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Dipolar superferromagnetism in monolayer nanostripes of Fe(110) on vicinal W(110) surfaces

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By epitaxial growth of Fe on a vicinal W(110) substrate, densely spaced and continuous monolayer stripes of Fe(110) were prepared, directed along [001]. They exhibit a sharp phase transition to ferromagnetic order, free from relaxations. The magnetic easy axis is in the plane, but along [110] that means across the stripes. This cross magnetization induces ferromagnetic dipolar coupling between the spin blocks in adjacent stripes, which are preformed by exchange interactions. The resulting superferromagnetic phase transition is therefore driven by dipolar interactions. [S0163-1829(98)52002-4]

Whereas magnetic phase transitions in three-dimensional (3D) crystals are dominated by exchange interactions, magnetic phase transitions in 2D systems are characterized by the interplay of exchange interactions, dipolar interactions, and surface-type anisotropies. In particular, magnetic long-range order has to be stabilized by magnetic anisotropies.[1,2] Motivated by experiments with films of perpendicular uniaxial anisotropy,[3,4] the theoretical literature has focused on the case of perpendicular surface anisotropy, where the competition with dipolar interactions (shape anisotropy) results in a rich variety of inhomogeneous magnetization structures.[5–8] However, it has been known for a long time that in the pseudomorphic monolayer Fe(110) on W(110),[9–11] outstanding structural quality is combined with strong uniaxial surface anisotropy, with an in-plane easy axis.[1–10] Because the competition of shape and surface anisotropy is avoided, critical behavior could be analyzed experimentally in this system with precision, and predictions of the 2D Ising model (2DIM) could be confirmed.[12] The structural quality of the system allowed a study of 2D magnetic percolation in the submonolayer regime, and to gain first data on magnetic ML stripes, grown from the (irregular) atomic steps of the W(110) substrate.[13] Recent advantages in preparing regular systems of parallel metallic nanostripes by growth on a vicinal substrate[14,15] open the opportunity to more systematically investigate magnetic order in monolayer stripes. In the present paper, we report on selected results of an extended study of Fe(110) monolayer stripes prepared on vicinal W(110), to be published in detail elsewhere. In order to obtain smooth coherent stripes, the substrate steps and the stripes on them were intentionally oriented along [001]. Contrary both to what is predicted by Monte Carlo (MC) simulations of the single stripes, and to results for Fe(111) stripes on Cu(111),[16] our samples show a quite sharp and relaxation-free ferromagnetic phase transition. This is a consequence of the easy axis being along [001], that means in the plane but across the stripes, which induces ferromagnetic coupling between adjacent stripes. As shown by MC simulations,[17–19] the single stripes are subdivided into full stripe width spin blocks with alternating sign of magnetization, and a length which continuously increases with decreasing temperature. The phase transition then developed from the combined action of exchange interaction which forms the blocks, with the anisotropy which enforces cross magnetization, and the dipolar interaction between spin blocks in adjacent stripes which finally drives the transition. Because these blocks take a similar role as the single domain particles of a superparamagnet, we call the ordering phenomenon dipolar superferromagnetism. To our knowledge, this type of magnetic ordering has not been observed before.

