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# A PNR study of the off-specular scattering during the asymmetric magnetization reversal in an exchange-biased Co/CoO multilayer

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**Abstract**

We report on the observation of the effects of exchange bias on the magnetization reversal processes in a [Co/CoO/Au]$_{20}$ system using polarized neutron reflectometry (PNR). The focus in this study is the investigation of the off-specular scattering of neutrons from magnetic domain structures during the magnetization reversal. In a previous PNR study on the same system, an asymmetry in magnetization reversal has been observed on opposite sides of the same hysteresis loop. For the decreasing field branch, the reversal was found to be dominated by domain wall motion of domains directed parallel or antiparallel to the applied field. In contrast, the reversal on the increasing field branch was characterized by rotation of magnetization. A significant loss of intensity was found for the specular reflected neutrons, while off-specular scattering experiments reveal that this magnetization reversal is not determined by coherent rotation but rather by a breaking up into smaller domains with different orientations.


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*Keywords:* Exchange bias; Polarized neutron reflectometry; Off-specular scattering

## 1. Introduction

The properties of thin ferromagnetic (FM)/antiferromagnetic (AFM) multilayer (ML) systems have been the subject of many studies in recent years. After field cooling FM/AFM systems below the Néel temperature $T_N$ of the AFM material, the

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magnetic hysteresis loop was found to be shifted with respect to the zero-field axis. This induced unidirectional anisotropy is called exchange bias (EB). In the EB state [1,2], one finds favorable conditions to observe correlated phenomena such as asymmetric reversal processes on opposite sides of the hysteresis loop (e.g. Ref. [3]). Polarized neutron reflectometry (PNR) has proved to be a powerful technique to study magnetization reversal processes in magnetic thin films. The present study focuses on off-specular ($\Theta_i \neq \Theta_f$) PNR measurements during the magnetization reversal in a [Co/CoO/Au] EB-ML. The experiments were performed in the EB state at 10 K, i.e. after field cooling the sample from the saturation state below $T_N$ of CoO. In a previous study, asymmetric magnetization reversals have been observed using specular PNR with spin analysis [4]. Off-specular scattering arises from structural and magnetic inhomogeneities. Therefore this type of experiment potentially provides information about magnetic domains. This has also been demonstrated recently in experiments on patterned samples with a dot size of 4 μm [5,6].

## 2. Experimental procedures

A $[Co(16.4\,nm)/CoO(2\,nm)/Au(3.4\,nm)]_{20}$ ML was prepared on an $Al_2O_3(0\,0\,0\,1)$ substrate by molecular beam epitaxy at a growth temperature of 300 K [4]. The base pressure of the ultrahigh vacuum chamber was $10^{-10}$ mbar. The CoO/Co bilayers are separated by Au spacer layers to avoid magnetic interaction between neighboring exchange-biased AFM/FM pairs. The purity of the substrate and the films was checked by Auger electron spectroscopy, while X-ray measurements revealed an FCC(1 1 1) surface orientation of Co. CoO layers of 2 nm thickness were obtained by an in situ oxidation method using a controlled exposure of high-purity oxygen gas [7,8]. STM measurements on as grown Co and CoO layers deposited on an $Al_2O_3$ substrate showed that the sample consists of granular Co and CoO layers with roundly or elliptically shaped grains with diameters of about 10–20 nm. Magnetic properties of the ML were characterized using a SQUID magnetometer on an as grown sample. The PNR experiments were performed at the reflectometer V6 at the Hahn-Meitner-Institut, Berlin with a neutron wavelength $\lambda = 0.466$ nm [9]. Off-specular scattering PNR experiments were carried out at 10 K using a two-dimensional position sensitive detector (PSD) after field cooling the sample in a field of 400 mT.

## 3. Results

In the biased state at $T = 10$ K, the hysteresis loop of the Co/CoO ML is considerably shifted away from zero field (EB field $H_E = -393$ Oe, see insets in Fig. 1) [4].

