Chapter 4

Antiferromagnetic Mn films on Fe(001)

Details of magnetic frustrations in an antiferromagnetic film that is grown on a ferromagnetic substrate have been mainly investigated theoretically as discussed in section 2.1.2. Due to the limited lateral resolution of real space magnetic imaging techniques only little experimental data are available. In our Sp-STM study, we focus on antiferromagnetic surfaces of thin antiferromagnetic Mn films grown on a ferromagnetic Fe substrate.

4.1 Properties of Mn on Fe(001)

Bulk Mn exists in a wide range of crystallographic structures with different magnetic behavior. Up to 1000 K, bulk Mn appears in a complex cubic phase ($\alpha$-Mn) with 58 atoms per unit cell and is antiferromagnetic below 95 K with a non collinear spin arrangement [97]. Between 1000 K and 1370 K, $\beta$-Mn is a stable cubic structure with 20 atoms per cell. A face centered cubic ($\gamma$-Mn) and body centered cubic ($\delta$-Mn) structure is found between 1370 K - 1410 K and 1410 K - 1518 K (melting point), respectively [98]. Because of the simple atomic structure of the high temperature phases ($\gamma$ and $\delta$ Mn), they have attract much attention in the last years. Many unsuccessful attempts were made to stabilize these phases directly at room temperature. Two ways were found to allow the stabilization: Alloying of Mn with a small amount of other metals or epitaxial growth on a suitable substrate. Mn can be stabilized in a body centered tetragonal (bct) structure on the (100) face of bcc Fe [99]. Many groups confirmed the bct structure of Mn in this system up to about 20 ML [100–102]. Performing strain analysis, Kim and coworkers [101] showed that the bct structure of Mn on Fe(001) could either originate from a deformation of the bulk $\gamma$ or the bulk $\delta$ phase which leads to a misfit of $-9\%$ or $+4\%$. LEED images and RHEED oscillations recorded during the growth of Mn showed that Mn grows in a layer-by-layer mode up to 10 to 25 ML [47,100,101]. Indications of some defects and
disorder were found [103–105] before three dimensional growth sets in. The thickness where the transition takes place is strongly influenced by the substrate quality, e.g. cleanliness, step and defect density, substrate temperature during evaporation, and growth rate [100]. For substrate temperatures between 420 and 470 K, Tulchinsky and coworkers [47] showed that the transition to three dimensional growth takes place between 15 and 23 ML and away from these so-called best growth conditions this transition occurs at thinner Mn film thicknesses. The onset of three dimensional growth is visible in the LEED pattern, where the intensity of diffraction spots fades away and a more complex diffraction pattern is observed [100]. RHEED oscillations vanish for the three dimensional growth [47]. Andrieu and coworkers found that the RHEED pattern taken above the critical thickness is the same as observed on thick α-Mn films on Ir(001) [102]. Thus, it is believed that thick Mn films deposited on Fe(001) relax in the α-Mn structure. The abrupt roughening is clearly visible in images taken with a scanning electron microscope. The transition from smooth to completely rough films happen within an increase of the Mn film thickness of about 4 ML [47]. This behavior is characteristic for the Stranski-Krastanov growth mode of Mn-films, in which the first 10 to 25 ML grow nearly layer-by-layer before a transition to three dimensional growth sets in. Because of the misfit between Fe and Mn the strain energy increases with increasing film thickness for pseudomorphic growth. At a critical thickness, the pseudomorphic epilayer becomes unstable and the film relaxes by forming defects such as dislocations and roughening of its surface. Likely, the driving force for the transition from layer-by-layer growth to three dimensional growth is the reduction of elastic strain energy induced by the lattice mismatch [106].

Thin Mn films grow pseudomorphically on Fe(001). However, a structural change is found between the second and third ML of Mn [100–102], which is interpreted as a modification of the out-of-plane lattice constant. The out-of-plane lattice constant increases after deposition of the second ML Mn, and it is possibly correlated with a magnetic transition from a ferromagnetic to an antiferromagnetic order of the Mn planes [102]. It is found that thick Mn films have an out-of-plane lattice constant of 0.323 nm [100,101]. When Mn was deposited at room temperature on Fe(001), no intermixing was found between Mn and Fe. The onset of intermixing was observed by AES for substrate temperatures during the growth between 420 and 440 K [46,102]. In STM and STS studies interdiffusion of Fe in the first ML Mn is found to start at substrate temperatures during Mn deposition of about 370 K [107]. At this substrate temperature, the intermixing was observed until the fourth Mn layer [105].

