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# Magnetic moments of fcc Fe overlayers on Cu(100) and Co(100)

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The magnetism of ultrathin fcc Fe overlayers on Cu(100) and Co(100) deposited at room temperature has been investigated at 110 K using magnetic circular dichroism in x-ray absorption spectroscopy (MCXD). Three regions of different magnetic behavior in dependence of the Fe thickness are distinguished in the element-specific MCXD investigation in agreement with the results of previous experiments. The Fe $3d$ magnetic spin and orbital moments are derived from the MCXD spectra by use of sum rules as a function of the Fe thicknesses. Up to 4 ML Fe the Fe spin moment is $3.0\mu_B$ on Co(100) and $2.8\mu_B$ on Cu(100) demonstrating that the whole Fe film is in a ferromagnetically ordered high-spin state. We find that between 5 and 10 ML Fe the average Fe spin moment is $1.1\mu_B$ on Co(100) and $0.8\mu_B$ on Cu(100). The surface region of the Fe films is ferromagnetically ordered on both substrates. The larger average moment on Co(100) originates from the additional contribution of the Fe atomic layer at the Fe-Co interface. For Fe thicknesses larger than 10 ML the average Fe moment increases again. [S0163-1829(99)01806-8]

## I. INTRODUCTION

Fcc Fe films are of particular interest in research on structure and magnetism because they exhibit a large variety of structural and magnetic properties which depend on the Fe overlayer thickness and the preparation conditions.

According to a structural model[1] based on low-energy electron diffraction (LEED) *I-V* (Refs. 2–4) and low-energy ion scattering[5] experiments, three regions occur for Fe overlayer thicknesses between 1 and 20 ML. In the first region, between 1 and 4 ML, the Fe film adopts the in-plane interatomic distance of the Cu(100) substrate (1.81 Å), whereas the interlayer distance is enhanced along the surface normal [1.88 Å (Ref. 2)] resulting in a tetragonally distorted structure for the whole Fe film. In the second region, between 5 and 10 ML, the Fe film has fcc structure. According to the LEED *I-V* results in Ref. 3 only the topmost two interlayer distances are enhanced ($d_{12}=1.84$ Å; $d_{23}=1.80$ Å) with respect to the interlayer distance in the interior (1.77 Å). In the third region, above 10 ML Fe, the structure gradually converts to bcc.[3–5] Reflection high-energy electron diffraction oscillations and LEED results observed for the growth of Fe on Cu(100) and Co(100) show a similar behavior on both substrates.[6] These results indicate that the fcc Fe overlayers on Co(100) have a similar structure as on Cu(100).[6] The similarity of the systems Fe/Cu(100) and Fe/Co(100) can be understood from the fact that Co grows on Cu(100) with the in-plane interatomic distances of the substrate up to 15 ML at room temperature[7] and thus provides similar structural conditions for the Fe overlayers as the Cu(100) substrate.

Theoretical studies of magnetovolume effects in bulk fcc Fe (Ref. 8) reveal that, depending on the lattice constant, bulk fcc Fe is nonmagnetic (NM), antiferromagnetic (AFM) or ferromagnetic, the latter one with high (HS) or low (LS) local spin moment. The lowest total energy has been found at a radius of the Wigner-Seitz cell of $r_{\mathrm{WS}}=2.54$ a.u. At this equilibrium radius a second-order phase transition from NM to AFM behavior with increasing unit-cell volume was predicted. When the volume is further enhanced a first-order phase transition from AFM to HS ferromagnetic behavior is predicted at a radius of 2.71 Å. From this strong dependence of the magnetic behavior on the lattice constant in bulk fcc Fe also the magnetic properties of the ultrathin Fe films on Cu(100) and Co(100) are expected to change with Fe thickness in correspondence with the structural modifications. In addition, in ultrathin Fe films also interface and surface effects can be important for the magnetic properties.