In Fig. 1, three ($\Theta_i$, $\Theta_f$) intensity maps, recorded at three different characteristic magnetic fields—for magnetic saturation (A, Fig. 1 top) and close to the coercive fields $H_{CA}$, at $-883$ Oe (B Fig. 2 middle), and $H_{CP}$, at $+112$ Oe (C, Fig. 1 bottom)—are shown for the case of polarized incoming (−) neutrons. For clarity, solid circles in the embedded SQUID curves in Fig. 1 mark the position in the hysteresis, where the neutron data were recorded. The intensity is plotted as a function of the incident and scattered angles $\Theta_i$ and $\Theta_f$ using a color scheme from white for the background noise to black for the maximum intensity of total reflection. The intensity maps in the selected $\Theta$ range feature the first- and second-order Bragg peak. Below each polarized intensity map the line scans of the specularly reflected intensity are plotted for incoming (−), full circles, and (+) neutrons, open circles. Solid lines are fits to the NSF reflectivity using a simulation program, which is based on the Parratt formalism [10]. The specularly reflected intensity profiles (specular ridge, $\Theta_i = \Theta_f$) are indicated in the intensity maps as dashed lines. It should be noted, that here no spin analysis was performed, i.e. $R^- = R^{--} + R^{-+}$ and $R^+ = R^{++} + R^{+-}$ are measured. Therefore the present off-specular intensity maps are not sensitive to in-plane rotations of the magnetization. Apart from the ridge at $\Theta_i = \Theta_f$, the most prominent non-specular features in Fig. 1 (A) are diffuse streaks extending perpendicular to the specular ridge and crossing the latter at the

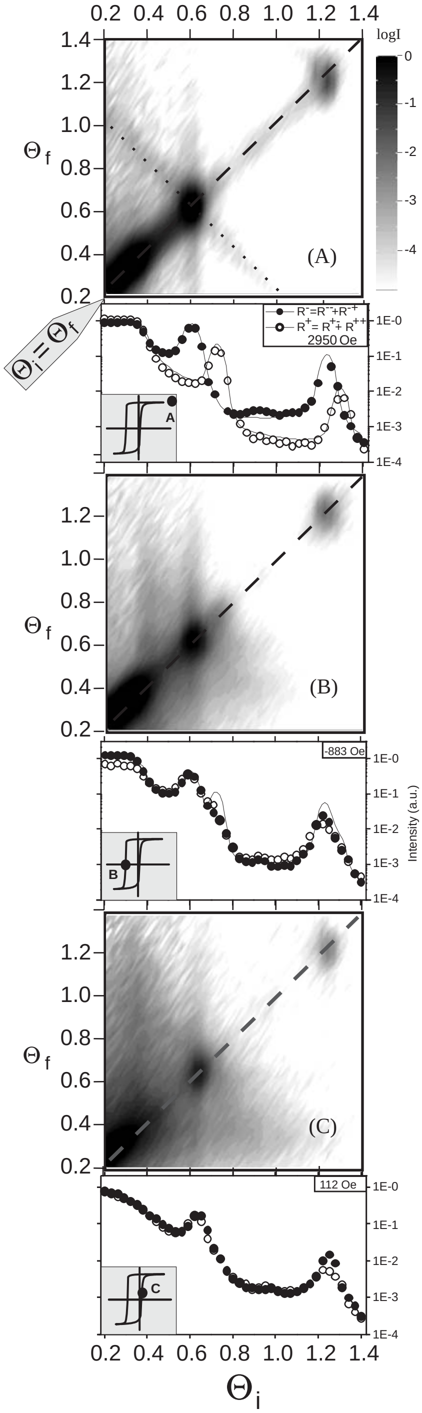


Fig. 1. Intensity maps (for incoming (–) neutrons) recorded at different characteristic fields. Top: saturation; middle: $H = -883$ Oe close to $H_{CA}$; $H = 112$ Oe close to $H_{CP}$. The dashed lines indicate the specularly reflected intensity, which is plotted below each map as a function of the incident angle.

