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Layer magnetization evolution in an Fe/Cr multilayer with uniaxial anisotropy

H.J. Lauter^{a,*}, V. Lauter-Pasyuk^{a,b,c}, B.P. Toperverg^{d,e}, U. Rücker^d, M. Milyaev^f, L. Romashev^f, T. Krinitsyna^f, V. Ustinov^f

> ^a Institut Laue Langevin, BP 156, Grenoble Cedex 9, F-38042, France b Physik Department, TU München, Garching D-85747, Germany Joint Institute for Nuclear Research, 141980 Dubna, Moscow Region, Russia d Forschungszentrum Jülich, IFF, Jülich D-52425, Germany Petersburg Nuclear Physics Institute, 188350 Gatchina, St. Petersburg, Russia ^f Institute of Metal Physics, Ekaterinburg 62019, Russia

Abstract

The direction of the magnetization of each Fe layer in an Fe/Cr multilayer with uniaxial anisotropy was determined with polarized neutron reflectometry. The vectors of the layer magnetization of the multilayer transit from an antiferromagnetic alignment into a nearly ferromagnetic one with increasing magnetic field. In the transition region the system consists of an antiferromagnetically aligned part and a ferromagnetically aligned part. The magnetization curve is characterized by the subsequent switching of the antiferromagnetically aligned bilayers into the nearly ferromagnetically aligned state. Via this mechanism the antiferromagnetic part of the multilayer reduces in favor of the ferromagnetically aligned part with increasing magnetic field.

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A detailed knowledge about the direction and the magnitude of the magnetization of single layers in a multilayer as a function of magnetic field or temperature is still missing and only recently the possibility of layer-by-layer magnetometry in magnetic multilayers was obtained with polarized neutron studies $[1,2]$. In the course of the discussion of the presented results the spin–flop transition [\[3\]](#page-3-0) or spin–flip transition [\[4\]](#page-3-0) arising in varying magnetic fields takes place depending on the ratio of the energy terms in the total energy. The

biquadratic term [\[5\]](#page-3-0) in the antiferromagnetic exchange coupling may play a role in the flip of the layer magnetization. We demonstrate with layer-by-layer magnetometry performed with polarized neutron scattering that the evolution of the layer magnetization structure as a function of the external magnetic field can be studied.

The investigated multilayer with the composition $[\text{Cr}(12.4 \text{ Å})/\text{Fe}(76 \text{ Å})]_{x12}/\text{Cr}(83 \text{ Å})$ was grown on a $MgO(112)$ substrate with molecular beam epitaxy. Its structure is schematically shown in [Fig. 1](#page-1-0) together with the scattering geometry. Here, only specular scattering will be discussed. The detected very small off-specular scattering does not influence the interpretation of the specular

^{*}Corresponding author. Tel.: $+33-476-207-239$; fax: $+33-$ 476-207-120.

E-mail address: lauter@ill.fr (H.J. Lauter).

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Fig. 1. Schematic presentation of the Fe/Cr multilayer sample with scattering geometry. Here, only four Fe layers are shown from the $[Fe/Cr]_{x12}$ structure. The sample has uniaxial in-plane anisotropy.

Fig. 2. Magnetization curve of the Fe/Cr multilayer determined with VSM. The crosses indicate the magnetic field at which neutron reflectometry experiments were performed and what values of the net magnetization were obtained from the reflectivity data. The inset shows the uniaxial behavior of the sample measured at \sim 100 G.

scattering. Prior to the reflectometry measurement the sample was characterized by vibrating sample magnetometry (see Fig. 2). The multilayer has uniaxial in-plane anisotropy as is shown in the inset of Fig. 2 in a measurement for a magnetic field $H \sim 100$ G. The easy axis runs along the 90– 270° orientation in Fig. 2. Systems with uniaxial symmetry have been studied theoretically [\[4\]](#page-3-0) and experimentally [\[2\]](#page-3-0) (see also the publications in Ref. [\[6\]](#page-3-0)). The Fe layers in the multilayer are expected to be antiferromagnetically coupled due to the Cr spacer layer of a thickness of 12.4 Å . The

magnetization curve is depicted in Fig. 2 and no evident antiferromagnetic (AF) behavior is exhibited. The reason for this behavior will be deduced from the analysis of the reflectometry data. Similar looking hysteresis loops are discussed in Ref. [\[7\]](#page-3-0) in view of oscillatory interchange coupling and it is noted that additional magnetocrystalline volume anisotropy may play a role. We will present here neutron reflectivity data taken along the hysteresis loop in increasing field from 20 G up to 700 G.

The sample has been saturated in a negative field (see Fig. 2) and neutron reflectivity data have been taken successively at the magnetic field values indicated in Fig. 2. The corresponding values of the total magnetization obtained from the neutron reflectivity measurements are also shown with these marks. The measurements were performed on HADAS $[8]$ at the FZ Jülich and the data with the model fits are shown in [Fig. 3](#page-2-0). The main features of the reflectivity curves measured with full polarization analysis are the half-order and full-order Bragg peaks at $Q_z \sim 0.038$ and \sim 0.076 Å⁻¹, respectively, being also present in the spin–flip (sf) reflectivity curves. However, in particular, part of the intensity of the Bragg peaks on the sf curves originates from the non-perfect polarization efficiency p of polarizer ($p_p = 0.94$) and analyzer ($p_a = 0.97$) which was taken into account in the fit to the data. In the fit the nuclear scattering length, density and the layer thicknesses were fixed and parameters of the fit were the directions of the magnetization of the Fe layers. The bulk saturation value for the Fe-layer magnetization was assumed. Different starting configurations of the angular distribution of the layer magnetization (LM) were tried and either the fit was not successful or was reaching the configuration of the results shown in [Fig. 4.](#page-3-0)

