Noncollinear Magnetization Structure at the Thickness-Driven Spin-Reorientation Transition in Epitaxial Fe Films on W(110)

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An in-plane spin-reorientation transition occurring during the growth of epitaxial Fe films on W(110) was studied *in situ* by using the nuclear resonant scattering of synchrotron radiation. The spin-reorientation transition originates at the Fe/W(110) interface and proceeds via a noncollinear spin structure resembling a planar domain wall that propagates towards the surface with increasing film thickness.

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The spin-reorientation transition (SRT) is one of the most fascinating phenomena observed for ultrathin ferromagnetic films. Despite the large number of experiments reporting on the occurrence of the SRT, the kinetics of this process was studied occasionally. In fact, only two different types of SRTs were identified, namely, the continuous second-order transition proceeding through the coherent magnetization rotation [1] and the discontinuous one manifested by formation of magnetic domains [2]. A third scenario that was recently proposed theoretically [3], in which SRT undergoes continuously via formation of an inhomogeneous noncollinear magnetic structure, has not been observed up to now for the $3d$ metal films. In rare cases when the SRT was studied in detail, its kinetics was concluded from the indirect determination of the thickness-driven trajectory in the anisotropy space spanned on the second- and fourth-order anisotropy constants [4,5]. In such an approach, the judgment between the coherent rotation picture and intermediate noncollinear magnetic structure is hardly possible, since the noncollinear magnetic structure cannot be distinguished from the homogenous magnetization model characterized by an average canting angle. The phenomenological higher-order anisotropies may arise from the noncollinear arrangement of the magnetic moments as pointed out by Farle *et al.* [6]. On the other hand, methods used until now in SRT studies, such as magnetometry [7], all kinds of magnetic microscopy [2], x-ray magnetic circular dichroism [8], etc., were unable to resolve depth magnetization dependence, by virtue of their integrating character or by their strictly surface sensitivity [1].

In the present paper, we report on the discovery of a noncollinear magnetic structure during the thickness-driven SRT process occurring in a chemically homogenous system, such as bcc-Fe, which is regarded as the archetypal collinear ferromagnet. The derived scenario of the in-plane SRT for Fe(110) films grown on W(110) proves inadequacy of the common SRT picture based on the balance of surface and volume anisotropies (that work within the homogenous magnetization approximation) and answers the question on the transition nature.

In the case of Fe/W(110), the magnetization switches during the film growth from the $[1\bar{1}0]$ to the [001] in-plane direction as the iron film approaches the critical thickness d_c. One of the two possible explanations for this process involves a large magnetic surface anisotropy preferring a $[1\bar{1}0]$ magnetization direction, which competes with favoring [001] easy magnetization axis magnetocrystalline and strain-induced magnetoelastic anisotropies [7,9]. The key role of the Fe(110) surface magnetic anisotropy is reflected in a strong dependence of the critical transition thickness on the adsorption of noble metals or gases [8], as well as the surface morphology that can be influenced by the growth mode [10]. Another explanation is connected with the strain-induced magnetoelastic anisotropy, originating from the large misfit between the Fe and W lattices, that prefers the $[1\bar{1}0]$ magnetization direction for the low thickness, as was proposed by Sander, Enders, and Kirschner [11]. In such an approach, with increasing film thickness SRT occurs due to the competition of the magnetocrystalline anisotropy and magnetoelastic anisotropy, which can change its sign due to the relaxation of strains for a certain film thickness. It is plausible that the two above pictures overlap and interplay, since the experimentally determined value of the critical thickness d_c strongly depends on the deposition protocol [7,12]. Similarly, the width δ of the thickness-induced transition that determines the kinetics of the SRT process [7,13] ranges from one [8] to a few monolayers [9], which indicates that the SRT scenario may not be unique. Theoretically, an instant SRT (small δ) means a discontinuous first-order transition via coexistence of magnetic phases (formation of magnetic

domains with the $[1\bar{1}0]$ and [001] magnetization directions), while larger δ's evidence a continuous process that proceeds through the coherent magnetization rotation [3]. The complex transition picture, as drawn above, becomes more complicated due to the sensitivity to the adsorption of residual gases, also under UHV conditions [14], resulting in the unavoidable superposition of the thickness- and adsorbate-induced effects when nominally uncoated films are investigated, especially when the Fe thickness approaches d_c. Moreover, usually an external magnetic field must be applied to observe the magnetization state, which may change the character of the transition [4]. All of this makes the intrinsic thickness dependence of the SRT process in the Fe(110)/W system hardly accessible. Indirectly, the formation of magnetic domains with the orthogonal magnetization near the critical thickness was concluded from the existing experimental data [8,14]. In the present Letter, we reveal an alternative scenario of the SRT in the Fe(110)/W.

