Nanoscale spatial switching of magnetic anisotropy in pseudomorphic Fe(110) on W(110)

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Pseudomorphic Fe(110) films, prepared on W(110) at room temperature, with a mean thickness of roughly 1.5 monolayers, consist of double-layer islands with a diameter of the order of 10 nm which are ferromagnetic (superparamagnetic) at room temperature, surrounded by a monolayer sea which becomes ferromagnetic below 222 K. Magnetic anisotropy is uniaxial in both components, but the easy axis spatially switches between perpendicular to the plane for the islands and in the plane for the monolayer sea. The intimate interconnection of exchange coupled components with orthogonal anisotropies induces a rich variety of new micromagnetic phenomena. [S0163-1829(97)01322-2]

Recently, considerable interest has been shown in lateral magnetic nanostructures (LMN's), which can be prepared either artificially[1–3] or by self-organization during film growth.[3–7] Whereas artificial LMN's are candidates for ultrahigh-density magnetic storage and sensor technologies, basic nanomagnetic phenomena can be studied in self-organized LMN's as well. Scanning tunneling microscopy (STM) offers the unique possibility to analyze the magnetic phenomena of these two-dimensional (2D) systems based on a detailed nanostructural information, which is not available in 3D cases.

We report on LMN's in which uniaxial ferromagnetic components with easy axes perpendicular and parallel to the film plane, respectively, are intimately interconnected on a 10-nm scale. The samples of our study are sesquilayers (sesqui = one and a half) of pseudomorphic Fe(110) on W(110), prepared at room temperature. Films of this type have been investigated before, and puzzling magnetic phenomena have been observed. Weber *et al.*[8] prepared them up to a thickness where T_c reached 300 K and then observed a striking sensitivity of T_c on submonolayer coverages of Fe, Pd, Ag, and O_2. Back *et al.*[9] verified in such films a 2D scaling of the critical properties with outstanding precision. Elmers *et al.*[7] observed a striking suppression of remanent magnetic long-range order between 1.2 and 1.5 monolayers (ML), Sander *et al.*[6] observed high coercivities of the order of 0.3 T at 140 K near 1.4 ML. It has been recognized before that one key for understanding these phenomena is given by the nanostructure of the films,[7,6,10] which are composed of ferromagnetic (superparamagnetic) double-layer (DL) islands, surrounded by a monolayer sea which becomes ferromagnetic below 222 K.[4,11] We will show that the easy axis of the magnetic DL islands is perpendicular to the plane, whereas that of the monolayer sea is [110] in the plane. This spatial switching of the magnetic anisotropy induces a new class of micromagnetic phenomena. It has been overlooked so far because both the ML and the extended DL (and thicker films) are magnetized along [110] in the plane.[4,7]

Our Fe films were prepared on atomically smooth and clean W(110) substrates held at room temperature, in two separate UHV systems for STM and magnetometry, respectively, with a growth rate of about 2 ML/min and a ratio of thickness to residual gas exposure better than 20 ML/L (1 L=1 langmuir= 10^{-6} Torr s). The structure of a typical film is shown in Fig. 1. The mean DL island area is 70 nm^2. Magnetic properties could be measured *in situ* at 120 K $\leqslant T \leqslant$ 500 K using torsion oscillation magnetometry (TOM),[12,13] supplemented by longitudinal and polar magneto-optical Kerr magnetometry (MOKE). In TOM, we follow small amplitude torsion oscillations of the sample, suspended on a thin torsion filament, and measure a magnetic torque constant R as a function of a homogeneous external field H. The interpretation is easy and quantitative for the case of a uniaxial ferromagnet or superparamagnet with anisotropy energy $F=KV\sin^2\Phi$, for the case of the ''normal orientation'' of the easy axis[12] (nearly) parallel to H. R and H are then connected with the magnetic moment m (or $m^*=\mu_0 m$) and the magnetic anisotropy field $H_K=2K/J_s$ by

$$R/H = m^*/(1+H/H_K). \tag{1}$$

For the ''anomalous orientation'' of the easy axis at right angles to H, one obtains negative values of R/H,[12] which