Experiments were done in UHV (base pressure below 10^{-10} Torr). As a substrate, we used a vicinal W(110) surface, polished *ex situ* and cleaned *in situ* in oxygen, resulting in an Auger-clean W(110) surface showing a $p(1\times1)$ low-energy electron diffraction (LEED) pattern. The surface normal deviates from [110] by $1.4°\pm0.1°$, resulting in atomic steps along the [001] direction ($\pm10°$), with a mean separation $w_0=9.1\pm0.6$ nm. The surface of the vicinal W(110) substrate therefore consists of terraces with an average width w_0, composed of $W_0=w_0/(2^{1/2}a_w)=40$ atomic rows ($a_w=0.316$ nm). However, there was a considerable distribution of widths, with a full width at half maximum given by $\Delta w_0/w_0=30\pm5\,\%$. Fe was deposited at room temperature from a resistively heated BeO crucible, with a rate of 0.3 ML/min, at a pressure of 2×10^{-10} Torr. Coverage Θ was monitored by a quartz balance and checked by scanning tunneling microscopy (STM), which could be done *in situ* at another stage of the system. As a consequence of the transferability of the sample, the thermocouple could not be fixed to the sample itself, but only to the sample holder, and the sample temperature had to be calibrated using the Curie temperature of an extended monolayer (on a separate sample). Absolute temperatures therefore could be obtained only with an error of about ±10 K. However, relative temperatures were measured with an accuracy of ±2 K. Annealing of the samples at 800 K for 3 min resulted in continuous nanostripes of monatomic height, as shown in the STM image of Fig. 1. All stripes are pseudomorphic with the W(110)

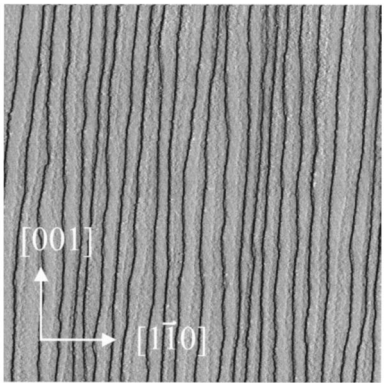

FIG. 1. STM image (250×250 nm) of a vicinal W(110) surface with steps along [001], covered by 0.5 pseudomorphic monolayer of Fe. Growth conditions as given in the text. In order to enhance the contrast the image has been differentiated. The lighter colored stripes adjacent to the step edges are the Fe nanostripes.

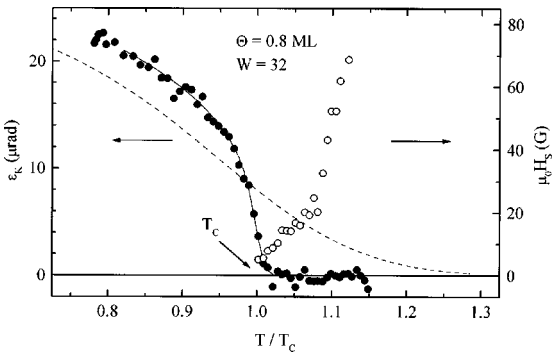

FIG. 3. Remanent ellipticity $\varepsilon_{K,r}$ (full circles, for $T<T_C$) and saturation field H_s (squares, for $T>T_C$) as a function of temperature T, for the sample of Fig. 2. The full line represents a fit by a power law of exponent $\beta=0.32$, convoluted with a Gaussian distribution of mean value $T_C=179$ K and width of $\Delta T_C=1.7$ K. The dashed line shows the same function but with a broader distribution of width $\Delta T_C=15$ K, as would be expected for noninteracting stripes.

substrate. Because the width w of the Fe stripes is given by $w=\Theta w_0$, it shows the same distribution width $\Delta w/w=30\pm5\,\%$ as the substrate terraces.