The LM distribution shown in [Fig. 4a](#page-3-0) for a magnetic field $H = 20$ G after saturation in positive field corresponds in Fig. 2 to the cross on the upper branch of the total magnetization hysteresis curve. Four pairs of Fe layers show AF configuration. A small deviation of the AF LM directions with respect to the outside field is noticed. Two pairs of Fe layers show that the field-antiparallel LM has switched into the field direction although no F alignment is reached. The

Fig. 3. Reflectivity curves (not corrected for polarization efficiency) taken from the Fe/Cr multilayer at various fields. The squares mark the measured $(++)$ and $(--)$ spin configuration, respectively. $(+)$ and $(-)$ denote that neutron spin and magnetic field are parallel or antiparallel, respectively. The spin–flip scattering from the sample (\pm) and (\pm) are represented for (\pm) in the lower curve of each figure. The fit to the data (solid lines) includes the polarization efficiency of the polarizer and the analyzer. The reflectivity curves were measured at the magnetic fields: (a) $H = 20$ G, (b) $H = 14$ G, (c) $H = 450$ G and (d) $H = 680$ G.

underlying feature is that multilayers with an even number of magnetic layers, like for the present sample, are in particular sensitive to the surface spin–flop transition [\[9\]](#page-3-0). Assuming that the LMs are all oriented in an AF alignment, one edge layer is oriented against the magnetic field direction.

This edge layer has only one neighbor with which it has AF exchange coupling. Thus, this edge layer can change its AF configuration more easily with respect to the field than LMs inside the multilayer. So, here in [Fig. 4a](#page-3-0) we see a surface effect due to the even number of layers; however, the surface effect results in a flip and not in a flop of the surface LM. This effect resembles the distortion mechanism theoretically treated in Ref. [\[4\];](#page-3-0) however, the evolution predicted to happen with increasing field is different from the one encountered here. Prediction is that the distortion moves from the surface into the multilayer and broadens its influence to neighboring LMs so that a homogeneous nearly F phase is reached. In our case, rather thick Fe layers have been chosen with an increased net magnetization with respect to thinner Fe layers, so that the Zeeman energy [\[4\]](#page-3-0) is dominating the exchange coupling energy. This argumentation, although needed to be confirmed by calculation, is contained in the description of the LM configurations measured at the other fields.

The LM measured at the magnetic field of 20 G (upper branch in [Fig. 2\)](#page-1-0) is equivalent to the LM at the magnetic field of -20 G (lower branch in [Fig.](#page-1-0) [2\)](#page-1-0). Therefore, the LM shown in [Fig. 4b](#page-3-0) for the magnetic field of 14 G (after negative saturation) is very close to the one in [Fig. 4a.](#page-3-0) The net magnetization of the measurement at 20 and 14 G is nearly the same in modulus but with different signs and agrees with the magnetization measurement in [Fig. 2.](#page-1-0)

The comparison of the LM in [Fig. 4c](#page-3-0) to the LM in [Fig. 4b](#page-3-0) shows that with increasing field the negative net magnetization disappears and that at 90 G already the edge layer is flipped into the direction of the external field. The change in the magnetization between 14 and 90 G corresponds to the flip of two LMs and so in [Fig. 2](#page-1-0) to the rather high step in the magnetization curve. The further increase in magnetic field to 450 G [\(Fig. 4d\)](#page-3-0) and 680 G [\(Fig. 4e](#page-3-0)) leads to the flip of two more LMs and then to the flip of one further LM in agreement with the magnetization measurement in [Fig. 2.](#page-1-0)

It seems that a complete AF state for the multilayer is nowhere stable along the magnetization

Fig. 4. Configuration of the layer magnetizations at five magnetic fields: (a) $H = 20$ G, (b) $H = 14$ G, (c) $H = 90$ G, (d) $H = 450$ G and (e) $H = 680$ G. The presentation of the layer magnetizations with full and broken arrows helps to identify couples of anitiferromagnetically coupled layers and their transition to the ferromagnetic-like alignment.

curve as seen from the configurations of the LMs in Fig. 4 and the magnetization measurement in [Fig. 2.](#page-1-0) However, the measurement of the virgin state could reveal this pure AF state.

The total energy minimization calculation [4] for the orientation of the LMs in the multilayer taking into account the Zeemann interaction, the antiferromagnetic exchange coupling and the in-plane anisotropy $[10]$ does not explain the sequential flip of LMs with increasing field. There might be two ways of solving the problem, either the model obtained from the fit should be revised or other energy terms should play a role in the minimization of the energy. It was attempted to choose the starting parameters so that they correspond to a symmetric model of collective rotation of the LMs similar to the model of the mentioned twisted canted state [1] adapted for the two-fold in-plane anisotropy of this sample, but the fits did not converge. An additional energy term would be given by the biquadratic term [5]. Indications of its influence might be found in the angular distribution of the LMs at the lower fields in Fig. 4. Here again, more precise measurements are required and model calculations including the biquadratic coupling should be performed.

In conclusion, it has been shown that it is possible to perform the layer-by-layer magnetometry of a magnetic multilayer with polarized neutron reflectometry. A coexistence of an anti-

ferromagnetic phase with a ferromagnetic-like phase was detected. A refined picture of these two phases is still needed, yet they can explain features along the magnetization curve like the remanence and the steps on the magnetization curve.

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