Also the magnetic properties of fcc Fe films on both substrates, Cu(100) and Co(100), display a large similarity. This has been investigated by means of the magneto-optical Kerr effect[6,9,10] (MOKE) and magnetic circular dichroism in x-ray absorption spectroscopy (MCXD).[11] According to the behavior of the MOKE signal of Fe/Cu(100) and Fe/Co(100) three regions can be distinguished as a function of the Fe thickness. Between 0 and 4 ML (region I) and above 10 ML (region III) the MOKE signal increases linearly with increasing Fe coverage. This behavior is expected for homogeneously magnetized films.[9] Contrary, between 5 and 10 ML (region II) the Kerr ellipticity is relatively small and constant indicating that the interior of the film does not contribute to the observed Kerr signal.[9] The changes in the behavior of the MOKE signal take place at exactly these Fe thicknesses where the structure changes[6,9,10] indicating that there is a correspondence between structure and magnetism. An exposure of the Fe films on Cu(100) in region II to 0.5 Langmuir CO lowered the Curie temperature by 40 K. This can be taken as evidence that the magnetism is essentially localized at or near the surface of the Fe films in region II.[9] Later a two-peaked behavior of the Kerr signal in region II as a function of the Fe film thickness has been observed at temperatures below 200 K.[10,12] Such a variation of the Kerr signal with Fe thickness suggests that the perpendicular component of the net magnetic moment of the whole film oscillates with thickness. This oscillatory behavior can be explained for example by a ferromagnetically ordered surface on Fe underlayers which are each ferromagnetically ordered and magnetically aligned layerwise antiparallel.[10] Moreover, it has been found that the development of the three magnetic regions critically depends on the growth conditions. If, for example, the Fe film is deposited at a substrate temperature of 190 K or lower the structural and magnetic properties of Fe/Cu(100) are qualitatively different.[10]

Comparing the Cu(100) and the Co(100) substrates differences are expected in the electronic and magnetic structure at the Fe-substrate interface because on Cu the Fe *d* states hybridize only weakly with Cu *sp* states whereas on Co they strongly hybridize with spin-polarized Co *d* states. As demonstrated by MCXD at room temperature[11] this leads to ferromagnetic order in the Fe layer at the Fe-Co interface. Also the magnetic anisotropies depend on the substrate. On Cu(100) the Fe films exhibit perpendicular anisotropy for low coverages and at about 11 ML the magnetization switches to in-plane orientation.[9] Contrary, on Co(100) the magnetization is in-plane for all Fe thicknesses studied.[6,11]

The magnetic properties of Fe/Cu(100) and Fe/Co(100) grown at room temperature have also been investigated by use of MCXD.[11] MCXD has the advantage over MOKE to be element specific due to the binding energy of the core levels involved in the x-ray-absorption process. Furthermore, the MCXD asymmetry at the maximum of the $L_3$ white line is proportional to the average magnetization of the element under consideration. This has been utilized in Ref. 11 in order to compare the magnetic properties of Fe on Co(100) with Cu(100) at *room temperature*. On Cu(100) no dichroism signal was observed for Fe films below 10 ML thickness indicating that the critical temperatures for magnetic order are below room temperature in this case. Above 10 ML Fe the MCXD asymmetry at the Fe $L_3$ maximum gradually increases. On Co(100) for Fe thicknesses between 1 and 4 ML the MCXD asymmetry of the tetragonally distorted fcc Fe is larger than for bulk bcc Fe which was ascribed[11] to an enhanced moment at the Fe-Co interface or at the Fe surface. At 5 ML the Fe asymmetry drops sharply and is small and slightly decreasing between 5 and 11 ML. The behavior of the asymmetry in region II was explained assuming that approximately one Fe layer at the Fe-Co interface carries a moment, the value of which is between the bcc Fe bulk moment ($2.2\mu_B$) and the maximum Fe moment in Fe-Co alloys ($\sim 3.0\mu_B$). The other layers have been assumed to be paramagnetic since the critical temperature in region II of Fe/Co(100) is likely to be similar to the Fe/Cu(100) system,[11] i.e., to be below room temperature.[9,10] Above 11 ML the MCXD asymmetry of Fe gradually increases.

Despite the extensive research on the magnetic properties of fcc Fe overlayers on Cu(100) and Co(100) deposited at room temperature the average magnetic moment in these films has not yet been determined. We investigated these systems by means of MCXD at a temperature of 110 K in order to estimate the magnetic moments, to draw conclusions on the layer-by-layer magnetic structure of the fcc Fe overlayers and to study the influence of replacing the Cu substrate by ferromagnetic Co. Thereby we concentrated on Fe thicknesses below the onset of gradual conversion to bcc structure at 10 ML Fe. Our MCXD measurements have been performed at a temperature of 110 K which is well below the Néel temperature of 200 K suggested in Ref. 10 but above 65 K given in Ref. 13.

