ordered Cr(1 1 0) surface with a defect-free length of ~600 Å. The latter was deduced from the measured halfwidth of the specular LEED beam amounting to 0.5% of the surface Brillouin zone along [0 0 1]. At this stage no residual nitrogen (which is the main bulk contamination [11]) was detectable by X-ray photoelectron spectroscopy. Co was deposited from an electron beam evaporator at a rate of ~0.5 Å/min and substrate temperatures between 250 and 500 K.

## 3. Results and discussion

As a result of the structure analysis carried out by high-resolution LEED (for more comprehensive discussion see Ref. [7]) Co is stabilized in pseudomorphic structure irrespective of the particular growth temperature employed. Fig. 1 shows an exemplary diffraction pattern for 5 ML Co deposited at 500 K which indicates that the crystallographic orientation of the epitaxial Co film corresponds to that of the Cr(1 1 0) surface, i.e., the integral-order spot positions of the overlayer (denoted by indices) coincide with those of the substrate. The present Co/Cr(1 1 0) films are characterized by two specific structural features: firstly, the overlayer adopts body-centered cubic arrangement[1] up to appreciable thickness. The lateral lattice parameters retain the pseudomorphic values of 2.88 Å along [0 0 1] and 4.08 Å along $[1\,\bar{1}\,0]$ (experimental error ±1%) over the entire coverage range investigated, that is up to 30 ML thickness. The interlayer spacing (estimated from characteristic electron energies associated with Bragg reflections of the specular beam) measures $d_{(110)} = 2.05$ Å (±4%). Secondly, the diffraction pattern shows a (3 × 1) superstructure consisting of two equivalent reconstruction domains. Each of these domains implies uniaxial three-fold superperiodicity along either one of the two in-plane close-packed atom row directions. Remarkably, the superstructure does not originate from a strictly two-dimensional surface reconstruction, instead, it shows periodic correlation normal to the film plane as well. This is evident from the observed intensity variation of superstructure spots as a function of the perpendicular scattering vector $\boldsymbol{K}_\perp$ which can be attributed to kinematic scattering from a coherent lattice reconstruction involving several atomic

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[1] The crystal structure of the Co overlayer is deduced from Bragg reflections due to dominant kinematic scattering observable for the specular LEED beam as well as for the first-order beams [7].

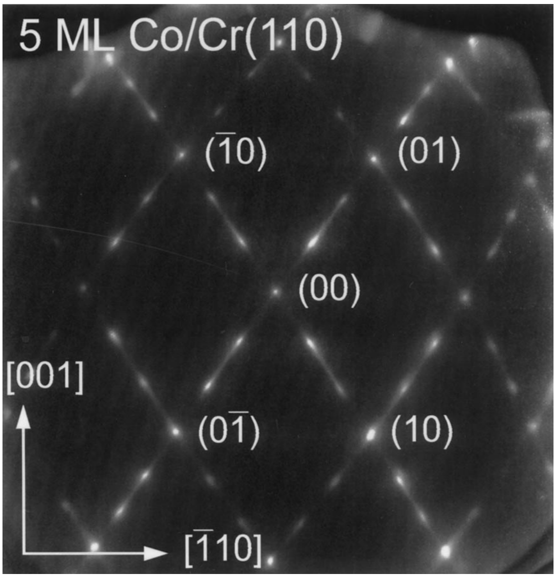

Fig. 1. LEED pattern of a 5 ML thick Co/Cr(1 1 0) film at 125 eV with a (3 × 1) superstructure in two equivalent domains.

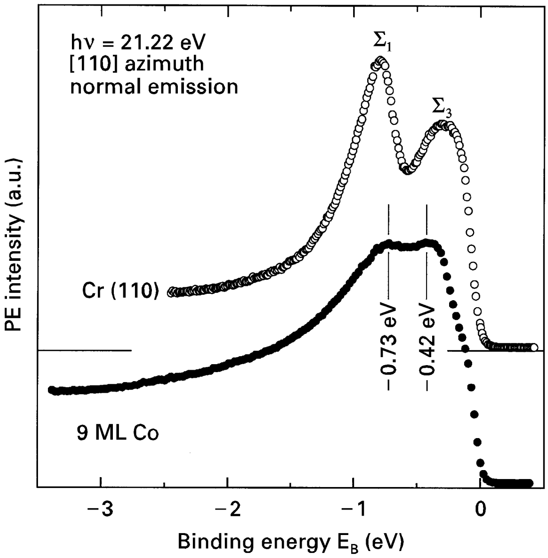

Fig. 2. PE spectra (unpolarized He I radiation, normal emission, light incidence along $[1\,\bar{1}\,0]$ azimuth and at $\Theta_i = -40°$ with respect to surface normal) of the initial Cr(1 1 0) surface (empty dots) and a 9 ML thick Co/Cr(1 1 0) film (full dots).