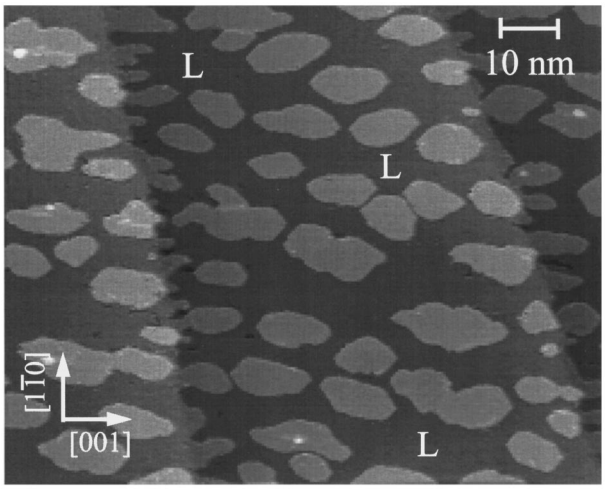

FIG. 1. STM image of a pseudomorphic Fe(110) film on W(110), prepared at 300 K, with a coverage θ=1.45, consisting of DL islands on a ML sea, with lagunalike sections indicated by (L). Incipient misfit dislocations are seen in a few islands only. The STM images were taken in a system separate from that of the magnetometric measurements shown in Figs. 2–4 under comparable UHV conditions.

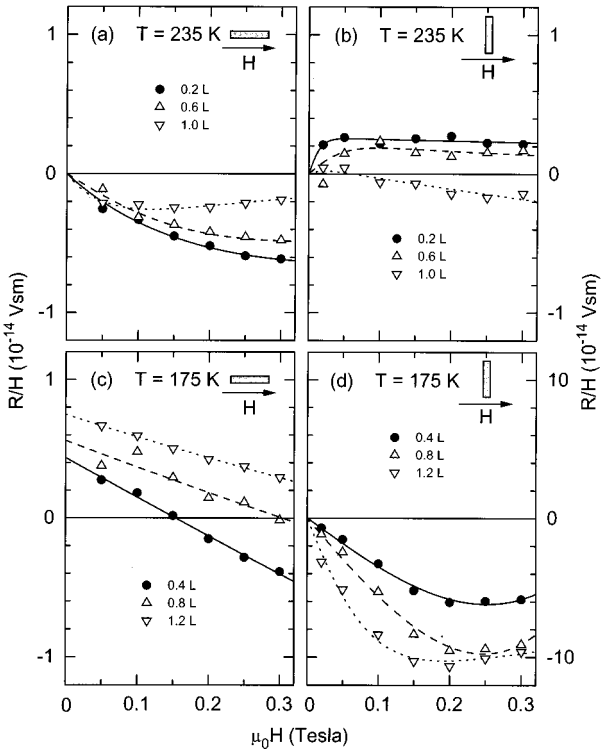

FIG. 2. TOM for a pseudomorphic Fe(110) film on W(110), prepared at 300 K, with a coverage $\theta=1.45$, with the sample parallel to H (a),(c) or at right angles to H (b), (d), respectively. The film was periodically switched between 235 K (a),(b) and 175 K (c),(d). Increasing exposure as indicated in langmuirs (L). The virtually clean film with the lowest exposure is indicated by solid symbols. Oscillation axis of the torsion filament along [001].

usually are much larger than m^*. Because their quantitative evaluation is difficult, they usually only can be taken as an indication of this anomalous orientation. Quantitative evaluation is possible for the case of a uniaxial superparamagnet for which m is connected with the axial field component H_{ax} by $m=m_0\tanh(H_{ax}/H_{sat})$, where $H_{sat}=k_BT/\mu^*$ is a saturation field, μ^* the magnetic moment of a uniaxial single-domain particle, and m_0 the saturation moment of the sample. If α is the angular amplitude of the oscillations ($1/\alpha=10$ in our case), one obtains, for $H\ll H/\alpha$,

$$R/H=-m_0^*(H/H_{sat}). \tag{2}$$

For the saturation field H_{sat}, of islands consisting of D atomic layers, at temperature T, one numerically obtains (using $J_s=2.2$ T)

$$\mu_0H_{sat}=430\ \text{G}\quad[(T/100\ \text{K})/(A/100\ \text{nm}^2)D]. \tag{3}$$