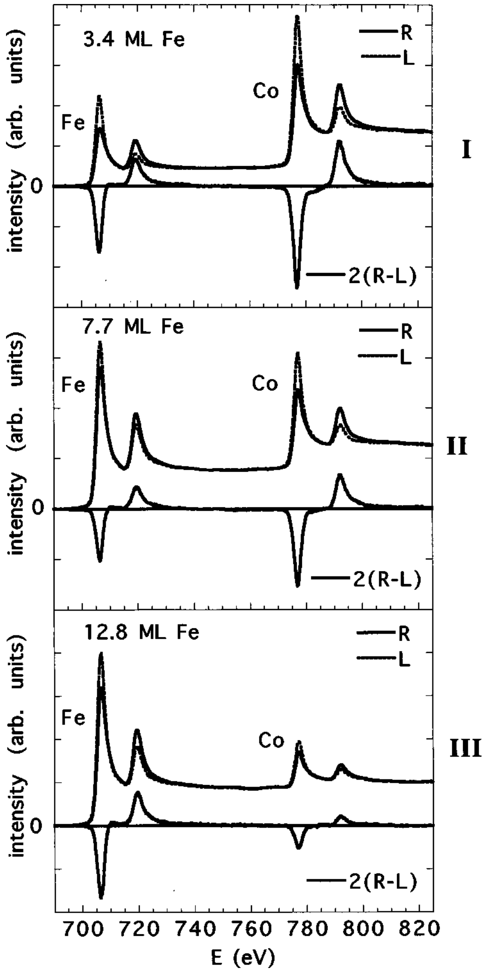

FIG. 1. MCXD spectra of 3.4, 7.7, and 12.8 ML fcc Fe on Co(100) at 110 K representing the magnetic region I, II, and III, respectively. The spectra have been measured with an angle of 23° between in-plane magnetization and photon polarization vector.

## II. EXPERIMENT

We used a Cu(100) single crystal as a substrate. Before the film deposition the substrate was prepared by cycles of sputtering and annealing until sharp diffraction spots were observed by LEED. Co and Fe were deposited *in situ* by e-beam evaporation with typical rates of 1 ML/min. The pressure in the chamber during evaporation of Co and Fe was below $3\times10^{-10}$ mbar and $8\times10^{-10}$ mbar, respectively. The deposition rates were determined using an oscillating quartz crystal thickness monitor. All Fe and Co films were deposited at room temperature at which a layer-by-layer growth mode has been observed over a wide range of Fe film thickness without significant interdiffusion at the Fe-substrate interface.[14] Using wedges in the experiments ensures identical growth conditions for different overlayer thicknesses. Thus Fe wedges with a slope of typically 1 ML/mm were prepared by moving the substrate behind a shutter. The Fe thicknesses were cross checked using x-ray-absorption spectroscopy (XAS) intensity ratios of the deposited materials and the substrate. The structural quality of the samples was checked by LEED. The samples could be cooled down to 110 K using liquid $N_2$.

