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Magnetism of nanoscale Fe islands studied by spin-polarized scanning tunneling spectroscopy

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We have performed low-temperature ($T = 15$ K) spin-polarized scanning tunneling spectroscopy of nanoscale Fe islands with a height of two atomic layers, grown on a stepped W(110) surface and surrounded by a closed monolayer (ML) Fe. These islands are single domain particles up to a coverage of 1.5 pseudomorphic ML and keep an antiferromagnetic out-of-plane ordering far beyond the onset of their coalescence. For small islands we observe a reorientation to in-plane magnetization between 2 and 3 nm island width due to exchange coupling to the in-plane magnetized ML.

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The investigation of magnetic nanostructures has been brought into focus both by the demand for high density magnetic data storage media as well as to gain improved insight into the magnetic phenomena governing systems of reduced dimension. In the past, nanomagnetic systems have been investigated intensively by laterally averaging techniques, often assisted by scanning tunneling microscopy (STM) for a structural and electronic analysis. With such an approach, however, details of the magnetic domain structure remain unknown and a correlation of structural and magnetic properties is difficult, if possible at all. In contrast, spin-polarized scanning tunneling spectroscopy (SP-STS) has been developed as a reliable tool to investigate magnetism down to the atomic level[1] and thus allows to simultaneously access and correlate the sample's structural, electronic, and magnetic properties on a nanometer scale.[2] As a model system for such an investigation we regard ultrathin pseudomorphic Fe films on W(110) in a coverage regime around 1.5 monolayers (ML).[3,4] Room temperature (RT) growth of Fe on W(110) leads to doublelayer (DL) islands surrounded by a single ML Fe, with island sizes of a few nanometers. For this system Weber *et al.*[5] suggested a model of spatially switching anisotropy where the easy axis of magnetization changes discontinuously from in-plane in the ML to out-of-plane in the DL islands. From the competition of anisotropy and exchange energy a complex magnetic ordering is to be expected, the details of which could, so far, not be investigated directly.

In the present work we report on the direct imaging of the magnetic domain structure of nanoscale Fe DL islands surrounded by a single ML Fe, grown on W(110) at RT in a total coverage regime between 1.2 and 2.1 ML Fe by means of SP-STS. We observe an antiferromagnetic (AFM) ordering of DL islands with a magnetization direction perpendicular to the surface. This ordering persists up to a coverage of 2.1 ML, well above the onset of island coalescence and strain relaxation. Whereas small islands are in a single domain state, extended islands beyond a length of ≈30 nm exhibit domain walls which are always oriented along the $[\underline{1}10]$ direction. A detailed analysis of the data reveals a vanishing of the magnetic out-of-plane contrast for islands below 2–3 nm island width. This result is explained by a reorientation transition of small DL islands to in plane, driven by exchange coupling to the in-plane magnetized ML.

The experiments have been performed in an ultrahigh vacuum system consisting of separate chambers for substrate preparation, metal vapor deposition, surface analysis, sample transfer, and cryogenic STM ($T_{\text{sample}} = T_{\text{tip}} = 15$ K).[6] The base pressure in each chamber is in the low 10^{-11} torr range. The W(110) substrate crystal is miscut by 0.5° leading to a terrace width of ≈25 nm. The Fe films were grown at RT (Ref. 7) at a rate of 0.6 ML per minute and transferred into the STM without annealing. We used etched W tips which were flashed *in vacuo*, then magnetically coated with 8 ± 1 ML Gd while held at RT, subsequently annealed at $T \approx 550$ K for 4 min, and transferred into the cryogenic STM. As already shown in Ref. 2 Gd coated W tips, prepared as described above, exhibit at $T = 15$ K a magnetization perpendicular to the surface plane. The tunnel current thereby becomes sensitive to the out-of-plane component of the surface magnetization.

