



Universität Hamburg

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Symmetry-breaking and topology in antiferromagnet-based heterostructures

DISSERTATION ZUR ERLANGUNG DES DOKTORGRADES
AN DER FAKULTÄT FÜR MATHEMATIK, INFORMATIK UND
NATURWISSENSCHAFTEN
FACHBEREICH PHYSIK DER UNIVERSITÄT HAMBURG

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Hamburg, 2026

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16.06.2026

Abstract

In recent decades spintronic devices such as magnetic random-access memory (MRAM), which make use of the electron spin to carry information, have emerged as an alternative to traditional charge-based technologies. Spintronic devices have the potential to be significantly more energy efficient as well as to offer higher information storage densities. This may for instance be achieved by storing information using magnetic quasi-particles such as skyrmions in racetrack memory. So far, spintronics research has mostly focused on ferromagnetic (FM) materials; however, in recent years investigations into antiferromagnetic (AFM) materials have increased. Antiferromagnets exhibit faster dynamics, are more robust against stray fields, and due to their net-zero magnetization exhibit no stray fields themselves. To develop next-generation AFM-based spintronic devices a deep understanding of the parameters governing AFM properties is required. Additionally, nano-scale fabrication methods, such as self-assembly, are needed. In this thesis different phenomena arising in ultra-thin AFM films are investigated using spin-polarized scanning tunneling microscopy (SP-STM).

In the first experimental part of this thesis, the role of magnetism in processes like adatom diffusion and nanostructure growth is investigated, as this may have implications for the design of AFM devices. These processes are often studied above the magnetic ordering temperature, so magnetic effects do not have to be considered. Even below the magnetic ordering temperature, where magnetic effects may exist, their strength is usually considered negligible compared to binding energies and diffusion barriers. In this thesis the motion of adatoms is investigated on a single fcc-stacked Mn layer on a Re(0001) crystal, hosting a row-wise AFM (RW-AFM) state. We observe one-dimensional adatom motion on the hexagonal Mn layer at 4.2 K, dictated by the underlying uniaxial magnetic state. Furthermore, a comparison of the growth of Co monolayer (ML) islands on Mn/Re(0001) above and below the magnetic ordering temperature reveals that the magnetic state causes the formation of 1D Co-nanostructures. For the latter case these findings demonstrate the existence of a reciprocal effect between structure and magnetism.

Also in the context of topological quasi-particles, AFM spin textures are believed to be superior to FM spin textures. FM skyrmions are topological particle-like knots in the magnetization that can be driven by current, although with additional transversal motion (skyrmion-Hall-effect). AFM skyrmions hosting ferromagnetic skyrmions on two sublattices are predicted to show no transversal motion when driven by current. The system of the Mn ML and Mn DL on Ta(110) is demonstrated to be a model-type system, hosting the first experimentally observed AFM skyrmions in a single magnetic layer. Magnetic frustration at the interface between the Mn ML and DL, hosting a non-collinear AFM spin-spiral and a collinear AFM state, respectively, is employed to stabilize the AFM skyrmions.

AFM on superconductors (SC) form magnet-superconductor-hybrid (MSH) systems, that can host topological superconducting phases. At the boundary to different topolog-

ical phases zero-energy edge modes emerge. Control over the emergence of these edge modes and their characteristics is desirable for future spintronic applications. In this thesis, the edge modes between two different MSH systems, the Mn ML and Mn DL on Ta(110), are investigated, and the edge mode at the [001] edge is found to be spin-polarized. Together with collaborative tight-binding calculations, both MSH systems are shown to host topological nodal point superconducting (TNPSC) phases. Furthermore, it is demonstrated that TNPSCs can be a platform to tune the emergence and properties of edge modes.

Inhaltsangabe

In den letzten Jahrzehnten haben sich spintronische Bauelemente wie magnetische Arbeitsspeicher (MRAM), die den Elektronenspin zur Informationsübertragung nutzen, als Alternative zu herkömmlichen ladungsbasierten Technologien etabliert. Spintronische Bauelemente haben das Potenzial, deutlich energieeffizienter zu sein und höhere Informationsspeicherdichten zu bieten. Dies kann beispielsweise durch die Speicherung von Informationen unter Verwendung magnetischer Quasiteilchen wie Skyrmionen in Racetrack-Speichern erreicht werden. Bislang konzentrierte sich die Spintronik-Forschung größtenteils auf ferromagnetische (FM) Materialien. In den letzten Jahren haben die Untersuchungen zu antiferromagnetischen (AFM) Materialien jedoch zugenommen. Antiferromagnete weisen eine schnellere Dynamik auf, sind robuster gegenüber Streufeldern und zeigen aufgrund ihrer Netto-Null-Magnetisierung selbst keine Streufelder. Um Spintronik-Bauelemente der nächsten Generation auf AFM-Basis zu entwickeln, ist ein tiefgreifendes Verständnis der Parameter, die die AFM-Eigenschaften bestimmen, erforderlich. Darüber hinaus werden Fertigungsmethoden im Nanomaßstab, wie beispielsweise die Selbstorganisation, benötigt. In dieser Arbeit werden verschiedene Phänomene, die in ultradünnen AFM-Filmen auftreten, mit Hilfe der spinpolarisierten Rastertunnelmikroskopie (SP-STM) untersucht.

Im ersten experimentellen Kapitel dieser Arbeit wird die Rolle des Magnetismus in Prozessen wie der Adatomdiffusion und dem Wachstum von Nanostrukturen untersucht, da dies Auswirkungen auf die Herstellung von AFM-Bauelementen haben kann. Diese Prozesse werden häufig oberhalb der magnetischen Ordnungstemperatur untersucht, sodass magnetische Effekte nicht berücksichtigt werden müssen. Selbst unterhalb der magnetischen Ordnungstemperatur, wo magnetische Effekte auftreten können, wird ihre Stärke im Vergleich zu Bindungsenergien und Diffusionsbarrieren in der Regel als vernachlässigbar angesehen. In dieser Arbeit wird die Bewegung von Adatomen auf einer einzelnen fcc-gestapelten Mn-Atomlage auf einem Re(0001)-Kristall, der einen reihenweisen AFM-Zustand (RW-AFM) aufweist, untersucht. Bei 4,2 K beobachten wir eine eindimensionale Adatom-Bewegung auf der hexagonalen Mn-Schicht, die durch den zugrunde liegenden einachsigen magnetischen Zustand bestimmt wird. Darüber hinaus zeigt ein Vergleich des Wachstums von Co-Monolage (ML)-Inseln auf Mn/Re(0001) oberhalb und unterhalb der magnetischen Ordnungstemperatur, dass der magnetische Zustand zu der Bildung von 1D-Co-Nanostrukturen führt. Für Letzteres belegen diese Ergebnisse die Existenz einer wechselseitigen Wirkung zwischen Struktur und Magnetismus.

Auch im Zusammenhang mit topologischen Quasiteilchen gelten AFM-Spintexturen gegenüber FM-Spintexturen als überlegen. FM-Skyrmionen sind topologische, teilchenähnliche Knoten in der Magnetisierung, die durch Strom angetrieben werden können, allerdings mit zusätzlicher transversaler Bewegung (Skyrmion-Hall-Effekt). Es wird vorhergesagt, dass AFM-Skyrmionen, die ferromagnetische Skyrmionen auf zwei Untergittern beherbergen, keine transversale Bewegung zeigen wenn sie durch Strom angetrieben werden. Das System aus der Mn ML und der Mn DL auf Ta(110) hat sich als Modellsystem erwiesen, das die ersten experimentell beobachteten AFM-Skyrmionen in einer monoatomaren

magnetischen Schicht beherbergt. Magnetische Frustration an der Grenzfläche zwischen der Mn ML und der DL, die eine nicht-kollineare AFM-Spirale bzw. einen kollinearen AFM-Zustand beherbergen, wird zur Stabilisierung der AFM-Skymionen genutzt.

AFM auf Supraleitern (SC) bilden Magnet-Supraleiter-Hybridsysteme (MSH), die topologische supraleitende Phasen beherbergen können. An der Grenze zwischen verschiedenen topologischen Phasen entstehen Nullenergie-Randmoden. Die Kontrolle über das Auftreten dieser Randmoden und ihre Eigenschaften sind für zukünftige spintronische Anwendungen wünschenswert. In dieser Arbeit werden die Randmoden zwischen zwei verschiedenen MSH-Systemen der Mn ML und der Mn DL auf Ta(110) untersucht, wobei die Randmode an der [001]-Kante spinpolarisiert ist. Zusammen mit begleitenden Tight-Binding-Rechnungen wird weiterhin gezeigt, dass beide MSH-Systeme topologische Nodal-punkt-Supraleitungsphasen (TNPSC) beherbergen. Darüber können TNPSCs eine Plattform sein, um das Auftreten und die Eigenschaften von Randmoden zu steuern.

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List of abbreviations

AFM	antiferromagnet
AL	atomic layer
ASE	anisotropic symmetric exchange
BZ	Brillouin zone
DFT	density functional theory
DL	double-layer
DMI	Dzyaloshinskii-Moriya-interaction
fcc	face-centred cubic
FFT	fast Fourier transform
FM	ferromagnet
hcp	hexagonal close-packed
IP	in-plane
LDOS	local density of states
LEED	low energy electron diffraction
MAE	magneto-crystalline anisotropy energy
ML	monolayer
MM	Majorana mode
NCMR	non-collinear magneto-resistance
NP	nodal point
OOP	out-of-plane
RMS	root-mean-square
RW-AFM	row-wise antiferromagnet
SOC	spin-orbit coupling
SP-STM	spin-polarized scanning tunneling microscopy
STM	scanning tunneling microscopy
STS	scanning tunneling spectroscopy
TAMR	tunneling anisotropic magneto-resistance
TMR	tunnel magneto-resistance
TNPSC	topological nodal point superconductivity
UHV	ultra-high vacuum
YSR	Yu-Shiba-Rusinov

1 Introduction

Current computing technologies are dominated by devices based on the charge of the electron, such as solid-state drives (SSDs) and dynamic random-access memory (DRAM). In recent decades a field of research and applications called "spintronics" has emerged, which makes use of the spin of the electron [1] and may offer superior energy efficiency in the future [2]. Examples of such spintronic devices are commonly-used magnetic hard-disk drives and magnetic random-access memory (MRAM), which are based on the GMR/TMR effect; the TMR effect was discovered by Julliere [3] and the GMR effect was discovered by Fert [4] and Grünberg [5]. Spintronics research investigates relevant phenomena such as spin-transfer-torque (STT), spin-Hall-effect (SHE), and spin-orbit-torques (SOT), which are important for continuous improvements to MRAM and other devices [6].

Hard drives offer superior long-term storage compared to SSDs but a lack in speed. However, for hard drives the improvements in recent years have been incremental. For a fundamental leap in storage density and/or efficiency, a change in the information-carrying magnetic unit is necessary. Currently, hard drives store their information in the magnetization of individual ferromagnetic domains. As potential smaller candidates for a smaller unit, magnetic domain walls or skyrmions and their potential application in race-track memory have been considered [7, 8]. These localized spin textures show increased energy barriers to the ferromagnetic state due to their topological character. Their creation and annihilation has been demonstrated and they can be driven by electrical currents [9–12]. However, challenges remain, as skyrmions show an additional transversal motion when driven by a lateral current (skyrmion-Hall-effect), leading to potential annihilation pathways at film boundaries.

The focus of these investigations has mainly been on ferromagnetic systems; however, more recently antiferromagnetic systems have increasingly come under consideration, as they offer certain advantages over their ferromagnetic counterparts [13, 14]. Antiferromagnets have a net-zero magnetization and therefore exhibit no stray fields. They are much more robust against external stray field perturbations and are expected to exhibit faster dynamics. Antiferromagnetic skyrmions, which can be considered as a combination of ferromagnetic skyrmions on different sublattices, see Fig. 1, show no skyrmion-Hall-effect when driven by electric current [15, 16]. However, unlike ferromagnetic topological spin textures, they cannot be stabilized by an applied magnetic field. Nevertheless, theoretical investigations show that they can be created using spin-polarized current pulses [16, 17].

For the application of topological antiferromagnetic spin textures in devices such as racetrack memory, a deep understanding of the interactions governing AFM thin films and the stabilization and behaviour of topological spin textures are necessary. Additionally, to further increase energy efficiency, a need for nanoscale fabrication methods for spintronic devices exists. While atom-by-atom assembly is possible with scanning tunneling microscopy (STM) [19], it is unrealistic for large scale device fabrication. Here, the self-assembly of nanostructures may offer the required control.

A further step beyond classical computing are quantum computers [20, 21], which in the future may allow for the solving of particular problems unsuitable for current technologies. However, the quantum computers of today require many qubits for error

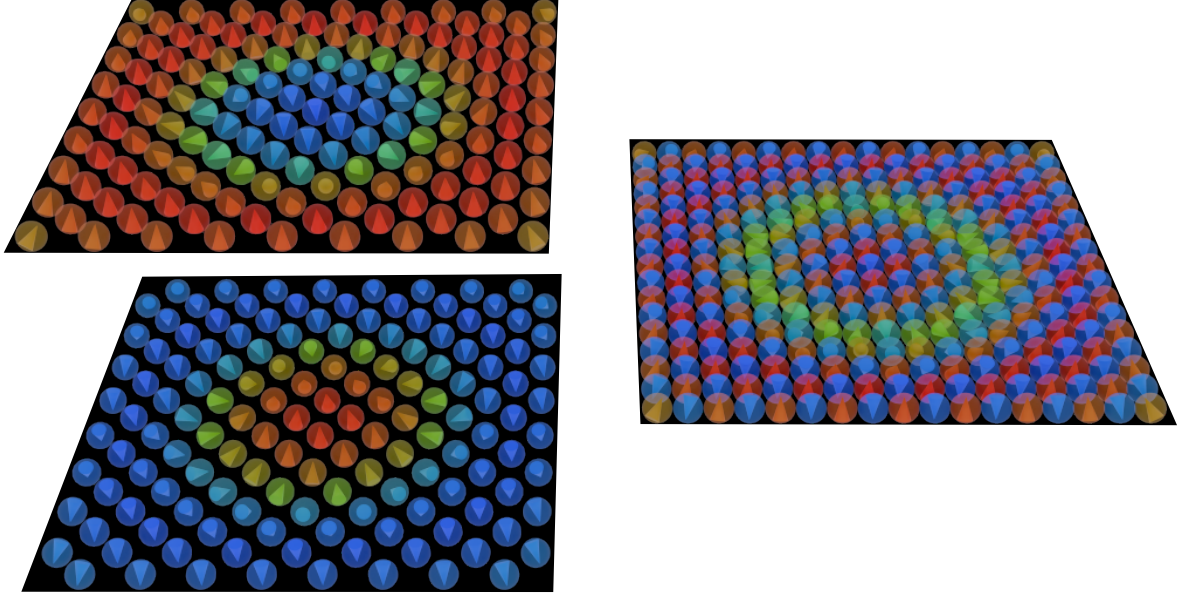


Fig. 1 Sketch of an AFM Néel skyrmion on a square lattice (right) and the two sublattices each hosting a ferromagnetic Néel skyrmion (left). Spins represented as cones with the colour corresponding to: red (up), blue (down), green (in-plane). The spin texture was simulated in Monte Crystal [18] with open boundary conditions.

correction, due to low qubit coherence times. A solution to this problem may be topological qubits, as they are more stable against perturbations [22, 23]. A candidate for such topological qubits are edge modes emerging at the boundaries of topological superconductors to trivial superconducting states, also known as Majorana modes (MM). In different experimental platforms, evidence for MM exist, such as in semiconducting nanowires proximity-coupled to a superconductor [24, 25], in vortices on p -wave superconductors [26], and magnet-superconductor-hybrid (MSH) systems, such as in magnetic chains and 2D-islands on s -wave superconductors [27–29].