Magnetic properties were studied in the preparation stage by Kerr magnetometry. Using a compensation technique,[19] we measured the Kerr ellipticity ε_K in absolute units, as a function of temperature T and external magnetic field H. It is crucial for the magnetic state of the system that the magnetic easy axis in the ML Fe/W(110) is along [110], with strong anisotropy fields of the order of 5 T[12,13] Accordingly, the external field was applied along [110], across the stripes. Magnetization loops were measured during warming up with a rate of roughly 1 K/min, after liquid nitrogen cooling to about 140 K. One loop was measured in 40 sec, thus limiting the temperature resolution to 1 K. Figure 2 shows loops for a sample with $W=32$ ($\Theta=0.8$). Temperatures are given in units of the Curie temperature $T_C=179$ K of the sample. At the lowest temperature, $T=0.82T_C$, we observe a typical easy axis loop, with switching between two single domain states at the coercive field $\mu_0 H_c=50$ G. The loop confirms that the easy axis is along [110], across the stripe, in spite of the shape anisotropy, which would induce parallel magnetization. The easy axis loop still is obtained at $0.96T_C$. Above a rather sharp transition at T_C, the loop cold be fitted by the law of a uniaxial superparamagnet, $\varepsilon_K(H)=\varepsilon_{K,S}\tanh(H/H_s)$, with surprisingly low saturation fields H_s which are shown in Fig. 3 as a function of T and will be discussed below. For $T<T_C$, the remanent value $\varepsilon_{K,r}(T)$ was fitted, following the ansatz of Ref. 12, by a power law of exponent $\beta=0.32$, with a Gaussian distribution of T_C values, with a mean value $T_C=179.1$ K and a surprisingly narrow width of $\Delta T_C=1.7$ K; see Fig. 3, which indicates a true equilibrium phase transition. For samples other than $W=32$, with $W=20$, 24, 28, the width of the transition was comparable. The Curie temperatures are presented in Fig. 4. To further check for the equilibrium nature of the transition, we searched for relaxations in the following experiment. After saturating the sample at 100 Oe for 1 sec, the field was switched off, and the remanent value $\varepsilon_{K,r}$ was measured at times of 500 and 1000 ms after switching. In the whole criti-

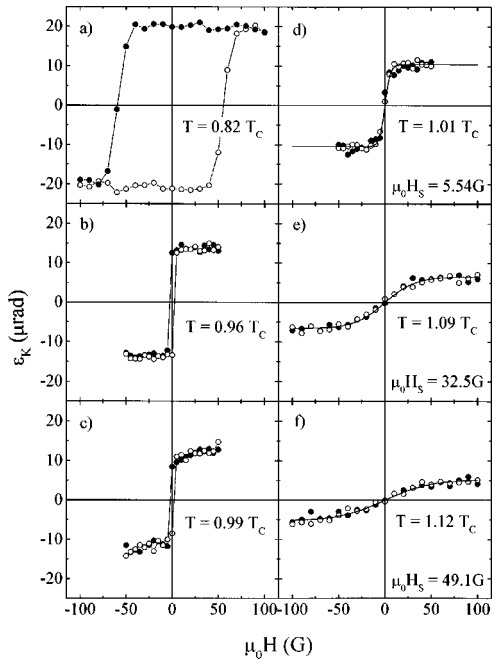

FIG. 2. Kerr ellipticity ε_K as a function of field H along [$\bar{1}$10] for a system of ML stripes of width $W=32$ (Fe coverage $\Theta=0.8$ ML), for various temperatures T which are given in units of the Curie temperature $T_C=179$ K of the sample. Full and open circles are for decreasing and increasing fields, respectively.

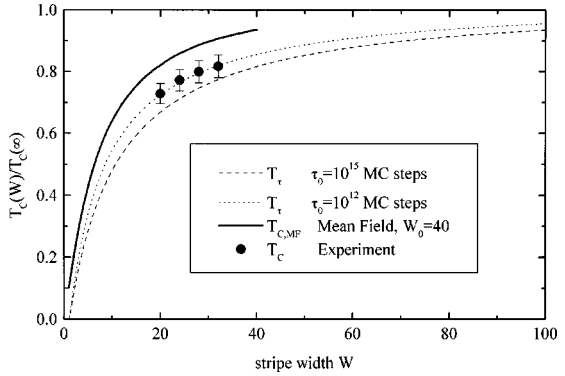

FIG. 4. Critical temperatures as a function of stripe width W, in comparison with the Curie temperature of the extended monolayer $T_C(\infty)$. Broken and dotted lines represent freezing temperature T_τ, for experimental time scales $\tau_0=10^{15}$ and 10^{12} sec, respectively. The full line represents the mean field T_C as determined from Eq. (6). Full points represent experimental data; relative temperatures with an error corresponding to the dot size; absolute temperatures with an error of ±10 K$=0.05T_C(\infty)$.

cal temperature regime, the difference was zero in our error limits of 5% of the saturation value, indicating the absence of relaxation in a window of relaxation times between 1 and 10 sec. This confirms the equilibrium nature of the transition.

