Direct observation of Cr spin polarization in epitaxial Cr/Co/Cr(100) trilayers

F. Brüssing, M. Ewerlin, R. Abrudan, and H. Zabel
Department of Physics, Ruhr-University Bochum, 44780 Bochum, Germany

(Received 11 June 2013; revised manuscript received 10 August 2013; published 24 September 2013)

The spin polarization of chromium at the interface next to a ferromagnetic layer is of general interest because of the competing ferromagnetic and antiferromagnetic exchange coupling. While the Fe/Cr interface has been well studied, information on the Co/Cr interface still remains scarce. Here we show for epitaxially grown Cr/Co/Cr(100) trilayers with smooth interfaces x-ray resonant magnetic scattering (XRMS) results in a saturation field of ±270 mT, recorded at the Co and Cr L3 edges, respectively. The XRMS results at the Co edge show the expected asymmetry and a ferromagnetic hysteresis for different incident angles θ. Furthermore, XRMS measurements with the energy tuned to the Cr L3 edge also exhibit an asymmetry, albeit much smaller than the one at the Co L3 edge. Moreover, the magnetic hysteresis of Cr taken at the L3 edge has a sign opposite to that of Co at the L3 edge over a broad range of incident angles. From these results we infer first that at the Co/Cr interface chromium is ferromagnetically polarized, and second that its spin structure is oriented opposite to the magnetization of Co.

DOI: 10.1103/PhysRevB.88.094431

PACS number(s): 75.25.—j, 75.70.Ak, 75.70.Cn

I. INTRODUCTION

The spin structure at the Fe/Cr interface has been well studied in the past because of its model character as a ferro-/antiferromagnetic (F/AF) heterostructure,1–3 its original importance for the discovery of the GMR effect,4,5 and its importance for spintronic devices.6 Fe and Cr share the same lattice symmetry and almost the same lattice parameter. In the [100] growth direction Cr on Fe(100) first develops an atomic layerwise commensurate antiferromagnetic structure up to about 24 monolayers (MLs), after which it transforms to an incommensurate spin density wave (SDW) state. In Fe/Cr/Fe(100) trilayers and in Fe/Cr(001) superlattices a short period of two ML interlayer exchange coupling is observed,7,8 and in addition a longer period comprising 12 MLs (±1.73 nm).9 Due to some intermixing at the interface, the 2 ML AF oscillation starts with some delay of about 2–3 MLs. All this has beautifully been demonstrated for Cr(001) grown on Fe whiskers and by imaging the domain structure via scanning electron microscopy with polarization analysis (SEMPA).7 X-ray magnetic circular dichroism (XMCD) experiments at the resonant Cr L3,2 edges showed that thin Cr films exhibit a ferromagnetic polarization with a magnetization direction opposite to the Fe magnetic moments.10–13 These results confirm theoretical predictions of a ferromagnetic state of Cr at the Fe/Cr interface and an antiparallel spin alignment, but are off as concerns the magnitude of the predicted Cr magnetic moment,14,15 which usually is overestimated by a factor of two to three as compared to experimental findings, which settle around 0.5–0.7 μB. Roughness at the top and bottom interface as well as interdiffusion may be invoked to explain this discrepancy.

The Co/Cr(100) interface is by far less well explored than the Fe/Cr(100) interface. This is partly due to its complexity as Co and Cr have different lattice symmetries and the lattice mismatch is considerable. Nevertheless it is worth investigating the spin structure at this interface as well and to compare it with the Fe/Cr interface. At the Co/Cr interface a reduction of the magnetic moment14,15 and the saturation magnetization was often observed,18 which appears to be directly related to the number of covering Cr monolayers,16 similar to the observation made in the case of Fe/Cr superlattices.17 A possible explanation for this phenomenon is a Cr magnetic moment at the interface that is polarized oppositely to the Co moment.17 In Co/Cr MLs it was found that the interlayer exchange coupling mediates antiferromagnetic coupling between the Co layers for thin Cr layers and ferromagnetic coupling for thicker Cr layers.19 The RKKY-like oscillatory interlayer exchange coupling has a period of 2.1 nm,19,20 but a short two ML period as in Fe/Cr(001) superlattices was never identified. This is in strong contrast to the behavior of the Fe/Cr systems, where the coupling properties are reversed19 and the long period oscillation (1.73 nm) is shorter by 0.4 nm. XMCD data on the polarization of Cr in Co/Cr heterostructures do not exist to the best of our knowledge. Here we present a detailed structural investigation of the heteroepitaxial Cr/Co/Cr(100) trilayer performed with synchrotron source hard x rays. In addition, we report our results on the magnetic properties as determined via soft x-ray resonant magnetic scattering and spectroscopy to scrutinize the spin polarization of Cr at the Co/Cr interface.