Circularly polarized x-ray radiation was provided by the

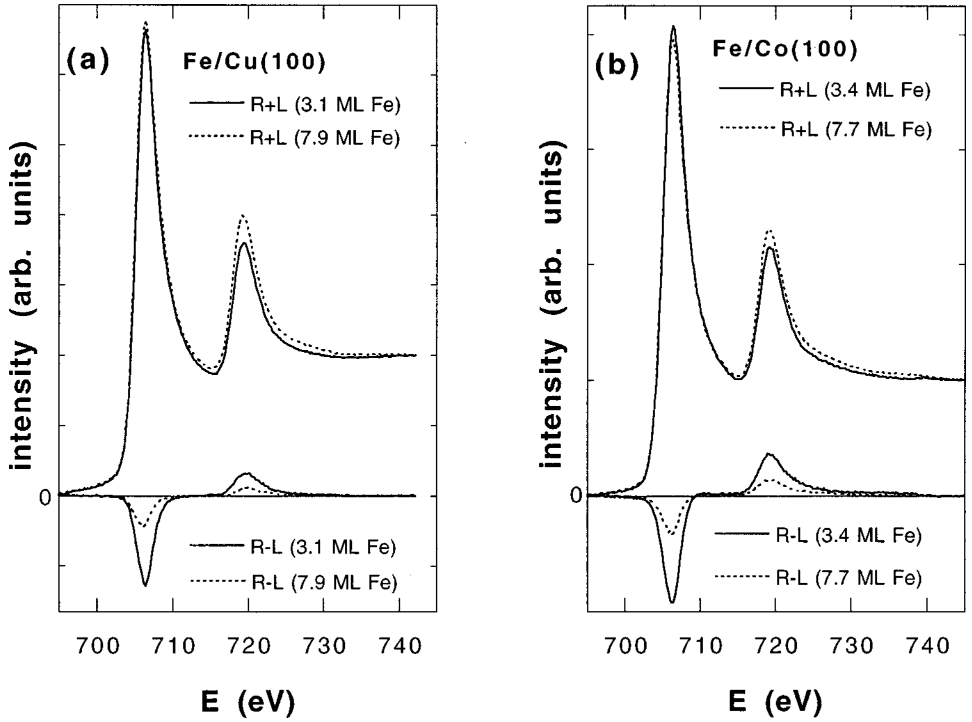

FIG. 2. Direct comparison of isotropic $(R+L)$ and dichroism spectra $(R-L)$ for (a) 3.1 ML and 7.9 ML Fe on Cu(100) and (b) 3.4 and 7.7 ML Fe on Co(100). The strong decrease in the asymmetry $|(R-L)/(R+L)|$ from region I to region II is clearly visible. The spectra were measured at a temperature of 110 K.

SX700/3 beamline at the synchrotron BESSY in Berlin. The samples were remanently magnetized by applying a field pulse along the fcc ⟨100⟩ direction, perpendicular to the surface plane, for Fe/Cu and along the in-plane fcc ⟨110⟩ direction for Fe/Co. The incidence angle of the light with respect to the surface normal was 67° for the Fe films on Co and 45° for the Fe films on Cu, thereby satisfying the condition for the observation of the MCXD effect that the magnetization must have a component parallel to the polarization vector of the incident x-ray beam. The dichroism spectra were measured by reversing the magnetization of the samples. The absorption was monitored by measuring the partial electron yield, with a detector positioned within the light-scattering plane, at a 45° angle with respect to the incoming beam. Simultaneously the photocurrent of a Au mesh located behind the exit slit of the monochromator was measured as a reference. In each spectrum the partial electron yield signal was normalized to the photon flux monitored by the photocurrent from the Au mesh.

## III. RESULTS

In Fig. 1 MCXD spectra of 3.4, 7.7, and 12.8 ML Fe on Co(100) are compared, each Fe thickness representing one magnetic region. For each Fe thickness absorption spectra are shown with the polarization vector of the incident circularly polarized x-ray beam parallel $(R)$ and antiparallel $(L)$ to the magnetization direction of the sample. The $L_{2,3}$ absorption edges of Fe appear at the low-energy side and of Co at the high-energy side of the spectra. The absorption edges of the two magnetic materials Fe and Co are well separated in energy, which allows element-specific x-ray-absorption measurements. With increasing Fe overlayer thickness both the x-ray absorption due to Fe and the attenuation of the Co photoelectron yield (EY) by the Fe overlayer increase. Thus the intensity ratio between Fe and Co increases with Fe overlayer thickness as can be seen clearly in Fig. 1. In the present experiment a retarding voltage of −66 V was used in front of the EY detector in order to enhance the surface sensitivity and to lower the sensitivity of the EY signal to magnetic stray fields. Fitting the attenuation of the Co intensity $I$ with increasing Fe overlayer thickness $d$ in our MCXD spectra with $I=I_0\exp(-d/\lambda)$ leads to an electron probing depth $\lambda$ of $12\pm1$ Å. The dichroism signals $(R-L)$ of Fe and Co have identical signs indicating that the corresponding net magnetic moments of the Fe and Co films are oriented parallel.

In Fig. 2 absorption $(R+L)$ and dichroism spectra $(R-L)$ of Fe on Cu(100) (a) and Co(100) (b) are directly compared for Fe coverages of ~3 ML (solid line) and ~8 ML (dashed line), respectively. Going from region I to II the absolute amount of the Fe $L_3$ asymmetry $[|(R-L)/(R+L)|]$, i.e., the average Fe magnetization decreases strongly on both substrates whereas in the system Fe/Co the Co magnetization is alike in both regions. Qualitatively the Fe films on both substrates behave in a similar way whereas a quantitative analysis of the spectra reveals differences caused by the substrates.[15] At a temperature of 110 K the absolute amount of the asymmetry decreases by a factor of 2.2 on Co and 3.3 on Cu from region I to II, i.e., the decrease is much stronger on Cu. At room temperature this factor on Co is ~10,[11] due to the fact that in region II only the Fe layer at the Fe-Co interface is ferromagnetically ordered.[11] In order to clarify which parts of the Fe films in region II contribute to the magnetization at 110 K we have used the sum rules[16,17] to determine the average Fe moments as a function of the Fe coverage.

