Transverse and Lateral Structure of the Spin-Flop Phase in Fe/Cr Antiferromagnetic Superlattices

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Direct evidence of the nonuniformly canted state of the spin-flop phase induced by a magnetic field applied to Fe/Cr(100) superlattices is obtained by polarized neutron reflectometry. It is unambiguously demonstrated that the magnetization of the alternating Fe layers is twisted through the multilayer stack proving a stable noncollinear configuration. The maximal tilt at the end layers progressively reduces towards the center of the multilayer. The set of tilt angles is deduced from a model-free data evaluation employing the supermatrix routine. Spin-flip off-specular scattering is determined by the in-plane magnetization fluctuations and is fitted by a theoretical model of domains.

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Since the discovery of the giant magnetoresistance (GMR) in Fe/Cr multilayers (ML) [1,2] it is well established that this prominent phenomenon existing for a wide variety of magnetic ML is intimately related with an antiferromagnetic (AF) exchange coupling between magnetic moments in adjacent layers and with their mutual reorientation under an applied magnetic field. However, as was emphasized in Ref. [3], current theoretical models explaining exchange coupling and the GMR effect invoke reliable and detailed experimental information on the magnetic moments' orientation in individual layers. This is especially important due to the fact that most of the models [4,5] assume a simple, usually homogeneous ground state, which as we show is not the case.

The problem of the ground-state spin configuration of an antiferromagnet subjected to a magnetic field parallel to its surface was realized long ago [6], but it gained much theoretical attention only after benchmarking experiments [7] on AF coupled Fe/Cr ML with strong in-plane uniaxial anisotropy. It turns out that a Fe/Cr ML is an excellent model system of an antiferromagnet, in which finite size and surface effects are much better pronounced than in bulk antiferromagnets. It was found that magnetic susceptibility shows at a certain in-plane magnetic field a peak interpreted as a signature of the *surface* spin-flop transition preceding the "bulk" phase transition at which magnetization in all layers flips almost perpendicular to the field. The experimental observations were supported by numerical calculations [7] performed for a simple one-dimensional model, which assumes that homogeneous magnetization of a Fe layer is displayed within the layer plane and is coupled via AF exchange interaction to that of neighboring layers. These results have triggered extensive theoretical debates [8] on the structure and stability of different inhomogeneous spin configurations in the spin-flop phase, finite size effects, discreteness, role of anisotropy, etc. The isotropic case was theoretically analyzed earlier [9].

Ongoing theoretical dispute employs rather sophisticated argumentation, but direct experimental information is still quite scanty. The magnetic ordering in magnetic ML is mostly experimentally studied by means of surface magnetometry, magneto-optics, and other methods [4], which give only integral characteristics of the ML, essentially use model assumptions, and do not yield the selective determination of magnetic properties of the buried layers. Such information can be retrieved from the data on polarized neutron reflectometry (PNR) by fitting specular reflection (for review, see Ref. [10]) by a theoretical model. However, in this case a possible contamination of the specular intensity by diffuse scattering, which is often seen from exchange coupled ML [11,12], should be accounted for. Then a complete evaluation [11] of the data is required, while valuable information such as the mean angle between sublattice magnetization and external field [13,14] can be obtained analyzing some particular features of off-specular scattering.

In the present Letter we report on the first direct experimental evidence of a twisted ground-state configuration realized in AF exchange coupled ML exposed to an in-plane external magnetic field above the spin-flop transition. This fact can be immediately established via the qualitative analysis of the line shape of the superstructure peaks on the PNR curve and the related off-specular Bragg sheet. However, quantitative evaluation of both specular reflection and off-specular scattering of polarized neutrons has been accomplished in order to deduce the layer-by-layer spin configuration through the ML stack, as well as within the plane of the layers. It

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is found that, in contrast to the assumptions [7–9], in-plane homogeneous magnetization is not stable and falls into a set of lateral domains, in which magnetic moments across the ML stack are arranged into a configuration consistent with the model calculations [7,9].

A series of (001) $[^{57}Fe/Cr]_N$ ML with various thicknesses of Fe and Cr layers and number of bilayers N was grown with molecular beam epitaxy on (110) Al_2O_3 substrates covered with a Cr buffer layer. *In situ* reflection high-energy electron diffraction and *ex situ* x-ray diffraction and reflection showed the single-crystalline structure of the samples with interface roughness of about 1 monolayer. The magnetization measurements show in-plane fourfold anisotropy [15]. Here we concentrate only on the results obtained for the sample $[Cr(9\ \text{Å})/{}^{57}Fe(67\ \text{Å})]_{12}/Cr(68\ \text{Å})/Al_2O_3$. The PNR experiments were carried out at the Institute Laue Langevin on the reflectometer ADAM [16] (wavelength $\lambda = 4.41$ Å; polarization 97%). Magnetic and nuclear scattering is separated using neutrons polarized parallel ("+" state) or antiparallel ("−" state) to the external field [10]. With additional polarization analysis spin-flip (+ − or − +) and non-spin-flip (++ or −−) components were measured.