II. SAMPLE DESIGN AND GROWTH PARAMETERS

In order to explore the Co/Cr interface and possible spin polarization of the Cr layer next to Co we have prepared two trilayer Cr/Co/Cr(100) heterostructures via molecular beam epitaxy (MBE) on a MgO(100) single crystalline substrate. The layer sequence of the samples is schematically shown in Fig. 1. For good epitaxial layer-by-layer growth an about 60 nm thick Cr(100) buffer layer was deposited on the MgO(100) substrate at a substrate temperature of 450 °C. The crystallinity of the Cr buffer layer was significantly improved by post-growth annealing at a temperature of 750 °C for 5 min. For the subsequent growth of the Co and Cr layers the substrate temperature was lowered to 300 °C as a compromise between good epitaxial growth and suppression of interdiffusion between the layers. The pseudomorphic growth of Co on Cr(100) is rather complex due to the different crystal structures (bcc for Cr and hcp for Co) and the lattice mismatch.
FIG. 1. (Color online) Layer sequence of epitaxial Cr/Co/Cr(100) trilayers grown by molecular beam epitaxy on MgO(100) substrates.

The growth properties are detailed in Ref. 21. In short, the Co growth starts with a (11\text{2}0) orientation on Cr(100), where the Co[0001] axis is parallel to the Cr[01\text{1}] axis. This results in a pseudomorphic bcc-like growth of Co on Cr with a 45° epitaxy and lattice misfits of 0.07% parallel to Co[0001] and 6.4% parallel to Co[11\text{00}]. Because of the two equivalent Cr[01\text{1}] in-plane axes, different Co(11\text{2}0) domains form with an angle of 90° with respect to each other.\textsuperscript{22} In addition to this a bcc-hcp martensitic transition takes place as a function of Co thickness within a thickness range up to 4.0 nm.\textsuperscript{21-23} The top Cr layer thickness is chosen such that it protects the sample from oxidation. At the Cr surface a stable oxide forms, which does not grow in time.\textsuperscript{24} Further details of the sample growth conditions are provided in Ref. 25.

Two samples I and II were grown, and the corresponding parameters of the samples are listed in Table I. Sample I has a total Co thickness of 5.8 nm and a top Cr thickness of 2.9 nm. For sample II the thicknesses are slightly different: 10 nm and 5.6 nm for the Co and Cr thicknesses, respectively. In sample II the Co martensitic transition is more complete and thus it is expected to have a weaker effect on the magnetization reversal than in the case of the thinner Co film. We have avoided purposely the Co thickness regime below 2 nm where in Co/Cr(100) superlattices a perpendicular anisotropy is observed.\textsuperscript{26}

III. STRUCTURAL CHARACTERIZATION

Layer thicknesses, interface quality, and epitaxial relationship between Co and Cr were investigated via hard x-ray diffraction (XRD) and x-ray reflectivity (XRR) measurements. For these investigations the beamline W1.1 of the synchrotron source DORIS at the HaSyLab was used with an incident energy of 10.5 keV (0.118 nm). The resulting reflectivity curves are presented in Fig. 2. Fits of these data using the standard modified Parratt formalism\textsuperscript{27} reveal that both samples have comparable layer quality, in particular the roughness of the buffer layer is similar in both cases. A splitting of the Co layer thickness in two parts increases the fit quality due to a better description of the martensitic transition of the Co layer. The first part of the Co layer mainly describes this transition and the second part represents the relaxed hcp-Co structure. The final roughness parameters of sample I (Co thickness 5.8 nm) and sample II (Co thickness 10.0 nm) are similar.