In the first experiment we investigated the spin-dependent electronic structure of the surface by means of SP-STS. Figure 1(a) shows a constant current image of 1.4 ML Fe/W(110). On top of a nearly closed ML anisotropic islands of the second layer have nucleated. Figure 1(b) displays averaged dI/dU spectra taken above the sites marked in Fig. 1(a). ML and DL spectra exhibit characteristic peaks at $U = +0.4$ V and $U = +0.69$ V, respectively, at the same energetic positions as measured with bare (nonmagnetic) W tips.[2] At negative bias the spectra are dominated by the tip's density of states, since here the spectra varied considerably from tip to tip.[8] On the DL we found two distinct types of spectra, represented by site *A* and *B*. Since these sites are identical in their (spin averaged) electronic structure—as known from measurements with bare W tips (not shown)—we can safely conclude that the difference between spectra of type *A* and *B* is a pure magnetic effect, caused by spin-polarized tunneling. From the known orientation of the tip magnetization we conclude that the two DL islands are magnetized up and down, respectively, perpendicular to the surface. In Fig. 1(c) we have plotted the lateral distribution of the dI/dU signal at $U = -0.25$ V using a linear gray scale. For this particular tip this bias voltage yields a maximum contrast with an effective spin-polarization of the tunnel junction of $P_{\text{eff}} = 0.45$,

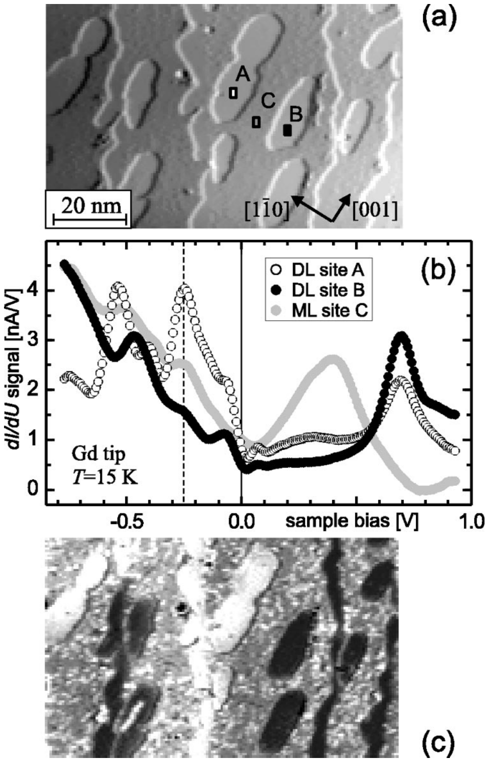

FIG. 1. (a) Constant current image (partially differentiated) of 1.4 ML Fe/W(110). (b) Averaged spectra taken at the marked sites in Fig. 1(a), tip stabilized at $U=0.9$ V, $I=1$ nA. On the DL two distinct types of spectra (A and B) exist due to spin-polarized tunneling, corresponding to parallel and antiparallel configurations of tip and surface magnetization. (c) Corresponding dI/dU signal at $U=-0.25$ V. The ML appears gray whereas the DL areas exhibit a two-stage contrast corresponding to an up and down magnetization.

defined as the (voltage dependent) asymmetry $P_{\mathrm{eff}}(U) :=[dI/dU_{\mathrm{A}}(U)-dI/dU_{\mathrm{B}}(U)]/[dI/dU_{\mathrm{A}}(U)+dI/dU_{\mathrm{B}}(U)]$. The ML appears gray, whereas the DL areas—islands as well as stripes at the W step edges—exhibit a two-stage contrast, corresponding to an up and down magnetization. Note that the intermediate dI/dU signal of the ML is not a result of its in-plane magnetization, but a consequence of its different electronic properties as compared to the DL. At $U=+0.69$, e.g., the DL areas exhibit an inverted contrast and the ML appears darkest, due to its low differential conductivity at this bias voltage.[2]