Bragg peak positions (dotted line in Fig. 1, top). The existence of the streaks implies a coherent interference of the scattered neutrons and can be attributed to correlated or conformal roughness (see also Refs. [11–13]), i.e. inhomogeneities of adjacent layers are replicated to some extent from one interface to the next, exhibiting the same vertical periodicity as the ML. This experimental finding is consistent with TEM images of the present ML revealing a waviness of the trilayers which is transmitted from the bottom to the top of the ML.

Furthermore, additional diffuse streaks appear at $\Theta_i = \Theta_1$ and $\Theta_i = \Theta_2$, where $\Theta_1, \Theta_2$ are the angles of the first- and second-order Bragg peaks. A similar ridge of diffuse intensity has been reported for $\Theta_i = \Theta_1$ on a W/C ML using X-ray scattering [12]. Its origin was attributed to a mechanism in which non-specular $q_x = 2\pi/\lambda^*(\cos\Theta_f - \cos\Theta_i)$ components couple the first- and second-order Bragg conditions. At $\Theta_i = \Theta_C$, i.e. if the incident angle equals the critical angle $\Theta_C$ of total reflection, a ridge of weakly increased intensity can be observed, which is due to Yoneda scattering [11,14]. None of these features would be observed for a ML structure with perfectly homogeneous layer planes.

However, without rigorous analysis it is impossible to obtain quantitative values of structural or magnetic attributes of the lateral inhomogeneities in this ML system. But at least some rough estimation can be made using the concept that a single-domain state would cause no diffuse magnetic scattering. Therefore the diffuse scattering features at saturation, shown in Fig. 1 (A), are assumed to originate from structural inhomogeneities.

In a previous study, the magnetization reversal at $H_{CA}$ was found to be dominated by the growth of domains with magnetization either parallel or antiparallel to the applied field using specular PNR with spin analysis [4]. In comparison with the case of magnetic saturation, there is an increased off-specular scattered intensity at $H_{CA}$ in a region of reciprocal space close to the specular rod and close to the first-order ML Bragg peak. The vertical diffuse streak intensities, observed at $\Theta_i = \Theta_1$, as well as at $\Theta_i = \Theta_C$ (Yoneda scattering),

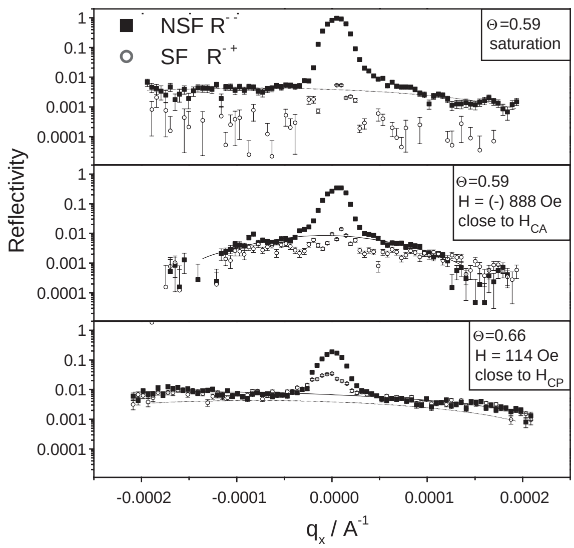


Fig. 2. Rocking scans with spin analysis at magnetic saturation (top), at $H = -888$ Oe close to $H_{CA}$ (middle) and at 114 Oe close to $H_{CP}$. Full square symbols represent NSF-reflectivities, open symbols SF reflectivities. Solid and dashed lines which schematically indicate background and diffuse intensities are guides to the eyes only.

are also enhanced at $H_{CA}$. This indicates that relatively large domains are present during this reversal process. This statement was further supported performing rocking scans with spin analysis, which are shown in Fig. 2. The solid squares correspond to NSF intensity, the open circles correspond to SF intensity. Assuming that the width of the distributions in $q_x$ is inversely proportional to a characteristic dimension in the $x-y$ plane we can get a rough estimate of an average size of the lateral inhomogeneities. However, for incident angles $\Theta_i$ between 0.59 and 0.7, the coherence length of the incident beam, which is a function of the divergence of the beam and the uncertainty in the neutron wavelength, is about 30 to 40 μm. Therefore we are not sensitive to in-plane inhomogeneities larger than 30 μm. For magnetic saturation, $2\pi/(\Delta q_x)$, with $\Delta q_x$ the FWHM, corresponds to lateral structures of about 30 μm or larger. Therefore the width of the peak at magnetic saturation is basically determined by the coherence of the beam.