We have also investigated the crystalline structure and quality of the samples with out-of-plane Bragg scans parallel to the surface normal (Cr and MgO [100] direction), see Fig. 3.\textsuperscript{28} The results indicate an excellent crystal quality by the appearance of the sharp Cr(200) and Co(11\text{2}0) peaks. Additionally, Laue oscillations are visible on either side of the Cr(200) peak (see inset of Fig. 3), confirming the high structural order of the buffer layers. The intensity of these peaks is much higher than that of the Co(11\text{2}0) peaks, which is due to the difference in thickness of the two layers. The difference of the Co peaks in respect to height, width,

![TABLE I. Layer thickness (t) and interface roughness (σ) of the used samples according to the fits of the x-ray reflectivity measurements plotted in Fig. 2. d_{Cr}\text{sum} is the sum of the bcc- and hcp-like Co thicknesses.](image)

<table>
<thead>
<tr>
<th>Layer</th>
<th>(t) [nm]</th>
<th>(σ) [nm]</th>
<th>(t) [nm]</th>
<th>(σ) [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>(d_{Cr}\text{buf})</td>
<td>74.4 ± 2.0</td>
<td>0.2 ± 0.2</td>
<td>56.3 ± 0.2</td>
<td>0.2 ± 0.1</td>
</tr>
<tr>
<td>(d_{Cr}\text{sum})</td>
<td>5.8 ± 0.4</td>
<td>1.7 ± 0.4</td>
<td>10.0 ± 1.0</td>
<td>2.0 ± 0.5</td>
</tr>
<tr>
<td>(d_{Cr}\text{top})</td>
<td>2.9 ± 0.6</td>
<td>1.2 ± 0.5</td>
<td>5.6 ± 0.9</td>
<td>1.1 ± 0.5</td>
</tr>
<tr>
<td>(d_{Cr}\text{ox})</td>
<td>3.0 ± 0.6</td>
<td>1.7 ± 0.4</td>
<td>3.0 ± 0.7</td>
<td>1.7 ± 0.4</td>
</tr>
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![FIG. 3. (Color online) X-ray scan in the out-of-plane direction parallel to the surface normal and to the [100] direction of MgO for samples I and II. The inset shows an enlargement of the Cr(200) peak with Laue finite thickness oscillations on both sides.](image)
and position of the two samples is a result of the different thicknesses and influenced by the martensitic transition of the Co layer.

Furthermore we have investigated the in-plane structural correlation of the different layers via surface x-ray scattering techniques at glancing incident and exit angles. For this purpose the incident and exit angles were kept constant slightly above the critical angle for total external reflection from the surface. Additionally, a line detector oriented parallel to the sample surface simultaneously measures the intensity for different in-plane Bragg peaks when the sample is rotated about its normal. To reveal the epitaxial relation of the layers the intensity distribution as a function of sample rotation by 360° is plotted in Fig. 4. The positions of the MgO(002), Cr(011), Co(1100), and Co(11001) peaks are marked, revealing the expected 45° epitaxial relation between Cr and MgO due to the coexistence of the Cr(011) and the MgO(002) peaks for one sample orientation. The pseudomorphic growth of Co on Cr with the two different Co(1120) domains, forming an angle of 90° with respect to each other, is revealed by the eight Co(T101) peaks appearing in the intensity map upon a complete rotation of the sample, see Fig. 4. The structural results completely agree with the earlier measurements reported by Metoki et al. and Donner et al. and in particular, confirm the martensitic phase bcc-hcp structural phase transition as a function of Co thickness.

IV. MAGNETIC POLARIZATION OF Cr

We have explored the magnetic polarization of Cr next to Co using x-ray resonant magnetic scattering (XRMS). This method is interface sensitive and allows us to analyze element selectively the magnetic properties of heterostructures, such as the magnetization profile across interfaces and the polarization at interfaces. For the more general magnetic properties of the trilayer including magnetic hysteresis measurements we refer to our earlier study reported in Ref. 25.

