Real-Space Observation of Dipolar Antiferromagnetism in Magnetic Nanowires by Spin-Polarized Scanning Tunneling Spectroscopy

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We have performed spin-polarized scanning tunneling spectroscopy of dipolar antiferromagnetically coupled Fe nanowires with a height of two atomic layers and an average separation of 8 nm grown on stepped W(110). Domain walls within the nanowires exhibit a significantly reduced width when pinned at structural constrictions. The lateral spin reorientation in the direction perpendicular to the wires has been studied with subnanometer spatial resolution. It is found that the spin canting in the Fe nanowires monotonously increases towards the step edges.

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Driven by the demand for ever higher density in data storage media in the past a strong effort has been undertaken in the preparation and characterization of magnetic nanostructures of the desired size and shape [1]. Recently, the interest in magnetic nanostructures has focused on perpendicularly magnetized Fe nanowires grown on slightly miscut single crystals [2-4]. While the structural and electronic properties have frequently been observed by means of scanning tunneling microscopy (STM) and spectroscopy (STS), i.e., with an experimental technique that allows real-space imaging with a resolution limit down to the atomic level, the magnetic properties have mostly been investigated by spatially averaging techniques like, e.g., magneto-optical Kerr effect. In particular, for perpendicularly magnetized Fe nanowires grown on stepped W(110) which exhibit a width of only 4 nm and a periodicity of 8 nm, a complex magnetic behavior has been found which was explained by a magnetic anisotropy which changes discontinuously on a nanometer scale [3-5]. However, the minimum distance required to change the magnetization direction is determined by the exchange length $L = \sqrt{A/k}$, where A is the so-called exchange stiffness and k the anisotropy constant. It was found that the spin rotation within Fe nanowires on such a narrow lateral scale cannot be explained on the basis of bulk properties [4,5]. Instead, it was assumed that the exchange stiffness A of the first and second Fe layer on W(110), i.e., A_{ML} and A_{DL} , is 1 order of magnitude smaller than in bulk Fe-an assumption which has so far not been verified by experimental results.

In this Letter we report on the direct observation of the magnetic domain structure of Fe nanowires grown on stepped W(110) by means of spin-polarized scanning tunneling spectroscopy (SP-STS) [6,7]. Our results unambiguously show that—in agreement with the explanation mentioned above [3-5]—the domain structure is governed by perpendicularly magnetized Fe nanowires with the magnetization pointing alternatingly up and down. Additionally, the high spatial resolution of SP-STS allows the evaluation of details of the spin reorientation both along and perpendicular to the nanowires, on a subnanometer scale. In particular, a detailed investigation of the domain wall width *w* revealed two different types of walls. While relatively broad domain walls were found in homogeneous double-layer (DL) wires ($w = 6 \pm 1$ nm), much narrower ones appear at structural constrictions ($w^* = 2 \pm 1$ nm). The broad domain walls of DL wires are consistent with a value of A_{DL} that is very close to the Fe bulk value of $A = 1 \times 10^{-11}$ J/m. The reduced width of domain walls that are pinned at structural constrictions is found to be in good agreement with a recent theoretical prediction [8].

The experiments have been performed in an ultrahigh vacuum (UHV) system with separate chambers for substrate preparation, sample transfer, metal vapor deposition (MVD), surface analysis, and cryogenic STM equipped with a 2.5 T magnet [9]. The base pressure in each chamber is in the low 10^{-11} torr range. The W(110) single crystal is miscut by 1.6° with respect to the (110) plane. This substrate is prepared by numerous cycles of long-term heating at 1500 K in an oxygen atmosphere of 10^{-7} -10⁻⁶ torr and subsequent flashing up to 2500 K [10]. We used etched W tips which were flashed *in vacuo* to remove oxide layers. In the MVD chamber the tips were magnetically coated with Gd while held at 300 K, subsequently annealed at $T \approx 550$ K for 4 min, and then transferred into the cryogenic STM. During the measurements, the tip and sample were at a temperature T = 16 K.