We selected a couple of films with a coverage $\theta=1.45\pm0.05$ (in substrate units), as measured by a quartz oscillator monitor. Figure 2 shows TOM measurements for one of these samples, the temperature T of which was periodically switched between 235 and 175 K, respectively, above and below $T_c(\text{ML})=222$ K. For $T=235$ K, where the ML is paramagnetic, we observe the DL signal only; see Figs. 2(a) and 2(b). We observe a strong dependence on residual gas exposure, which is a general property of the DL component and forms a severe restriction for our measurements. Only a few TOM measurements, using about 20 min each, were therefore possible for a virtually clean film on which we focus. Data for the virtually clean film with the lowest exposure are represented by the solid symbols. The most important result is that the pure DL data in Figs. 2(a) and 2(b) clearly show the signature of perpendicular magnetization, with positive values of R/H for the sample at right angles to the field [Fig. 2(b)] and negative values for the sample parallel to the field [Fie.d 2(a)]. In fitting the data, we used the supplementary information, taken from polar MOKE, that the remanence of the polar loops was always zero, e.g., Fig. 4(c), below. For the virtually clean film, we obtain $m_0^*=0.26\times10^{-14}$ V s m, which is lower than what would be expected from a simple model, $0.45m_{DL}^*=0.9m_{ML}^*=0.86\times10^{-14}$ V s m (the monolayer moment is given by $m_{ML}^*=0.95\times10^{-14}$ V s m in the present geometry). This indicates a reduced magnetization of the DL and blocking of some larger islands which will be discussed below. The magnitude of the anisotropy field can only roughly be estimated as $|\mu_0H_L|=1$ T. m^* decreases with increasing exposure. This is the beginning of a process which was observed in all samples and always resulted finally in a switching of the magnetization into the plane, at exposures of the order of 5 L. Dedicated work on this adsorption-induced spin reorientation transition will be published elsewhere.[14] A rough estimate only is possible for H_{sat}. A mean value of $\mu_0H_{sat}=0.07$ T from 0.2 and 0.6 L in Fig. 2(b) fits the expectation from Eq. (3) (with $A=70$ nm^2), $\mu_0H_{sat}=0.072$ T, thus confirming the superparamagnetic nature of the DL signal.

At $T=175$ K [Figs. 2(c) and 2(d)], the signal of the now ferromagnetic ML component is superimposed. It consists in positive values of R/H for the film parallel to the field [Fig. 2(c)] and strong negative values in the perpendicular case [Fig. 2(d); note the enlarged scale on the ordinate] which completely mask the weak positive DL signal. The data of Figs. 2(a) and 2(b) show that an exposure of up to 0.6 L can be tolerated without strong changes. During this 0.6-L period, the sample signal R/H could be switched back and forth between both temperatures. The data for 0.4 L in Fig. 2(d) were taken between those for 0.2 and 0.6 L in Fig. 2(b), which nearly coincide. The switching between the patterns in Figs. 2(a), 2(b), and 2(c), 2(d) therefore is clearly driven by temperature, not by adsorption. The transition between $T>T_c$ (ML) and $T<T_c$ (ML) is shown in more detail in Fig. 3 for a virtually clean film (exposure $\leqslant$0.7 L). For the film parallel to H [Fig. 3(a)], we observe at 235 K the negative DL signal, as in Fig. 2(a). For $T\leqslant T_c(\text{ML})=222$ K, the positive ML signal is superimposed. The extrapolated axial section which represents the ML magnetic moment, disappears near T_c(ML) as expected and shown in the inset. The sample signal is a superposition of contributions from ML and DL, which, however, strongly interact, as will be shown below. For the film perpendicular to H [Fig. 3(b)], the positive signal from the DL only is seen for $T>T_c$(ML). For $T<T_c$(ML) it is masked by the strong negative ML signal, like in Fig. 2(d) (note the changed vertical scale).