The XRMS measurements were performed with the ALICE chamber, using the synchrotron source BESSY II of the Helmholtz Zentrum Berlin. Reflectivity scans with the energy tuned to the Co and Cr L₃ edges were taken as a function of the scattering vector $q_z$,

$$ q_z = \frac{4\pi}{\lambda} \sin(\theta), $$

and are shown in Fig. 5 for the samples I and II, respectively. The reflectivity scans were carried out in a saturation field of $H_{sat} = \pm 270$ mT. The difference in the reflected intensity is due to the ferromagnetic state of the sample. The difference in signal for $\pm H_{sat}$ becomes more pronounced by taking the asymmetry

$$ \frac{I + H_{sat} - I - H_{sat}}{I + H_{sat} + I - H_{sat}}, $$

which is plotted in the bottom panels of Fig. 5. The asymmetry of Co is clearly visible up to $q_z = 1.25 \text{ nm}^{-1}$; the asymmetry of Cr is much weaker. To enhance the visibility of the asymmetry and for better comparison within the same panel, both asymmetries for Co and for Cr are color coded: red for Co and black for Cr in the upper and lower panel. The
stripe pattern in the lower panel represents the sign of the asymmetry. Furthermore, the asymmetry of Cr is scaled by a factor of 20 to improve its visibility. We recognize that over a large $q_z$ range the asymmetry of Co and Cr have opposite signs. For sample II the asymmetry for Cr is generally smaller than for sample I. Unlike in XMCD experiments, in XRMS measurements the sign of the asymmetry may change as a function of $q_z$, due to the interference between the charge and spin part of the structure factor. However, it is important to notice that over the entire $q_z$ range scanned, the asymmetries of Co and Cr are mainly opposite to each other, yielding strong evidence for an antiparallel spin alignment.

To follow up on the relative sign of the Co and Cr asymmetries we have carried out energy scans for a fixed incident beam angle of $\theta = 15^\circ$ and measuring the reflected intensity as a function of energy in the range of the Co and Cr $L_3$ and $L_2$ edges. The incident beam angle is typical for XMCD experiments in absorption, fulfilling the selection rule for magnetic sensitivity. In reflection geometry, however, the intensity detected is folded with the interference from the charge and spin part of the form factor, as is expressed in XRMS and already mentioned before. By changing the energy, $q_z$ also changes:

$$q_z = \frac{4\pi E}{hc} \sin(\theta),$$

from $q_z \approx 0.14 - 0.21$ nm$^{-1}$ for energies between 550 and 810 eV at an incident angle of 15$^\circ$, respectively, covering the energy range of the Co and Cr $L_2,3$ edges. The energy scans are shown in Fig. 6 at the Cr (left) and Co (right) $L_3$ and $L_2$ edges of sample I, respectively. In the top panel right circular polarized light ($\sigma_+$) was used and the energy scan was performed in magnetic saturation fields of $H_{sat} = \pm 270$ mT at 300 K. Again, for a better visibility of the intensity difference, the asymmetries are plotted in the lower panels of Fig. 6 (black solid squares). We first notice that the asymmetry at the Co $L_3$ and $L_2$ edges are equivalent to the respective XMCD signals of Co.\textsuperscript{29,36} The asymmetry of Cr is much weaker than for Co, but still clearly visible at the Cr $L_2$ edge, whereas it is barely noticeable and within the noise level at the Cr $L_3$ edge. More important is the fact that the asymmetries of Co and Cr have opposite sign. This is a clear indication of a Cr local moment that is aligned antiparallel to the Co moment at the Co/Cr interface.

We have confirmed the opposite asymmetry for Co and Cr by measurements at 80 K (black open squares). For plotting the results within the same panel, we used left circular polarized light ($\sigma_-$) and performed again the same measurements in magnetic saturation fields of $H_{sat} = \pm 270$ mT. The results for Co show a stronger asymmetry as expected from the increasing order parameter of Co and also a clear asymmetry for the Cr $L_3$ edge, in sign opposite to the Co $L_3$ edge.

The inset in Fig. 6 shows magnetic hysteresis curves taken at the Co and Cr $L_3$ edges and at a sample temperature of 300 K. These hysteresis curves have the same shape, coercive field, and squareness, but opposite signs, and confirm again the antiparallel alignment of the Co and Cr moments at the Co/Cr interface.