Developing an in-depth understanding and control of the emergence and properties of such edge modes in MSH systems may allow for an application in future spintronic devices. For instance, it has been shown that in an MSH systems, hosting a ferromagnetic spin-spiral on an s -wave superconductor, the dispersion of the edge modes depends on the termination angle of the spin-spiral at the edge [30]. So far, the research on edge modes has been focused on the boundary between a topological superconducting state and the superconducting substrate; however, investigations of the edge modes at the boundary between different MSH systems may provide a more versatile approach to edge mode engineering.

In this thesis antiferromagnetic ultra-thin films were investigated using STM and spin-polarized (SP-)STM. I will discuss the effects of symmetry-breaking and frustration in these systems and the relation to diffusion and growth, and the stabilization of local topological spin textures. In addition I will discuss the boundary modes emerging in the magnet-superconductor-hybrid system of Mn/Ta(110).

In Chapter 2 the theoretical background of the relevant measurement techniques, STM and LEED, the magnetic interactions governing the magnetic states in the experimentally investigated systems, as well as magnet-superconductor-hybrid systems will be discussed.

The general experimental setup as well as broadly applicable measurement details will be discussed in the Methods Chapter 3.

The first experimental chapter of this thesis, Chapter 4, focuses on the system of fcc-stacked Mn on Re(0001) and the symmetry-breaking AFM state. Specifically, the effects of magnetism on the diffusion and growth of ultra-thin films below the magnetic ordering temperature will be discussed. I will show how the deposition of material can be employed to map the local AFM domain structure.

The second experimental chapter, Chapter 5, will report on the results obtained on the system of Mn/Ta(110). First, the collinear, non-collinear, and non-coplanar spin textures observed on the Mn monolayer (ML) and double-layer (DL) will be discussed, followed by the results of the investigation on the interaction of magnetism in the Mn layers and superconductivity of the Ta substrate. Both edges between the magnetic superconductor hybrid (MSH) systems and the superconducting substrate as well as the between the two MSH systems will be analysed.

Appendix A describes the investigation into the structure and magnetic state of ultra-thin transition metal (Fe, Co, Mn) oxide films on Ir(111), while Appendix B briefly discusses the growth and magnetism of ultra-thin films of Fe, Co, and Mn on the unreconstructed Ir(001) surface.

2 Theoretical Background

In this section, we will discuss the theoretical background necessary to understand the contents of this thesis. This includes the theoretical descriptions of the measurement techniques used, mainly scanning tunneling microscopy and low-energy electron diffraction, as well as the physics governing the behaviour of the ultra-thin film systems investigated during this thesis, mainly magnetism and superconductivity.

2.1 Scanning tunneling microscopy

Scanning tunneling microscopy, short STM, is a measurement technique used to measure the structural, electronic, and sometimes magnetic properties of conducting surfaces down to the atomic scale [31–33]. Here, a metallic tip is brought in close vicinity to the sample surface and a bias voltage is applied. The exponential distance dependence of the tunnel current flowing between tip and sample is exploited to measure precise changes in sample topography, as the tip is scanned across the surface.

2.1.1 The tunnel effect

In this section, the tunneling process of wave-like particles such as electrons through a potential barrier is discussed. The STM makes use of the tunnel effect, with electrons tunneling from tip to sample or vice versa, depending on the polarity of the applied bias voltage. This section bases its mathematical description of the tunnel effect on [34].

A wave-like particle in 1D evolves according to the Schrödinger equation:

$$\frac{-\hbar^2}{2m} \frac{d^2\psi}{dx^2} + E_{\text{pot}}\psi(x) = E\psi(x) \quad (1)$$

The space, in which the particle moves, is separated into three regions by a tunneling barrier of width a and height E_0 , see Fig. 2. In the two regions outside the barrier (I) and (III) our particle can be described by an oscillating wavefunction with a deBroglie wavelength λ and a wavevector k , while it is described as an exponentially decaying wave function in region (II). Assuming tunneling from left to right allows for the writing of the wave function as follows in equation (2):

$$\psi(x) = \begin{cases} e^{ikx} + Re^{-ikx} & \text{if } x < 0 \\ Ce^{\kappa x} + De^{-\kappa x} & \text{if } 0 < x < a \\ \tau e^{ikx} & \text{if } a < x \end{cases} \quad (2)$$

with

$$k = \sqrt{2mE}/\hbar \quad (3)$$

$$\kappa = \sqrt{2m(E_0 - E)}/\hbar \quad (4)$$

Due to reflections at the interfaces, the wavefunction in region (I) and (II) consists of two terms describing waves moving in opposite directions, while this is not necessary for

region (III). The amplitude of the wavefunction decays exponentially for the region inside the barrier with the decay length κ , as the particle energy is smaller than the barrier height E_0 . The size of κ and therefore the decay inside the barrier increases for larger barrier heights.

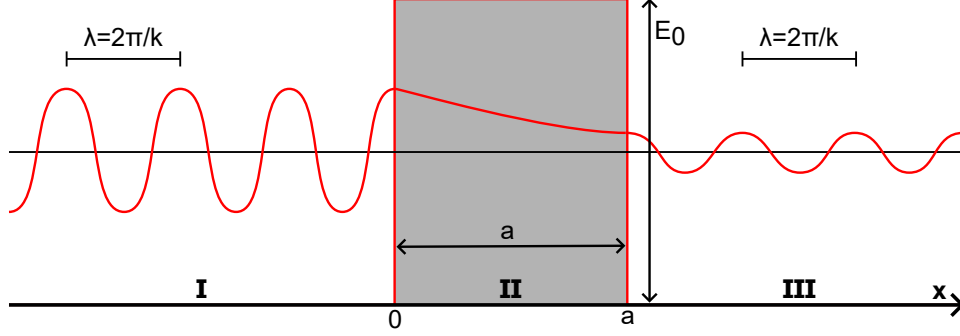


Fig. 2 Sketch of a wave-like particle tunneling through a potential barrier of height E_0 and width a . Figure adapted from [35].

From the fact that the overall wavefunction across the entire region is required to be continuously differentiable, boundary conditions for the four coefficients R, C, D , and τ can be derived. The transmission amplitude can then be written as follows:

$$\tau(E) = \frac{2ike^{-ika}}{2i\kappa k \cosh(\kappa a) + (k^2 - \kappa^2) \sinh(\kappa a)} \quad (5)$$

This transmission amplitude is directly related to the tunnel probability $T(E)$.

$$T(E) = |\tau(E)|^2 = \left(1 + \frac{E_0^2}{4E(E_0 - E)} \sinh^2(\kappa a)\right)^{-1} \approx \left(\frac{16E(E_0 - E)}{E_0^2} e^{-2\kappa a}\right) \quad (6)$$

The tunnel probability is always finite as the amplitude of the wavefunction in region (III) is always non-zero. The simplification in equation (6) holds for $\kappa a \gg 1$, which applies for the typical tunnel parameters in STM (i.e. $a = 0.8 \cdot 10^{-9}$ m, $\kappa = 10^{10} \frac{1}{\text{m}}$). One finds that the tunnel probability decreases exponentially with the barrier width a and as the square root of the effective barrier height $E_0 - E$ increases.

2.1.2 Tunneling in STM

As the tunnel current observed in STM is directly proportional to the tunnel probability, equation (7) for the tunnel current follows:

$$I \propto e^{-2\kappa a} \quad (7)$$

In an STM the electrons tunnel from the tip into the sample through the vacuum tunnel barrier for $U > 0$. The height of the tunnel barrier is related to the work functions ϕ of the tip and the sample, see sketch of the tunnel process in Fig. 3a. The effective tunnel barrier height $\bar{\Phi}$ can be approximated by averaging the work functions, see equation (8),

while the decay constant κ is related to the effective barrier height as written in equation (9):

$$E_0 - E = \bar{\Phi} = \frac{\phi_{\text{tip}} + \phi_{\text{sample}}}{2} \quad (8)$$

$$\bar{\Phi} = \frac{\hbar^2 \kappa^2}{2m_e} \quad (9)$$

κ and therefore the effective barrier height can be obtained by measuring tunnel-current vs tip-sample distance $I(z)$ curves for any particular tip and sample configuration. Here, the tip-sample distance z can be equated to the barrier width a .

In the tunnel process, as sketched in Fig. 3a, the electrons tunnel from the occupied states near the Fermi level of the tip to the unoccupied states of the sample. A simple model to describe the tunnel current in an STM tunnel junction for the low bias regime is the Tersoff-Hamann model [36] based on Bardeen's transfer Hamiltonian approach [37]. The model further assumes low temperatures, a tip with a spherical tip apex, as well as an s -wave tip wavefunction. The resulting approximation for the tunnel current is shown in equation (10):

$$I \propto n_t(E_F) e^{2\kappa r} n_s(E_F, \mathbf{r}_0) U \quad (10)$$

where n_t represents the LDOS of the tip at the Fermi level, $n_s(E_F, \mathbf{r}_0)$ represents the LDOS of the sample at the Fermi level, and κ represents the decay rate. Further, $\mathbf{r}_0$ represents the position of the centre of the spherical tip apex, with the bold font being used to indicate vectors, r represents the radius of the tip apex, and U the applied bias, see also sketch in Fig. 3b. Notice that the tunnel current can be treated as following Ohm's law for small bias values.

This model can be extended for larger bias values where one can sum over the range of available states as given by [32], see equation (11). An applied bias to the sample shifts its Fermi level relative to the tip Fermi level by eU , see Fig. 3a. One finds that the tunnel current depends on the LDOS of tip and sample in this energy range.

$$I \propto \int_0^{eU} n_s(\mathcal{E}) n_t(\mp \mathcal{E} \pm eU) T(\mathcal{E}, eU) d\mathcal{E} \quad (11)$$

with

$$\mathcal{E} = E - E_F \quad (12)$$

$$T(\mathcal{E}, eU) = \exp \left\{ -2(d+r) \left[\frac{2m}{\hbar^2} \left(\frac{\phi_{\text{tip}} + \phi_{\text{sample}}}{2} + \frac{eU}{2} - \mathcal{E} \right) \right]^{\frac{1}{2}} \right\} \quad (13)$$

T is the energy- and bias-dependent transmission coefficient, $\mathcal{E}$ is the energy in relation to the Fermi level, and d is the tip-sample distance. Here, however, $n_s(\mathcal{E})$ represents the LDOS of the sample at the sample surface, with the relation to the LDOS at the position of the tip apex $n_s(\mathcal{E}, \mathbf{r}_0)$ given by equation (14).

$$n_s(\mathcal{E}, \mathbf{r}_0) \propto n_s(\mathcal{E}) T(\mathcal{E}, eU) \quad (14)$$

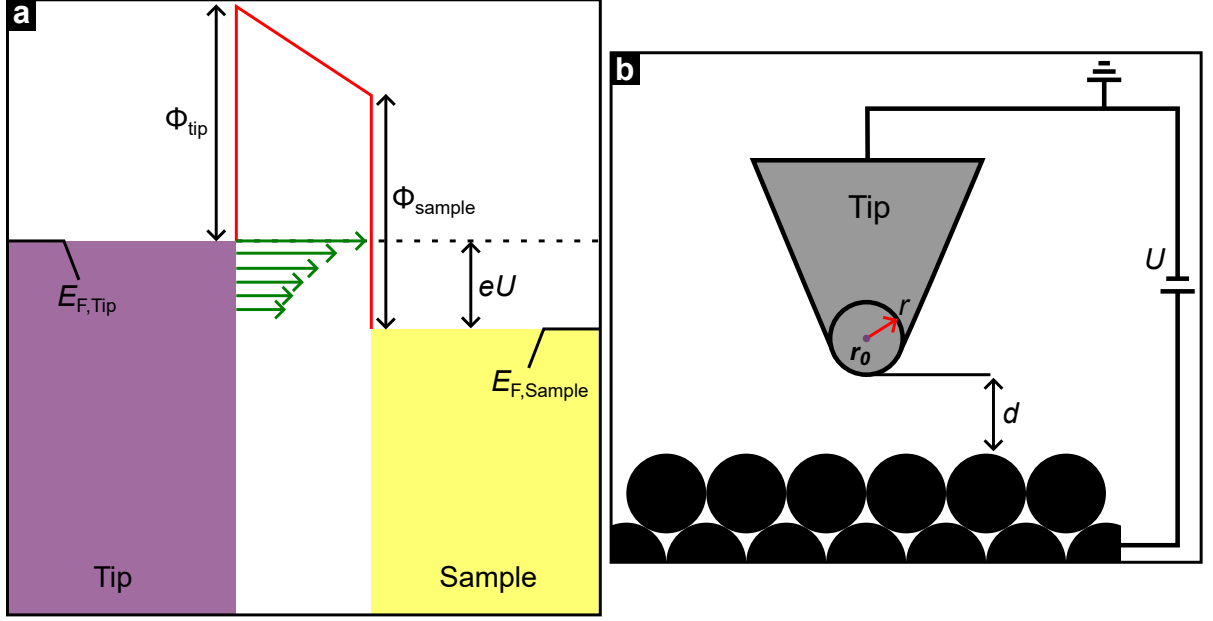


Fig. 3 **a**, Sketch of the tunnel process between tip and sample for $U > 0$. Electrons tunnel from the occupied states near the Fermi level of the tip to the unoccupied states above the Fermi level in the sample. **b**, Sketch of the junction between tip and sample, where d is the distance between tip and sample, r is the radius of the tip apex, $\mathbf{r}_0$ is the position of the centre of the tip apex, and U is the bias voltage applied to the sample.

The transmission coefficient T is similar to the tunnel probability described in Sec. 2.1 and encodes the distance between tip and sample, as well as an approximation of the height of the tunnel barrier, which is both bias- and energy-dependent. Among the states in the range E_F to $E_F + eU$, the states with the highest energy see the lowest tunnel barrier height, which results in a tunnel current contribution which is exponentially larger than for lower energy states, see green lines in Fig. 3a.

Additional information can be gathered by measuring the differential tunneling conductance dI/dU , which is given by equation (15) [32], assuming a constant tip LDOS:

$$dI/dU \propto en_t(0)n_s(eU)T(eU, eU) + \int_0^{eU} n_t(\pm eU \mp \mathcal{E})n_s(\mathcal{E})\frac{dT(\mathcal{E}, eU)}{dU}d\mathcal{E} \quad (15)$$

The transmission coefficient scales with the bias voltage in a monotonous way, providing a smooth background, and signals in the dI/dU can often be related to the LDOS of the sample as described by the first term in equation (15). However, one has to consider that changes in the dI/dU signal can also be related to changes in the transmission coefficient, as is the case for changes in the surface work function, for instance. Additionally, at negative bias the tunneling is dominated by the LDOS of the tip, so variations in the tip LDOS can become visible.

2.1.3 Measurement modes

An STM can be operated in various different measuring modes, and both imaging and spectroscopy are possible. As mentioned previously, variations in the tip-sample distance during tunneling can be used to estimate the effective tunnel barrier height, which is used in local barrier height imaging modes; but these will not be the focus for our applications. For topographic imaging modes, the STM tip is scanned laterally across the sample using a piezo-scanner, which deflects laterally depending on an applied voltage. For spectroscopy modes, the tip is positioned at a particular lateral position above the sample surface, and the bias-dependence of the tunnel current is studied.