The samples of both Figs. 2 and 3 were prepared on a W(110) substrate with the torsion filament axis along [001]; hence torque signals both from the ML (with in-plane easy axis [110]) and the DL (with perpendicular easy axis [110]) were superimposed. In order to isolate the signal from the

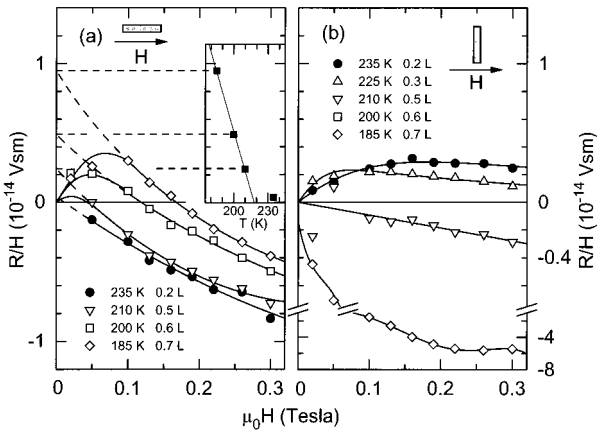

FIG. 3. TOM as in Fig. 2, but for monotonically changed temperatures between 235 and 185 K. The measurement at 235 K is repeated finally (small circles) at a final exposure of 1.0 L. Oscillation axis of the torsion filament along [001]. The inset of (a) shows the extrapolated value of R/H vs temperature T.

perpendicularly magnetized DL, we prepared further samples on a rotated W(110) substrate with the torsion axis now along [110], thus inactivating the ML component. Results are shown in Figs. 4(a), 4(b), and 4(d). The suppression of the ML signal and the resulting isolation of the DL signal is most clearly seen from a comparison of Figs. 2(d) and 4(d). However, the striking dependence of the polar MOKE loops on exposure, shown in Fig. 4(c), in particular the appearance of the Perminvar loop at 2.8 L, and the nonmonotonic evolution of the data Fig. 4(d) with increasing exposure indicate the superposition of complicated coupling effects between

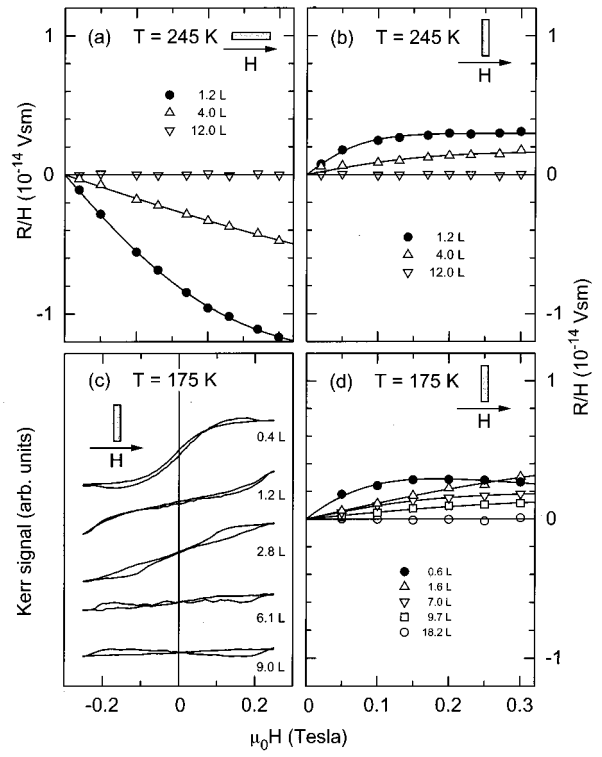

FIG. 4. TOM in (a), (b), and (d) as in Figs. 2(a), 2(b), and 2(d), but with rotated oscillation axis of the torsion filament along [110] and at slightly different temperatures of 245 and 175 K, respectively. Polar Kerr loops taken at 175 K in (c).