Both the narrow width and the absence of relaxations are in contradiction to what would be expected for independent stripes. First, MC simulations[18,20] of the 2DIM confirm that any stripe of finite width becomes ferromagnetic not by a true phase transition but by freezing at some freezing temperature T_τ, where the relaxation time τ, increasing with decreasing temperatures, becomes equal to some experimental time scale. The relaxation window checked above should necessarily be met, which is not the case. But even if T_τ were observed as a kind of effective Curie temperature, its considerable dependence on stripe width w,[13] in connection with the wide distribution $\Delta w/w$, would induce, for the case of independent stripes, a broadened transition with a width of $\Delta T_C = 15$ K, as indicated in Fig. 3 by the dotted line. A width of this order has been observed for the widely spaced stripes investigated previously.[13,12] In the present case of narrowly spaced stripes, we observe a narrowing of the transition by one order of magnitude. In the following we show that this narrowing is a result of the dipolar coupling, which is induced between the narrowly spaced stripes by their cross magnetization.

Any explanation of the coupling between neighboring stripes must be based on an understanding of the spin dynamics in a single stripe. Because these internal dynamics are governed by exchange field of the order of 10 to 100 T, they are independent of a superimposed dipolar coupling with coupling field below 0.1 T. Because the phase transition of the extended monolayer could be described quite well by the 2DIM,[13,12] we use this model for our uniaxial stripes too. Monte Carlo simulations for a 12×1440 stripe[17] (i.e., stripe of 12×1440 spin sites) showed that below $T_C(\infty)$ the stripe decomposes into spin block of full width W, with a length L which increases with decreasing T. These full width spin blocks act in a similar manner as the (macro)moments of an Ising chain. The magnetization decays exponentially at any finite temperature, and no remanent order is left. However, the relaxation time τ exponentially increases with decreasing T, thus giving rise to an effective freezinglike transition towards a permanently magnetized state, at a (freezing) temperature $T_\tau(W)$, where τ equals the experimental time scale τ_0. It has been show previously[18] that the observed ordering temperatures in widely separated monolayer stripes can be understood in terms of $T_\tau(W)$. Recent MC simulations of $W\times10\,000$ stripes[20] result in an interpolation formula for τ which is given, in units of MC steps per site (about 1 ps), by

$$\tau = C_\tau \exp\{a\{W[(T_C(\infty)/T) - p] - (T_C(\infty)/T)q\}\}, \quad (1)$$

with $C_\tau = 658$, $a = 3.13$, $p = 0.97$, $q = 1.06$. For an interpretation we use $2J = 0.884 k_B T_C(\infty)$ (nearest-neighbor coupling constant J).We find that the activation energy $[3.13(W - 1.06)k_B T_C(\infty)] = [3.54(W - 1.06)2J]$ is not far from the energy of a half circle domain wall of radius W $[\pi W 2J]$. Such a half circle is not unreasonable as a model for the critical nucleus for a new spin block. The last term, $1.06 \times 2J$, can be interpreted as a boundary correction. We conclude that τ is governed by the activation energy for spin block generation. In our experiment the typical time scale is 1 sec, corresponding to $\tau_0 = 10^{12}$ MC steps. Equating τ_0 with τ, we find from Eq. (1) the freezing temperature $T_\tau(W)/T_C(\infty) = 3W/[\ln(\tau_0/C_\tau) + 3W]$. For large W, this equals $(1 - 7/W)$, in agreement with the finite-size scaling form proposed earlier.[13] $T_\tau(W)/T_C(\infty)$ is visualized in Fig. 4 by the broken lines.