The spatial distribution, shape, and size of Mn islands during growth of Mn on Fe was analyzed by STM and STS measurements [104,105]. In the sub-monolayer range, the growth of Mn at a substrate temperature of 430 K is characterized by small islands with about 10 to 25 nm diameter, an average spacing of the order of 10 nm and predominant step edge orientation along ⟨100⟩ [104]. Larger, rounder, and more widely spaced islands are found for a film thickness above 2 ML. At the Mn surface of films between 4 to 10 ML small regions with rectangular cross-shaped
Figure 4.1: a) Line scans obtained by SEMPA measurements on a bare Mn wedge on an Fe-whisker [47]. The Mn thickness varied from 0 to 16 ML. The magnetization component collinear ($M_x$) and perpendicular ($M_y$) to the Fe-whisker magnetization as a function of the Mn film thickness is shown. b) presents the line scan $M_x$ of a) after substraction of the exponential background (indicated by the gray dotted line in a)) caused by the Fe substrate.

patterns start to form [104, 105]. It was speculated that these small rectangular islands are local reconstructions and a precursor to three-dimensional growth, the phase transition to α-Mn which was found in thicker films.

The first evidence that Mn on Fe(001) forms ferromagnetic planes which order antiferromagnetically to each other with a period of two ML was presented by Walker and Hopster [46]. More recently, the layer-wise antiferromagnetic order between adjacent Mn atomic layers on Fe(001) was confirmed by scanning electron microscopy measurements with polarization analysis (SEMPA) [47]. Fig. 4.1 shows line scans of the magnetization component collinear to the magnetization of the Fe substrate ($M_x$) and perpendicular to it ($M_y$) as a function of the Mn film thickness [47]. An exponentially decay with superimposed oscillations is visible in the $M_x$ component while no $M_y$ component is observed (Fig. 4.1a)). Fig. 4.1b) shows the $M_x$ component after subtracting an exponential background, which is related to the Fe substrate, to clearly demonstrate the oscillations having a period of two ML with the beginning of the fourth Mn layer. This shows the layer-wise antiferromagnetic order. The growth
temperature was 420 K. The onset of the two layer oscillations varied depending on the substrate temperature during the Mn film growth [47]. The oscillations started with the third Mn layer when evaporating the Mn film at 520 K. The absence of the $M_y$ magnetization component is ascribed to two reasons. The first one is that Mn couples solely collinearly to the magnetization of the underlaying Fe substrate. The second is that the resolution of SEMPA during this measurement was about 100 nm. If non-collinear coupling is present in the form of equal numbers of small domains they may be averaged out.

Contradicting results exist concerning the magnetic coupling of the first and second Mn layer on Fe(001). Some groups confirm a ferromagnetic alignment of the magnetic moments of the first Mn layer [108,109] whereas other found hints for an antiferromagnetic alignment [110,111]. The investigation of the magnetic structure performed with SEMPA [47] suggest that the magnetic orientation of the first few layers is sensitive to the quality and crystallographic nature of the underlaying Fe. Andrieu and coworkers [112] found that the sign of the coupling of the first Mn layer strongly depends on the amount of O on the Fe substrate surface. They found a transition between ferromagnetic and antiferromagnetic orientation of the first layer with increasing the amount of O on the Fe surface. The following discussion of Sp-STM measurements performed on Mn films on Fe(001) will be similar in case of ferromagnetic or antiferromagnetic order of the first Mn layer and the Fe substrate. No qualitative differences occur.

### 4.2 Experimental results

Before each measurement, the Fe-whisker was cleaned by Ar-sputtering and annealing with the procedure described for the investigation of pure Fe-whisker surfaces in section 3.2. Mn was thermally evaporated and the growth rate was determined by the monolayer period oscillations obtained by MEED. Fig. 4.2 shows such MEED oscillations measured during the growth of Mn on Fe at a substrate temperature of 310 K. Eight MEED oscillations were observable in this measurement. After two strong oscillations, the intensity drops at a coverage of 1.8 ML and the further oscillations have much lower intensities. The coverage of 1.8 ML is determined by the period of the oscillations in the low intensity regime assuming structural homogeneity. A similar behavior of the oscillations were reported in the literature [47] and the drop in intensity occurred in the region where other groups observed structural changes in the Mn film [100–102].

To increase the accuracy to fractions of a ML even in thicker films, STM images of the topography were used. STM images yield a quite exact determination of the coverage between n and n+1 ML. Thus, the integer coverage n was defined by MEED and the fractional coverage between n and n+1 ML by topographic STM images. In the following, the uncertainty of the Mn coverage is defined by the accuracy of MEED measurements and all Mn film thicknesses presented in this work are given
4.2 Experimental results

Figure 4.2: MEED oscillations obtained during the growth of Mn on Fe(001) at a substrate temperature of 310 K.

with an estimated error of 10%.