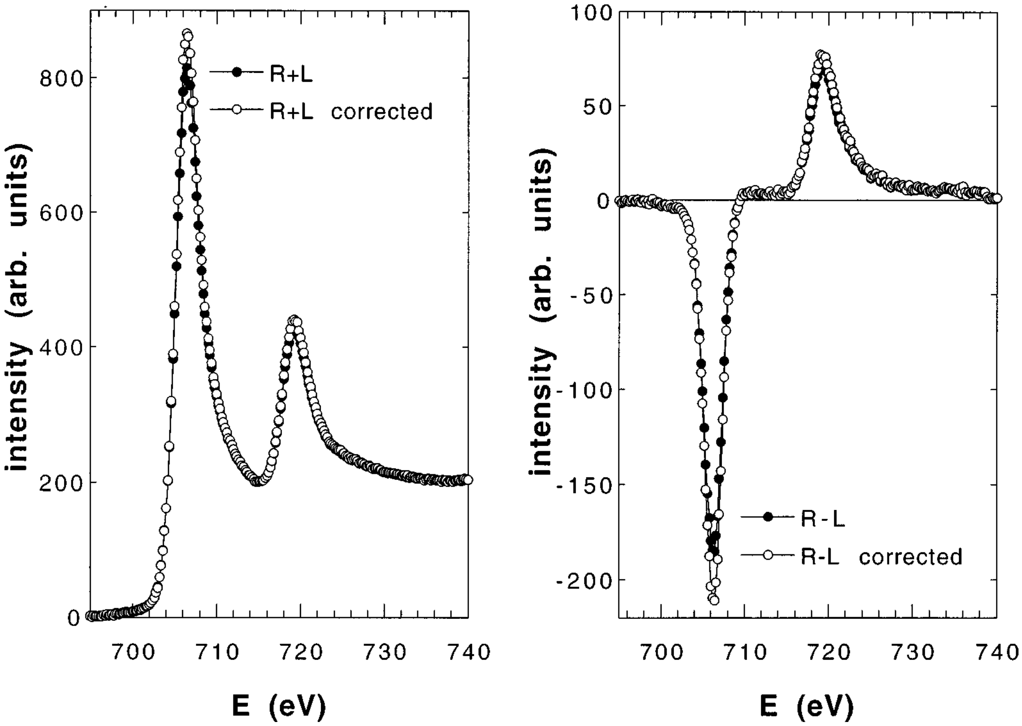

FIG. 3. Isotropic (left panel) and dichroism spectra (right panel) of 3 ML Fe/Co(100) before (solid symbols) and after (open symbols) self-absorption correction. The spectral contrast is enhanced by the self-absorption correction. At $L_3$ the dichroism intensity is enhanced more than at $L_2$.

Before the application of the sum rules it has to be ensured that the intensity in the absorption spectra is proportional to the energy-dependent absorption coefficient $\mu(E)$. It is known that the electron yield (EY) is affected by the so-called *self-absorption effect.* This effect manifests itself by the fact that at grazing incidence angles the EY is strongly enhanced and no longer proportional to the x-ray absorption coefficient.[18–20] The physical origin of this effect is that at grazing incidence the effective photon penetration depth becomes of the same order of magnitude as the electron probing depth. Consequently, all photoelectrons and secondary electrons are emitted from the sample independent of the photon energy above the threshold because in this case the photoelectrons are not longer *self-absorbed.* Thus the EY is saturated and the variations of the absorption coefficient are no longer detectable in the EY.

The absorption spectra were corrected for the self-absorption effect using the formalism described in Ref. 20 thereby following essentially the procedure described in Ref. 21 where the self-absorption correction had been performed in the thick sample limit.

The effect of the self-absorption correction is demonstrated in Fig. 3 for 3 ML Fe/Co(100). In Fig. 3 the original (full circles) and self-absorption corrected (open circles) experimental isotropic (left panel) and dichroism spectra (right panel) are compared. The intensity of the white lines is increased in the corrected spectra with respect to the edge jump. The absolute value of the dichroism signal is significantly enhanced at the $L_3$ but only slightly enhanced at the $L_2$ resonance. These enhancements have a large effect on the values for the orbital moment but only a substantially smaller effect on the values for the spin moment as determined by the sum rules given in Refs. 16,17. Since the orbital moment is proportional to the area under the $L_3$ dichroism signal minus the area under the $L_2$ dichroism signal (both areas are taken positive), its value is strongly enhanced by the self-absorption correction. In contrast, the spin moment is only enhanced by a few percent by the correction, because it is proportional to the area under the $L_3$ dichroism signal plus two times the area under the $L_2$ dichroism signal.