At magnetic field values close to the coercive field $H_{CA}$, the NSF-intensity for (−) neutrons of the first-order Bragg peak is reduced by a factor of 2–3 with respect to the corresponding peak at magnetic saturation. The FWHM of the corresponding peak is not larger than for magnetic saturation. Furthermore at $H_{CA}$ both NSF ($I^{++}$ and $I^{--}$) intensities are about equal and about 50 times larger than the SF ($I^{-+}$ and $I^{+-}$) intensities in the specular regime around $q_x = 0$, confirming that large parts of the sample are aligned either parallel or antiparallel to the applied field.

Weakly enhanced diffuse scattering extending to $q_x$ values of about $\pm 0.0001\,\text{Å}^{-1}$ can be observed more or less pronounced for all four cross sections. The width of the enhanced diffuse intensity distribution is of the order of $\Delta q_x \approx 0.0001\,\text{Å}^{-1}$, which would correspond to lateral sizes of about 6 μm. This indicates the presence of some smaller domains in addition to the predominantly large domains (> 30 μm), which basically give rise to specular scattering.

In contrast, the diffuse scattering at $H = 112$ Oe close to $H_{CP}$ (see Fig. 1 (C)) extends far into the off-specular region, implying the existence of smaller domains. At the same time the specular intensity is considerably reduced. Specular PNR experiments with spin analysis [4] revealed significant SF-intensities of the same order of magnitude as the NSF-intensities. This suggests that a large part of the magnetization has rotated during the reversal process.

Note that the Bragg peak position is different for $H_{CA}$ (for saturation, respectively) and $H_{CP}$ due to the fact that the reflectivity curve looks different for small and large domains (see $\Theta$–$2\Theta$ scans in Fig. 1). For more details please see Ref. [4].

Rocking scans recorded at $H = 114$ Oe close to $H_{CP}$ (see Fig. 2 bottom) show significantly enhanced diffuse scattering extending to $|q_x| = 0.0002\,\text{Å}^{-1}$.

The off-specular intensity at that field is equally distributed to NSF and SF reflections (ratio NSF:SF—1:1) and extends to the borders of the recorded $q_x$ range. Apart from the slightly enhanced diffuse scattering between $q_x = \pm 0.00015$ and $q_x = \pm 0.00016$, which arises due to Yoneda scattering, no distinct features in the off-specular scattering regime were observed. Within the accessible $q_x$ range, we are not sensitive to lateral sizes smaller than $\approx 1\,\mu m$. These results suggest the existence of a distribution of small domains of different magnetic orientation during the reversal at $H_{CP}$.

This result agrees with the finding of a FM domain distribution during the magnetization reversal in a thin Co/CoO bilayer [15], where a model-free way was used to determine the mean square dispersion of the magnetization vectors of lateral domains.

## 4. Conclusions

Off-specular PNR experiments on a $[Co/CoO/Au]_{20}$ EB ML at fields close to the coercive fields were presented for the biased state at $T = 10\,K$. A rough estimation on the relative domain size is given here. For magnetic fields close to $H_{CP,}$ diffuse scattering experiments indicate the existence of a distribution of smaller domains ($<1\,\mu m$) with statistically distributed magnetic in-plane orientations. During the magnetization reversal dominated by domain wall motion ($H \approx H_{CA}$), the relatively small amount of off-specular scattering observed indicates the presence of a few domains with domain sizes of the order of a few μm, while most of the reflected intensity arises due to specular scattering from large domains ($>30\,\mu m$) aligned parallel or antiparallel to the field. The results on the present ML provide further details on the mechanisms during the asymmetric magnetization reversal in an EB ML consisting of granular FM and ultrathin granular AFM layers.

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