In Fig. 4.3a) and b) two STM images show the topography of 8.9 ML and 9.2 ML Mn. The substrate temperature during evaporation for the film presented in a) was 310 K and in b) 370 K. Each gray level corresponds to a change in thickness by one ML. The structure observed at the surface of the Mn films, evaporated at different substrate temperatures show a completely different behavior. Note that image Fig. 4.3a) has a magnified scale of a factor of 4 compared to image Fig. 4.3b). The film evaporated at lower substrate temperature forms small islands and four different levels are exposed at the surface. Small brighter dots are visible on the different levels (~3 nm in diameter) which may have two possible origins. They can arise from intermixing in the first few Mn layers shining through the complete film due to the different lattice constants of Fe and Mn. As the substrate temperature during evaporation was close to room temperature, no intermixing should occur [107]. We believe that this structure is more likely due to the onset of the phase transition to α-Mn. The Mn film is already in the thickness range where this phase transition can set in.

The film deposited at 370 K (Fig. 4.3b) shows a much smoother surface with rounder, larger, and more widely spaced islands. Weakly two steps of subatomic height are visible, indicated by black arrows. These steps are caused by steps of the underlying Fe substrate. The difference in the out-of-plane lattice constant between Fe and Mn, for Mn films thicker than 5 ML is 0.018 nm hence, n+1 ML Mn are higher by 0.018 nm compared to n ML Mn plus 1 ML Fe. Thus, where Mn overgrows an Fe step edge, steps of subatomic height are formed at the surface of the Mn film.
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Figure 4.3: STM images of a) 8.9 ML and b) 9.2 ML Mn on Fe(001). The films were deposited at a substrate temperature of 310 K (a)) and 370 K (b)), respectively. In a) a bias voltage of -0.2 V was used and in b) 0.4 V, in both cases the average tunneling current was 3 nA. Note the different scale of these images.

Pierce and coworkers found that such buried Fe steps are visible through the Mn films up to a film thickness of 10 ML.

Beside the large Mn terraces and the round Mn islands, small rectangular islands are visible which are found frequently along buried Fe step edges and defects. This observation is in agreement with the results found by Pierce and coworkers [104]. They related these islands to local reconstructions being a precursor to the phase transition to $\alpha$-Mn. We found that these islands have predominantly edges along $\langle 100 \rangle$. The analysis of the size of these islands showed no special unit cells. Every size within atomic distances was found. The height is about $\frac{2}{3}$ of the height of monatomic steps between Mn terraces.

All Mn films investigated with the Sp-STM in the following study were grown at 370 K.

4.2.1 Magnetic order in Mn films

As already discussed, thin Mn layers on Fe(001) couple ferromagnetically within one Mn atomic plane while normal to the surface an antiferromagnetic coupling of neighbored Mn planes with in-plane spin polarization was observed. Fig. 4.4a) shows a schematic model of the topographic and magnetic behavior of Mn layers on Fe(001). The arrows represent the direction of the magnetic moments of the atomic Mn planes and the Fe substrate, also visualized by the different gray levels. In the schema, ferromagnetic order between the first Mn layer and the Fe substrate was assumed, but antiferromagnetic order would not change the following results except the sign of the spin polarization. When several Mn layers are exposed at the surface, in adjacent Mn layers the magnetization points into opposite directions.
4.2 Experimental results

Figure 4.4: a) Sketch of the magnetic order of Mn films on Fe(001). The ferromagnetic Fe substrate and the antiferromagnetic order between adjacent Mn layers is indicated by different gray levels and arrows pointing in opposite directions. b) Topography and c) corresponding spin signal of 11.9 ML Mn on Fe (001) measured with the Sp-STM.

due to the layer-wise antiferromagnetic order, as schematically shown (Fig. 4.4a)). In Fig. 4.4b) and c) the topography and the corresponding spin signal taken with the Sp-STM on a 11.9 ML Mn film on Fe(001) are presented. Three different Mn layers are exposed at the surface, a nearly closed layer with some rounded holes and islands (Fig. 4.4b). Again small rectangular islands are also present.