By applying the sum rules to the self-absorption corrected MCXD spectra the Fe spin and orbital moments have been derived. In the sum-rule analysis the steplike contribution to the absorption spectrum of the 4*s* states, which have less structure in the valence band than 3*d* states, has been described by arctan functions. The incomplete degree of circular polarization, $P_{circ}=0.73$,[22] and the angle between the polarization vector of the incident radiation and the magnetization direction of the sample have been taken into account. The resulting Fe spin moments on Co (solid circles) and Cu (open triangles) are plotted versus the Fe thickness in Fig. 4. For 1–4 ML Fe the spin moments are large, $3.0\mu_B$ on Co and $2.8\mu_B$ on Cu. Between 4 and 5 ML Fe there is a sharp decrease in the Fe moment. For 5–10 ML Fe the spin moments of Fe averaging over region II are $1.1\mu_B$ on Co and $0.8\mu_B$ on Cu. With respect to region I they are reduced to ~40% on Co and to ~30% on Cu. For Fe/Co the orbital moments of Fe are $0.18\mu_B$ in region I and $0.08\mu_B$ in region II. The observed dependence of the Fe moment on the Fe coverage is discussed in the following section.

## IV. DISCUSSION

The large Fe spin moments in region I correspond to a ferromagnetically ordered high-spin state of the tetragonally distorted fcc Fe. A ferromagnetically ordered high-spin state

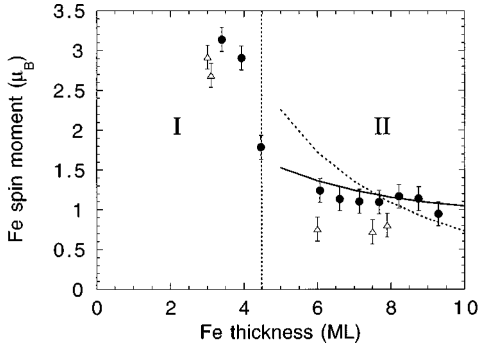

FIG. 4. Experimental spin moments of Fe on Co(100) (solid circles) and on Cu(100) (open triangles) as a function of the Fe thickness resulting from the application of the sum rules to the MCXD spectra. The solid (dashed) line indicates the magnetic moment resulting from the assumption that 2.5 (4.5) ML Fe at the surface (Fe-Co interface) are ferromagnetically ordered with an enhanced spin moment of $2.6\mu_B$.

is expected to form in bulk fcc Fe for enhanced unit-cell volumes[8] and in ultrathin Fe films on Cu(100).[23–25] Indeed, the Fe spin moment in region I on Cu of $2.8\mu_B$ resulting from our MCXD measurement agrees (i) with the result of a total-energy augmented-spherical-wave spin-polarized band calculation on magnetovolume effects in bulk fcc Fe (Ref. 8) and (ii) with the result of a self-consistent, all-electron total-energy full-potential linearized-augmented-plane-wave local spin-density-functional calculation[23] which is $2.85\mu_B$ for 1 ML Fe/Cu(100) and in average $2.73\mu_B$ for 2 ML Fe/Cu(100). For Fe films on Cu(100) deposited at a reduced substrate temperature of 120 K and subsequently annealed to 300 K larger Fe spin ($3.40\mu_B$) and orbital ($0.24\mu_B$) moments have been observed by means of MCXD.[26] These larger moments do not contradict our results since the structural and magnetic properties of the Fe overlayers on Cu are known to drastically depend on the preparation conditions.

The strong reduction of the Fe spin moment between 4 and 5 ML Fe on both substrates indicates that the magnetic behavior of the Fe films abruptly changes and that the Fe films are not longer in a ferromagnetically ordered high-spin state in region II. Moreover, in region II the average Fe moments are larger on Co than on Cu indicating that replacing the Cu substrate by ferromagnetic Co leads to an additional ferromagnetic contribution in the Fe overlayers. In order to investigate which parts of the Fe films on Cu(100) contribute to the net magnetization in region II we studied the dependence of the Fe dichroism on $O_2$ exposure. It was possible to suppress the dichroism signal of 6 ML Fe/Cu(100) at a substrate temperature of 110 K by exposing the sample to only 2 L $O_2$. This experimental observation indicates that on Cu(100) the surface region of the Fe films is ferromagnetically ordered, in line with former MOKE experiments[9] in which the surface has been contaminated with CO. Our experimental observations furthermore indicate that on Cu neither the interior of the fcc Fe overlayer nor the Fe layer at the Fe-substrate interface are ferromagnetically ordered in region II, otherwise the dichroism signal would not have dropped to zero after $O_2$ exposure. The latter finding is surprising since it is expected from theory[25] that not only the Fe bilayer in the surface region but also the Fe bilayer at the Fe-Cu interface is ferromagnetically ordered.