Constant-current mode The most widely used STM imaging technique used for imaging complex sample topographies is the constant-current mode. Here, a feedback loop is used to continuously adjust the tip-sample distance to keep the current at a particular value. The topography signal can then be obtained by translating the voltage applied to the z -electrode of the piezo-scanner during the imaging process to the z -coordinate of the tip. The values needed for a particular translation are dependent on the dimensions of the piezo-scanner and the temperature, and have to be calibrated using a reference sample with known lattice constants.

The obtained topography-signal at every pixel position of the scan area can then be used to construct a topography map $z(x, y)$, which is then mapped onto a range of colour values (i.e. a colour-map) and plotted as a 2D image. Note that the STM tip is not fixed/stopped at discrete pixel positions but moves continuously. For each pixel the data is captured and averaged for a distance δx and small time δt . The topography map does not exactly represent the structural height differences of the sample surface, but instead the z -position of the tip also depends on the electronic structure of the surface it is scanning on; the topography is therefore also called the apparent height, which can be equated to the actual height only for sample regions that are electronically (and magnetically) identical.

The finite response time of the feedback loop introduces feedback artefacts during the imaging process; their severity depends on the scan speed across the surface. At crystallographic step edges or other sharp changes in apparent height, the current is not constant and is above or below the set-point value depending on the scan direction. If the tip moves too quickly across the sample surface, tip-sample collisions may occur.

In addition to the topography signal, the current signal is also recorded, and depending on the scan speed the current map can show features of the sample in the fast scan direction. For periodic signals, the current channel can often provide better contrast than the topography signal. Additionally, current maps can also provide easier comparability between regions with large apparent height differences.

Constant-height mode A second method of imaging a surface is the constant-height mode in which the tip is scanned in a plane parallel to the surface with the feedback loop switched off. The resulting current map is then a measure of the sample topography. This measurement mode is typically only used on atomically flat surfaces.

Compared to constant-current images, constant-height images are limited in their measurement duration depending on the creep of the piezo-scanner and thermal drift of the sample. However, they do not exhibit any feedback artefacts and can therefore be the imaging mode of choice in some cases.

dI/dU-maps Maps of differential tunnel conductance can be recorded simultaneously to the topography and current signal in both constant-current and constant-height mode. The dI/dU-signal is measured by applying a sinusoidal modulation to the bias voltage at a frequency typically in the range of 3-5 kHz, which is fast enough to not affect the feedback loop. This allows a lock-in amplifier to directly measure the dI/dU-signal (i.e. the effective slope of the current) in a bias voltage range $U \pm \sqrt{2}U_{\text{mod}}$ given by the amplitude of the bias modulation U_{mod} , typically written as an RMS value, where U is the DC bias value applied to the tunnel junction. For a good signal-to-noise ratio in a dI/dU-map, the scanning speed is required to be slightly slower than for pure topography imaging, as for every pixel one or more periods of the modulation signal have to be included.

In Sec. 2.1.2 we have seen that the dI/dU-signal can be a measure for the local density of states of the sample, therefore these maps are good for differentiating between regions that differ significantly in their LDOS at the measured bias voltage. This can allow for easier identification of different materials on the surfaces, different stacking of the same thin film, or even between magnetically different regions such as between anti-ferromagnetic domains and domain walls, as will be discussed in Sec. 2.1.4. In this thesis, constant-height imaging is employed together with dI/dU-maps to separate features in the LDOS at the measurement bias from features in the apparent height.

Multipass constant-current mode Multipass imaging is a measurement mode that can be used for scanning complex sample morphologies while allowing for scanning at different bias voltages and while the tip traces identical paths across the sample. It can even allow for measurements at zero bias.

It works by first measuring and recording the topography of a single line in the image at a set non-zero bias value and then retracing that topography again while applying a different bias value and recording the current- and/or dI/dU-signal. In this thesis, it is used together with the dI/dU-signal to map the LDOS at the Fermi energy across a sample region.

Constant-height bias spectroscopy In constant-height bias spectroscopy the tip is placed at a particular lateral position of the surface and stabilized using the feedback loop for a set of tunnel parameters (i.e. bias voltage and tunnel current), which determines the tip-sample distance. The feedback loop is then disabled and the bias voltage is ramped between two bias values. Note that this can include values near and at zero bias. Both the current signal and the dI/dU-signal are typically recorded during this sweep.

Multiple of these spectroscopy measurements can be performed together in the form of a line or a grid. Similar to dI/dU-maps, spatial variations of the LDOS across the sample can be probed this way and additional energy resolution is provided. However,

due to much longer measurement times per pixel, only much lower resolution grids in smaller areas of the sample are feasible.

2.1.4 Spin-polarized STM

Using spin-polarized tips, magnetic order can be observed at the atomic scale [33]. Antiferromagnetic tips allow for spin-polarized tunneling while minimizing stray fields. In contrast, for ferromagnetic Fe/W-tips the tip magnetization direction can be controlled by an external magnetic field, but can disturb the sample with stray fields. As will be discussed in Sec. 2.3, this does not affect the antiferromagnetic states investigated in this thesis, which are stable against stray field perturbations. Magnetic contrast in SP-STM is obtained using the tunnel magnetoresistance effect (TMR).

An extension of the Tersoff-Hamann model to include spin-polarized tunneling has been derived by Wortmann *et al.* [38], see equation (16):

$$I(\mathbf{r}_0, U, \theta) = I_0(\mathbf{r}_0, U) + I_P(\mathbf{r}_0, U, \theta) \propto n_t \tilde{n}_s(\mathbf{r}_0, U) + \mathbf{m}_t \tilde{\mathbf{m}}_s(\mathbf{r}_0, U) \quad (16)$$

Here, I_0 is the non-spin-polarized current, while I_P represents the spin-polarized current that depends on the angle θ between the magnetization of tip and sample at the centre of the tip apex $\mathbf{r}_0$. The total LDOS of the tip and sample is given by $n_t = n_{t\uparrow} + n_{t\downarrow}$ and $n_s = n_{s\uparrow} + n_{s\downarrow}$, while the local magnetization is represented by $\mathbf{m}_t$ and $\mathbf{m}_s$, respectively. The tilde mark denotes an integration over the energy.

The dot product of the magnetization vectors $\mathbf{m}_t$ and $\mathbf{m}_s$ indicates a cosine(θ) dependence of the spin-polarized contribution I_P to the tunnel current. Meaning for parallel and anti-parallel alignment of tip and sample magnetization, one finds maximum and minimum spin-polarized current, respectively, while the contribution is zero at $\theta = 90^\circ$.

This difference in the magnitude of the tunnel current shows up as a change in apparent height in constant-current topography images, but can also appear in current-maps depending on the scan speed, and in dI/dU -maps depending on the bias.

As the magnetic state can also result in a change to the LDOS of the system, some magnetic features can also be resolved using non-magnetic tips. For instance, changes in the non-collinearity (i.e. changes in the nearest-neighbour angles) can manifest themselves as changes in the LDOS due to mixing of spin channels, resulting in contrast for domain walls and skyrmions [39, 40]; this mode of electronic contrast is named non-collinear magneto-resistance (NCMR). Note that homogeneous spin-spirals cannot show this effect, because, although they are non-collinear, the angle between spins never changes.

Such a state can, however, show changes in the LDOS resulting in a mode of electronic contrast called tunneling anisotropic magneto-resistance (TAMR). This effect is based on the mixing of electronic states of different orbital character due to SOC, which can differ between regions with different magnetization axes. For instance, it can result in contrast between IP and OOP regions of the homogeneous spin-spiral [41] or between ferromagnetic domains and the domain walls [42].

2.2 Low energy electron diffraction

Low energy electron diffraction (LEED) is a surface-sensitive technique to characterize the surface lattice structure of crystalline samples. Here, electrons are accelerated onto the sample at an incident angle normal to the surface with energies in the range of 20-200 eV. As electrons exhibit characteristics of both particles and waves, one can assign them a deBroglie wavelength λ_e :

$$\lambda_e = \frac{h}{p} \quad (17)$$

Here, h is the Planck constant, and p is the electron momentum. With an electron wavelength on the order of the atomic spacings, diffraction at the atomic lattice becomes relevant. The elastically backscattered electrons are separated from the inelastically scattered electrons using a retarding electric field and are then accelerated onto a fluorescent screen. The resulting LEED pattern represents the reciprocal lattice of the surface and can provide information regarding the relative size and angle of surface reconstructions and superstructures formed by adatoms compared to the atomic lattice.

The diffraction of the low-energy electrons at the surface layers of the crystalline substrate is described by Bragg's law or by the Laue equations. The resulting reciprocal lattice is an indication of the possible wavevectors $k = 2\pi/\lambda$ for the diffracted electrons. Each point of the reciprocal lattice (LEED spot) corresponds to a particular wavevector; the length of this vector is proportional to the electron momentum $\vec{p} = \hbar\vec{k}$. These possible wavevectors are described by the Laue equation:

$$\vec{k}_{\parallel, \text{out}}(n_1, n_2) = \vec{k}_{\parallel, \text{in}} + n_1 \vec{a}_1^* + n_2 \vec{a}_2^* \quad (18)$$

Here, $k_{\parallel, \text{out}}$ is the length of $\vec{k}$ of the back-scattered electrons parallel to the surface, and $k_{\parallel, \text{in}}$ corresponds to the incoming electrons. $\vec{a}_1^*$ and $\vec{a}_2^*$ are the reciprocal lattice vectors. n_1 and n_2 are integers and can be used to index the observed LEED-spots, see also Fig. 4.

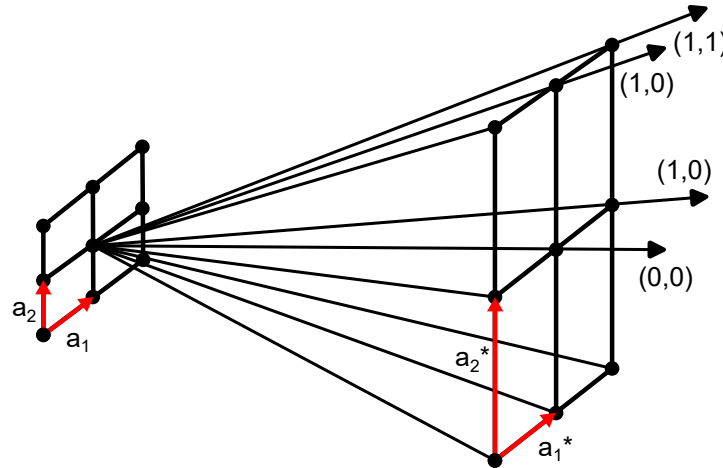


Fig. 4 Sketch of the LEED spots, which represent the reciprocal lattice of a square lattice based on [43].

The (0,0)-spot represents electrons with the same $k_{\parallel}$ as the incoming electron beam. Because we are looking at elastically scattered electrons, the overall magnitude of the

wavevector needs to be conserved:

$$k_{\perp,out}(n_1, n_2) = \left[\frac{2m_e E_{kin}}{\hbar^2} - |\vec{k}_{\parallel,out}(n_1, n_2)|^2 \right]^{\frac{1}{2}} \quad (19)$$

As the energy of the incoming electrons increases, $k_{\perp,out}$ needs to increase as $\vec{k}_{\parallel,out}$ cannot, resulting in the LEED spots moving closer to the (0,0)-spot. As a consequence of this, more LEED spots become visible at higher energies.

Because the diffraction process occurs within only a few atomic layers at the surface, no well-defined Laue condition exists for the perpendicular component of the wavevector and the resulting LEED spots are relatively broad. Additionally, there are still interference effects from these different atomic layers and as a result the LEED spot intensity is not constant with changing beam energy. The information encoded in these fluctuations can be extracted by measuring Intensity-vs-Voltage curves and can provide information about atomic distances. However, this is not relevant for this thesis.

A sketch of the LEED pattern of a $p(1 \times 2)$ -superstructure on a square lattice is shown in Fig. 5. Note how a larger unit cell in real space results in a smaller unit cell in reciprocal space. The relative size and angle of the superstructure unit cell compared to the atomic unit cell, as measured in LEED, directly reveals the size and angle of the unit cells in real space.

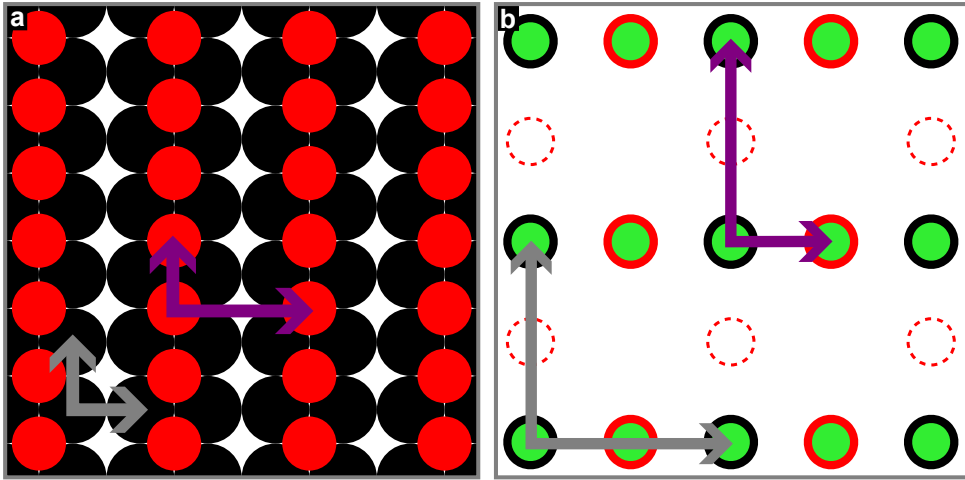


Fig. 5 Sketch of a $p(1 \times 2)$ superstructure on a square lattice in **a** and the corresponding LEED pattern in **b**. Red/purple indicate spots, atoms and unit cells related to the superstructure, while black/grey indicate the same for the atomic lattice. The red dotted circles indicate spots related to the second rotational domain.

In cases where the superstructure does not follow the symmetry of the underlying substrate, rotational domains can exist on the surface. If the spot of the electron beam is much larger than the typical domain size, which is usually the case, the LEED pattern includes a superposition of the LEED patterns of all individual rotational domains. In Fig. 5b, one would therefore expect additional spots at the position of the dotted red circles.

2.3 Magnetism

To understand magnetism in ultra-thin films one needs to understand the mechanisms by which the atomic magnetic moments couple to each other and by which long-range magnetic order can arise. For the magnetism of $3d$ transition-metals, which are investigated in this thesis, the magnetic moments are generally dominated by the intrinsic angular momentum of electrons (spin), while the orbital angular momentum is quenched due to the crystal field of the surrounding atoms. The magnetic moment of an atom is therefore often referred to as spin. In this section, the different possible interactions between neighbouring spins relevant for the systems in this thesis are discussed briefly. A more extensive discussion on magnetism in condensed matter systems can be found in [44].

2.3.1 Heisenberg exchange

The most relevant interaction leading to long range magnetic order is the exchange interaction. It arises as a result of the Coulomb interaction and the Pauli exclusion principle, by which the overall wavefunction of a system is required to be antisymmetric under the exchange of two electrons. This requires either an antisymmetric spin part (singlet) or spatial part (triplet) of the wave function. For a two-electron system it becomes clear that a singlet state corresponds to anti-parallel aligned spins while the triplet state corresponds to parallel aligned spins. The exchange coupling strength or exchange integral J can then be related to the energy difference between the singlet and triplet state.