The Fe substrate was homogenously magnetized in one direction over the whole imaged area, as determined by Kerr-microscopy. The direction of sensitivity of the ring was chosen collinear to the magnetization of the Fe substrate. Thus, the imaged spin signal shows the projection of the spin component collinear to the Fe magnetization. In the spin signal (Fig. 4.4c), clearly the layer-wise antiferromagnetic order between the three different Mn layers is visible. The spin polarization between adjacent Mn layers is opposite, indicated by the black and white areas. The spin signal for n and n+2 ML Mn is the same. This observation is in agreement with results found by Yamada and coworkers [6]. The Sp-STM measurement was performed with a bias voltage of 0.1 V at an average tunneling current of 3 nA. Under these conditions, the highest spin contrast was found, as will be discussed in
section 4.2.2. All following Sp-STM images were taken under these conditions. The spin signal between oppositely spin-polarized Mn layers is about 1.2%. In spite of this low contrast, the signal to-noise-ratio is high. This magnetic order of the Mn film was found in images taken on one Fe terrace.

Around each island and hole, a contrast is visible in the spin signal close to the edges (Fig. 4.4c)). In general, such a cross talk of the topographic signal in the spin signal is always visible at the position of step edges but the size and the spatial extension changes depending on the tip apex. This will be discussed in more detail in section 4.2.1.

**Topologically induced magnetic frustrations**

The unperturbed layer-wise antiferromagnetic order is disturbed if a step of the underlaying Fe substrate is present. Fig. 4.5 presents schematically the topological and magnetic situation of Mn layers overgrowing a step edge of the Fe substrate underneath. The nomenclature is the same as presented in Fig. 4.4a). The thickness of the Mn layers on both sides of a monatomic Fe step differs by one ML. This means n layers Mn are grown on the upper side of the Fe substrate step edge and n+1 layers on the lower side. Due to the vertical lattice mismatch, subatomic steps are formed at the Mn film surface at the position of Fe step edges, as already shown in Fig. 4.3b). The situation of the magnetic order above such step edges is more complicated. An undisturbed layer-wise antiferromagnetic order within the Mn film is not possible when the Mn moments at the interface, on both sides of the step edge are aligned in the same direction by the Fe substrate. Thus, Mn layers which meet at the position of the Fe step edge have an opposite spin polarization. This leads to a magnetic frustration. When the Mn film thickness is smaller than the distance between two Fe steps, it is likely that the frustration reaches the Mn film surface, as schematically indicated in Fig. 4.5 (see section 2.1).

Fig. 4.6 shows Sp-STM images of the topography and the corresponding spin signal of a 6.9 ML Mn film grown over monatomic steps of the Fe substrate. In the topography, three buried Fe steps are visible indicated by the black arrows. The
Mn coverage changes by one ML on both sides of the step edges which is clarified by the numbers presenting the different Mn layers. In the spin signal, the layer-wise antiferromagnetic order of neighboring Mn layers separated by monatomic Mn steps is visible. In addition, magnetic frustrations are visible in the regions above the buried Fe step edges, each separating the Mn film into two domains. Along the three buried Fe step edges, a reversal of the spin contrast appears. In these regions the spin polarization of the Mn rotates by 180°. In the areas indicated by A and B, the coverage changes from 6 to 7 ML Mn and from 7 to 8 ML along the same buried Fe step edge. As a consequence of the layer-wise antiferromagnetic order the contrast is reversed. Identical situations occur at other areas in this image. The magnetically frustrated regions are similar to 180° domain walls in the antiferromagnetic film but they are pinned at the Fe substrate step edges. For thin films, these regions are much narrower than bulk domain walls, as will be shown later.

The observation of the magnetically frustrated regions at the surface of thin Mn films at the position of buried Fe step edges indicates that the magnetic frustrations are extended throughout the whole Mn film up to the interface, as schematically shown in Fig. 4.5. This means that the coupling energy at the interface between Fe and Mn is higher than the domain wall energy in the Mn film which is likely for thin films. However, Sp-STM is only surface sensitive so that the behavior within the Mn film is not accessible.

A closer look at the region above a buried Fe step edge allows to study the
Figure 4.7: Sp-STM image of a) the topography and b) the corresponding spin signal of 11.9 ML Mn on Fe(001). One buried Fe step edge is running almost vertically through the center of the images, indicated by arrows. c) Sketch of the cross section along the black line in a). d) Line profile taken along the black line in a) showing a monatomic Mn step and a step of subatomic height formed because of a buried Fe step. e) Line profile (averaged over 70 lines) across the magnetically frustrated region in the Mn over-layer at the position of the green box in b). The solid line represents a fit of a \( \tanh \)-function to the wall profile. In all images, line profiles shown in green are line profiles across magnetic frustrations.