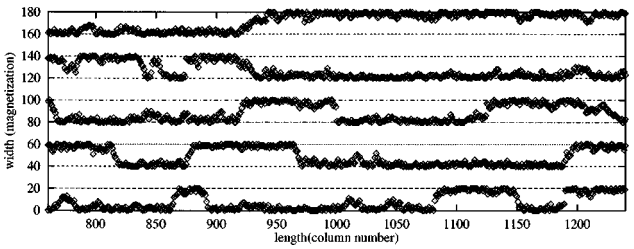

FIG. 5. Section (500×180) from a MC snapshot of a sample with $W = 20$, in a periodic model of noninteracting stripes. Alternating rows of width 20 represent either the Fe stripes or the bare tungsten stripes between. The Fe stripes are structurized by a representation of their magnetization; see text.

The formation of spin blocks provides a qualitative explanation for the low saturation fields (5 mT at $T = 1.1T_C$), which are two orders of magnitude smaller than those observed for extended Fe monolayers on smooth W(110) [400 mT at the same relative temperature (see Ref. 10) or 2DIM simulations]. Let $m_L = WL\mu_{\mathrm{Fe}}$ be the magnetic moment of one full width spin block of length L, with μ_{Fe} being the average moment per site in this block. Considering the spin blocks as independently fluctuating elements, the thermal average m of the component along H follows the law of a uniaxial superparamagnet,

$$m/m_L = \tanh\{m_L H/k_B T\}. \quad (2)$$

For the saturation field $H_s = m_L/(dm/dH)$ we obtain $H_s = k_B T/m_L = (k_B T/\mu_{\mathrm{Fe}})/(WL)$, which qualitatively explains the low values. However, the proportionality to $(T - T_C)$, which is seen in Fig. 3 just above T_C, is not reproduced. This is a result of dipolar coupling between the stripes and the spin blocks in them. They are visualized in Fig. 5 by the MC snapshot of a (480×180) section of a sample with $W = 20$, in a periodic model of noninteracting stripes. The figure consists of rows of width 20, alternatingly representing either the Fe stripes of width $W = 20$, or the bare tungsten stripes of width $W_0 - W = 20$ between. The Fe stripes are structurized by a representation of the ''magnetization,'' more precisely speaking the magnetic moment per atomic row of width 20, switching between ±10 for complete up and down magnetization, respectively. The cross magnetization results in magnetostatic coupling of adjacent stripes. A quantitative treatment of this coupling in the heavily fluctuating system is beyond the scope of this paper. For a qualitative understanding, we note that the length L of the blocks is of the order of 10^2 to 10^3, much larger than $W < 40$. The coupling of neighboring blocks in one stripe, therefore, can be neglected in comparison with the coupling between neighboring stripes, which dominates the system. What can be easily calculated is the dipolar (stray) field $H_{D,0}$, which one stripe in a magnetically saturated film would feel from all other (parallel magnetized) stripes. $H_{D,0}$ is roughly proportional to W/W_0. For the example of Figs. 2–4, $W = 32$ on $W_0 = 40$, one calculates $\mu_0 H_{D,0} = r191$ G, where r

$=\mu_{\mathrm{Fe}}/2.2\mu_B$ is a correction factor taking into consideration the variation of μ_{Fe}. We roughly estimate the reduced coupling field H_D in the fluctuating system by a mean-field ansatz $H_D=(m/m_L)H_{D,0}$, resulting in

$$m/m_L=\tanh\{m_L[H+(m/m_L)H_{D,0}]/k_BT\}. \tag{3}$$

Using standard procedures of mean-field theory, this results in a saturation field

$$H_s=H_{D,0}[(T-T_{C,\mathrm{MF}})/T_{C,\mathrm{MF}}], \tag{4}$$

where $T_{C,\mathrm{MF}}=m_LH_{D,0}/k_B$ is a mean-field Curie temperature. This is just the mean-field behavior observed in the critical regime of Fig. 3, where we find $\mu_0H_s=(240\ \mathrm{G})\ [(T-T_C)/T_C]$ (experimental Curie temperature T_C), in good qualitative agreement with $\mu_0H_{D,0}=r\times 191$ G. This clearly confirms our dipolar interpretation of the ordering. Quantitative agreement could not be expected in view of the crude mean-field approximation.