Such an interaction can be generalized to apply between all neighbouring atoms in the Heisenberg model and takes the form of equation (20):

$$E = - \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \quad (20)$$

where $\mathbf{S}_{i/j}$ is the spin at atomic site i/j , respectively, and J_{ij} represents the exchange integral between the two spins. For $J > 0$ the spins favour ferromagnetic alignment, while for $J < 0$ antiferromagnetic (AFM) alignment is favoured, see Fig. 6a-b. In the AFM case, the axis, that the neighbouring, oppositely-oriented spins are aligned with, is called the quantization axis or the Néel vector. The above Hamiltonian not only includes

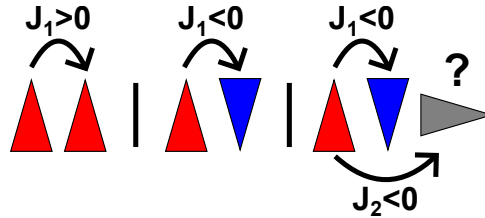


Fig. 6 Sketch of Heisenberg exchange resulting in ferromagnetic/antiferromagnetic alignment in the left and middle panel, respectively and exchange frustration in the right panel.

exchange between nearest-neighbours J_1 , but also next-nearest-neighbours J_2 , and in fact all possible combinations of spins on atomic sites $J_{>2}$. However, the direct exchange interaction is a very short-range interaction as the orbitals of site i and j need to overlap;

so considering only J_1 can be sufficient. In some cases, indirect exchange mechanisms need to be considered as well, which includes mechanisms like the RKKY interaction in metallic systems, where neighbouring sites can couple via the conduction electrons, leading to a larger interaction range.

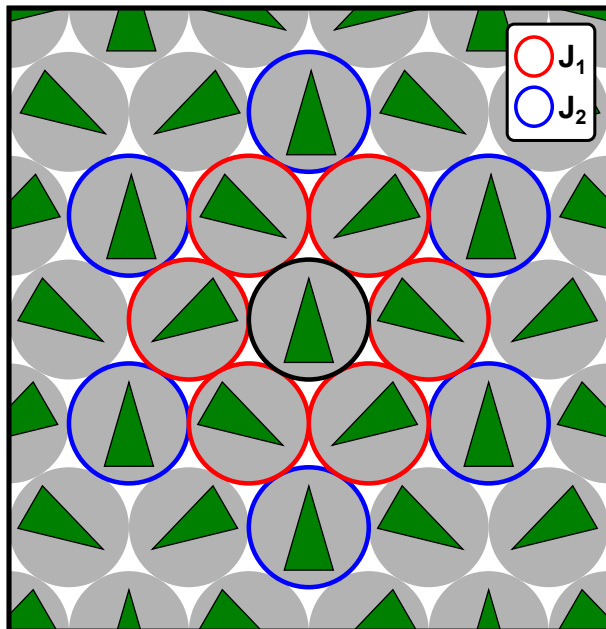


Fig. 7 Sketch of a 120° Néel state. Nearest/next-nearest-neighbours are marked by red/blue circles, respectively.

Magnetic frustration can give rise to non-collinearity, for instance consider purely nearest-neighbour exchange with $J_1 < 0$ on a hexagonal lattice. It is not possible to align all magnetic moments anti-parallel in a collinear fashion, and therefore the resulting AFM state shows spins at 120° angles instead and is called the Néel state, see Fig. 7. This magnetic state was observed experimentally in systems such as Mn/Ag(111) [45] and Cr/Pd(111) [46]. If additionally $J_2 < 0$ and of large enough magnitude, the magnetic ground state will be a row-wise AFM state. This state is relevant for some of the experiments in this thesis and will be discussed in more detail in Sec. 2.3.7. Exchange frustration can arise not just geometrically as for hexagonal lattices, but also when considering different relations between J_1 and next-nearest-neighbour exchange J_2 or higher, see Fig. 6c, with the possibility of the formation of non-collinear spin-spiral states [47].

2.3.2 Higher order exchange interactions

Higher order exchange interactions (HOI) are an extension of the Heisenberg model and consider interactions between more than two spins, which can include terms with 4 spins (4th order), 6 spins (6th order) and higher [48]. For instance, this includes the more commonly considered bi-quadratic exchange interaction:

$$E_{biq} = - \sum_{ij} B_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j)^2 \quad (21)$$

Here, B_{ij} is the bi-quadratic exchange constant and denotes the strength of the coupling. For positive/negative values, this results in a preference for collinear/perpendicular alignment. This interaction is of fourth order and other terms include also the 4spin-3sites and 4spin-4sites interactions.

Overall, while these HOI are typically small, they can play a role in cases where the energy between different magnetic states is very close. This can be the case for multi-Q states that are superpositions of spin-spiral states, as these are degenerate with their constituent states when only considering Heisenberg exchange. HOI can lift this degeneracy. For the case of an Fe-ML on the Rh-ML or Rh-DL on Ir(111), a significant contribution of the HOI affecting the overall spin-structure was found; the HOI exchange constant was determined to be quite large and to be up to ≈ 5 meV [49].

2.3.3 Dzyaloshinskii-Moriya-interaction

Another interaction that can lead to non-collinearity in magnetic films is the anisotropic exchange interaction also called Dzyaloshinskii-Moriya-Interaction (DMI) [50, 51]. It requires spin-orbit coupling and broken inversion symmetry. Inversion symmetry in thin-film systems is broken at the interface. The DMI can be mediated by a third lattice site, with increasing interaction strength for mediating sites with large SOC [52, 53]. The contributions from all the mediating sites $\mathbf{D}_{ijn}$, see Fig. 8, can be combined into the DMI vector $\mathbf{D}_{ij}$. The DMI energy is then given by [51]:

$$E = - \sum_{ij} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) \quad (22)$$

Here, $\mathbf{S}_i/\mathbf{S}_j$ represent the spins on lattice sites i/j , respectively. The DMI favours perpendicular spins, oriented in the plane perpendicular to the DMI vector. In magnetic ultra-thin film systems, it can introduce non-collinearity by leading to a coherent spin rotation with a particular rotational sense, depending on the direction of the DMI vector [54], see also Fig. 8. The resulting magnetic states are called spin-spirals, where a ferromagnetic spin-spiral indicates a coherent rotation of the magnetization across the spin-spiral period. AFM spin-spirals also show a continuous spin rotation of the quantization axis (Néel vector); however, nearest-neighbour angles $> 90^\circ$ are possible. This direction can be determined according to the symmetry rules described by Dzyaloshinskii and Moriya [50, 51]. At surfaces, this vector is typically oriented in-plane leading to Néel type domain walls with an out-of-plane component.

2.3.4 Magneto-crystalline anisotropy

SOC also leads to magneto-crystalline anisotropy, where the spins interact with the crystal field of the surrounding lattice. This makes certain orientations of the spins energetically favourable. Such a direction is called an easy-axis. The magneto-crystalline anisotropy energy (MAE) can be described in first approximation as uniaxial for surfaces, see equation (23) [44, 55]:

$$E_{\text{MAE}} = - \sum_i K (\mathbf{S}_i \cdot \mathbf{n})^2 \quad (23)$$

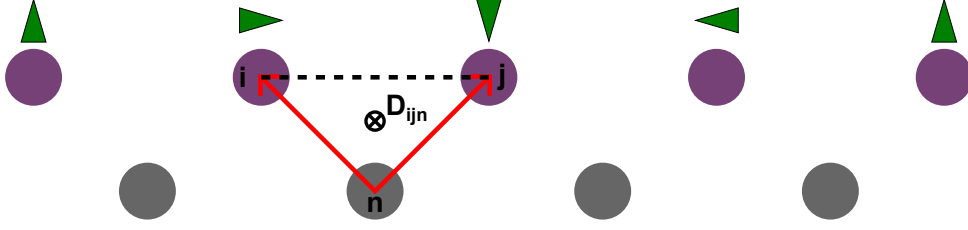


Fig. 8 Sketch of the DMI leading to a 90° spin-spiral with a particular rotational sense. Atomic sites i and j represent magnetic atoms at the surface while site n indicates one of the mediating atoms in the layer below.

with K being the anisotropy constant, and $\mathbf{n}$ the normal vector of the surface. For $K > 0$ the spins preferentially align out-of-plane, while they align in-plane for $K < 0$, also referred to as easy-plane anisotropy. Note that the energy of spins oriented parallel or anti-parallel to the anisotropy axis is degenerate. The differences in anisotropy energy for the alignment of spins along the different crystallographic directions inside the plane of the film is typically small and can be neglected.

2.3.5 Anisotropic symmetric exchange

The anisotropic symmetric exchange interaction (ASE) is related to SOC as well. It is also referred to as a pseudo-dipolar interaction and can be written as shown in equation (24) [56] and has a form reminiscent of the dipole-dipole interaction, see equation (25).

$$E_{\text{ASE}} = - \sum_{ij} J_{ij}^{\text{ASE}} (\mathbf{S}_i \cdot \mathbf{d}_{ij}) (\mathbf{S}_j \cdot \mathbf{d}_{ij}) \quad (24)$$

Here, $\mathbf{d}_{ij}$ is the unit vector between site i and j , and J_{ij}^{ASE} is the ASE exchange constant. For $J_{ij}^{\text{ASE}} > 0 / J_{ij}^{\text{ASE}} < 0$ the spins preferentially orient parallel/perpendicular to $\mathbf{d}_{ij}$. ASE is typically neglected, but was found to play a role for the RW-AFM state of Mn/Re(0001), see Sec. 2.3.7. The same is true for the dipole-dipole interaction, where the energy of two magnetic dipoles has the form described in equation (25) [44]:

$$E_{\text{dipolar}} = \frac{\mu_0}{4\pi|\mathbf{r}|^3} [\boldsymbol{\mu}_i \cdot \boldsymbol{\mu}_j - \frac{3}{|\mathbf{r}|^2} (\boldsymbol{\mu}_i \cdot \mathbf{r})(\boldsymbol{\mu}_j \cdot \mathbf{r})] \quad (25)$$

2.3.6 Zeeman energy

In an applied magnetic field, the energy of the magnetic moments can be described by equation (26) [44]:

$$E_B = - \sum_i \mu \mathbf{S}_i \cdot \mathbf{B} \quad (26)$$

with μ corresponding to the magnitude of the magnetic moment at atomic site i and $\mathbf{B}$ corresponding to the applied magnetic field. The spins gain energy, called the Zeeman energy, by aligning themselves with the direction of the magnetic field. This energy term can be neglected for the antiferromagnetic systems investigated in this thesis as it

will not lead to any change in the magnetic state at experimentally achievable magnetic field strengths. However, it is relevant for the soft-magnetic Fe/W-tips employed during some of the measurements shown in this thesis, as their magnetization is changed using a magnetic field.

2.3.7 The RW-AFM state of Mn/Re(0001)

As Sec. 4.2 and Sec. 4.3 discuss results obtained on the RW-AFM state of fcc-stacked Mn/Re(0001), this system will be briefly introduced here. RW-AFM states exist for hexagonal magnetic layers with a nearest-neighbour coupling $J_1 < 0$ and next-nearest-neighbour coupling $J_2 < -\frac{1}{8}J_1$ [47]. The spins along one of the close-packed rows are all oriented in the same direction, called the $\uparrow\uparrow$ -row direction, while the spins one row across are inverted, see left sketch Fig. 9. This magnetic state can also be referred to as the 1Q state, because it can be described as a spin-spiral with a single Q-vector. In SP-STM images a magnetic contrast of bright and dark lines is visible, see Fig. 10a.

For the system of fcc-Mn/Re(0001), the spins further align along the $\uparrow\uparrow$ row direction due to an easy-plane anisotropy and due to the dipole-dipole interaction, as well as the ASE, which are of similar magnitude [56].

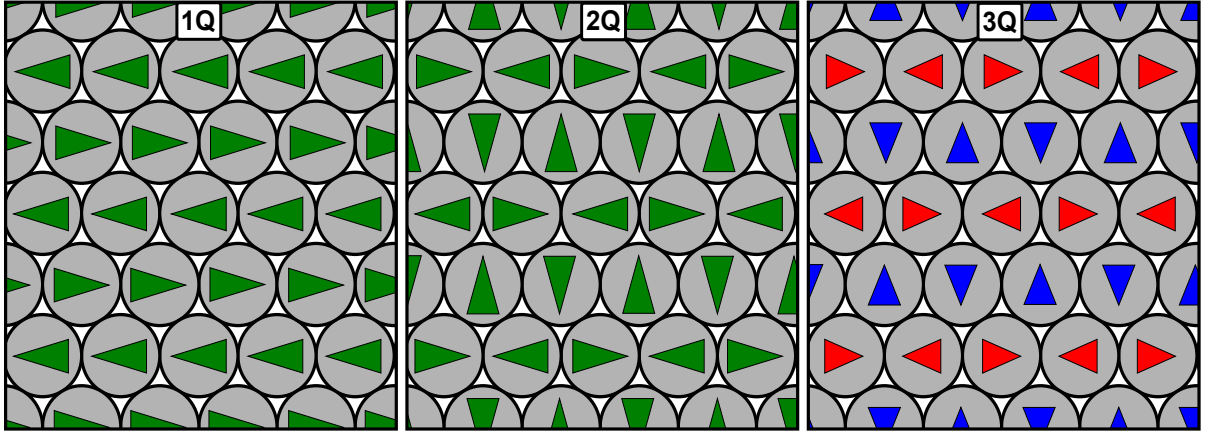


Fig. 9 Sketch of the RW-AFM (1Q) state (left), 2Q state (middle), and 3Q state (right). The spins are represented by cones and the color corresponds to: red (up), blue (down), green (in-plane).

As the RW-AFM state breaks the symmetry of the hexagonal lattice one finds three rotational domains of this magnetic state, see Fig. 10a. At the interface between rotational domains, domain walls are observed, which host a magnetic 2Q state in their centre [57]. This state is a superposition of two RW-AFM states and can be described using two Q-vectors, see middle sketch in Fig. 9. Neighbouring spins are rotated by 90° or 180° resulting in a (2×2) magnetic unit cell.