In the case of the Co substrate the MCXD measurements at *room temperature* in Ref. 11 imply that approximately one atomic Fe layer at the Fe-Co interface is ferromagnetically ordered with a spin moment between the bcc Fe bulk moment ($2.2\mu_B$) and the maximum Fe moment in FeCo alloys ($\sim 3.0\mu_B$). At a temperature of 110 K our MCXD measurements of Fe/Co reveal an average Fe spin moment of $1.1\mu_B$ in region II. In order to draw conclusions on the magnetic structure of Fe/Co it is important to note that the probing depth ($\lambda = 12$ Å) of the electrons which serve as a measure for the energy-dependent absorption coefficient is comparable in size with the Fe film thickness. If it is assumed that only Fe layers at the surface and the Fe-Co interface contribute to the magnetization with an enhanced spin moment of $2.6\mu_B$ two extreme cases can be distinguished. In the first case all ferromagnetically ordered layers are assumed to be located at the Fe-Co interface and in the second case in the surface region of the Fe film. In the first case 4–5 ML Fe at the Fe-Co interface would be ferromagnetically ordered and a fast decrease of the Fe moment with Fe thickness would be observed as indicated in Fig. 4 by the bold dashed line. In contrast, in the second case 2–3 ML Fe in the surface region would be ferromagnetically ordered and a behavior as indicated in Fig. 4 by the solid line would be observed. Clearly, the experiment is described much better by the second extreme case. In Fig. 4 only a small deviation from the solid line is observed if it is assumed that 2 ML in the surface region and 1 ML Fe at the Fe-Co interface are ferromagnetically ordered. The reason for this small deviation is that the electron yield signal from deeper layers is significantly damped by the layers above. Thus in the average Fe moments given in Fig. 4 the contributions from Fe layers located in the surface region have more weight than deeper Fe layers. This damping effect is described by $Y_n = Y_1 e^{-(n-1)/\lambda}$ (with $n \geqslant 1$) where $Y_n$ is the damped partial electron yield from the *n*th layer. For the atomic layer at the surface $n=1$. Because of $\mu_n \propto (R_n - L_n)/(R_n + L_n)$ the following equation for the average magnetic moment $\mu_{\text{average}}$ of the Fe films results:

$$\mu_{\text{average}} = \frac{\sum_{n=1}^{N} \mu_n e^{-(n-1)/\lambda}}{\sum_{n=1}^{N} e^{-(n-1)/\lambda}}, \tag{1}$$

where $\mu_n$ denotes the magnetic spin moment in the *n*th atomic layer, $N$ is the total Fe film thickness and $\lambda$ is the probing depth. Note that for homogeneously magnetized films the damping has no effect on the average moment. In our analysis leading to the bold dashed line and the solid line in Fig. 4 the damping has been taken into account by Eq. (1). Thus comparing to the room-temperature results in Ref. 11 our MCXD measurements demonstrate that on Co in region II at 110 K not only $\sim 1$ ML Fe at the Fe-Co interface but also $\sim 2$ ML Fe in the surface region of the Fe films are ferromagnetically ordered. Also our recent spin and angle-resolved photoemission investigation[27] demonstrates that the surface region in Fe films on Co(100) is magnetically alive. Furthermore, from the observed strong dependence of the average Fe magnetization on $O_2$ exposure in our MCXD experiment at 110 K, and from the enhanced average Fe magnetization at 110 K compared to the room-temperature results in Ref. 11, it can be concluded that the Fe film in region II is not homogeneously ferromagnetically ordered with a reduced moment.

The statistical error in the determination of the fcc Fe spin moment by the MCXD experiment is about $\pm 0.15\mu_B$ as indicated in Fig. 4. Therefore an oscillation in the magnetic spin moment as a function of the Fe thickness with an amplitude of $\geqslant 0.3\mu_B$ could have been resolved. Models on the magnetic structure of the Fe films in which the magnetic moments are high ($2.2$–$3.0\mu_B$) at the interfaces, the magnetization direction is ''pinned'' at the Fe-Co interface and the surface layers are coupled to the Fe-Co interface layers by layerwise antiferromagnetically aligned layers in the interior, lead to oscillations in the average moment with an amplitude of $\geqslant 1\mu_B$. Simple models of this kind can be excluded by our MCXD results. Also this finding is in agreement with our spin- and angular-resolved photoemission investigation[27] which reveals that the overall spin polarization of the photoelectrons originating from the surface region of the Fe films does not change sign as a function of the Fe thickness in region II at 110 K.