Our thickness-induced SRT was monitored *in situ* by using grazing-incidence nuclear resonant scattering (NRS) of synchrotron radiation. The continuous SRT process was clearly identified, as evidenced through its large width, $\delta \sim 3$ ML, and several intermediate magnetization states could be concluded from the NRS measurements. The numerical analysis of the NRS data indicated that, for this extremely clean experimental run, a noncollinear magnetization structure is formed in the vicinity of the critical thickness, with a strong surface magnetization pinning along the $[1\bar{1}0]$ direction. With increasing thickness, the transition is initiated at the bottom atomic layers, neighboring with the tungsten substrate, and finally is completed at the surface layer. We are convinced that after many experiments [7–12,14,15] the true thickness dependence of SRT could be obtained for the Fe(110)/W system.

NRS [16] is a synchrotron analogue of Mössbauer spectroscopy, in the sense that it involves a recoilless resonant excitation of the nuclear energy levels (induced by the x rays with energy 14.4 keV for ^{57}Fe), split due to the hyperfine interactions. The analysis of the characteristic quantum beat pattern seen in the time evolution of the intensity of nuclear resonant scattering (the so-called time spectrum) allows the site- and layer-selective determination of hyperfine magnetic fields and electric field gradients, including information about their orientation. NRS is capable of distinguishing among ferromagnetic, antiferromagnetic, or noncollinear order, thus allowing the determination of the sublattice and, in the case of layered systems, the sublayer magnetic [17] and thermoelastic [18] properties. Moreover, a virgin magnetic state is accessible, since no magnetic field is necessary for the determination of the local magnetization vector probed by the magnetic hyperfine interactions. The intrinsic depth sensitivity of NRS arises from the evanescent penetration depth of the x-ray radiation field for the grazing-incidence geometry at the incidence angles close to the Fe critical angle [17].

The measurements were done at the Nuclear Resonance beam line ID18 [19] at the European Synchrotron Radiation Facility in Grenoble, with the use of a recently installed multichamber UHV system [20]. ^{57}Fe was evaporated on a freshly cleaned W(110) crystal that was prealigned to the x-ray beam. The preparation was made at room temperature, with the exception of the first monolayer, which was deposited at 500 K to ensure a good nucleation [21]. The evaporation rate was 0.5 ML/min, and the final nominal thickness reached about 30 ML, as was checked *in situ* by x-ray reflectivity measurements. The morphology of the resulting Fe film was studied separately with STM. A rooflike surface modulation could be clearly seen in the STM image taken for a 50-Å-thick Fe film, indicating that on the top of the continuous 40-Å-Fe base layer, 12-Å-high islands, elongated along the $[1\bar{1}0]$ direction, with triangular cross sections, are formed, in agreement with previous low energy electron diffraction data [10]. For the NRS measurements at the grazing-incidence geometry (grazing angle 0.23°, close to the Fe critical angle), the x-ray beam was vertically focused to about 15 μm, which resulted in about a 3-mm-long beam path on the sample surface. Such conditions ensure high thickness homogeneity (on average better than 0.1 Å) on the investigated sample area as checked by *in situ* x-ray reflectivity measurements at various places on the sample. The k vector of the incident radiation was parallel to the $[1\bar{1}0]$ in-plane Fe direction. Directly during the preparation, a set of the NRS time spectra was collected in thickness steps corresponding to a fraction of the Fe monolayer. It should be noted that the deposition of Fe was not interrupted from the beginning up to the completion of the SRT process, and the spectra were accumulated on-line during the film growth, ensuring a contamination-free Fe surface. The accumulation time of a single spectrum was typically 10 sec, which means that, for the given evaporation rate, the magnetic state reflected in the spectra is averaged over a thickness smaller than 0.1 of the Fe monolayer. The numerical analysis of the measured time spectra was performed by using the software package CONUSS [22] based on the dynamical theory of the nuclear resonant scattering.