The domain walls are electronically different to the domains and can be imaged directly using a non-magnetic tip, likely due to NCMR/TAMR effects, see Fig. 10b. In this system, the uniaxial magnetic state breaks the symmetry of the electronic states. This in turn results in an asymmetry in the shape of some of the defects. As this asymmetry is directly linked to the magnetic state, the appearance of the defects, which can be imaged using a

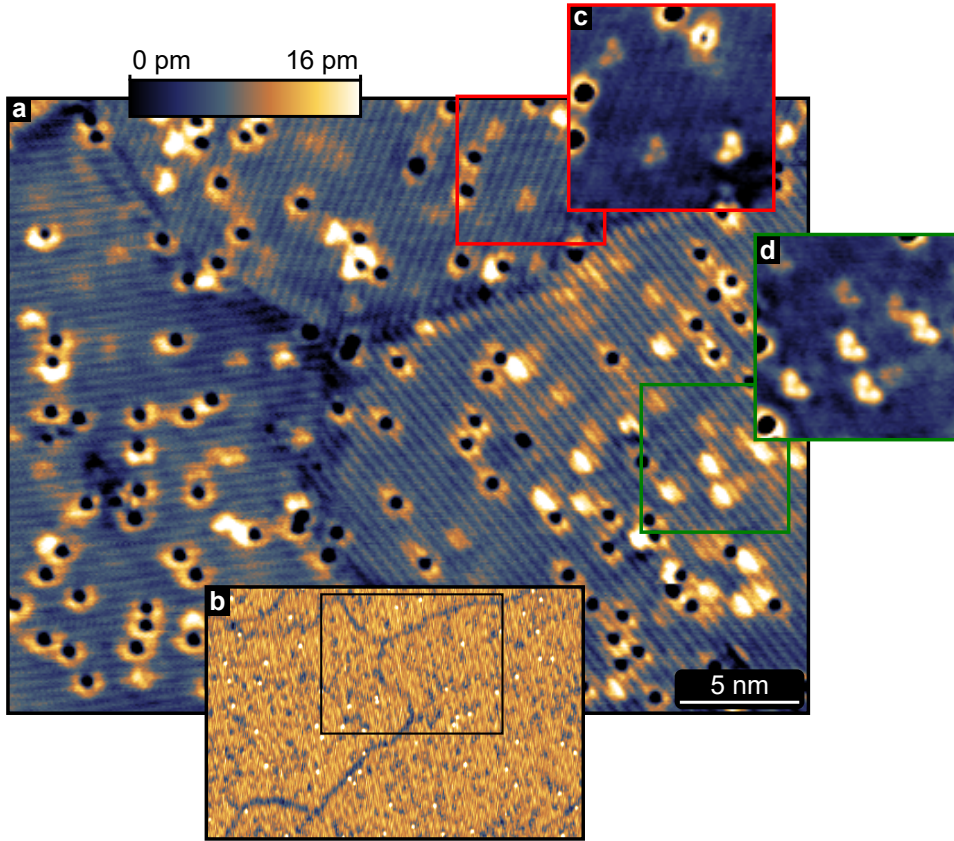


Fig. 10 **a**, SP-STM image of the three rotational domains of the RW-AFM state on Mn/Re(0001). $U = -30$ mV, $I = 7$ nA. **b**, dI/dU -map of a large area surrounding the scan frame of **a**, see black rectangle. $U = 500$ mV, $I = 3$ nA. **c-d**, Close-up views of the regions in the respectively coloured rectangles in **a**. The tip magnetization changed to show a much-reduced magnetic contrast, while the defects clearly indicate the direction of the $\uparrow\uparrow$ -rows. The colour scale has been adjusted. $U = -30$ mV, $I = 7$ nA. all: $T = 4.2$ K. All images were measured by Jonas Spethmann.

non-magnetic tip, can be used to determine the orientation of the $\uparrow\uparrow$ -rows, see Fig. 2.3.7c-d.

A superposition of three RW-AFM states results in the magnetic state shown in the right sketch of Fig. 9. It can be constructed using three Q-vectors and is therefore also referred to as the 3Q state. Here, neighbouring spins are rotated by the tetrahedron angle with the same unit cell as the 2Q state.

When considering the RW-AFM state and its superposition states using only Heisenberg exchange, one finds them to be energetically degenerate. HOI can differentiate between them. This state is experimentally observed for the hcp-stacked Mn monolayer (ML) on Re(0001) [56] or at junctions between three rotational domains of the RW-AFM state in the system of Mn double-layer (DL) on Ir(111) [58].

2.3.8 Topology in magnetism

Topology in condensed matter systems is a tool to sort states into groups based on the possibility of continuously deforming one into the other. The classic example to illustrate this is given by a cup and a donut, as a cup can be moulded into the shape of a donut without the need for any cutting or ripping (i.e. they can be continuously deformed) and vice versa. One can then define a topological invariant, in this case the number of holes, which is the same for all systems that can be continuously deformed into each other and these groups are called topological phases. States in a topological phase with a different topological invariant (e.g. objects with two holes), cannot be continuously deformed into a donut.

In magnetism a plethora of topological states exist as windings in the magnetization, akin to a knot in a rope that cannot be easily unwound. In 2D this includes skyrmions, which have particle-like properties and are also referred as topological defects, see Fig. 11. The topological invariant that can be defined for these systems is called the winding number or topological charge w and it describes the number of times the spins wrap around the unit sphere [59].

$$w = \frac{1}{4\pi} \int_M \mathbf{m} \cdot \left(\frac{\partial \mathbf{m}}{\partial x} \times \frac{\partial \mathbf{m}}{\partial y} dx dy \right) \quad (27)$$

with

$$|\mathbf{m}| = 1 \quad (28)$$

$$\mathbf{m}(\mathbf{r} \rightarrow \text{inf}) = \text{const} \quad (29)$$

Here, $\mathbf{m}$ is the magnetization at point $\mathbf{r}$ and M represents the 2D manifold, which is the compactified plane $\mathbb{R}^2$ or a 2-sphere when using spherical coordinates [59]. The topological charge of a collinear ferromagnetic state is zero and it is therefore considered topologically trivial. For skyrmions in an OOP ferromagnetic background a topological charge of ± 1 can be calculated, while merons live in an IP ferromagnetic background, have a topological charge of ± 0.5 and can form pairs known as bi-merons. Any continuous deformation of the magnetization configuration of a skyrmion will not change the topological charge, resulting in an infinitely high energy barrier to the ferromagnetic state. Note that this only applies for continuous magnetization fields, as the discrete nature of real systems results in a finite energy barrier. Additionally, the anisotropy constant and the size of real systems are finite as well, allowing for pathways to create and destroy these topological defects.

Skyrmion lattices have been experimentally found in chiral magnets [60, 61], and monolayer films [62], while individual skyrmions were first found in ultra-thin films [9, 12] and subsequently in multilayer systems [63, 64]. DMI typically plays an important role in their stabilization together with Heisenberg exchange, magnetic field (i.e. Zeeman energy) and easy-axis OOP anisotropy. The dipolar interaction becomes more relevant as the skyrmion size increases. Depending on the DMI vector, skyrmions come in two types, Néel- and Bloch-type, corresponding to cycloidal and helical rotation of the spins, respectively, see also Fig. 11a-b. In ultra-thin films, Néel-type skyrmions are typically favoured due to the orientation of the DMI vector at the surface, see Sec. 2.3.3.

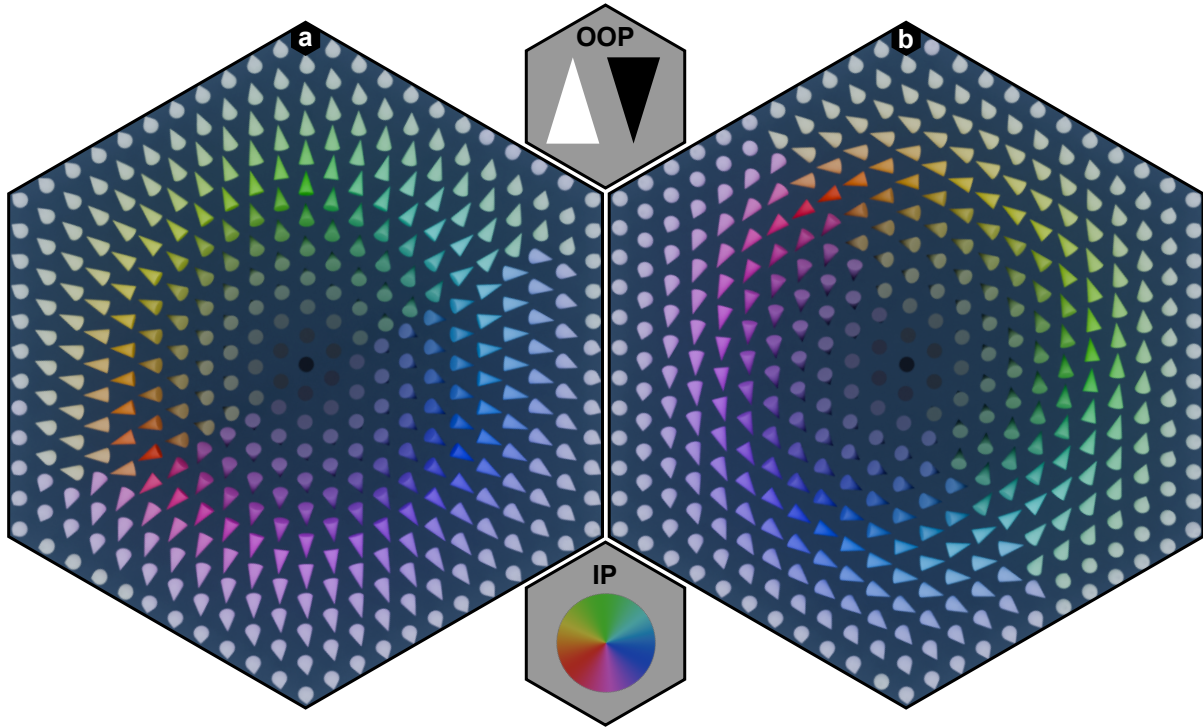


Fig. 11 **a**, 3D sketch of a Néel-type skyrmion. **b**, 3D sketch of a Bloch-type skyrmion. The cones represent the atomistic magnetic moments. White/black colouring represents OOP up/down, respectively, while the IP-directions are represented in the colours of the hue wheel as indicated in the legend. The skyrmions were simulated in Monte Crystal [18] with periodic boundary conditions, resulting in a small amount of distortion.

The principles of topology discussed so far can also be applied to antiferromagnetic systems; here it is useful to separate the AFM lattice into ferromagnetic sub-lattices. Such localized topological spin textures can then exist on each sub-lattice, with opposite topological charge, resulting in a cancellation when considering all sublattices similar to the net magnetization in AFM systems. With the topological charge being always zero, AFM skyrmions do not show the topological Hall-effect [15, 16]. Experimentally, they have so far been mainly observed in synthetic antiferromagnets, which are layered ferromagnets, with weak AFM interlayer coupling between ferromagnetic layers [65, 66]. AFM merons and anti-merons have been observed at the interface between an AFM Fe_2O_3 thin film and a Pt overlayer, where the ferromagnetic sublattices are still located in different layers [67]. While theoretical predictions for AFM skyrmions or merons within a single AFM layer, where the ferromagnetic sublattices are interlaced, exist [68], no experimental results have been reported so far.

2.4 Superconductivity

Superconductors exhibit two special traits: Below a critical temperature T_c , their resistance to electrical current disappears, as discovered by H. Kamerlingh Onnes in 1911 [69]. Secondly, they expel magnetic field lines when cooled below the same critical temperature

while inside a magnetic field. This effect is also called the Meissner-Ochsenfeld-effect [70] and is also referred to as perfect diamagnetism. The behaviour one would expect of a perfect conductor is decidedly different, here magnetic field lines may be prohibited from entering the material but the expulsion of magnetic flux would not occur. For superconductors a critical magnetic field H_c exists, where superconductivity is destroyed. The strength of this critical field is related to the condensation energy of the superconducting state (i.e. the difference in free energy between normal-conductor and superconductor).

The two traits of a superconductor were described phenomenologically by F. and H. London in the London equations [71] and are given by [72]:

$$\mathbf{E} = \frac{\partial}{\partial t}(\Lambda \mathbf{J}_s) \quad (30)$$

$$\mathbf{h} = -c(\nabla \times (\Lambda \mathbf{J}_s)) \quad (31)$$

with

$$\Lambda = \frac{m_e}{n_s e^2} \quad (32)$$

Here, m_e , and e are the mass and charge of the electrons, c is the speed of light, and n_s the density of electrons. ∇ is the Nabla operator, $\mathbf{J}$ describes the current density, $\mathbf{E}$ the electric field, and $\mathbf{h}$ the magnetic field. The first equation describes the perfect conductivity of a superconductor; notice how in the absence of an electric field all currents are unchanging. The second equation together with the Maxwell equation:

$$\nabla \times \mathbf{h} = \frac{4\pi \mathbf{J}}{c} \quad (33)$$

gives

$$\nabla^2 \mathbf{h} = \frac{\mathbf{h}}{\gamma_L^2} \quad (34)$$

where γ_L is referred to as the London penetration depth. It describes the phenomenon of perfect diamagnetism and the exponential screening of a magnetic field from the inside of the superconductor.

2.4.1 BCS theory

The first microscopic theory of superconductivity was developed by Bardeen, Cooper, and Schrieffer in 1957 and was named BCS-theory accordingly [73]. In prior years, an energy gap between the ground state of the superconductor and the quasi-particle excitations with a size of $\approx 3 - 4k_B T_c$ had been observed experimentally [74–77]. The BCS theory explains this with two electrons with opposite momentum and spin forming a Cooper pair due to their coupling through phonons (i.e. vibrational excitations of the crystal lattice). This electron-phonon coupling has been verified by measurements of an isotope effect [78, 79]. A minimum energy 2Δ is needed to break this Cooper pair with Δ for $T \ll T_c$ given by:

$$\Delta(0) = 1.764 k_B T_c \quad (35)$$

Here, k_b is the Boltzmann constant. This gap in the quasi-particle excitation spectrum as described by [80] shows itself as a gap in the electronic DOS of the superconductor. The DOS can be written as:

$$D_s(E) = \begin{cases} D_n(E_F) \frac{E}{\sqrt{E^2 - \Delta^2}} & \text{for } E > \Delta \\ 0 & \text{for } E < \Delta \end{cases} \quad (36)$$

D_s/D_n represent the DOS of the superconductor and the normal conductor respectively. The latter can be typically assumed to be constant in the vicinity of E_F for metals. No states exist between $-\Delta$ and $+\Delta$ as Δ represents the smallest possible quasiparticle excitations. For realistic systems one has to consider the finite lifetimes of the quasiparticles as well, which leads to an intrinsic broadening of features. This is described by the Dynes function [81]:

$$\frac{D_s(E)}{D_n} = \text{Re} \left\{ \frac{E - i\Gamma}{\sqrt{(E - i\Gamma)^2 - \Delta^2}} \right\} \quad (37)$$

Here, Γ is the intrinsic broadening parameter. The resulting LDOS for a gap $\Delta = 1$ meV and a broadening $\Gamma = 0.03$ meV at 0 K is shown in Fig. 12b. For the experimental results presented in this thesis, we measure the LDOS of the superconductor using tunnel spectroscopy, see Sec. 2.1.2.

Here, electrons tunnel from a metallic tip into the superconducting sample or vice versa. The occupation of states around the Fermi level is smeared out, as described by the Fermi function, see red curves in Fig. 12a. Tunneling from the occupied states in the tip to the unoccupied states in the sample or vice versa therefore leads to additional broadening of features due to temperature see Fig. 12a. In Fig. 12b simulated dI/dU -spectra are shown for different temperatures. Broadening due to the bias modulation has been neglected here. In general, the broadening due to temperature and modulation bias is given by a convolution of the LDOS with a Gaussian of FWHM [82]:

$$\sqrt{\left(\frac{3.2k_B T}{e}\right)^2 + \left(2\sqrt{2}V_{\text{mod}}\right)^2} \quad (38)$$

This indicates that at a measurement temperature of 1.3 K a maximum resolution of $\approx 360 \mu\text{eV}$ is achievable and this does not take into account additional broadening due to bias modulation and noise such as RF-noise.

The energy resolution can be further enhanced beyond the temperature limits by using superconducting tips, as the sharp coherence peaks of the tip are used to probe the electronic features of the sample instead [83]. However, this is not relevant for this thesis as we will be using metallic/spin-polarized tips for all spectroscopy measurements, because our emphasis is on an atomic scale correlation of magnetic and superconducting properties.

2.4.2 Unconventional superconductivity

In the BCS theory only a conventional pairing mechanism, namely via phonons, leading to the formation of pairs of electrons with opposite spin (i.e. singlet pairing) is considered.