layers of the overgrowth. Obviously, the reconstruction observed here is an inherent feature connected with the forced pseudomorphic structure of the Co overlayer. It is noted that lattice reconstructions are also well-known to occur for other forced structures such as pseudomorphic FCC Fe films stabilized on Cu(1 0 0) [12].

To explore the electronic and magnetic properties of the pseudomorphic Co/Cr(1 1 0) films under investigation we measured UV photoemission (PE) spectra and magnetic dichroism (MD) in the valence band region. In the latter case one probes the asymmetry $A(M_\pm) = [I(M_+) - I(M_-)]/[I(M_+) + I(M_-)]$, i.e., the normalized difference in emission intensity for opposite magnetizations of the specimen. MD in valence band photoemission yields sizable effects even for unpolarized light [13,14] as used in the present study. The upper curve in Fig. 2 shows the PE intensity of the initial Cr(1 1 0) surface measured as a function of the binding energy with respect to the Fermi edge (He I radiation, light incidence along $[1\,\bar{1}\,0]$ azimuth and 40° off the surface normal). Owing to the employed normal emission geometry, relevant final states have to be totally symmetric with respect to the surface normal, i.e., they are characterized by $\Sigma_1$ symmetry [15]. Dipole allowed transitions will thus occur for initial states with $\Sigma_1$, $\Sigma_3$, or $\Sigma_4$ symmetry only. Accordingly, the two distinct emission peaks at $-0.31$ and $-0.77$ eV binding energy are identified as direct transitions from $\Sigma_3$ and $\Sigma_1$ initial-state bands near the Fermi edge, as has been demonstrated previously by Persson and Johansson [16]. After depositing a 9 ML thick Co film onto the Cr(1 1 0) host the PE spectrum represented by the lower curve in Fig. 2 is obtained. The emission intensity in the valence band region shows up as a faint double peak structure with particular contributions located at $-0.42$ and $-0.73$ eV binding energies. Since the present Co/Cr(1 1 0) overlayers under consideration are strained to pseudomorphic BCC structure it is obvious to compare their PE spectra with electronic features previously reported for Co in BCC arrangement. Prinz et al. [8] performed band structure calculations and spin-resolved PE studies to investigate BCC Co films stabilized on GaAs(1 1 0). Pronounced spin-polarized emission from minority and majority bands was found, corroborating previous theoretical findings [17,18] that the band structure of BCC Co is significantly different for minority and majority spins. The separate minority and majority peaks observed in the spin-resolved PE spectra were assigned to direct transitions involving the uppermost occupied minority and majority bands with $\Sigma_1$ and $\Sigma_3$ symmetry. Their relative peak positions (which were measured for photon energies between 20 and 50 eV) resemble those of the double peak structure in the spin-averaged valence band emission found in the present case (as denoted by vertical lines in Fig. 2). This coincidence suggests that Co films on Cr(1 1 0) exhibit electronic properties similar to those known for Co/GaAs(1 1 0) in BCC symmetry [8]. Hence, it seems justified to qualitatively discuss our data on the basis of band structure calculations previously reported for BCC Co [8,17]. Potential initial states to contribute to the peak located at $E_B = -0.42$ eV would then correspond to the uppermost occupied $\Sigma_1$ and $\Sigma_3$ minority bands, whereas the energetic position of the second peak at $E_B = -0.73$ eV is consistent with emission from the uppermost occupied $\Sigma_1$ and $\Sigma_3$ majority bands.