The fitted time spectra are shown in Fig. 1 for the selected Fe film thicknesses. A regular beat structure that is exemplified in Fig. 1, curve *a*, reflects, according to the theoretical fits, the uniform magnetization state along the $[1\bar{1}0]$ direction, with the hyperfine magnetic field close to bulk Fe ($B_{\mathrm{HF}} = 32.9$ T and that is almost constant for other discussed spectra). Such a state persists up to the thickness of about $d = 51$ Å. It has to be noted that, although no external magnetic field was applied during the Fe growth, a small in-plane residual magnetic field, with components of about 1 Oe along the $[1\bar{1}0]$ and [001] directions, was measured at the sample position. Such a field may easily erase the domains of the virgin state especially that, during the growth, the Fe film undergoes thickness-induced

paramagnetic-ferromagnetic phase transition, at which the magnetic susceptibility is infinite.

Similarly, the spectra for the coverages above 56 Å (see Fig. 1, curve *f*) can be easily fitted by assuming a homogeneous magnetization but now parallel to the [001] direction, with the bulklike magnitude of B_{HF}. It is clear that the SRT process is not abrupt but extends over a relatively large thickness range of $\delta \sim 6$ Å, corresponding to 3 ML. The most unique and also challenging to fit were the time spectra accumulated during the progress of SRT (Fig. 1, curves *b*–*e*). Theoretically, the possible ways of the magnetization transition from $[1\bar{1}0]$ to [001] are (i) coherent rotation and (ii) coexistence of [001] and $[1\bar{1}0]$ magnetized domains and (iii) a formation of the laterally or vertically inhomogeneous noncollinear magnetic structure. The first two models that assume a homogeneous magnetization depth profile across the Fe(110) films produced distinctly different spectra; however, neither of them could satisfactorily fit the experimental data in the transition, as is exemplified in Fig. 2 for the selected Fe film thickness ($d = 53.2$ Å) in the middle of SRT. On the other hand, the laterally inhomogeneous noncollinear magnetic structure that can be considered as a combination of the above models could be also excluded, as such magnetization structures did not lead to the measured time spectra. Successful fits, as shown in Fig. 1, curves *b*–*e*, by the solid lines, could be obtained only when a depth-dependent magnetization structure was assumed. The distribution of the magnetization directions was modeled by dividing the film with nominal thickness *d* (a fixed parameter of the fit for the given thickness) into *N* equivalent sublayers, with the thickness $d_N = d/N$. For each sublayer, an in-plane orientation of the hyperfine magnetic field (sublayer magnetization M_N) was defined by the angle φ_N with respect to the $[1\bar{1}0]$ in-plane direction. Accordingly, the only free parameters of the fits for increasing film thickness were the φ_N values. The results shown in Fig. 1 were obtained for $N = 5$, which appeared to be a minimum number providing high quality fits, whereas higher numbers of sublayers did not significantly improve the matching of the fits and the measured spectra. The orientation of the sublayer magnetizations $\vec{M}_{1-5}$, as derived from the analysis of the NRS data, is shown schematically in Fig. 3 for the successive Fe thicknesses. The onset of the transition was noticed for the thickness of 51.6 Å. SRT from $[1\bar{1}0]$ to [001] is initiated at the deepest layers (neighboring the tungsten substrate), which switch first, while the magnetization of the remaining sublayers forms a fanlike structure. Then the spin structure evolves in a rather smooth and continuous manner. With increasing thickness, the magnetization of the subsequent sublayers rotates, and finally the transition is completed by the topmost surface layers. Apparently, the initial phase of the thickness-induced SRT took place in such an abrupt way that we could not access its details, especially that the contribution to the time spectrum coming from the deepest layers is an order of magnitude smaller than that from the topmost one. It is possible that the small residual magnetic field, discussed above, forbids formation of antiparallelly magnetized domains before and after the SRT, while in the absence of the magnetic field, the SRT process could be realized via formation of fanlike magnetization with the opposite sense of rotation.