The results of the PNR experiment performed in the external magnetic field of 19.5 mT applied along an in-plane easy axis (001) after a saturation in a field of 1 T are shown in Fig. 1(a). The scattering pattern for incoming neutrons in the − state is presented as a function of $(p_i + p_f)$ and $(p_i - p_f)$, with $p_{i(f)} = 2\pi \sin\alpha_{i(f)}/\lambda$ the normal to the surface component of the incoming (outgoing) wave vector and $\alpha_{i(f)}$ the angle of incidence (scattering). The specularly reflected intensity along the line $p_i = p_f$ shows the total thickness oscillations and the first order Bragg peak at $Q_z = 0.0826\ \text{Å}^{-1} = 2\pi/d$ ($Q_z = p_i + p_f$ is the wave vector transfer component normal to the surface; $d = 76$ Å is the bilayer thickness). The absence of measurable diffuse scattering around the first order Bragg peak confirms the negligible interfacial roughness. The intensity around the 1/2-order superstructure Bragg peak position at $Q_z = 0.0413\ \text{Å}^{-1}$ indicates a kind of AF ordering across the ML stack.

The intensity map in Fig. 1(a) has several peculiar features well reproduced in model calculations in Fig. 1(b). The first is that the superstructure Bragg reflection on the specular line is split into two maxima. The second one is strong off-specular scattering forming a Bragg sheet along the line $Q_z d \approx \pi$ and crossing the specular ridge at exactly the position of the minimum of the split Bragg reflection. Measurements with polarization analysis are shown in Fig. 1(c) for the − state. The analyzed area framed by the dashed lines indicates the limited cross section of the analyzer operating in transmission mode. The intensity of the non-spin-flip specular reflection is transmitted by the analyzer, but the analyzed part of the Bragg sheet (close to the specular