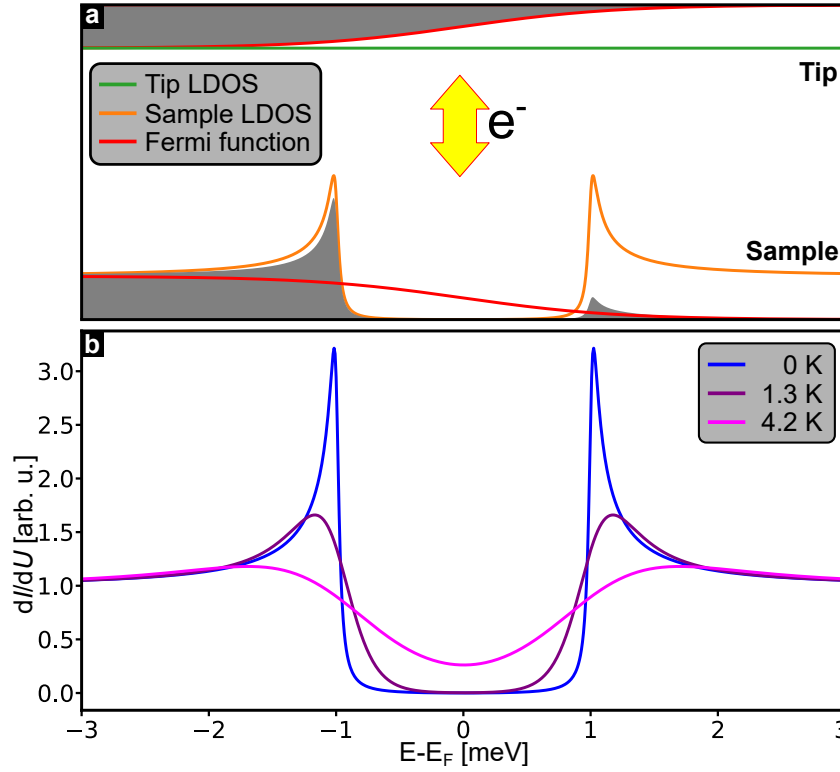


Fig. 12 a, Sketch of the tunneling between a metallic tip and a superconducting sample. The LDOS of tip and sample are coloured in green/orange, respectively. The LDOS of the sample was calculated using the Dynes function with $\Gamma = 3 \times 10^{-2}$ meV, $\Delta = 1$ meV. The Fermi function is shown in red and for illustration purposes a smeared-out distribution at $T = 7$ K was chosen. The grey-coloured regions under the LDOS of tip and sample represent occupied states according to this Fermi function. **b**, Simulated dI/dU -spectrum around the Fermi level for different temperatures with the same parameters $\Gamma = 3 \times 10^{-2}$ meV, $\Delta = 1$ meV.

This is the case for elemental superconductors like Tantalum and Rhenium for instance. Unconventional superconductors are superconductors that cannot be described by the BCS theory. Differently mediated pairing mechanisms exist, for example for cuprates, which have much higher critical temperatures. Here, antiferromagnetic spin-fluctuations are proposed to play a role [84].

Superconductors are grouped together based on the symmetry of their superconducting order parameter Δ and named according to the symmetries of the atomic Hydrogen orbital (s , p , d , f , ...). For spin singlet pairing the superconducting order parameters is of even parity and of s -wave or d -wave symmetry. Here, s -wave superconductors refer to the $\mathbf{k}$ -independent order parameter of conventional superconductors as described by BCS theory, while evidence for d -wave superconductivity exists in cuprates [84], where the superconducting order parameter is dependent on $\mathbf{k}$.

In some cases, pairing can occur between electrons with the same spin (i.e. triplet pairing). These superconductors are also considered to be of p -wave (or f -wave) type and exhibit odd-parity. Intrinsic p -wave pairing appears to be very rare as indications have so far been found only for Sr_2RuO_4 [85] and UTe_2 [86]. However, p -wave pairing can be

induced in magnet-superconductor-hybrid systems.

2.4.3 Type-I and type-II superconductors

The spatial extent of Cooper pairs is described by the characteristic length called the coherence length ξ . This corresponds, according to Ginzburg and Landau, to the distance across which the superconducting order parameter can vary [87]. Together with the London penetration depth γ_L they define the Ginzburg-Landau parameter κ .

$$\kappa = \frac{\gamma_L}{\xi}. \quad (39)$$

This dimensionless parameter is approximately temperature independent. Abrikosov found that superconductors can show different behaviour regarding increasing magnetic fields [88].

For the case of $\kappa \ll 1$ they behave as expected, we find a critical magnetic field h_c where the superconductivity breaks down to the normal-conducting state; this type of superconductor is called type-I.

However, for $\kappa \gg 1$ the coherence length becomes smaller than the penetration depth and the surface energy can become negative, which should allow the formation of interfaces between the superconducting and normal-conducting state to become favourable. Indeed, starting at a magnetic field $h_{c,1}$, the flux lines can penetrate the superconductor and form an array of flux tubes also called the Shubnikov phase, indicating that these superconductors are no longer perfect diamagnets. Only at a field value $h_{c,2}$ is the superconductivity completely destroyed. These superconductors are type-II superconductors and the crossover between type-I and type-II occurs at $\kappa = 1/\sqrt{2}$.

2.4.4 Magnet-superconductor-hybrid systems

In the previous section we have seen that superconductivity can be suppressed by a magnetic field. Similarly, magnetic impurities inside the superconductor can lead to a decrease in the critical temperature and a change in the DOS as found by Abrikosov and Gor'kov [89]. What happens at the limit of a single magnetic impurity was investigated by Yu, Shiba and Rusinov [90–93].

YSR-states The single magnetic impurities act as a pair-breaking potential, locally reducing the energy required to break Cooper-pairs apart. This is because the impurity spin acts as a scattering centre with an exchange component, which tends to misalign the spins of the Cooper pairs. The results are particle-hole symmetric states inside the superconducting gap (YSR-states), with their energy depending on the coupling strength to the superconductor. The strength of this coupling can be related to the coupling of the impurity spin to the conduction electrons in a normal conductor (i.e. the Kondo effect) [94]. In normal conductors the impurity spin can be screened by a cloud of polarized conduction electrons, forming a many-body singlet ($S = 0$) ground state.

In superconductors the Kondo coupling competes with the superconducting ground state. For an impurity on a superconductor, we find the formation of a many-body ground

states with a dependence on the relative strength of the Kondo coupling $k_B T_K$ and the superconducting condensation energy Δ , where T_K is the Kondo temperature and k_B is the Boltzmann constant [95, 96].

For $k_B T_K \gg \Delta$ (i.e. strong coupling between impurity spin and superconductor, we find the screened impurity spin and the formation of a many-body singlet ground state. A YSR excitation to the first excited state, through tunneling electrons for instance, shows up as a state very near the coherence peaks in the DOS of the superconductor. For $k_B T_K \ll \Delta$ (i.e. weak coupling) the Kondo correlations cannot fully screen the spin and the many-body ground state is magnetic ($S > 0$), also called free-spin state. The YSR excitation to the screened state ($S \pm 1/2$) similarly lies very close to the coherence peaks.

Only for regimes where both energies are of a similar scale $k_B T_K \approx \Delta$ do we find YSR states clearly inside the superconducting gap. The quantum phase transition between excitations from the free-spin ground state to the screened excited state to excitations from the screened ground state to the free-spin state occurs at $k_B T_K \approx 0.3\Delta$ [96]. The spectral-weight of the particle/hole-like excitations is not necessarily symmetric around the Fermi level, which can be attributed to an influence of the Coulomb potential and asymmetries in the DOS of the normal state [96].

The spatial extent of the YSR states shows a $1/(k_F R)$ dependence for short distances and an exponential decay dependent on the superconducting coherence length for large distances [92].

Topological superconductivity A general requirement of topological superconductivity in MSH systems is effective p -wave pairing. To achieve this, s -wave superconductivity of the substrate, time-reversal symmetry-breaking due the magnetic moments in the magnetic layer, and Rashba spin-orbit coupling are necessary [97]. At surfaces inversion symmetry is broken, resulting in Rashba SOC which itself breaks the spin-symmetry, allowing for a mix of s -wave and p -wave pairing [98].

In the last decade 1D chains of magnetic atoms have been built on s -wave superconductors. Here, hybridization leads to the formation of bands inside the superconducting gap and gapped topological superconducting phases have been found [99]. Similarly, for 2D-magnetic layers on an s -wave superconductor evidence for topological superconductivity [29] and nodal-point topological superconductivity (TNPSC) [100] have been found.

In gapped systems a topological invariant, called the Chern number, can be defined and phases with different Chern numbers cannot be continuously deformed into one another, instead the gap has to close at some point between the two phases. Spatially, this leads to edge modes between regions of different Chern numbers and this is also referred to as bulk-boundary correspondence. In 1D systems, this leads to edge modes at the ends of the chains, while for 2D islands this leads to a chiral edge mode propagating around the island edge, see Fig. 13a. These edge modes are topologically protected against perturbations by the topological gap. In both cases, edge states are possible candidates for Majorana modes (MM) [27, 29], which are of interest for fault-tolerant quantum computing applications; however, definitive proof remains challenging.

For topological nodal-point superconducting systems [101, 102], the Chern number is not defined, as the system is not gapped. Here, the nodal-points (i.e. points in the

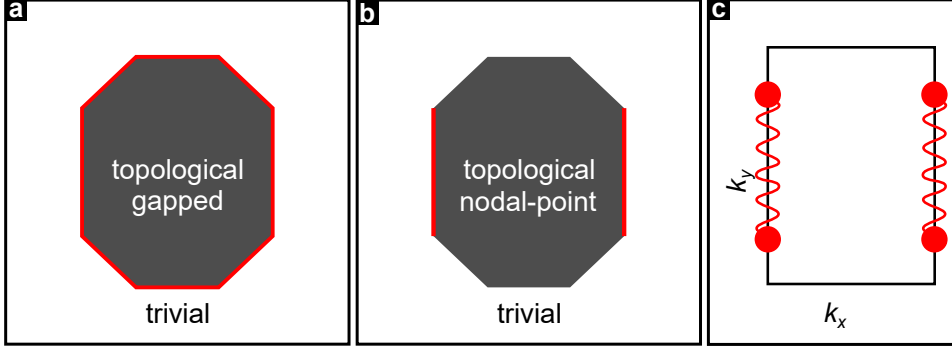


Fig. 13 **a**, Sketch of an edge mode propagating at the edge between a gapped topological superconducting phase and the trivial superconductor. **b**, Sketch of an edge mode propagating only at the vertical edge between a nodal-point topological superconducting phase and the trivial superconductor. **c**, Sketch of the Fermi surface with potential nodal points marked in red with edge modes propagating between nodal points as indicated by the red lines, corresponding to the nodal-point phase in **b**.

Brillouin zone where the band gap is closed) are of topological character. They always exist as pairs with opposite topological charge and may only detach from the Fermi surface if they merge and are located at the same point in the Brillouin zone.

Similar to gapped systems, at the boundary between different topological phases (i.e. different number of nodal-points) edge modes exist [100]. An edge mode always propagates between two nodal points and depending on the layout of nodal points and edge modes between them, edge modes in real space are not required to exist for all edge directions and can only appear on edges with certain crystallographic directions, see Fig. 13b-c.

Collinear AFM systems are effectively time-reversal symmetric, because the state can be reproduced by performing a time-reversal operation followed by a translation operation. They therefore exhibit a TNPSC for a large part of their phase space [102]. Similar arguments can be made for coplanar spin-spiral states, therefore, to search for gapped topological superconducting systems in AFM layers, non-coplanar magnetic states are more suitable [103–105].

2.5 Diffusion and growth

Diffusion plays a fundamental role in the growth of thin films. The standard picture represents diffusion as a random-walk of nearest-neighbour jumps. However, longer jumps have been found to play a role at temperatures as low as $< 0.1T_M$, where T_M is the melting point of the metal substrate, and at higher temperatures they play an increasingly larger role [106]. For self-diffusion processes (i.e. Ir adatom on Ir substrate), additional processes such as exchange with substrate atoms may need to be considered as well. The diffusivity is given in transitional state theory as [106]

$$D = \nu l^2 \exp\left(\frac{S_D}{k_B}\right) \exp\left(-\frac{E_D}{k_B T}\right) \quad (40)$$

where ν is the effective vibrational frequency of the adatom, l is the jump length, and S_D/E_D are the entropy/activation energy for diffusion. The expected mean-square displacement $\langle \Delta x^2 \rangle$ is then given by:

$$\langle \Delta x^2 \rangle = 2Dt, \quad (41)$$

with t representing time. With increasing temperature, the diffusivity becomes larger and adatoms diffuse more quickly on the surface.

At high enough temperatures, the energy of the atoms is sufficient and islands can form with the thermodynamic equilibrium shape. At lower temperatures the island shape can be driven kinetically and one has to differentiate between free diffusion on a terrace and diffusion along edges, as they can show different temperature dependencies.

The relevance of these different diffusion processes was demonstrated for the system of Pt/Pt(111) [107]. Michely *et al.* showed that the Pt growth at 200 K is kinetically driven and that Pt grows in a fractal shape, because edge diffusion is not possible and the Pt atoms "stick" at the first position they "hit" at an edge (hit and stick mechanism). Additionally, they show that different temperature-dependent diffusion along the triangularly coordinated edges and the rectangularly coordinated edges, see Fig. 14a, leads to a change in island shape for Pt islands grown at 400 K, 455 K, and 640 K. In this regime the growth is still kinetically driven, only at a temperature of 710 K has a local 2D equilibrium been reached and the growth is thermodynamically driven.

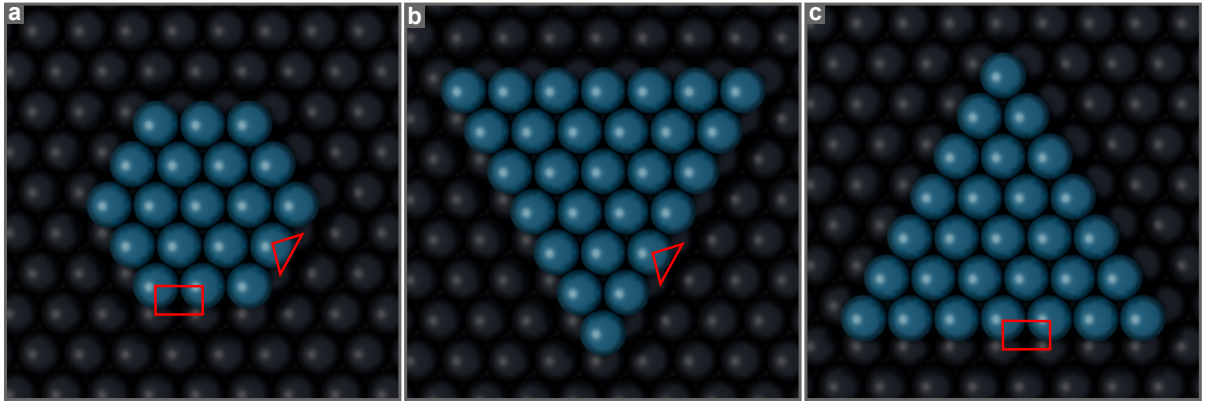


Fig. 14 **a**, Sketch of a hexagonal island on a hexagonal lattice, with the two different edge types marked by the red square/triangle, respectively. **b**, Sketch of a triangular island on a hexagonal lattice with only the edge with triangular coordination present. **c**, Sketch of a triangular island on a hexagonal lattice with only the edge with square coordination present.

The change in the island shape at different temperatures can be explained by the fact that the edges that grow the slowest end up dominating in the final island shape. Fig. 14b corresponds to fast growth at the squarely coordinated edges, so only the triangularly coordinated edges remain, while Fig. 14c corresponds to the opposite behaviour. This difference in the growth speed for the two differently coordinated edges could be related to different diffusion rates on one hand, but also possibly to different rates of adatoms at the edges evaporating back onto the terraces. The latter effect was first discussed by Mo *et al.* for Si on Si(001) [108] but was excluded in the case of Pt on Pt(111).