From the above data, the following picture of the magnetic structure during the studied SRT emerges: (i) The Fe layer neighboring the *W* substrate, about 20 Å thick, switches first to the bulklike easy direction along the [001] direction, (ii) the central part of a similar thickness is characterized by a noncollinear magnetization state, and (iii) the surfacemost layer, with the thickness corresponding to the amplitude of the vertical roughness, has its magnetization pinned to the $[1\bar{1}0]$ direction. Such a com-

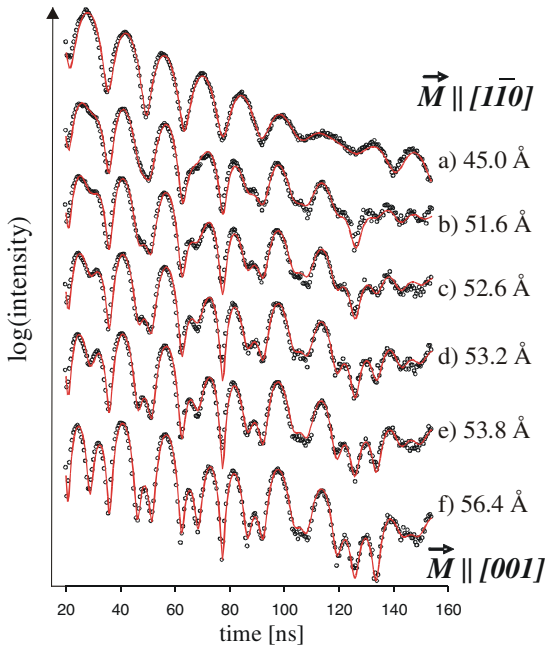

FIG. 1 (color online). The time spectra accumulated during continuous Fe evaporation labeled with the corresponding Fe thicknesses are shown. The top and bottom spectra reflect according to the theoretical fits (continuous line) homogenous magnetization states with the easy axis along $[1\bar{1}0]$ and [001], respectively.

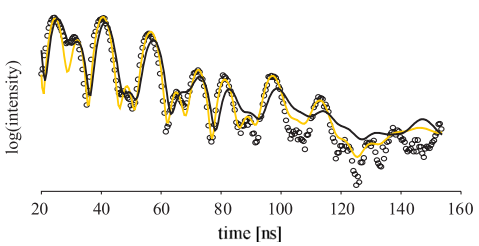

FIG. 2 (color online). The best fits to the time spectrum measured for $d = 53.2$ Å, obtained within the domain (black line) and coherent rotation (orange [gray] line) models.

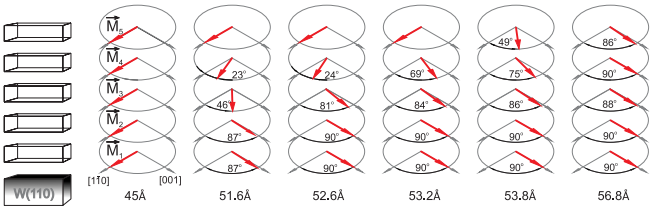

FIG. 3 (color online). The magnetization structure during the thickness-induced SRT for the Fe/W(110) system, as derived from the NRS measurements using a five sublayer model. The sublayer magnetization vectors are labeled as $\vec{M}_{1-5}$.

plex behavior cannot be explained within the basic phenomenological picture of SRT, in which, at the critical thickness, the effective magnetic anisotropy constant $K_{\mathrm{eff}} = K_v + K_s/d$ zeroes, due to opposition of the volume magnetocrystalline and magnetoelastic anisotropies (described by K_v) and magnetic surface anisotropy (described by K_s). The fact that the SRT process is initiated at the bottom of the Fe film points to the conclusion that most probably magnetoelastic anisotropy at the vicinity of the Fe/W(110) interface evolves during the film growth with the increasing thickness in such a way that the easy magnetization direction of the bottom part of the film changes from the $[1\bar{1}0]$ to the [001] direction. Our experiment then explains the apparent contradiction concerning the decisive factor for the magnetic anisotropy in the Fe(110)/W system. A strong surface anisotropy pins the magnetization to the $[1\bar{1}0]$ direction, as postulated by Gradmann, Korecki, and Waller [9], while the SRT itself can be attributed to the changes of strain-induced magnetoelastic anisotropy evolving with the thickness during the film growth, in agreement with the interpretation by Popescu *et al.* [23]. In fact, both magnetic surface anisotropy and magnetoelastic anisotropy are responsible for the transition, as they become competing at the vicinity of the critical thickness only, while for the lower thickness they can cooperatively favor the $[1\bar{1}0]$ magnetization direction. In conclusion, the nature of the thickness-induced SRT was investigated for the Fe(110)/W system. Our studies clearly verify that SRT is not purely of local origin in the surface atomic layer but comes as an intricate consequence of concurring electronic [24] and stress-driven [25] effects. The noncollinear, exotic magnetic phase of epitaxial Fe films was found in the vicinity of a critical SRT thickness. Such a magnetic structure resembles a planar domain wall with its center propagating towards the surface as the thickness increases. In the original paper on SRT in Fe on W(110) [9], it was argued that the magnetization direction is homogenous across the film depth. In view of the present results, such a model must be revised, and noncollinear magnetization states, never before observed for such thick bcc-Fe films, have to be explicitly allowed. Generally, such noncollinear magnetization states imply a strong modification of the exchange interaction, which in our case can be simply explained by epitaxial strains, taking into account a considerable tensile stress recently found in an analogous Fe film as thick as 9 nm [26].