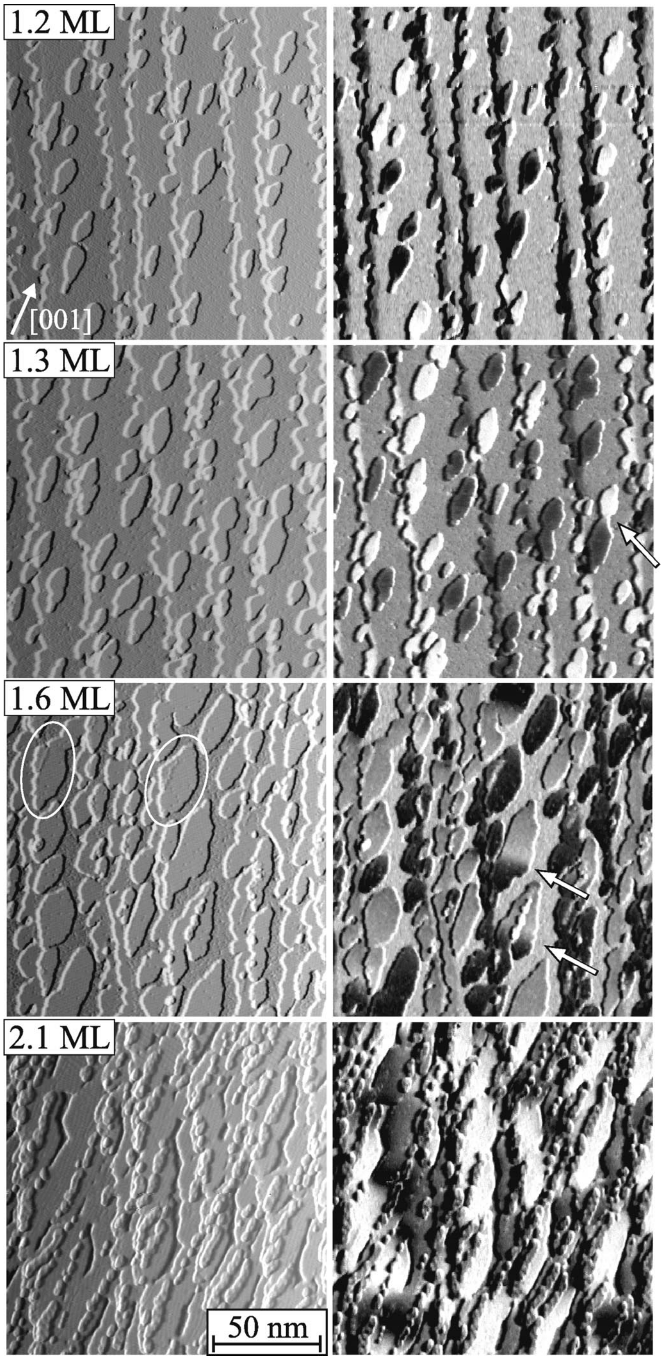

FIG. 2. Constant current images (left) and simultaneously recorded dI/dU maps (right). 1.2 ML ($U=-0.4$ V) and 1.3 ML ($U=-0.8$ V): all DL islands are in a single domain state with the magnetization pointing up or down. An exception is marked by an arrow. Small islands exhibit intermediate gray levels due to a reorientation to in plane. 1.6 ML ($U=+0.7$ V): Dislocation lines are visible (see ovals) as well as small patches of the third layer. Two domain walls oriented along $[1\bar{1}0]$ are marked by arrows. 2.1 ML ($U=-1.0$ V): The DL is nearly closed and the third layer contribution is substantial. Nevertheless, an out-of-plane AFM ordering is present in the DL.