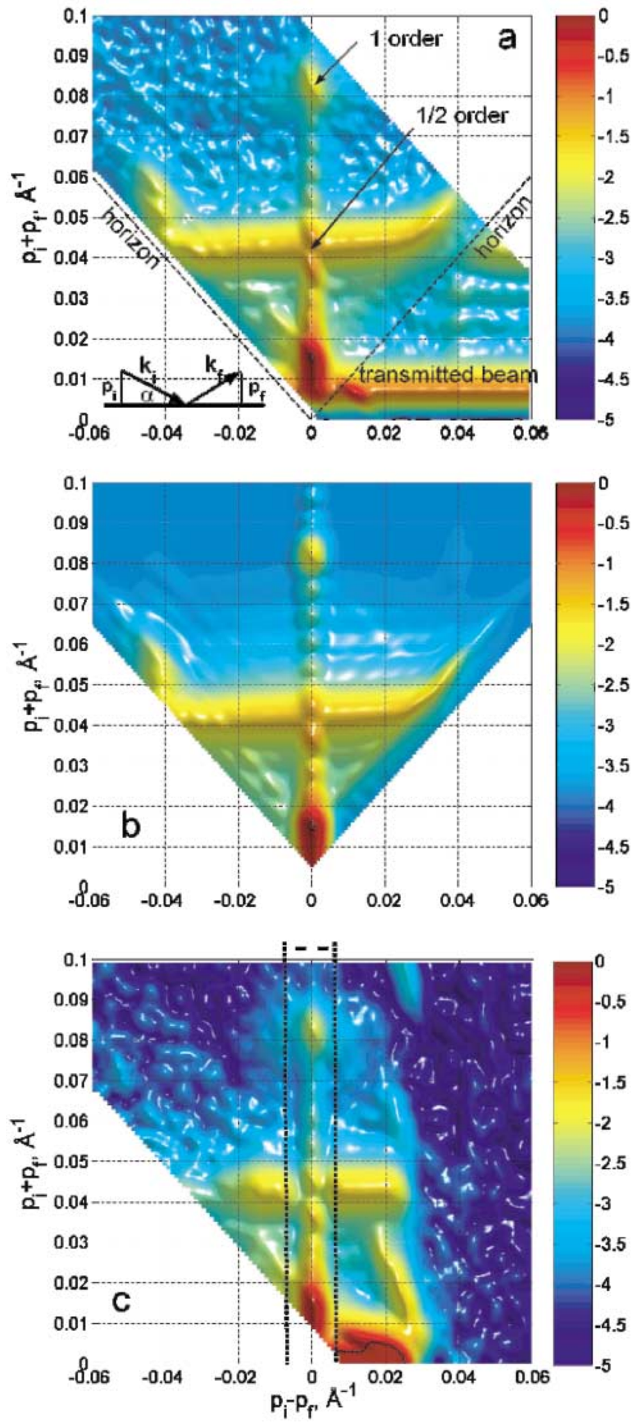

FIG. 1 (color). (a) Experimental two-dimensional (2D) map of the intensity scattered from the Fe/Cr ML as a function of $(p_i - p_f)$ and $(p_i + p_f)$, with p_i and p_f the perpendicular to the sample surface components of the incoming and the outgoing wave vector, respectively (see inset). The logarithmic intensity color scale is shown on the side. The incoming neutrons are in the − state. (b) 2D model fit to the experimental data in (a). (c) Experimental 2D intensity map as in (a) with the analyzed region ("−−" state) between the dashed lines.

ridge) is not transmitted which implies that the off-specular scattering flips the neutron spin.

The presence of magnetic off-specular scattering (vanishing at saturation) means that the layer magnetization is laterally not homogeneous, but rather decomposed into domains, the size of which is smaller than the lateral projection $l_\| \sim 2\pi/(\delta Q_\|)$ of the neutron coherence length, where $\delta Q_\|$ is the uncertainty in the lateral momentum transfer $Q_\| \approx (\lambda/4\pi)(p_i^2 - p_f^2)$. The neutron spin can be flipped due to scattering from domains with magnetic moment components $\mathbf{M}_n^\perp$ perpendicular to the external field $\mathbf{H}$. If these components are AF correlated in different layers n, then spin-flip off-specular scattering is mostly concentrated into the superstructure Bragg sheet [11,12,17] at $Q_z d = \pi$.

Absence of spin-flip specular reflection signifies that the mean magnetization $\overline{\mathbf{M}}_n$ averaged over domains smaller than $l_\|$ has in each magnetic layer n only the projection $M_n^\| = (\overline{\mathbf{M}}_n \mathbf{H})$. A superstructure Bragg peak on the reflectivity line means that this projection varies from layer to layer with alternation in the deviations $\delta M_n^\| = M_n^\| - M^\|$ from the magnetization $M^\| = \langle M_n^\| \rangle$ averaged over the sample. From Fig. 1(a) it follows that the alternation of $\delta M_n^\|$ is not perfect and has a "stacking fault" in the middle, dividing the ML into two parts. The reflection from those parts results in destructive interference, so that the reflection exactly at $Q_z = \pi/d$ is almost forbidden, the 1/2-order Bragg reflection is suppressed in the middle and the superstructure peak appears as a double peak as seen in Fig. 1. The off-specular Bragg sheet is not doubled and passes through the minimum of the split superstructure Bragg reflection. So, there is no stacking fault in the alternation of the projections $M_n^\perp$. Hence all main features in Figs. 1(a) and 1(c) are well understood and we launched a two-dimensional (2D) fitting routine (see below) providing remarkable agreement between Figs. 1(a) and 1(b). The fit quality is illustrated by Fig. 2, where the intensity along the specular line (vertical cut) and a horizontal cut through the minimum of the 1/2-order Bragg peak are depicted. The routine converges to a set of parameters collected in Fig. 3(a), which shows an inhomogeneously canted state in the magnetization distribution with the canting angles φ_n varied from layer to layer for the two types of lateral domains with mean size of 2800 Å. $M_n^\|$, $M_n^\perp$, and $M^\|$ are plotted in Fig. 3(b) and are expressed in units of the magnetic neutron scattering length density Nb_m directly proportional to the layer magnetization [10]. It is clearly seen that $\delta M_n^\|$ alters the sign for neighboring layers except in the middle of the ML, where it has the same sign, i.e., a stacking fault. The physics of this phenomenon can be easily understood: the angles φ_n for the end layers, missing one of the neighbors, should be smaller than for the interior layers, having both neighbors. This provides a gain in the Zeeman energy under the condition of the reduced symmetry at the boundaries of the ML stack, where the end

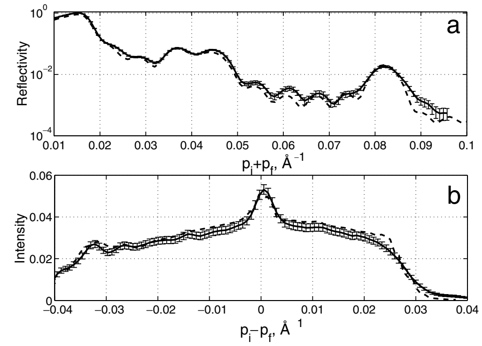

FIG. 2. (a) Measured reflected intensity (solid line) multiplied by $\sin(\alpha_i)$ along the specular line together with the fit (dashed line) extracted as a vertical cut from Figs. 1(a) and 1(b), respectively. (b) Measured off-specular intensity (solid line) along the 1/2-order Bragg sheet together with the fit (dashed line) extracted as a horizontal cut from Figs. 1(a) and 1(b).