The sample temperature during growth can not only affect the shape of the islands but also their density. At low temperature, adatoms meet each other before reaching the step edges and form immobile clusters, resulting in the growth of many individual islands. In contrast, at higher temperatures the atoms move to the step edges quickly and spend less time on the terraces while small clusters may still be mobile, resulting in step-flow growth.

3 Methods

All experiments were carried out in an ultra-high vacuum (UHV) system with a base pressure $2 \cdot 10^{-10}$ mbar. This pressure was achieved after a bake-out using a mix of turbo pumps, ion getter pumps, and Ti sublimation pumps and allows for the preparation of clean surfaces that remain uncontaminated for hours/days. Note that the pressure in the STM chambers used for measurements is typically better.

Multiple different chambers were used for sample cleaning, sample preparation, LEED measurements, and STM measurements, with in-situ transfers between them being possible. We will discuss general preparation details in this section, but the details regarding specific sample systems will be discussed in the corresponding sections.

3.1 Experimental setup

The single crystal samples were bound on W sample plates with W wire before being introduced to the UHV. Cleaning was done using a combination of sputtering (i.e. Argon ion bombardment) and annealing on an electron-beam heating stage (e-beam stage). The sputtering was done by dosing Argon atoms using a finely adjustable leak valve at a chamber pressure of $5 \cdot 10^{-6}$ mbar resulting in a sputter rate of approximately 4 atomic layers (AL) per hour. Similar leak valves were also used for dosing Oxygen and Hydrogen gas.

Crystals like Ir(111), Ir(001), and Re(0001) were additionally annealed in an Oxygen atmosphere to remove Carbon impurities. Oxygen partial pressures in the range of $5 \cdot 10^{-8}$ mbar to $2 \cdot 10^{-7}$ mbar and a temperature-ramp up to 900 – 1000°C for Ir samples and 1100°C for Re samples were used. Before metal deposition, a flash annealing was performed and the deposition temperature was controlled by the time before the start of the deposition. The estimated sample temperature 10 min after the flash is $\approx 100 - 200^\circ\text{C}$ and a complete cool-down to RT is only achieved after ≈ 90 minutes.

Mn was evaporated from a pyrolytic boron-nitride (PBN) Knudsen cell of volume 2cm^3 held at a temperature of 690°C, resulting in a deposition rate of ≈ 0.2 AL per minute. Fe and Co were evaporated from e-beam evaporators with a 2 mm rod of each material. One of the Co evaporators was mounted to allow deposition onto a sample in one of the STMs without requiring a large retraction of the tip, which in principle enables measurements of the same sample area before and after evaporation.

The prepared samples were then moved to one of the two STMs for measurement, where they were also cooled down to the respective measurement temperatures. The measurements were performed in two home-built STMs. One operates at a base temperature of 4.2 K with the capability of cooling down to 1.3 K using a Joule-Thompson cycle. Additionally, a magnetic field up to 9 T can be applied out-of-plane. All measurements in this STM were performed using antiferromagnetic Cr-bulk tips. The second STM operates at a base temperature of 8.4 – 8.8 K but can be operated at 80 K using liquid nitrogen instead of liquid helium. It allows for a perpendicular magnetic field up to 2.5 T. In this STM both antiferromagnetic Cr-bulk tips, as well as ferromagnetic Fe/W-tips were used for spin-resolved measurements.

The Cr-bulk tips were prepared via etching in 1M HCL solution, followed by cleaning

inside the STM by using field-emission on a W(110) surface. Subsequently, the tip was shaped using voltage pulses up to 6 V and gentle tip-surface collisions. This allows for tailoring of the tip-magnetization, with both IP and OOP configurations being possible.

The W-tips were electrochemically etched in a saturated solution of NaOH in distilled water. In UHV the tip was flash annealed before $\approx 20 - 50$ AL of Fe were deposited on the tip apex. Afterwards, the tip was annealed at a temperature of $200 - 300^\circ\text{C}$ for 10 minutes. This typically results in a ferromagnetic tip with an IP magnetization, that can be aligned OOP using magnetic fields of $\approx \pm 2$ T.

3.2 Measurement methodology

For dI/dU -maps measured at bias voltages > 1 meV, modulation amplitudes $U_{\text{mod}}(\text{RMS})$ of 10% of the applied DC bias voltage were used, while for the dI/dU -maps < 1 meV and spectroscopy measurements inside the superconducting gap of Ta a modulation amplitude of $U_{\text{mod}}(\text{RMS}) = 50 \mu\text{V}$ was used.

At temperatures of 1.3 K for superconductivity measurements, the thermal broadening due to temperature is approximately $360 \mu\text{V}$, while the addition of a $50 \mu\text{V}$ bias modulation leads to an additional broadening of $\approx 25 \mu\text{V}$, resulting in a total broadening of $385 \mu\text{V}$ according to equation (38). One has to find a balance between large modulation bias giving a better signal-to-noise ratio and keeping the additional broadening effects sufficiently small. The chosen modulation affects the overall energy resolution only slightly, while providing an acceptable signal-to-noise ratio in the spectroscopy measurements.

An additional factor regarding signal-to-noise ratio is the time-constant τ of the lock-in amplifier. The value of τ also determines an optimal scanning speed for maps and the optimal averaging duration for each bias point in spectroscopy measurements. For dI/dU -maps the scan speed is adjusted, so the signal is averaged over a duration $> \tau$ for every pixel of the image. This limits the number of pixels for zero-bias dI/dU -maps where a time-constant of 10 ms was used. This is especially true for constant-height maps where due to thermal drift and piezo creep the measurement duration is limited, as the feedback loop is disabled. However, multipass maps are also limited in the number of pixels, because the measurement duration can become very long, especially because they take twice the time compared to ordinary constant-current maps measured with the same number of pixels and with the same scan speed.

Here, it is important to mention the lock-in delay that exists for the lock-in amplifier. This needs to be accounted for when relating simultaneously recorded topography and current maps. For the lock-in parameters used (time-constant $\tau = 10\text{ms}$, sensitivity $200 \mu\text{V}$), a delay between topography and dI/dU -signal of $\approx 40\text{ms}$ was observed. This was found by comparing the shift along the fast scan axis between the fwd/bwd topography images with the shift between the fwd/bwd dI/dU -maps. The resulting shift in pixels was translated into a delay time by relating it to the scan speed.

For the spectroscopy measurements of the superconducting gap, a time-constant of 30 ms was used, while each voltage step was also averaging over three time-constants. Additionally, usually three spectra were taken back-to-back and averaged. This way if the tip was unstable during one of the three spectra, that one could be excluded.

For the STM images, extracted line-profiles, and 2D-FFTs the software Gwyddion [109] was used.

4 Anisotropic Motion and Growth on Mn/Re(0001)

STM, with its capabilities to manipulate individual atoms, allows for the true bottom-up fabrication of nanostructures. However, atom-by-atom assembly is heavily limited regarding the size and number of these structures. The self-assembly of atoms into nanostructures is another bottom-up approach to growth that should be more feasible to deploy for many larger scale applications. We therefore need to develop a deep understanding of the surface diffusion of atoms and clusters, including free diffusion on terraces, as well as diffusion along edges and around corners. Structural effects on the motion of atoms on surfaces have been previously investigated; this includes the effect of low-symmetry surfaces such as fcc(110) [110, 111] and uniaxial reconstruction [112, 113]. Highly anisotropic diffusion on such a uniaxial reconstruction can be used to grow self-assembled one-dimensional bi-atomic chains [114, 115]. The effect of edge diffusion for different edge types and its temperature dependence can be exploited to grow ultra-thin films with different island shapes [107, 116].

Diffusion on an unreconstructed hexagonal surface is expected to be quasi-isotropic (Fig. 15a), while on low-symmetry surfaces it can be anisotropic (Fig. 15b). However, a symmetry-breaking uniaxial magnetic state could also lead to anisotropic diffusion, see Fig. 15c. The role that magnetism could play in these diffusion processes has so far not been explored. Ultra-thin film growth is usually done at room temperature or at elevated temperature and therefore the adatoms may be too mobile to reveal any subtle magnetic effects or the magnetic material involved, especially a single atomic layer, is above its respective magnetic ordering temperature. In section 4.2 we will discuss the effect magnetism can have on the motion of adatoms in the system of a single fcc-stacked Mn monolayer on Re(0001), in-part based on our manuscript "Anisotropic atom motion on a row-wise antiferromagnetic surface" published in May 2025, see [Publications](#). Afterwards section 4.3 deals with the anisotropic growth of Co islands on the same system, based in-part on the preprint manuscript "Reciprocity of Magnetism and Nanostructure Growth" available on arXiv, see also [Publications](#).

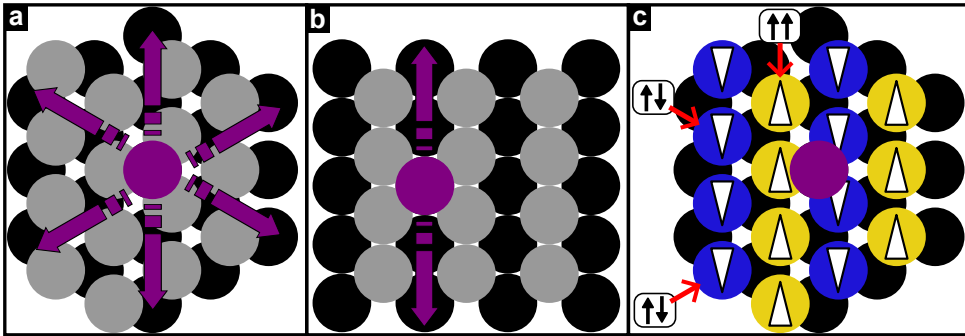


Fig. 15 **a**, Sketch of the quasi-isotropic diffusion of an adatom on a hexagonal surface. Purple arrows indicate the possible hopping directions. **b**, Sketch of the anisotropic diffusion of an adatom on a fcc(110) surface. **c**, Sketch of a magnetic adatom on a hexagonal surface hosting a uniaxial magnetic state. Red arrows indicate the different close-packed row directions named the $\uparrow\uparrow$ -rows and the $\uparrow\downarrow$ -rows. Figure adapted from our manuscript [117].

4.1 The preparation of ultra-thin films of Mn on Re(0001)

For more information about the general UHV and STM setup see Sec. 3. The Re crystal is first cleaned via cycles of annealing in an Oxygen atmosphere of $10^{-7} - 10^{-8}$ mbar at temperatures of up to 1400 K [118]. The crystal is then flashed to a temperature of ≈ 1800 K before Mn deposition. The Mn is evaporated from a Knudsen cell held at 690° , resulting in a deposition rate of ≈ 0.2 ML/min. The Re sample is still at elevated temperatures of $\approx 100 - 200^\circ\text{C}$ from the final flash. The Mn grows as large pseudomorphic fcc-stacked regions attached to the step edges of the Re single-crystal, see Fig. 16a,b.

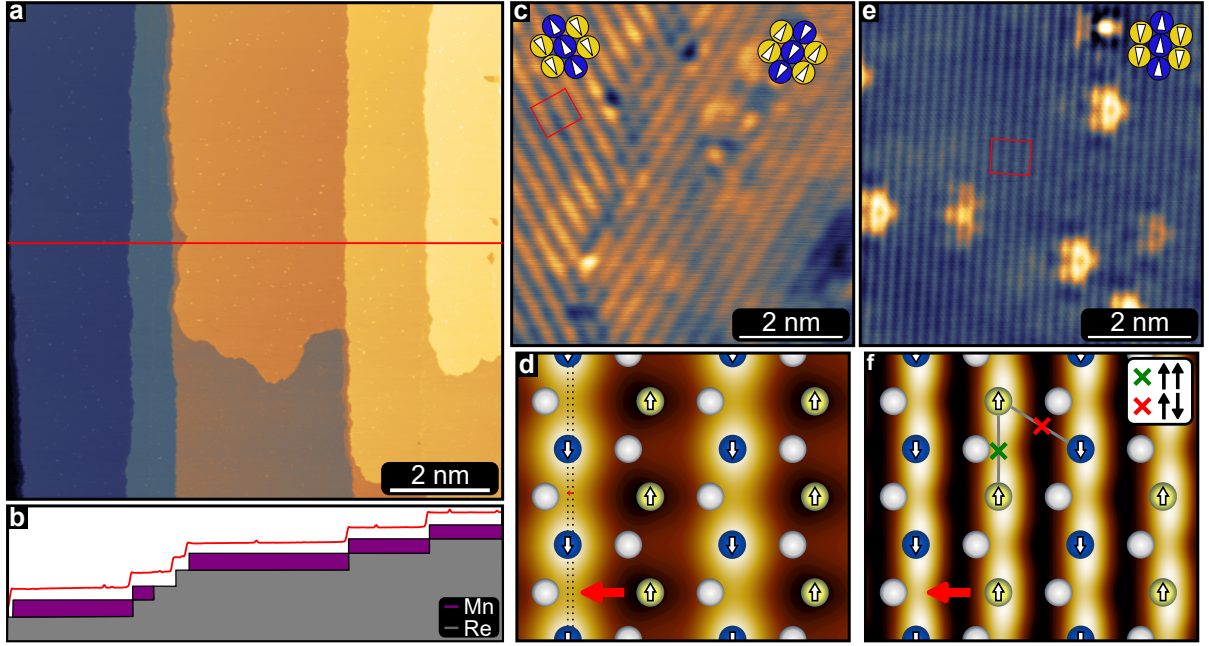


Fig. 16 **a**, STM image of ≈ 0.4 atomic layers (AL) of Mn on Re(0001). $U = 500$ mV, $I = 300$ pA. **b**, Sideview sketch of Mn layers (purple) on Re (grey) according to the line-profile, red line, extracted along the red line in **a**. **c**, SP-STM image of a Mn layer on Re. The blue-yellow sketches indicate the $\uparrow\uparrow$ -row direction. $U = 14.8$ mV, $I = 1$ nA. **d**, Calculated SP-STM image at $U = 50$ mV, $3\text{\AA}$ based on DFT, overlaid by the Mn atoms (blue/yellow) and the top-layer Re atoms (grey). **e**, STM image of a Mn layer on Re(0001) measured with a non-magnetic tip. $U = 30$ mV, $I = 1$ nA. **f**, Calculated (non-magnetic) STM image at $U = 50$ mV, $3\text{\AA}$ based on DFT. Green/red crosses mark the $\uparrow\uparrow$ - and the $\uparrow\downarrow$ -bridge site, respectively. **d,f**, Mn layer shift direction indicated by red arrows. All images measured at $T = 4.2$ K. DFT calculations were done by Soumyajyoti Haldar.

4.2 The anisotropic motion of adatoms on Mn/Re(0001)

STM can be used to study the motion of single atoms on surfaces [119]. In standard STM imaging, fast moving atoms are not visible and only practically stationary atoms can be imaged. Studying thermally activated diffusion is only possible if the diffusion processes occur on the time scale it takes to scan a line of the image or on the time scale of the imaging duration, which is on the order of seconds to minutes. The time scale of the

diffusion processes can be controlled by adjusting the measurement temperature using variable temperature STMs. Adatom motion can also be initiated using the STM tip and the tunneling electrons at a temperature where the atoms are immobile otherwise. However, the presence of the STM tip can affect the local potential landscape, so this process is not identical to the thermally activated diffusion of atoms. Nevertheless, information about the underlying potential landscape can be obtained.