DL and ML elements, which remain to be investigated. The disappearance of the DL signal in Fig. 4(d) at 18 L must not be explained in terms of vanishing magnetization, but by rotation to the [110] axis. This will be shown elsewhere.[14]

The perpendicular magnetization of the DL components is surprising because both the ML and films with $D \geqslant 2$ are magnetized in the plane.[4,15] We suggest that it is a result of the pseudomorphic strain which persists in room-temperature-prepared films up to $\theta = 1.5$ and then starts to be relaxed by the formation of misfit dislocations.[7,16] In order to check in how far the perpendicular DL anisotropy can be explained as strain anisotropy, we compare the observed anisotropy with its components in a simple model. We consider the observed anisotropy energy per area, written as $L(\mathrm{DL})2d\cos^2\vartheta$, where $L(\mathrm{DL})$ is a volume-type anisotropy constant which can be represented by an anisotropy field $H_L = 2L(\mathrm{DL})/J_s$ [see Fig. 2 and Eq. (1) $(K = |L|)$], $d = 0.2$ nm is the distance of atomic layers, and ϑ is the polar angle of the magnetization with respect to the surface normal. In this notation, perpendicular anisotropy is indicated by $H_L < 0$. H_L could be measured in few cases only, with a wide scatter of the observed values, which is not surprising in view of the decrease of $|H_L|$ with exposure and its supposed considerable dependence on island size. The maximum values we observed were given roughly by $|\mu_0 H_L| = 1$ T. They must be taken as a lower limit for the anisotropy of the clean, extended pseudomorphic DL, which is given in a rough estimate as $\mu_0 H_L(\mathrm{DL}) = -2 \pm 1$ T, or $L(\mathrm{DL})2d < -0.35 \pm 0.17$ mJ/m^2. In our simple model, $L(\mathrm{DL})$ is composed of the well-known[17] surface anisotropies $K_s^{\mathrm{W/Fe}} = +1.92$ mJ/m^2 and $K_s^{\mathrm{Fe/UHV}} = -0.97$ mJ/m^2, the shape anisotropy $(J_s^2/2\mu_0)2d = +0.77$ mJ/m^2, and the postulated magnetoelastic strain anisotropy $K_{\mathrm{me}}d$. In including the strain anisotropy of one ML only, we make use of the fact that the first ML remains pseudomorphic in the interface, as shown by Mössbauer spectroscopy[18] and confirmed by STM.[16] The strain contribution of the first ML is therefore contained in $K_s^{\mathrm{W/Fe}}$. We then result in $K_{\mathrm{me}}d = L(DL)2d - K_s^{\mathrm{W/Fe}} - K_s^{\mathrm{Fe/UHV}} - (J_s^2/2\mu_0)2d \leqslant -2.1$ mJ/m^2. This is an enormous amount. It is 8 times larger than what is calculated using linear bulk magnetoelastic data,[19] using an in-plane strain of 10.4% and a normal contraction of 10%, $K_{\mathrm{me}}^* d = -0.26$ mJ/m^2. In our strain model of the strong perpendicular anisotropy of the DL, the strain anisotropy therefore is enhanced in comparison with the predictions of bulk elasticity by more than a factor of 8. Even without interface pseudomorphism, this would be more than a factor of 4.

The experimental values $\mu_0 H_L(\mathrm{DL}) = -2 \pm 1$ T and $\mu_0 H_L(\mathrm{ML}) = +4$ T (Ref. 20, with a correction $H_s^{\mathrm{Fe/UHV}} - H_s^{\mathrm{Fe/Ag}}$ from Ref. 17) enable us to numerically determine blocking areas of the DL islands and exchange lengths in both components, which open detailed insight into the magnetic state of our nanomagnetic system. Using standard theory of superparamagnetism,[21] we find that the DL islands are blocked for

$$|\mu_0 H_L(\mathrm{DL})| \geqslant 1.2 \text{ T } [(T/100 \text{ K})/(A/100 \text{ nm}^2)]. \quad (4)$$

The mean DL island area in Fig. 1 is given by 70 nm^2, the maximum one by about 300 nm^2. We conclude that both superparamagnetically fluctuating and thermally stable

(blocked) islands are contained in typical samples. Because blocked islands cannot be remagnetized by the available fields, they are ineffective both in TOM and in MOKE. This explains in part the comparatively low values of m in Figs. 2(b), 3(b), 4(b), and 4(d). Because the details of the island size distribution depend sensitively on the step density and step morphology of the substrate and on the preparation conditions, and so does the fraction of blocked islands, strong differences between different preparations, in particular from different laboratories, are quite natural. During exposure, $|\mu_0 H_L|$ decreases and blocked islands therefore can enter the magnetometric window, resulting in an increase of m during exposure, which was actually observed in several samples (not shown).