For Fe/Co(100) the ratio of the orbital moment to the spin moment is between 0.06 and 0.07 and does not change significantly from region I to region II. This value is in excellent agreement with the result of a first-principles calculation in Ref. 28.

In the application of the spin sum rule the existence of the magnetic dipole term has been ignored. Therefore the values for the spin moment include the contribution from the magnetic dipole term which is expected to become important for atoms in a noncubic environment, such as at surfaces[29] and in the tetragonally distorted structural Fe phase in region I. In principle it is possible to determine the pure spin moment without magnetic dipole contribution by averaging over measurements with magnetization along the three Cartesian axes.[30] This could not be realized in our experiment because the magnetic field provided by the coils designed to remanently magnetize the sample along the easy axis by a short current pulse is not sufficient to magnetically saturate the sample along all three Cartesian axes during the absorption measurements.

Interestingly, also the $L_3$-$L_2$ intensity ratio in the isotropic spectra changes from region I to II (Fig. 2). For Fe/Co(100) the isotropic $L_3$-$L_2$ intensity ratio defined in the inset of Fig. 5 as $a/b$ is plotted versus the Fe overlayer thickness in Fig. 5. It decreases from 2.95 to 2.50 between 3 and 6 ML Fe, stays constant between 6 and 10 ML Fe and increases again above 10 ML Fe. According to the theory of the atomic branching ratio [the branching ratio in our notation is defined as $a/(a+b)$, see Fig. 5] presented in Ref. 31 the $L_3$-$L_2$ intensity ratio in the isotropic spectra depends on both the $3d$ spin-orbit splitting and electrostatic interactions between the core hole produced in the x-ray-absorption event and the valence electrons. Furthermore, it was found[31] that high-spin states have on average a larger branching ratio than low-spin states, if the spin-orbit splitting in the valence band can be neglected. These findings are consistent with the dependence of $a/b$ on the Fe thickness (Fig. 5) observed in our MCXD experiment.

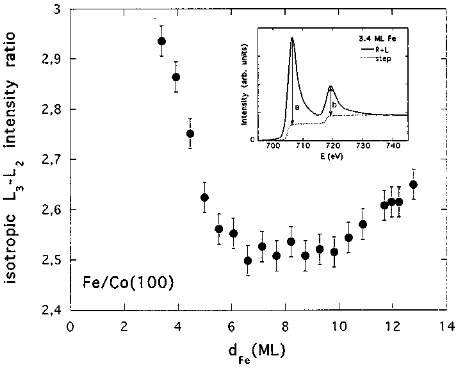

FIG. 5. Isotropic $L_3$-$L_2$ intensity ratio $a/b$ of Fe/Co(100) at a temperature of 110 K as a function of the Fe thickness. Inset: Definition of $a$ and $b$. The $L_3$-$L_2$ step height ratio is 2:1.

In summary, the magnetism of ultrathin fcc Fe overlayers on Cu(100) and Co(100) deposited at room temperature has been investigated element specifically at 110 K using magnetic circular dichroism in x-ray-absorption spectroscopy. Three regions of different magnetic behavior in dependence of the Fe thickness which have previously been found by use of the magneto-optical Kerr effect are qualitatively confirmed. Element- and shell-specific magnetic spin and orbital moments have been derived by use of sum rules from the self-absorption corrected MCXD spectra. Up to 4 ML Fe the average Fe spin moment is $3.0\mu_B$ on Co(100) and $2.8\mu_B$ on Cu(100) demonstrating that the Fe films are in a ferromagnetically ordered high-spin state. Between 5 and 10 ML Fe the average Fe spin moment is $1.1\mu_B$ on Co(100) and $0.8\mu_B$ on Cu(100). In this thickness regime the surface region is ferromagnetically ordered on both substrates. On Co additionally the Fe layer at the Fe-Co interface is ferromagnetically ordered resulting in the larger Fe spin moment on Co in region II. Indications for simple layerwise antiferromagnetism in the interior of the Fe films on Co(100) with pinning of the magnetization direction at the Fe-Co interface are not observed.

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