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[1] R. Allenspach, M. Stampanoni, and A. Bischof, Phys. Rev. Lett. **65**, 3344 (1990)
[2] R. Allenspach and A. Bischof, Phys. Rev. Lett. **69**, 3385 (1992)
[3] P. J. Jensen and K. H. Bennemann, Surf. Sci. Rep. **61**, 129 (2006)
[4] Y. T. Millev, H. P. Oepen, and J. Kirschner, Phys. Rev. B **57**, 5848 (1998)
[5] D. Wilgocka-Ślęzak *et al.*, Phys. Rev. B **81**, 064421 (2010).
[6] M. Farle, A. N. Anisimov, W. Platow, P. Poulopoulos, and K. Baberschke, J. Magn. Magn. Mater. **198–199**, 325 (1999).
[7] H. J. Elmers and U. Gradmann, Appl. Phys. A **51**, 255 (1990).
[8] I. G. Baek, H. G. Lee, H. J. Kim, and E. Vescovo, Phys. Rev. B **67**, 075401 (2003).
[9] U. Gradmann, J. Korecki, and G. Waller, Appl. Phys. A **39**, 101 (1986).
[10] M. Albrecht, T. Furubayashi, M. Przybylski, J. Korecki, and U. Gradmann, J. Magn. Magn. Mater. **113**, 207 (1992).
[11] D. Sander, A. Enders, and J. Kirschner, J. Magn. Magn. Mater. **200**, 439 (1999).
[12] F. Gerhardter, Y. Li, and K. Baberschke, Phys. Rev. B **47**, 11 204 (1993).
[13] C. Klein, R. Ramchal, A. K. Schmid, and M. Farle, Phys. Rev. B **75**, 193405 (2007).
[14] D. Yu *et al.*, Surf. Sci. **601**, 5803 (2007).
[15] P. Baumgart, B. Hillebrands, and G. Güntherodt, J. Magn. Magn. Mater. **93**, 225 (1991).
[16] *Nuclear Resonant Scattering of Synchrotron Radiation*, edited by E. Gerdau and H. DeWaard, Hyperfine Interact. Vol. 123–125 (Springer, New York, 1999).
[17] S. Couet *et al.*, Appl. Phys. Lett. **94**, 162501 (2009).
[18] T.Ślęzak *et al.*, Phys. Rev. Lett. **99**, 066103 (2007)
[19] R. Rüffer and A. I. Chumakov, Hyperfine Interact. **97–98**, 589 (1996).
[20] S. Stankov *et al.*, Rev. Sci. Instrum. **79**, 045108 (2008).
[21] H. Bethge, D. Heuer, Ch. Jensen, K. Reshöft and U. Köhler, Surf. Sci. **331–333**, 878 (1995).
[22] W. Sturhan, Hyperfine Interact. **125**, 149 (2000).
[23] R. Popescu *et al.*, Phys. Rev. B **68**, 155421 (2003).
[24] M. Cinal and D. M. Edwards, Phys. Rev. B **50**, 3754 (1994)
[25] D. Sander, Rep. Prog. Phys. **62**, 809 (1999).
[26] S. Stankov *et al.*, Phys. Rev. Lett. **99**, 185501 (2007).