magnetic behavior of the magnetic frustration in more detail. A magnified image of such a region is shown in Fig. 4.7 (a) topography and b) corresponding spin signal). Here, one buried Fe step edge is running almost vertically through the center of the imaged area (black arrows as guideline). The line profile in Fig. 4.7c) taken along the black line in Fig. 4.7a) shows a step of monatomic height between two different Mn layers (\( \approx 0.16 \) nm) and a step of subatomic height (\( \approx 0.020 \) nm) at the position of a buried Fe step edge due to the different lattice constants of Fe and Mn. In Fig. 4.7b) the layer-wise antiferromagnetic order between the Mn islands and the
Mn layer underneath is clearly visible. Following the way of the buried Fe step edge, a magnetically frustrated region is present in the spin signal. Fig. 4.7d) presents an averaged line profile across the topologically enforced magnetic frustration at the position of the green box in Fig. 4.7b)). The measurements indicate that the magnetic frustration has a certain lateral extension. The difference of the spin signal is about 0.9%. To estimate the wall width at the surface, the experimental profile is fitted with a tanh-function (see section 2.1.1). This function is the exact analytical solution for a profile across a one dimensional domain wall in an infinite uniaxial system [48], and it is given by equation 2.2. This tanh-function is plotted as a blue line in Fig. 4.7d) and it reproduces the shape of the transition region well. Due to the good agreement, this function is used to determine the wall width which is in this case $4.6 \pm 0.2$ nm. In the fitting procedure, the experimental data is weighted by the errors obtained by the standard deviation of averaging over 70 line scans at every individual position. The weighted fit assigns greater importance to less noisy data points allowing a more accurate estimate for the fit parameters. The given error of the wall width only includes the statistical error as determined by the fitting routine. In the following, the presented wall widths are always given within this error. Errors arising for example from lateral calibration of the piezo of the scanner and errors caused by lateral drift during the measurement are not included.

The weak, regular pattern visible in the topography and in the spin signal (Fig. 4.7) is caused by noise. The frequency of this noise level is between 20 and 30 Hz and originates most likely from mechanical vibrations of the STM. The mean amplitude is about 23 pm. In addition, again the contrast at the step edges is visible. Nevertheless, no cross talk is visible in the spin signal at the position of subatomic steps at the position of buried Fe step edges.

In the following, the behavior of the magnetically frustrated regions with increasing Mn film thickness is investigated.

**Dependence of the width of the magnetically frustrated regions on the Mn film thickness**

The width of the magnetically frustrated regions was studied for several Mn film thicknesses. A Sp-STM image of the thinnest Mn film (2.7 ML) on which a spin contrast was obtained, is presented in Fig. 4.8. Andrieu and coworkers observed that the layer-wise antiferromagnetic order starts between the second and third ML Mn [102]. Tulchinsky and coworkers showed that the beginning of the two ML oscillations depends on the substrate temperature during Mn deposition. As visible in Fig. 4.1, magnetization changes occur already in the sub-monolayer range of Mn films.

In the Sp-STM image, four Mn terraces and two buried Fe steps are visible in the topography (Fig. 4.8a)), the latter are indicated by arrows. In this case, n Mn

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1Note, this width is not determined by the lateral resolution of the Sp-STM. The resolution is much better, as will be shown later.
layers plus 1 ML Fe appear higher in the topography than n+1 ML Mn, because the out-of-plane lattice constant for thin Mn films, up to 2 or 3 ML is smaller than that of Fe. This reverses for thicker Mn films (see Fig. 4.6c)), where the out-of-plane lattice constant is larger than that of Fe. This observation is in agreement with the increase of the out-of-plane lattice constant of Mn for thicker films [101]. Beside the monatomic steps between Mn terraces and subatomic steps, formed by underlying Fe steps, small patches are imaged on the Mn terraces (Fig. 4.8a)). They have a width of about 10 nm and a height of about 50 pm and are more frequently found close to the edges of Mn terraces. From literature it is known that Mn intermixes with Fe for substrate temperatures above 370 K during Mn deposition [107]. Thus, most likely these islands are created by interdiffusion. Note on this thin Mn film, the small patches are likely due to intermixing, whereas the small rectangular islands of thicker films are related to the phase transition of Mn.

Since the spin signal contains only changes in the spin polarization, the spin polarization at the sample surface of the intermixed and alloyed layers can be imaged. In the corresponding spin signal (Fig. 4.8b)), the layer-wise antiferromagnetic order and the formation of a magnetic frustration along the buried Fe step edges are visible. We analyzed the width of the magnetically frustrated region between the second and third ML Mn. The line profile presented in Fig. 4.8c) was taken at the
4.2 Experimental results

Figure 4.9: Six line profiles across magnetically frustrated regions in Mn films of different film thicknesses. The film thickness is presented by \( n + 0.5 \) ML and the wall width in nm, both are indicated in the images.

marked area in Fig. 4.8b) where no changes of the contrast due to intermixing was observed. For this a thin Mn film, we found a narrow wall of only 1.2 nm across a buried Fe step edge. This is the sharpest magnetic feature we found on Mn films on Fe(001). From the line profile one can see that the lateral resolution of the Sp-STM is at least 1.2 nm.