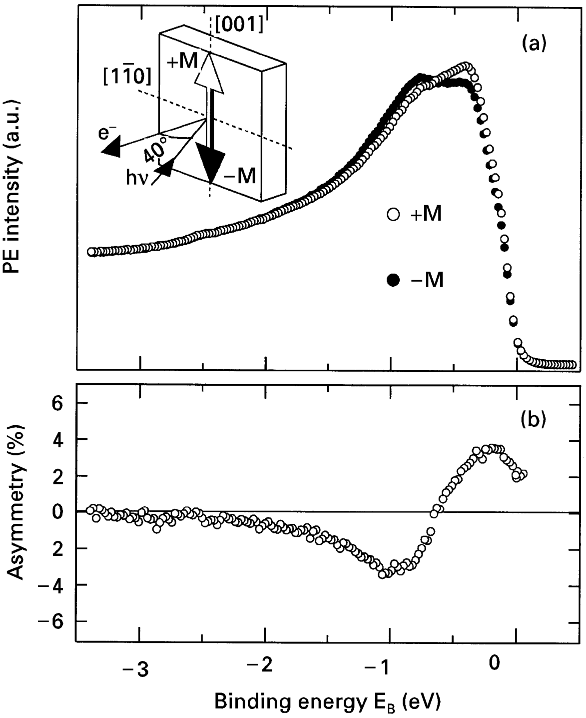

Fig. 3. PE intensity of a 9 ML thick Co/Cr(1 1 0) film measured at room temperature (experimental geometry same as in Fig. 2) for sample magnetization parallel (empty dots) and antiparallel (full dots) to the [0 0 1] direction (a) and corresponding asymmetry $A(M_\pm) = [I(M_+) - I(M_-)]/[I(M_+) + I(M_-)]$ (b); the inset in the upper panel illustrates the employed measuring geometry.

Furthermore, the Co films under investigation exhibit pronounced magnetic dichroism in the valence band region. Fig. 3a shows related spectra for a magnetically saturated 9 ML thick Co film (He I radiation, normal emission geometry, angle of incidence[2] with respect to surface normal $\Theta_i = -40°$). Maximum dichroic effect was obtained for light incidence along the [1 $\bar{1}$ 0] azimuth; this case is shown in Fig. 3a. Owing to the known angular dependence of magnetic dichroism in photoemission [19,20] this suggests a spontaneous in-plane magnetization which is aligned along the [0 0 1] azimuth, in agreement with previously reported easy axis behavior of BCC Co/GaAs(1 1 0) films [4,9]. Before discussing the present azimuthal dependence of the dichroic effect in detail, we focus on the observed asymmetry. Open (full) data points in Fig. 3a denote the valence band emission for sample magnetization parallel (antiparallel) to the [0 0 1] direction. Fig. 3b shows the corresponding asymmetry $A(M_\pm)$ which adopts positive values close to the Fermi edge with maximum magnitude of +4.0% at $E_B = -0.25$ eV, changes sign at about $-0.65$ eV binding energy, and reaches the largest negative value of $-3.0\%$ at $E_B = -1.0$ eV. The sign reversal as observed here suggests a definite correlation between the asymmetry and the spin polarization of particular initial states contributing to the PE process. Experimental evidence for this correspondence was first reported by Getzlaff et al. [13], who studied dichroic effects in valence band photoemission from ultrathin Fe(1 1 0)-, Co(0 0 0 1)-, and Gd(0 0 0 1) films on W(1 1 0) with unpolarized light. Following qualitative arguments based on Cherepkov's atomic model [19], these authors demonstrated that the

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[2] The sign of $\Theta_i$ refers to the experimental geometry as defined in Ref. [19].