In a second set of experiments we investigated the dependence of the magnetic structure upon Fe coverage. Instead of taking full spectra as described above, we measured maps of the dI/dU signal at specific bias voltages only, which were chosen separately to optimize the magnetic contrast. We thereby increased the lateral resolution and reduced the measurement time by a factor of 20. Simultaneously we recorded constant current images to allow a direct comparison with the sample's topography. Figure 2 shows a sequence of constant current images (left) and simultaneously acquired dI/dU maps (right) with an Fe coverage from 1.2 up to 2.1 ML. At 1.2 ML all DL islands are in a single domain state. They display a two-stage contrast corresponding to an out-of-plane AFM ordering. Small islands, however, show an intermediate dI/dU signal, a finding we will discuss in detail later. Also at 1.3 ML we observe an out-of-plane AFM ordering with the islands as single domain particles. There is, however, one exception to this rule for an island with a constriction (see arrow). Here a domain wall is found right at the constriction where the domain-wall energy is minimal. Furthermore, the magnetic structure of the island ensemble is not completely random. Chains of parallelly magnetized islands are found along the step edges, reminiscent of the AFM coupled DL wires investigated in Ref. 2. This ordering is obviously a result of ferromagnetic (FM) coupling between the islands which is mediated by exchange coupling to the DL stripes that have grown at the *W* step edges. This kind of magnetic structure becomes even more pronounced at 1.6 ML. At this coverage island coalescence has started and extended islands display misfit dislocation lines oriented along [001]. We marked two such islands by ovals in the topography image. The dislocation lines are also visible as white lines in the dI/dU map due to their local influence on the electronic structure. In contrast to lower coverages domain walls are present in islands beyond a length of ≈30 nm even in the absence of constrictions. They are always oriented along [110] with a wall width of 7±1 nm. We suggest these domain walls to be Bloch-type walls with the magnetization within the wall pointing along [110] which is the easy direction of the closed DL at elevated temperatures.[3] Two domain walls are marked by arrows. The lower one crosses a dislocation line on which third layer patches have nucleated. Surprisingly, the appearance of dislocation lines and the accompanied partial release of misfit strain has no detectable influence on the magnetic domain structure. We observe neither a loss of out-of-plane contrast nor any local interaction of dislocation lines and domain walls. At 2.1 ML the second layer is nearly closed except for trenches running roughly along [001]. Lines of third layer patches have nucleated along the same direction. Nevertheless, the dI/dU map reveals that out-of-plane AFM ordering is still present in the DL, with domain sizes governed by the substrates terrace width, i.e., 25 nm. These results are different from those in Refs. 3 and 4. Elmers *et al.* observed in-plane FM ordering for coverages $\Theta > 1.5$ ML at $T > 115$ K and Sander *et al.* found in-plane FM ordering for the full range of coverage from 0.8 to 2 ML at $T = 140$ K. The perpendicular magnetization we observe at 2.1 ML despite island coalescence and presumably magnetic percolation is, however, in agreement with perpendicular domains we observe for annealed films of the same coverage (not shown). A possible explanation for the discrepancy is an enhanced perpendicular anisotropy at low temperatures.

Utilizing the high spatial resolution of SP-STS we have performed a detailed analysis of the size dependence of the magnetization of the DL islands. The inset of Fig. 3(a) shows two small islands displaying an intermediate dI/dU signal (left) and the corresponding topography (right).[9] From the model of spatially switching anisotropy[5] a reorientation of island magnetization to in plane is expected below a critical island size, driven by exchange coupling to the in-plane magnetized ML. In order to verify this prediction quantitatively we plotted the average dI/dU signal of ≈140 free standing DL islands vs their width along [110] for the sample of 1.3 ML coverage presented in Fig. 2. The result is shown in Fig. 3(a) with each gray square corresponding to a particular island. The error bar represents the standard deviation over the island area. Beyond 4 nm width a strong two-stage contrast is displayed corresponding to $P_{\rm eff} \approx 0.45$. This contrast vanishes between 2 and 3 nm width and the dI/dU signal then stays constant within our measurement accuracy at an intermediate level.

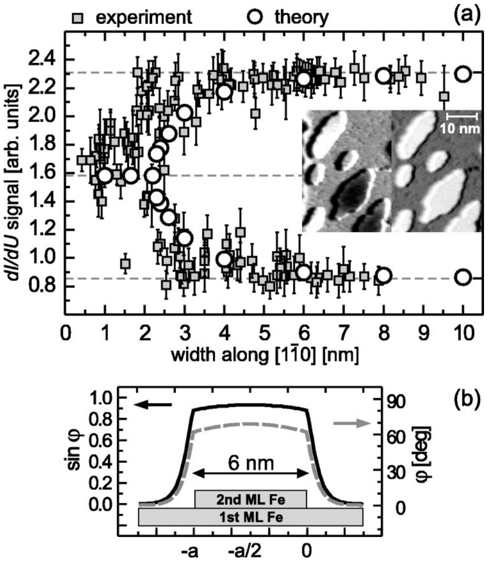

FIG. 3. (a) Average dI/dU signal of DL islands (gray squares) vs island widths along [110]. A vanishing of magnetic out-of-plane contrast is observed with decreasing width at 2–3 nm. Calculations (circles) suggest a transition to in-plane magnetization at 2.4 nm. The inset shows dI/dU map and topography for islands with three distinct magnetization directions. (b) Calculated magnetization direction $\varphi(x)$ (gray curve) and its sine (black curve) as a function of lateral distance across a DL.