The small darker areas (lateral extension of about 10 nm) which are only visible in the spin signal are most likely caused by intermixing of Fe and Mn resulting in a slightly different spin polarization. This measurement shows the capability of Sp-STM to investigate the spin polarization of intermixed regions independently of changes in the topography. The short, sharp white lines interrupting the spin signal are likely due to changes of the tip apex during scanning.

A selection of several line profiles obtained across magnetically frustrated regions in Mn films having different thicknesses (Fig. 4.9) clearly shows a widening of these regions with increasing Mn film thickness. The smallest width of 1.2±0.1 nm was imaged between the second and third ML Mn and the widest one of 6.9±0.3 nm between 18 and 19 ML (±2 ML) Mn. Thicker Mn films could not be investigated due to the phase transition to \( \alpha \)-Mn resulting in a three dimensional growth and rough surfaces.

The wall width across buried Fe step edges was determined for the six different Mn film thicknesses presented in Fig. 4.9. The widths of the magnetically frustrated regions were always extracted by fitting the experimental line profiles averaged over 25 to 70 lines with a tanh-function (equation 2.2). Fig. 4.10 shows the width of
Figure 4.10: The width of magnetically frustrated regions of Mn surface layers as a function of the Mn film thickness in ML (bottom scale) and equivalent in nm (top scale). The solid line is a linear fit to the experimental data points.

magnetically frustrated regions as a function of the Mn film thickness. Since the wall occurs between two different Mn layers, i.e. n and n+1, we followed the nomenclature of Stoeffler and coworkers [62] and plotted the value of the wall width at the position of n+0.5 ML Mn. The error bars in the Mn thickness result from the uncertainty of the evaporation rate. For the wall width the statistical errors are indicated, as described above. Fig. 4.10 shows that the wall broadens linearly with increasing Mn film thickness, as indicated by a linear function fitted to the experimental data [8]. This behavior of the widening of magnetically frustrated regions with increasing Mn film thickness will be discussed in more details in section 5.1.

**Deviations from the layer-wise antiferromagnetic order of Mn**

Normally, we observed the layer-wise antiferromagnetic order of Mn films on Fe(001) and magnetically frustrated regions in the Mn film at the position of buried Fe step edges. At some areas, we found no formation of a magnetic frustration at
4.2 Experimental results

Figure 4.11: Sp-STM image of a) the topography and b) the corresponding spin signal of 9.2 ML Mn on Fe(001). Deviations of the magnetic order at the buried Fe step edges, as described so far are visible. The red arrows indicate a buried Fe step edge where a magnetic frustration is formed in the lower part of the image which is absent in the upper half. The black arrows indicate again buried Fe step edges. The surface of Mn films, though a buried Fe step edge was present. No change of the projection of the spin polarization along the direction of the sensitivity of the ring appeared. Thus, the magnetic order in this case has to be different from the above discussed case. One example is presented in Fig. 4.11. In the topography (Fig. 4.11a)), three buried Fe step edges are visible running vertically through the image. In the corresponding spin signal, a clear spin contrast is visible, showing for example the alternating spin signal on both sides of Mn terraces separated by monatomic steps. Along the buried Fe step edges on the left and right side of the image, no magnetically frustrated regions are visible. Following the way along the buried Fe step edge running through the middle of the image, a magnetically frustrated region is formed in the lower part which disappeared in the upper half of the image. This is pointed out by the two red arrows. The transition between the two kinds of magnetic orders at this Fe step edge results in the formation of an extended magnetically frustrated region in one Mn layer, in this case visible on the Mn terrace A (Fig. 4.11a)). These changes of the magnetic order along buried Fe step edges occurred mainly in regions where an accumulation of the rectangular islands was found. It seems likely that the rectangular islands influence the order in the Mn film. Because a magnetic frustration has to occur somewhere when the Mn film grows over an Fe step edge, it might be that in this case a closed wall is formed near the interface as schematically shown in Fig. 2.4c).