The exchange length is given as $\lambda_{ex}=(2A_{ex}/J_s|H_L|)^{1/2}$ $=2.8$ nm/$(|\mu_0 H_L|/\mathrm{T})$. Using the exchange constant of bulk Fe, $A_{ex}=2\times10^{-11}$ J/m, and the anisotropy fields given above, we obtain $\lambda_{ex}(\mathrm{ML})=2.4$ nm and $\lambda_{ex}(\mathrm{DL})=3.4$ nm for ML and DL, respectively. The latter represents the DL as prepared, and it increases with increasing exposure. The accidental agreement in order magnitude of λ_{ex} with the lateral dimensions of the nanostructure as seen in Fig. 1, in combination with the rough coincidence of the DL island area with the blocking area, induces a rich variety of possible micromagnetic phenomena in the samples. Due to exchange coupling, the magnetization direction rotates continuously by 90° in an edge region of the width $\lambda_{ex}(\mathrm{ML})+\lambda_{ex}(\mathrm{DL})$. This reduces the mean value of the DL moment. In small islands or for enlarged values of $\lambda_{ex}(\mathrm{DL})$, the island magnetization remains nearly in plane. There is much similarity to the case of indirect antiferromagnetic coupling in a trilayer with finite size of the spacer, where a collapse to perfect ferromagnetic alignment despite antiferromagnetic coupling takes place below some critical size.[22] We similarly suggest in our present samples for islands below some critical size of the order $\lambda_{ex}(\mathrm{DL})$ a collapse to perfect in-plane magnetization, despite perpendicular anisotropy. We suggest that the strong changes of polar MOKE loops in Fig. 4(c) is caused by such effects via the increase of $\lambda_{ex}(\mathrm{DL})$ during exposure. Similar effects are expected in narrow ML channels, the magnetization of which is forced into the perpendicular direction. They therefore are expected to transfer ferromagnetic coupling, resulting in ferromagnetically coupled DL rafts. We suggest further that such narrow ML channels virtually decouple the attached ML sea regions. Laguna-like ML-sea parts might thus be formed, indicated in Fig. 1 by L, which then could behave superparamagnetically. It is further evident that the switching barrier of a ML laguna depends on the magnetization state of the fluctuating system of surrounding DL islands and vice versa. And the wall energy in the ML sea not only depends on the position of the wall between the islands, as has been discussed by Sander *et al.*[6] for the case of in-plane magnetized DL islands, but on the fluctuating magnetization state of the surrounding DL system too. We expect in general a complex dynamic coupling of the fluctuations in the ML and DL systems. The ML remagnetization then may take place in part by a kind of jumping driven by the superparamagnetic DL fluctuations. The high coercive fields up to 0.2 T observed by Sander *et al.*[6] would indicate a comparatively high fraction of large thermally stable DL islands. In contrast, we observed comparatively low longitudinal coercive fields (<0.02 T) using longitudinal MOKE, which can be explained by smaller islands. The frustration of long-range remanent magnetic order which we observed previously in Fe sesquilayers by spin-polarized low-energy electron diffraction (Ref. 7) and explained there tentatively by a virtually antiferromagnetic interaction between DL islands is now explained quite naturally from the spatially switching anisotropies. Apparently, the resulting interconnected fluctuations of in-plane ML and perpendicular DL elements continuously demagnetize the film in zero field.

In conclusion, we observed in pseudomorphic sesquilayers of Fe(110) on W(110) a spatial 90° switching of the anisotropy between easy plane in the ML to perpendicular in the DL, on a 10-nm scale, which coincides with the exchange lengths in both media. The perpendicular anisotropy in the DL apparently is caused by the pseudomorphic strain, strongly enhanced in comparison with expectations from linear magnetoelasticity. The exchange interaction between the partly fluctuating, partly blocked DL islands and the monolayer sea induces a rich variety of new micromagnetic phenomena which remain to be analyzed in detail. It would certainly be interesting to study magnetoresistance in these or related structures. The recently reported preparation of ultrathin W(110) substrates on Al_2O_3 (Ref. 23) might open a way for such investigations.

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