The growth of Fe/W(110) has been intensively investigated in the past [11–14]. Our Fe films were grown at T =300 K at a rate of 0.6 monolayers per minute (ML/min) and subsequently annealed at $T \approx 520$ K for 4 min leading to step flow growth. The Fe film grows pseudomorphically, i.e., expansively strained by about 10%, as long as the terrace width remains below the critical Fe double-layer width for misfit dislocation formation of 9 nm [3,13]. The miscut of the W(110) substrate used in this study results in an average terrace width of 8 nm. Indeed, the constant current topograph of 1.5 ± 0.1 ML Fe/W(110) as shown in the inset of Fig. 1(a) is dominated by atomically flat terraces and monatomic steps of a nominal height of 2.24 Å. The sample surface is free of any misfit dislocations. Schematically, the structure of the sample is represented below the line section of Fig. 1(a). It consists of



FIG. 1. (a) Line section and topography (inset) of 1.5 ML Fe/W(110) showing terraces with an average width of 8 nm. The structure of the sample is schematically represented. (b) The tunneling spectra of ML and DL stripes exhibit peaks at U = +0.40 and U = +0.68 V, respectively. While no differences in the spectra were found between different Fe DL stripes with W tips two quantitatively different spectra were measured when using Gd-coated tips. (c) A map of the dI/dU signal (U = 0.68 V) reveals that the spectra alternate between adjacent Fe DL stripes being caused by an AFM dipolar coupling. Two ferromagnetically coupled nanowires and domain walls within single nanowires are marked by black and white arrows, respectively.

alternating ML and DL Fe stripes. Since in our experiment the substrate step direction deviates from the [001] direction, the width of the nanowires is not homogeneous but slightly fluctuates along the step edges.

By using black and white arrows we have also represented the magnetic structure of mono- and double-layer stripes as recently proposed by Elmers, Hauschild, and Gradmann [3-5]. Combining longitudinal and polar Kerreffect measurements, an onset of perpendicular magnetization for Fe coverages $\Theta \ge 1.1$ ML was found [4]. The coverage range between 1.4 and 1.8 ML Fe/W(110) is characterized by magnetic saturation at relatively low external perpendicular fields combined with the absence of a hysteresis, i.e., zero remanence [3]. It has been argued [3] that this experimental result is caused by the perpendicularly magnetized Fe DL stripes which prefer to occupy a demagnetized ground state by antiferromagnetic (AFM) dipolar coupling, i.e., by periodically changing the magnetization direction between adjacent DL stripes. Since, however, all available magnetic information was based on spatially averaging experiments, many interesting questions on nanomagnetic details remained unanswered.

Before we turn our attention to SP-STS measurements we have to understand the electronic properties of the sample. The upper panel of Fig. 1(b) shows tunneling dI/dU spectra measured with a bare (nonmagnetic) W tip taken above Fe ML and DL stripes. As already mentioned in an earlier STS study performed at 300 K, both the Fe ML as well as the DL exhibit characteristic tunneling spectra [14]. In the present low-temperature experiment, we found peaks at U = +0.40 V for the ML and U = +0.68 V for the DL [15]. We would like to emphasize that—within our measurement accuracy—we found the same spectra above any ML or DL stripe, respectively.

In a second set of experiments we investigated the nanomagnetic structure of the Fe DL stripes by means of SP-STS. According to Refs. [3,4] the Fe DL stripes are perpendicularly magnetized. One crucial requirement for obtaining a magnetic contrast in SP-STS measurements is an appropriate magnetization direction of the tip. For the purpose of this study we used W tips which were coated by 8 ± 1 ML Gd. Bulk Gd is ferromagnetic below its bulk Curie temperature $T_{CB} = 292.5$ K. It is well known that Gd films with a thickness of 8 ML are ferromagnetic for $T < 0.7T_{CB}$ [16] and exhibit a perpendicular easy axis at $T < 0.6T_{CB}$ [17]. Since both phase transition temperatures are far above our measurement temperature of 16 ± 1 K we can safely conclude that the tip is ferromagnetic with the magnetization vector pointing along the tip axis, i.e., perpendicular to the surface plane. As already shown in previous publications [6,7], the conductivity between the two magnetic electrodes depends on the electron density of states within a particular energy range given by the applied bias voltage and on the sign and the magnitude of the electron spin polarization.