We also observed a change from one configuration to the other by scanning the same area more than once. This case is presented in Fig. 4.12. Fig. 4.12a) and b) shows the topography and the corresponding spin signal measured during the first scan. Only one buried Fe step edge is running vertically through the image.
Figure 4.12: Sp-STM images, a), c) topography and b), d) corresponding spin signal of 11.9 ML Mn on Fe(001). Both images are taken at the same area but images c) and d) were taken 2 hours later than a) and b). The red arrows indicate a region where a change in the magnetic order along a buried Fe step edge was observed. Deviations in the shape of the imaged structures between a), b) and c), d) are caused by a different lateral drift of the sample during scanning. In image a) and b) a drift mainly in the horizontal direction occurred.

(indicated by the black arrows). In the upper part of the spin image a magnetically frustrated region is formed in the Mn layer whereas in the lower part no reversal of the spin polarization is observed at the step edge. Again, the behavior of the frustration changes in the vicinity of an accumulation of rectangular islands. The region of interest is indicated by the red arrow. Fig. 4.12c) and d) shows the same scan area imaged 2 hours later. In the mean time, images were taken by zooming into the area of the upper part of Fig. 4.12a). The magnetic frustration along the buried Fe step edge is now formed in the entire imaged area. The red arrow shows the position to which the domain wall shifted. The position of the buried Fe step edge is difficult to see in Fig. 4.12c), but the comparison to Fig. 4.12a) shows that the wall in Fig. 4.12d) is really formed along the buried Fe step edge. Comparing the two spin images (Fig. 4.12b) and d)) the whole area left from the red arrows changed the spin polarization by $180^\circ$. A change of the magnetization by $180^\circ$ at the
4.2 Experimental results

A sharp transition between the two different magnetic configurations occur (see green arrow). There, the contrast changes abruptly from one scan line to the other. The spin signal was imaged for both scan directions, which means for scanning from right to left and from left to right, called forward and backward scan. When comparing the images of forward and backward scan, the contrast in the discussed region is opposite in only one scan line (not shown here). This means that the change occurred during scanning that particular line. In this case, no normal wall is visible. The wall has no extension and is as sharp as the transition between two adjacent line scans. Most likely, the wall position has changed at one lateral scan point. Magnetically frustrated regions were predominantly observed in the Mn surface layer above buried Fe step edges. This indicates that this configuration is energetically more stable than the absence of magnetic frustrations at these positions. However, our measurements show that the system can stay in this metastable configuration long enough to be observed.

The experiment indicated that an energetic barrier had to be overcome to move the magnetic frustration to the position above the Fe substrate step. The simplest assumption is that a magnetic frustration in form of a 180° domain wall had to be formed. We assume that a wall of a length of about 17 nm, corresponding to the length of the region between the two islands indicated by the red arrow in Fig. 4.12b), had to be formed. Once it is established, it can freely move to the step edge. To estimate the probability for that process we used the wall energy of a 180° bulk domain wall according to \( \frac{\sqrt{2}}{4} AK \) [113]. The thermal switching probability can be expressed by an Arrhenius law of the form [114]:

\[
\tau = \tau_0 e^{\frac{E_B}{k_B T}}
\]

where \( \tau \) is the inverse of the switching rate and \( \tau_0 \) is the inverse of the attempt frequency which is of the order of \( 5 \times 10^{-9} \) s [115]. \( k_B \) is the Boltzmann constant and \( E_B \) the energy barrier. It is difficult to estimate the energy barrier accurately and a rough estimation suggests a value of 0.5 eV which results in a high probability of thermal switching processes. However, our observations show that switching events are rare. We imaged only this particular one. From this estimation, we can only say that thermal activation provides one possible explanation.

Alternatively, the wall could be moved by effects induced by the Sp-STM electrode. We used a current density of \( \approx 10^6 \) Acm\(^{-2}\). From literature it is known that with a spin-polarized current of a density of \( 10^7 \) Acm\(^{-2}\), a current induced switching of magnetic particles is possible [116]. Therefore, it cannot be excluded that the movement of the magnetic frustration is triggered by the spin-polarized current flowing between the SP-STM electrode and the sample while scanning.
Cross talk in the spin signal

As already mentioned, a cross talk of the topography is observed in the measured spin signal. Two different explanations for this observation seem reasonable. If the reaction of the feedback loop of the z-piezo is chosen too slow, a higher tunneling current is measured when the STM-tip has to go a step upwards (tip is for a short while too close to the surface) and a lower current is measured when it scans a step downwards (tip is for a short while too far away from the surface). Thus, at step edges, the tunneling conditions are different which causes changes in the spin-polarized tunneling current as well. This mechanism implies that the contrast of the cross talk reverses when changing the scanning direction. This was indeed observed, especially when scanning fast over large areas (of the order of $\mu$m).