observed sign of asymmetry $A(M_{\pm})$ is a qualitative measure for the initial state polarization. Accordingly, the presently observed sign reversal of asymmetry in Fig. 3b may be interpreted as an indication for the difference in spin polarization of particular initial states involved, i.e., the uppermost $\Sigma_1$ and $\Sigma_3$ minority and majority bands of Co in BCC structure. This conclusion, however, has to be taken with caution since recent investigations [14] have revealed that the correlation between asymmetry and initial state polarization is not of general validity for MD in valence band photoemission. Experimental studies in conjunction with band structure calculations showed that the main ingredient necessary to produce MD in valence band photoemission corresponds to the hybridization of initial states induced by spin–orbit coupling (SOC). Pronounced MD was observed for Co/Cu(1 0 0) [14], Co/W(1 1 0) [21] and Fe/W(1 1 0) [22] due to excitation from SOC-induced hybridization zones in the band structure. In addition, Henk et al. [20] have investigated magnetic dichroism in valence band photoemission from low-indexed magnetic surfaces by calculating spin density matrix elements within a one-electron model. They derived analytical expressions for the spin-averaged intensity and the spin polarization of photoelectrons for various surface orientations. For cubic (1 1 0) surfaces with an in-plane magnetization parallel (antiparallel) to the [0 0 1] direction (which corresponds to our situation) and p-polarized incident light, it is evident from their results that the asymmetry $A(M_{\pm})$ is determined by products of matrix elements which involve initial state wave functions of both $\Sigma_1$ and $\Sigma_4$ character. For s-polarized light, on the other hand, no magnetic dichroism in the sense of intensity asymmetry upon magnetization reversal was found (in accordance with earlier work [19] based on a pure atomic picture). This theoretical description which applies to linearly polarized light is also applicable to our experimental situation, since unpolarized light is equivalent to an incoherent superposition of s- and p-polarized light. In the framework of these theoretical considerations, SOC-induced band gaps due to hybridization of $\Sigma_1$ and $\Sigma_4$ initial state bands may be responsible for the presently observed MD effect (provided that an appropriate final state is available under excitation with He I radiation). According to calculated BCC Co band structures [8,17] a crossing point of $\Sigma_1$ and $\Sigma_4$ initial state bands indeed exists at a binding energy of about $-1.3$ eV. This value, however, is too low compared to the experimentally observed energy range in which significant MD occurs [cf. Fig. 3b]. To verify the origin of the observed MD effect in detail future band structure calculations are required which take into account the actual structural data of the pseudomorphic Co/Cr(1 1 0) films.

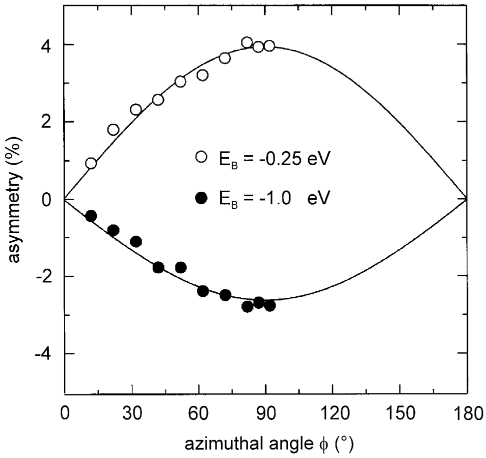

Fig. 4. Azimuthal dependence of the magnitude of asymmetry at binding energies of $-0.25$ eV (empty dots) and $-1.0$ eV (full dots); the azimuthal angle $\varphi$ refers to the angle between the [0 0 1] direction and the azimuth of light incidence, the data are fitted according to $\sin(\varphi)$-like azimuthal dependence (solid lines).

Finally, the dependence of the MD effect on the azimuthal angle $\varphi$ is addressed. According to theory [19,20], p-polarized incident light and an in-plane-oriented magnetization yields an angular variation which follows $\sin(\varphi)$-like behavior, with $\varphi$ the angle between the in-plane magnetization direction and the azimuth of light incidence. Hence, maximum dichroic effect is obtained if the magnetization is perpendicular to the incident direction of light ($\varphi = \pi/2$), whereas no dichroism occurs if the magnetization is parallel to the azimuth of light incidence ($\varphi = 0$). Fig. 4 shows the angular dependence of the magnitude of asymmetry measured at binding energies of $-0.25$ eV (empty dots) and $-1.0$ eV (full dots), respectively. The $x$ axis refers to the angle between the [0 0 1] direction and the