Similar experiments as detailed above were performed for sample II, but at an incident angle of $\theta = 8.5^\circ$ and low temperature measurements at 50 K. The results are shown in Fig. 7. Because of the larger sample thickness, interference effects are here more pronounced than for sample I. Nevertheless, the opposite signs of the asymmetry for Co and Cr at 300 K as well as 50 K are confirmed. The corresponding hysteresis measurements shown as an inset in Fig. 7 are indeed more noisy, but overall agree with the results from sample I. The smaller overall Cr signal in sample II as compared to sample I can be related to the difference in the depth profile and to a smoother Co/Cr interface, which may result in a reduced polarization of the Cr layer, because less Cr atoms are in direct contact with Co atoms. The magnetic hysteresis measurements of this sample shown in the upper right hand panel of Fig. 7 agree with those of sample I: The shapes of the Co and Cr hysteresis curves are the same but their signs are opposite.

Simultaneously with the energy scans in reflection mode we have also recorded the total electron yield signal in order to determine the XMCD signal from our samples. However, the XMCD signals in particular for Cr turned out to be even weaker than the asymmetry in reflection mode, yielding inconclusive results. This shows that the reflection geometry in some cases can be more sensitive to weak magnetic signals than the absorption geometry.

![FIG. 6. (Color online) Top panel: Energy scans in the region of the Cr $L_{2,3}$ edges and Co $L_{2,3}$ edges taken in reflection geometry at 300 K in a saturation field of $\pm 270$ mT. Bottom panel: Asymmetries for scans at 300 K and 80 K of sample I. The asymmetries for 300 K and 80 K are plotted with opposite signs as to make their differences better visible. Inset: Hysteresis loops of sample I recorded at the Co and Cr $L_3$ edges.](094431-4)
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V. SUMMARY AND CONCLUSION

We have investigated the crystal structure and epitaxial relationship of the heteroepitaxial trilayer Cr/Co/Cr(100) by hard x-ray scattering using a synchrotron based diffractometer in combination with a linear detector. In the out-of-plane and in-plane directions we could confirm an excellent pseudomorph growth of Co on Cr(100) with the Co [0001] axis parallel to the Cr[011], forming two Co(1120) structural domains. While the lower Cr/Co interface is well defined with a low roughness, the top Co/Cr interface is much more rough. The roughness is most likely caused by the Co-domain formation and the martensitic bcc-hcp transformation with increasing Co thickness. This is most likely the reason why a two monolayer oscillation of the interlayer exchange coupling was never observed in the case of Co/Cr(100), in contrast to the Fe/Cr(100) system. Using this heteroepitaxial Cr/Co/Cr trilayer, we have investigated the Cr polarization at the Co/Cr interfaces by means of soft x-ray resonant magnetic scattering. The asymmetries determined at the Co and Cr L3 edges and the element specific hysteresis loops undoubtedly reveal a weak but clear and temperature independent polarization at the Cr L3 edge which is opposite to the Co magnetization direction. Two different Cr/Co/Cr trilayers were investigated with different layer thicknesses and interface roughnesses. However, the overall results concerning the polarization of Cr at the Co/Cr interface and its sign are equivalent and confirm an antiparallel alignment of the Co and Cr magnetic moments. X-ray circular magnetic dichroism in absorption geometry via total electron yield detection did not yield conclusive results.

ACKNOWLEDGMENTS

We gratefully acknowledge financial support by the DFG through SFB 491. Furthermore we would like to thank Jürgen Podschwadek for technical support of the sample preparation and Wolfgang Caliebe and Oliver Seeck for their help at the Beamline W1.1 of the HaSyLab. The soft x-ray magnetic scattering was performed with the ALICE chamber at BESSY II of the HZB, which is funded through BMBF 05K10PC2.


FIG. 7. (Color online) Top panel: Energy scans for sample II in the region of the Cr L2,3 edges and Co L2,3 edges taken in reflection geometry at 300 K in a saturation field of 270 mT. Bottom panel: Asymmetries for scans at 300 K and 50 K of sample II. The asymmetries for 300 K and 50 K are plotted with opposite signs as to make their differences better visible. Inset: Hysteresis loops of sample II at the Co and Cr L3 edges.
27. For fitting x-ray reflectivity data, see http://panalytical-x-pert-reflectivity.software.informer.com.
28. The coordination system is set by keeping the z axis parallel to the in-plane Co c axis.