The lower part of Fig. 1(b) shows dI/dU spectra which were measured using a Gd-coated probe tip. With the Gd

tip positioned above the Fe ML we found spectra which are qualitatively identical with the spectra measured with bare W tips. In contrast, we found two different types of spectra above the DL stripes which will hereafter be referred to as $\uparrow\uparrow$ and $\uparrow\downarrow$. Both spectra exhibit a peak at the same energetical position already mentioned above, i.e., U = +0.68 V, but differ in intensity. While the differential conductivity at the peak position amounts to only dI/dU = 1.3 nA/Vfor the spectra of type $\uparrow\uparrow$ it is enhanced by about 40% to dI/dU = 1.8 nA/V for type \downarrow . The relative intensities between both types of spectra invert for U < 0.5 V. These differences in the tunneling spectra of the DL Fe stripes are caused by spin-polarized tunneling between the magnetic tip and Fe DL stripes being magnetized either parallel or antiparallel to the tip. In Fig. 1(c) we have plotted a map of the differential conductivity dI/dU at the peak position (U = 0.68 V). Different intensities of the dI/dU signal show up as different grey levels. Since the dI/dU signal at U = 0.68 V is much lower for the monolayer than for double-layer stripes the former appears black. Furthermore, the data reveal that most double-layer stripes exhibit only one type of spectrum, either $\uparrow\uparrow$ or $\uparrow\downarrow$, and that the type alternates between adjacent stripes. This observation is consistent with the proposed AFM out-of-plane coupling of adjacent stripes [3-5]. In Fig. 1(c) we have marked some exceptions from this rule by arrows demonstrating the impact of the high spatial resolution of SP-STS. Approximately in the middle of the image one can recognize two adjacent stripes which exhibit the same dI/dU signal (black arrows). Obviously, these stripes are so close together (d < 1.5 nm) that the exchange coupling overcomes the energy gain due to an AFM coupling. Furthermore, at the very right edge of the image we can find two stripes which change the type of spectrum from the bottom to the top part of the image, i.e., both stripes exhibit a domain wall (white arrows).

We have shown so far that the DL peak position (U =0.68 V) is particularly suited for the imaging of magnetic domains since the contrast between the spectra $\uparrow\uparrow$ and $\uparrow\downarrow$ is maximum. Another bias voltage which allowed imaging of the magnetic domain structure with high contrast was U = -0.3 V. In the following we have not taken full tunneling spectra at every pixel of an image which requires a measurement time of about 10 h per image. Instead, we have only measured the dI/dU signal at the voltage values given above. This reduces the measurement time to about 30 min for an image with (500 \times 500) pixel. Figure 2 shows two different types of domain walls which have been observed within the DL stripes. Relatively broad domain walls with a width $w_0 = 6 \pm 1$ nm were found in homogeneous DL stripes [18]. This finding is in strong disagreement with a recent publication, in which the DL exchange length was estimated to $L_{\rm DL} = 0.5$ nm based on Kerr-effect measurements [4]. Instead, using the definition $w_0 = 2L = 2\sqrt{A/k}$ [8] our results suggest that $L_{\rm DL} = 3$ nm. With $k_{\rm DL} = 1 \times 10^{-6}$ J/m³ [4] this leads to $A_{\rm DL} = 9 \times 10^{-12}$ J/m which almost perfectly agrees



FIG. 2. Line sections showing the change of the dI/dU signal when crossing a domain wall being located in a smooth (upper panel) or constricted (lower panel) Fe DL stripe. Maps of the dI/dU signal are shown in the inset. The positions at which the line sections were drawn are marked by solid black lines.