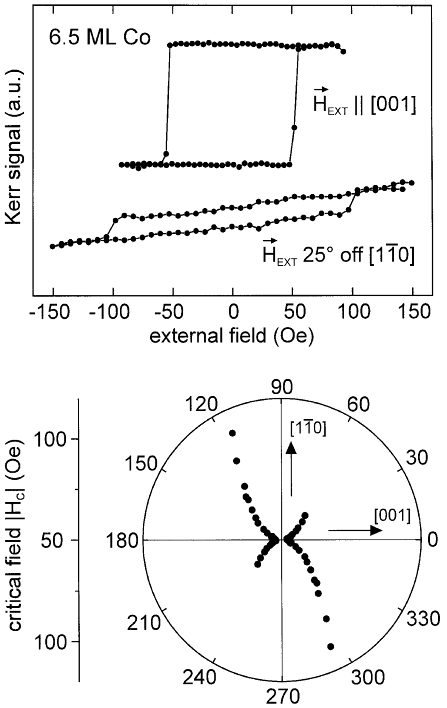

Fig. 5. Upper panel: Kerr hysteresis loops for a 6.5 ML thick Co film at 340 K with the external field oriented in the film plane and aligned along the [0 0 1] azimuth (top) and 25° off the [1 $\bar{1}$ 0] azimuth (bottom); lower panel: polar plot of the critical field $|H_C|$ depending on the azimuthal direction of the external field (azimuthal angle measured with respect to the [0 0 1] direction).

azimuth of light incidence. The data are fitted according to sin($\varphi$)-like angular behavior (solid lines). As apparent from the observed angular dependence, the spontaneous magnetization is aligned along the [0 0 1] azimuth since light incidence along the [1 $\bar{1}$ 0] azimuth leads to a maximum MD effect while no MD occurs for incidence along the [0 0 1] azimuth. We verified the easy axis behavior as deduced from the present MD effect by complementary MOKE measurements. The upper panel of Fig. 5 shows Kerr hysteresis loops measured for a 6.5 ML thick film at 340 K with the external field oriented in the film plane and aligned along the [0 0 1] azimuth (top) and 25° off the [1 $\bar{1}$ 0] azimuth (bottom). The polar plot in the lower panel shows the magnitude of the critical field $|H_C|$ depending on the azimuthal direction of the external field (azimuthal angle measured with respect to the [0 0 1] direction). It is evident from the data in Fig. 5 that rectangular hysteresis loops with minimum coercivity are obtained with the external field along [0 0 1], whereas a strong reduction in squareness and a continuous increase in coercivity are observed when the external field direction is changed towards the [1 $\bar{1}$ 0] azimuth. In particular, the sample could not be saturated with maximum external field applied along the [1 $\bar{1}$ 0] azimuth. Hence, the present Co/Cr(1 1 0) films exhibit a uniaxial in-plane orientation of the spontaneous magnetization along [0 0 1], in accordance with the observed angular dependence of the MD effect. Furthermore, a linear increase of the remanent Kerr signal with Co coverage was observed which indicates ferromagnetic behavior of the entire film.

## 4. Conclusions

The electronic and magnetic properties of pseudomorphic Co/Cr(1 1 0) films as deduced from UV photoemission and MOKE measurements are in accord with those previously reported for BCC Co stabilized on GaAs(1 1 0). The measured spin-averaged PE intensity distribution agrees with principal features of the spin-dependent band structure of BCC Co as determined by experiment [8] and theory [17]. The sign of PE intensity asymmetry upon magnetization reversal is consistent with the expected spin polarization of contributing initial states, if one accepts the qualitative application of the atomic model for MD [19]. The validity of this concept for the system investigated here and the detailed origin of the observed MD effect have to be verified by future band structure calculations based on the structural data of these Co/Cr(1 1 0) films. Furthermore, the spontaneous magnetization (deduced from Kerr hysteresis loops and the azimuthal dependence of the MD effect) is oriented in the film plane and corresponds to the [0 0 1] direction, in agreement with the uniaxial easy axis behavior observed for Co/GaAs(1 1 0) by magnetometry [4] and Brillouin light scattering [9]. This tight correspondence in primary magnetic properties is remarkable, since the Co/Cr(1 1 0) films investigated here exhibit specific structural details: due to pseudomorphism their lateral lattice constants are larger by 2% compared to those of Co/GaAs(1 1 0) [4] and they are characterized by a superimposed (3 × 1) reconstruction. We associate this circumstance with the theoretical finding [18,23,30] that in the case of BCC Co physical quantities like magnetic state or magnitude of magnetic moments are rather insensitive to the actual lattice parameters. This behavior is in clear contrast to the widely investigated case of metastable FCC Fe which shows a delicate interrelationship between magnetic state and pseudomorphic strain [12,24].

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