To understand this result we performed micromagnetic calculations in the spirit of Ref. 10. Taking into account exchange and anisotropy energy only and neglecting the finite island length along [001], we searched for one dimensional functions of the magnetization angle $\varphi(x)$ that minimize the energy integral across a single DL area of width *a*. With the DL between $x = -a$ and 0 and $\varphi = 0$ corresponding to in-plane magnetization, the relevant integration extends from $-a/2$ to ∞:

$$\Gamma_{\rm DL}=2\int_{-\frac{a}{2}}^{0}\left\{A_{\rm DL}2t\left(\frac{d\varphi}{dx}\right)^2+K_{\rm DL}2t\cos^2\varphi\right\}dx \qquad (1)$$

$$\Gamma_{\rm ML}=2\int_{0}^{\infty}\left\{A_{\rm ML}t\left(\frac{d\varphi}{dx}\right)^2+K_{\rm ML}t\sin^2\varphi\right\}dx. \qquad (2)$$

Here *A* is the exchange stiffness, *K* the anisotropy constant, and $t = 2$ Å is the layer thickness. We parametrized test functions in a way that they automatically satisfied the boundary conditions and numerically calculated the set of parameters minimizing $\Gamma_{\rm DL}+\Gamma_{\rm ML}$ for a number of widths *a*. We used the boundary conditions $\varphi'(-a/2)=0$, $\varphi(\infty)=0$, $\varphi(0^-)=\varphi(0^+)$, and $\varphi'(0^+)=2A_{\rm DL}/A_{\rm ML}\cdot\varphi'(0^-)$. The last condition ensures a continuous torque at the boundary between ML and DL, with a factor of 2 arising from the different layer heights. The anisotropy constants have been determined experimentally to $K_{\mathrm{ML}}=5\times10^{6}$ Jm^{-3} and $K_{\mathrm{DL}}=1\times10^{6}$ Jm^{-3}.[11] For an estimate of A_{DL} we utilize the domain-wall width $w_{\mathrm{DL}}=7\pm1$ nm as observed by SP-STS. With the equation $w_{\mathrm{DL}}=2\sqrt{A_{\mathrm{DL}}/K_{\mathrm{DL}}}$ we get $A_{\mathrm{DL}}=1.25\times10^{-11}$ Jm^{-1}. The exchange stiffness of the ML is chosen to $A_{\mathrm{ML}}=1.25\times10^{-12}$ Jm^{-1} to achieve a good agreement with the experimental data. In Fig. 3(a) we have plotted the sine of the magnetization angle in the middle of the DL area, $\sin\varphi(-a/2)$, vs the width a (white circles), with the dashed grid lines corresponding to 0 and ±1, respectively. The calculated values display a transition to perfect in-plane magnetization in the DL at $a_c=2.4$ nm, in good agreement with the experimental data. The scattering of experimental values between 2 and 3 nm is not surprising, since the energy difference between in-plane and a slightly tilted magnetization becomes very small in the vicinity of the critical width, and the actual angle therefore depends on the exact local configuration, like, e.g., island shape and local stray field. Furthermore, we only considered a symmetric boundary condition with the ML magnetized parallel on both sides of the DL. In an antiparallel configuration, however, the magnetization must rotate via 90° in the DL which might explain some deviations observed for small islands.[12] Figure 3(b) shows the calculated magnetization direction $\varphi(x)$ (gray curve) and its sine (black curve) for a width of $a=6$ nm. Since for an out-of-plane magnetized tip the dI/dU signal is proportional to the sine of the magnetization angle the black curve is the one as measured by SP-STS. It agrees with the experimental data in the way that the dI/dU signal appears rather flat on the DL and we do not observe any magnetic contrast in the ML in the vicinity of the DL islands. We therefore assume that the rotation takes place in a very narrow region at the interface of ML and DL.

In summary, we have imaged the magnetic domain structure of Fe DL islands which are surrounded by a single ML Fe. We observed out-of-plane AFM ordering in the full range of coverage from isolated DL islands at 1.2 ML up to a nearly closed DL at 2.1 ML. For small islands we observed a vanishing of the magnetic out-of-plane contrast at 2–3 nm island width, a result we explain by a reorientation transition to in plane, driven by exchange coupling to the in-plane magnetized ML.

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