with the bulk value $A = 1 \times 10^{-11}$ J/m. At structural constrictions which often serve as pinning centers for domain walls we found, however, narrower domain walls as can be seen in the line section shown in the lower panel of Fig. 2. Typically, the width of domain walls being pinned at structural constrictions amounts to $w^* = 2 \pm 1$ nm. This behavior has recently been proposed theoretically by Bruno [8]. We have applied Bruno's approach by modeling the width of the DL stripe S(x) by the quadratic approximation $S(x) = S_0(1 + \frac{x^2}{d^2})$ (model II in Ref. [8]). Here, S_0 is the minimum width of the constriction positioned at x = 0, x is the distance from minimum, and d is a fit parameter. Indeed, in our case the constriction could well be fitted by using $S_0 \approx 0.8$ nm and d = 1 nm which results in a reduced domain width $w^* = \frac{8d}{\pi} \approx 2.5$ nm being in fair agreement with the experimental observation.

Another point of interest is how the spin orientation changes when crossing the DL stripes perpendicularly. Figures 3(a) and 3(b) show details of the topography and dI/dU signal (U = -0.3 V) of 1.5 ± 0.1 ML Fe/W(110). The AFM coupling between adjacent Fe DL stripes appears as different grey levels in Fig. 3(b). Since we have never found any magnetic contrast in the monolayer stripes with Gd-coated tips, we have adjusted the z scale of the line section in Fig. 3(c) to allow a high sensitivity on the dI/dU signal of the DL stripes only. Obviously, the dI/dU signal of the dark DL stripes monotonously decreases when moving away from the ML-DL transition while the dI/dU signal of the bright DL stripes monotonously increases. Since the electronic



FIG. 3. (a) Topography and (b) dI/dU signal at U = -0.3 V as measured with a Gd-coated probe tip. Because of the fact that at this particular sample bias the dI/dU signal of the $\uparrow\uparrow$ DL stripes is almost equal to the ML, both appear with a similar grey level [cf. Fig. 1(b)]. (c) Line section drawn across the nanowires. Schemes of the lateral spin reorientation are shown in (d) (taken from Ref. [5]) and (e) (new model proposed).

properties of all double layers are identical (cf. Fig. 1), this behavior can be explained only by a magnetic effect. In this context it is worthwhile to compare the result of Fig. 3(c) with a detailed model of ultrathin films with laterally modulated anisotropies which has been proposed by Elmers and co-workers [4,5]. As schematically represented in Fig. 3(d), it has been suggested that for a coverage of 1.5 ML Fe/W(110) and a DL stripe width of 4 nm a continuous spin rotation occurs from in plane in the middle of the ML stripe to out of plane in the middle of the DL stripe. The line section of Fig. 3(c) reveals, however, that the maximum contrast is not obtained in the middle of the DL stripe but at the descending step edges. Based on the available data we propose the following picture of the spin structure which is also schematically represented in Fig. 3(e): (i) The Fe ML stripes exhibit no out-of plane magnetization even very close (<3 Å) to the transition to the DL stripes. (ii) Even very close to the transition to the ML (<3 Å) the spins in the DL stripes

are canted by about 30° to the surface plane. (iii) The canting increases monotonously towards 90° with respect to the surface plane when approaching the step edges. We can conclude that in spite of the fact that $A_{DL} \approx A_{vol}$ the AFM coupling between adjacent DL stripes being separated by ML stripes of only 4 nm width becomes possible because the spin rotation occurs close to the ML-DL transition.

In summary, we have imaged the domain structure of antiferromagnetically coupled Fe nanowires with a thickness of only 2 ML. Two different types of domain walls were found within the wires: though smooth nanowires exhibit walls with a width of about 6 nm, structural constrictions lead to a much smaller wall width of about 2 nm. The lateral spin reorientation in the direction perpendicular to the wires has been investigated in detail and compared to a theoretical model. Our SP-STS results reveal that the spin canting within the Fe nanowires is nonsymmetric and monotonously increases towards the step edges.

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