We use the average value $\langle L(T)\rangle=\Sigma L_1^2/\Sigma L_i$ as a measure for the length L of a full width spin block. The relation $T_{C,\mathrm{MF}}=m_LH_{D,0}/k_B$ then provides an implicit equation,

$$k_BT_{C,\mathrm{MF}}=r^2 2.2\mu_BH_{D,0}W\langle L(T_{C,\mathrm{MF}})\rangle \tag{5}$$

for an estimate of $T_{C,\mathrm{MF}}$ from the temperature dependence of $\langle L\rangle$. From MC simulations,[20] we obtained numerical values of $\langle L\rangle$ which could be fitted in analogy to Eq. (1) by

$$\langle L(T)\rangle=C_L\ \exp\{0.884W[(T_C(\infty)/T)-1]\}, \tag{6}$$

with $C_L=50\pm 30$. Note that this is an Arrhenius law for the density of walls of energy $[0.884k_BT_C(\infty)W]$. Numbers for $T_{C,\mathrm{MF}}(W)$ are shown in Fig. 4 in comparison with freezing temperatures T_τ and experimental values T_C, which agree with $T_{C,\mathrm{MF}}$ only roughly. This is not surprising in view of both the crude mean-field approximation and of the uncertainty of T_C, as discussed above and indicated by the error bars. The crucial point is that $T_{C,\mathrm{MF}}$ is larger than T_τ. This means that for decreasing temperatures the system orders by dipolar coupling before it could freeze, and the ordering phenomenon is really dipolar superferromagnetic, as stated in the title. The narrow width of the transition is reasonable as a consequence of the long-range order of the dipolar coupling fields.

In conclusion, we observed a new type of magnetic phase transition in narrowly spaced monolayer stripes of Fe on W(110). The transition is sharp and free from relaxations. Due to the magnetic easy axis being in the plane but across the stripes, there is magnetostatic interstripe coupling of ferromagnetic sign between the full stripe width spin blocks in adjacent stripes, which in turn are preformed by intrastripe exchange coupling. The dipolar interstripe coupling finally drives the transition. Because the length of the blocks rapidly increases with decreasing temperature, and the coupling is proportional to their length, the phase transition takes place when the block length reaches some critical length, typically of the order of 1000 atomic distances. The ordering phenomenon is related to previously observed superferromagnetism in nanosized, exchange-coupled island systems.[21] However, it shows clear differences, both because the coupling entities, given by the spin blocks, are not structurally preformed, and even fluctuate above the transition both in position and in size, and because the coupling is not given by exchange but is pure dipolar. Despite a considerable scatter in stripe width, the transition is sharp, due to the long-range nature of the dipolar coupling fields. Our case is quite different from that of Fe stripes on a Cu(111) vicinal surface,[16] where well-expressed relaxations were observed in a wide temperature range. The differences in the critical behavior result from differences of morphology and of magnetic anisotropy. The Fe strips on Cu(111) show relaxations because they are formed by chains of weakly coupled segments, and because their perpendicular magnetization results in antiferromagnetic magnetostatic coupling of adjacent stripes, whereas our Fe stripes on W(110) are relaxation free because they are continuous, and because they are subject to ferromagnetic magnetostatic coupling, due to their in-plane, cross stripe magnetization. We consider the transition in our stripe system as a model prototype for the interplay of magnetostatic coupling with exchange coupling and anisotropies in 2D systems, which has only recently been attacked by MC simulations,[7] though it is always present, but difficult to handle and therefore neglected in many theoretical models.

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