In the case of small area scans, the cross talk was found not to depend on the scan direction so that the influence of the feedback loop can be excluded. In this case, large changes of the size, the sign and the spatial extension of the cross talk were found. Variations from day to day or even within one day have been observed. Therefore, this effect is likely influenced by local changes at the tip apex. In accordance with that, in topographic line profiles across islands with monatomic steps, changes in the profiles were found on different island sides (smoother or sharper transitions). Thus, the shape of measured line profiles across a step strongly depends on the exact shape of the tip apex.

The tunneling position at the tip apex may change when the tip is crossing a step edge and sidewise tunneling may occur producing different tunneling conditions. These differences are likely producing changes in the spin-polarized tunneling current and by this may cause a cross talk in the topography and in the spin signal. Even a dependence of the cross talk on the bias voltage was observed which supports the above ideas. This kind of phenomena is inherent to the method and cannot be suppressed easily.

### 4.2.2 Measurement of the voltage dependence of the spin contrast

The difference of the spin-dependent tunneling current between adjacent Mn layers on Fe(001) strongly depends on the bias voltage. All presented spin images were obtained at a bias voltage of 0.1 V, because at this voltage, a high spin contrast was observed. In the following, we show how the spin contrast changes by changing the bias voltage.

The measurements were carried out on Mn films thicker than 4 ML, were a constant electronic structure of Mn films was found [105]. In these measurements, the ring was scanned over the sample surface and at each point the bias voltage was ramped. When the measurements were performed the feedback was on during the ramping of the bias voltage. The spin signal detected in this experiment corresponds to the signal obtained in constant current mode images in our Sp-STM
4.2 Experimental results

Figure 4.13: Spin contrast between oppositely spin polarized Mn layers normalized to the tunneling current $2\bar{I}$ as a function of the bias voltage. The measurement was performed between 11 and 12 ML ($\pm 1$ ML) Mn.

measurements. This mode was selected to find the maximum spin contrast for the measurement shown before. By changing the bias voltage, the distance between ring and sample changed to keep the average tunneling current constant. Changes of the distance influence the transmission probability of states between ring and sample. For smaller distances, states with $k_\parallel \neq 0$ are suppressed less than for larger distances. In our experiment, the voltage was reduced by about a factor of 10 which results in a distance change of the order of 1 Å [67]. In experiments on Co(0001), no influence of the barrier properties on the spin polarization of the tunneling electrons was found for these conditions [117].

Fig. 4.13 presents the result for ramping the bias voltage from +0.8 V to 0.06 V and from −1.4 V to −0.1 V. The general trend can be described by a positive spin contrast above 0 V, a change of sign and a negative spin contrast below −0.3 V. The spin contrast becomes more negative when the applied voltage is decreases down to −1 V. For positive voltages, the spin contrast is highest at about 0.1 V. For different tips, the size of the spin contrast may change but qualitatively similar results for the voltage dependence were observed.
STS measurements were performed to compare the behavior of the spin contrast with the electronic structure of the Mn films. For the STS measurements, the tunneling current was stabilized at 1 nA and a bias voltage of 1 V. In this case, the magnetization of the ring was not switched and the spectroscopic measurement was performed on only one Mn terrace. During the measurement, the bias voltage was modulated by 30 mV with a frequency of about 6 kHz and changes of $dI/dU$ were detected with a phase sensitive lock-in amplifier. In Fig. 4.14, the normalized $dI/dU$ spectrum is presented. Two small shoulders are visible, one at about $+0.25$ V and one at about $-0.5$ V. As discussed in section 2.2, $dI/dU$ is assumed to be proportional to the local density of states. In this approximation, the two shoulders indicate that bulk band edges or surface states/resonances are present at these voltages. The shoulder around $+0.25$ V is not far away from the enhanced spin contrast shown in Fig. 4.13. Thus, they are possibly caused by the same electronic properties. However, no direct correlation can be found between the shoulder at about $-0.5$ V and features in the spin contrast. Sp-STS measurements performed by Yamada and coworkers [6] on the same system for Mn films thicker than 4 ML show two features in $(dI/dU)/(I/U)$ spectra. In agreement with our data, one feature is present at about $-0.5$ V, but the other, a pronounced peak at about 0.8 V.
is absent in our data. These differences may be caused by differences in the local density of states of the sample or the tip. Differences on the sample surface can be caused by different substrate temperature during Mn deposition or cleanliness of the substrate or the Mn film.

Comparison between our experimental data of the spin contrast as a function of the bias voltage and our STS data and theoretical calculations are given in the discussion in section 5.2.