layers experience only half of the AF exchange interlayer coupling. The latter drives the magnetization vector of the next to the outermost layers farther away from the field direction. Canting angles φ_n relax towards the bulk values in depth of the ML, where surface influence is less essential. A striking feature of this relaxation is that for an even number of magnetic layers it leads to a symmetric configuration with an unavoidable stacking fault in the middle. Indeed, both of the outermost layers are under symmetric conditions, but belong to different antiphase parts (A and B). Therefore, if the magnetization in the top layer is tilted clockwise for an angle φ_0^A, then in the

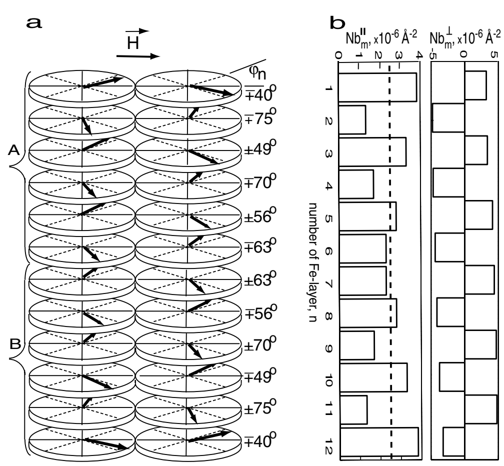

FIG. 3. (a) Configuration of the magnetization $\mathbf{M}_n$ in the Fe/Cr ML in the external field $\mathbf{H}$ applied in plane along one of the easy axes; dashed lines mark the hard axes. Only Fe layers are shown. The only possible two types of domains (left and right) are depicted. Brackets A and B indicate the two transverse antiphase parts of one lateral domain. The canting angles φ_n between $\mathbf{M}_n$ and $\mathbf{H}$ are shown on the right-hand side. (b) Magnetic parts of the neutron scattering length density $Nb_m^\|$ (left) and $Nb_m^\perp$ (right) proportional to $M_n^\|$ and $M_n^\perp$. The dashed line indicates $M^\|$.

bottom it is tilted anticlockwise for the angle $\varphi_N^B = -\varphi_0^A$, and generally, $\varphi_n^A = -\varphi_{N-n}^B$ [18]. The resulting configuration in Fig. 3 has no net magnetic moment component perpendicular to the field and therefore is stable. However, the mirrored configuration is equally probable, so the sample falls into lateral domains sketched in Fig. 3(a). Domains reduce the dipolar energy via demagnetization of each individual layer, and the mean value $\overline{M_n^\perp} = M\overline{\sin\varphi_n}$ averaged over domains is zero.

Because of the kinematic restrictions the domain size cannot immediately be deduced from the 1/2-order Bragg sheet extension [12], and we usually measure also the Bragg sheet of the 3/2 order [11]. It covers a greater range in lateral momentum transfer $Q_\parallel$ and is less distorted by dynamical effects [13,14] close to the total reflection edges. Nonetheless, the domain size can still be determined from our data presented here via quantitative 2D analysis, being carried out simultaneously for both PNR and off-specular scattering. It was performed using a formalism based on the distorted wave born approximation [19] developed earlier [13,14,20,21]. Within this approach $M_n^\perp$ components of the domain magnetization are treated as a source of a perturbation of the reference neutron wave functions. The latter are determined by layer optical potentials, which account also for the contributions from $\overline{M_n^\parallel}$. Then the scattering amplitude is just proportional to the matrix elements between the reference wave functions for incident and scattered neutron waves propagating inside the mean ML potential. In fact, the problem is reduced to the calculation of the reflectance and transmittance matrices for each layer using, for instance, the supermatrix routine [20,21]. Finally, the magnetic scattering cross section is determined by the domain lateral form factor $F_\parallel(Q_\parallel)$, the ML structure factor, and a combination of reflectance and transmittance amplitudes for both neutron spin states [21]. Under these circumstances off-specular scattering carries the information not only on domains, but also on mean magnetization averaged over domains. Consequently, off-specular scattering is to be fitted along with the specular reflection, which depends solely upon the mean magnetization of the layers.

In conclusion, the experimental evidence of the nonuniform twisted canted state in Fe/Cr ML in the spin-flop phase has been obtained. The canting angles are maximal in the end layers and progressively relax towards the middle of the ML from both sides. The magnetic moments in the two end layers are tilted in antiphase, which creates an unavoidable stacking fault in the middle. The ML is decomposed into two types of vertical domains with the mirrored configuration of magnetic moments.

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