4.2.1 Broken symmetry in Mn/Re(0001)

The fcc-stacked Mn monolayer hosts a row-wise antiferromagnetic state. This uniaxial magnetic state breaks the three-fold symmetry of the hexagonal surface and therefore exists as three symmetry equivalent rotational domains on the Mn surface, refer to Sec. 2.3.7 for a more detailed introduction. Using a spin-polarized STM tip, stripes with a period of twice the atomic row distance are visible, see Fig. 16c. The two different orientations of the stripe pattern correspond to two different rotational domains. The magnetic contrast on the left domain is stronger than on the right domain because the tip magnetization is aligned more with the AFM quantization axis of the left domain. At the intersection of two rotational domains, a domain wall hosting a superposition spin texture can be seen as a hexagonal pattern [57].

Interestingly, we can observe the orientation of the underlying magnetic domains also with a non-magnetic tip as stripes with a period of one atomic row distance, half the period of the magnetic contrast, see Fig. 16e. This is because the symmetry of the electronic structure is also broken, due to the RW-AFM state. The two possible bridge sites, the $\uparrow\uparrow$ and the $\uparrow\downarrow$ -bridge sites, in the system are no longer equivalent, see green and red crosses in Fig. 16f.

Calculated STM images based on the DFT results can reproduce the magnetic and electronic contrast, see Fig. 16d-f. Important to note here is that the DFT calculations find that the Mn layer does not just relax its interlayer distance but also relaxes laterally by moving the atoms closer towards the bridge site positions by 15 pm. This magnetism-induced shift is included in the simulations in Fig. 16d,f and the shift direction is marked by the red arrows. For this system, the shift has not been observed experimentally as the effect is quite small. However, for the system of the Mn bilayer on Ir(111), which hosts the same RW-AFM state, an even larger magnetism induced shift has been predicted by DFT and also found in the experimental data [58]. Additionally, in the system of Mn/Re(0001) the symmetry of the LDOS around subsurface defects is broken and their asymmetric shape can be used to identify the orientation of the magnetic state in scenarios where no magnetic or electronic stripe contrast is visible, see also defects in Fig. 16e.

4.2.2 Tip-assisted diffusion

We start by depositing single Co atoms at a temperature of $\approx 20 - 30$ K on the Mn monolayer. At the measurement temperature of 4.2 K the adatoms are immobile on the surface, however, they are easily movable using an STM tip during standard STM imaging using typical imaging parameters (1 nA, 10 mV). In Fig. 17a, a single Co atom is moving with the STM tip during scanning. As the tip is moving vertically upwards

(slow scan direction), a Co atom jumps strictly upwards as well, and is imaged multiple times, appearing as a ridge in the image similar to a chain of atoms. In the middle of the vertical ridge a kink is visible; this is the only time the adatom moves one atomic row to the right. This measurement appears to be similar to the "tip-assisted diffusion" experiments performed by Li *et al.* using Ag atoms on Ag(110)[111, 120]. They found that Ag atoms move preferentially along the $[1\bar{1}0]$ direction in the presence of an STM tip due to the structure of this low-symmetry fcc-surface. In our case, this dragging of the Co atom on the RW-AFM state may therefore be an indication that the magnetic state dictates the direction of motion for Co atoms.

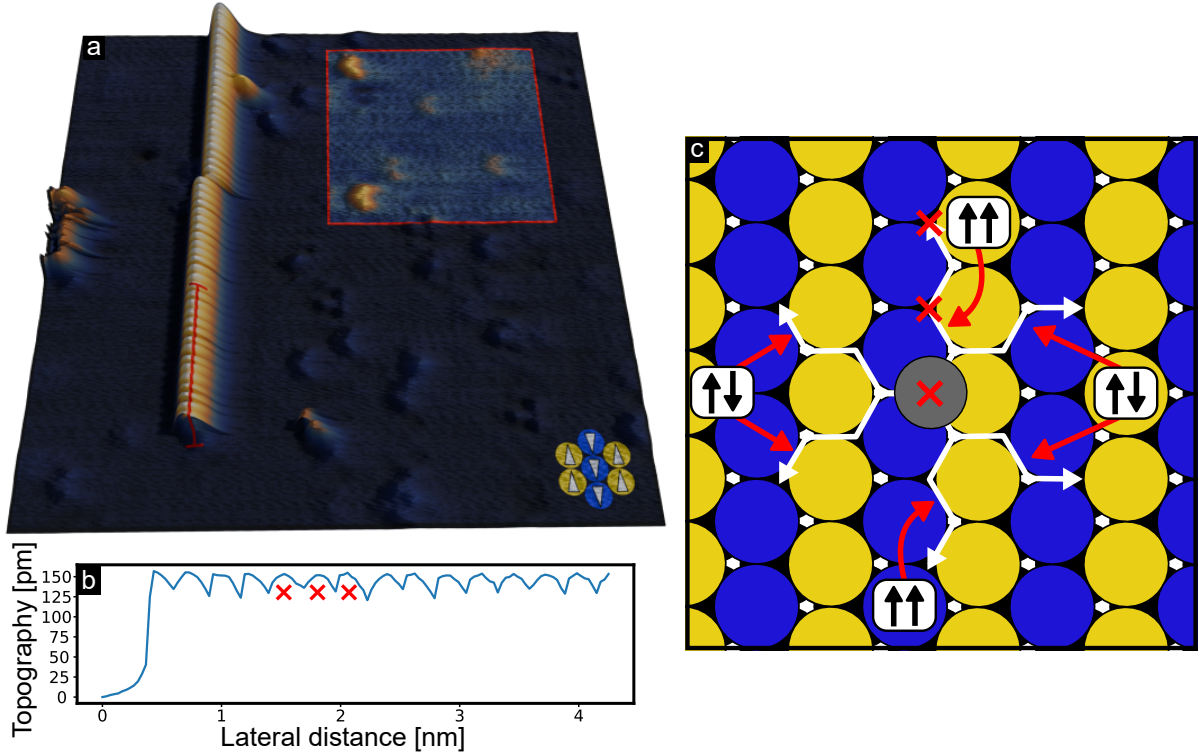


Fig. 17 **a**, 3D view of an STM image of Co atoms on Mn/Re(0001). The bright vertical ridge is a single Co atom imaged multiple times as it moves vertically during scanning. The colour-map in the red square was adjusted to allow for better contrast of the subsurface defects. The slow scan direction is upwards. $U = 7$ mV, $I = 1$ nA, $T = 4.2$ K. **b**, Line-profile along the red line in **a**. The red crosses indicate the stable hollow-site position of the Co atom. **c**, Sketch of possible diffusion paths along either the $\uparrow\downarrow$ -rows or the $\uparrow\uparrow$ -rows. The red crosses indicate the hcp site positions along a $\uparrow\uparrow$ -row and correspond to **b**.

The line-profile in Fig. 17b was taken in the vertical direction along the top of the ridge. We can see that the Co atom is imaged repeatedly, spaced a distance of one Re lattice constant apart. This means, that we find the Co atom only in hollow-sites with one particular stacking; the residence time of the Co atom in the other stacking is too short to be measured using STM. The DFT calculations, that will be discussed at a later point, indicate that the hcp-site is energetically more favourable than the fcc-site. From the line-profile, we also see that the tip has moved past the atom vertically before the

atom jumps. Interestingly, the atom jumps from behind the tip to in front of the tip, which is reminiscent of the pulling mode in atom manipulation imaging [19].

The vertical direction in Fig. 17a corresponds to the close-packed row direction where the spins of the Mn atoms all point in the same direction. We will refer to this direction as the $\uparrow\uparrow$ -row direction. Adatoms moving along a close-packed row direction can either move along the $\uparrow\uparrow$ -rows or the $\uparrow\downarrow$ -rows, see also Fig. 17c. Our measurements indicate that the Co atom preferentially moves along the $\uparrow\uparrow$ -rows. However, an anisotropy in the movement direction of Co adatoms could in principle also originate from an asymmetric tip apex.

4.2.3 Atom kicking of Co

To clearly attribute the observed anisotropy to the underlying magnetic state, one would have to observe anisotropic movement of the adatoms in two different directions on two different magnetic domains using the same micro-tip. To reduce the involvement of the STM tip in the motion of the atoms and improve reproducibility, we switch to using a stationary tip and inducing motion using voltage pulses; we will call this process "atom kicking". This sudden increase in voltage at the same tip-sample distance leads to an increased tunnel current flowing through the atom until it jumps away from the tip and the tunnel current drops sharply, see inset in Fig. 18a. Before and after the pulses, the atom position is measured with a standard STM image, where one has to adjust the tunneling parameters until no atom moves during imaging. For repeatability, the tip is moved to the centre of the atom via atom-tracking feedback using a gradient ascent method [121]. The tip moves in a circle with a radius of 10 pm at a frequency of 5 Hz, and the slopes in the different directions are calculated using the topography signal. The tip is then moved to the local extremum (i.e. to the centre of the atom). To ensure reproducibility and comparability, the tip is stabilized at the same tunnel parameters resulting in the same tip-sample distance for each of these kicking processes.

Fig. 18a shows a Mn monolayer with two different rotational domains and a domain wall between them. Note that no magnetic contrast is visible, indicating that either the tip magnetization direction is nearly perpendicular to the quantization axis in both domains, which is only possible for a mostly OOP tip magnetization, or the tip is non-magnetic, due to some non-magnetic material at the tip apex. The orientation of the magnetic state in the rotational domains can be identified by the asymmetric LDOS of the defects, and is marked by the blue-yellow sketches. The Co atoms were kicked (i.e. their motion was initiated) by voltage pulses with a magnitude of +200 mV. Here, Fig. 18b,d show the atom positions after the pulses at the locations marked by red crosses in Fig. 18a, while the atom positions in Fig. 18c,e were measured after the pulses at the positions of the red cross in Fig. 18b,d. We find that on the left domain (red square) the atoms move diagonally while on the right domain (green square) they move vertically, see respectively framed images in Fig. 18b-e.

We find the same movement direction even for adatoms not directly under the tip but at a distance of up to 5 nm away, although they move only a lattice site or two, compare the current atom positions with their previous ones marked by black circles. In all cases the Co atoms move strictly one dimensionally along the $\uparrow\uparrow$ -rows, with an accuracy of one

lattice site. We can exclude any tip asymmetry effects here, as the atoms move in different directions with the same micro-tip. This clearly demonstrates that the anisotropic motion of Co is directly related to the underlying magnetic state.

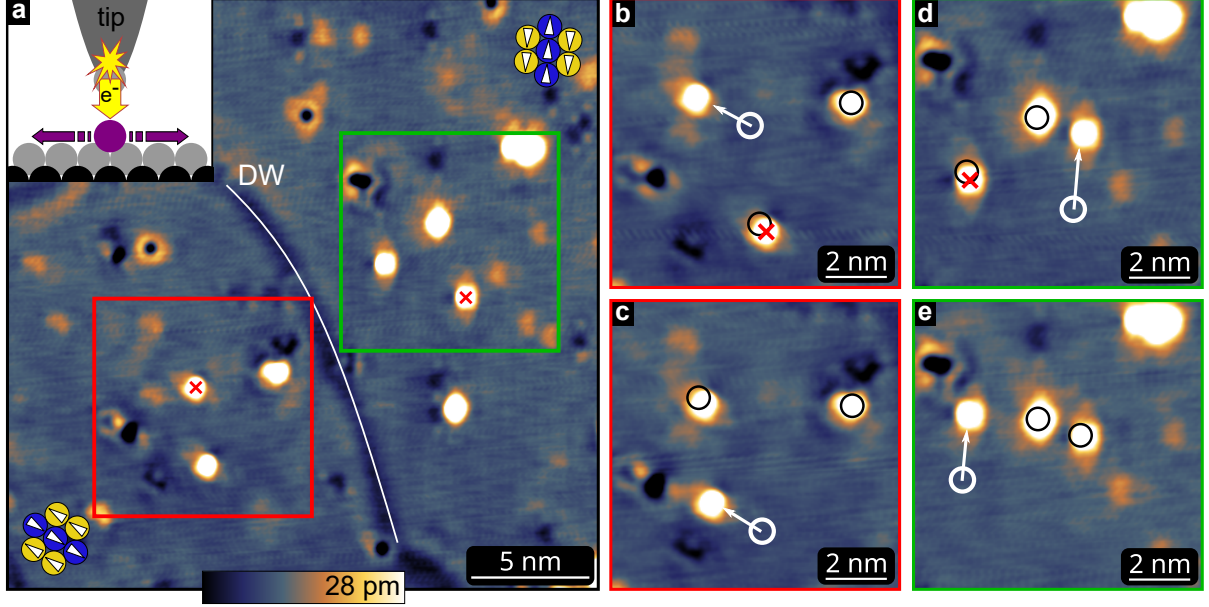


Fig. 18 **a**, Overview STM image of two rotational domains on Mn/Re(0001). The domain wall is visible due to a reduced LDOS and marked by the white line. The domain orientation can be obtained from the defect shape and is marked by the blue-yellow sketch. Inset shows a sketch of the kicking process. **b**, STM image in the area marked by the red square in **a** after a voltage pulse at the location of the red cross in **a**. The upper left Co atom moved diagonally along the $\uparrow\uparrow$ -rows. The white/black circles mark the positions of the Co atoms before the voltage pulse directly under the tip and further away from the tip, respectively. **c**, The bottom Co atom moved in the same direction after a voltage pulse marked by the red cross in **b**. **d**, STM image in the area marked by the green square in **a** after a voltage pulse at the location of the red cross in **a**. The bottom right Co atom moved vertically along the $\uparrow\uparrow$ -rows. **e**, The bottom left Co atom moved along the same direction after a voltage pulse marked by the red cross in **d**. All images: $U = +8$ mV, $I = 1$ nA, $T = 4.2$ K. All voltage pulses: $U_p = +200$ mV, $\Delta t = 1$ s.

In the data shown in Fig. 18, both atoms on the left domain move in the same direction, diagonally to the top-left, while both atoms on the right domain move vertically upwards. Along the $\uparrow\uparrow$ -row direction the atoms can move in two symmetry equivalent directions, see Fig. 17c, and the adatoms should therefore be equally likely to move in either direction. We found that specific micro-tips break this symmetry, likely due to an asymmetry in the tip apex, as was for instance the case for the tip used in Fig. 18. Other micro-tips show symmetric movement distributions along the $\uparrow\uparrow$ -rows.

Additionally, the direction of the induced motion can also be controlled via precise placement of the STM tip before the voltage pulse. When we first position the tip at the centre of the adatom and then move the tip 1-2 lattice sites away from the centre along the direction of the $\uparrow\uparrow$ -rows before applying the voltage pulse, we observe that the Co atom moves exclusively in the direction away from the tip. Although we cannot exclude the

possibility that this effect is also related to the specific micro-tip and other tips may not reproduce this behaviour. In addition, however, the average movement distance appears to be reduced slightly, as less current is flowing directly through the atom. Interestingly, when moving away from the centre of the atom in the direction perpendicular to the $\uparrow\uparrow$ -rows before kicking, we still observe the Co atoms moving along the $\uparrow\uparrow$ -rows.

For voltage pulses of +200 mV magnitude, the typical kicking distance for the data in Fig. 18 is ≈ 10 lattice sites. In general, we find that single site hopping of the Co atoms can be induced starting at a bias voltage of 8 – 50 mV, depending on the micro-tip. This strong variance in the threshold voltage for inducing motion of adatoms indicates that the tip shape/symmetry has an influence on this process and only data captured using the same micro-tip can be compared quantitatively. This effect does appear to become less relevant as the bias voltage of the pulses during atom kicking increases. At +200 mV the average kicking distances for different micro-tips were similar at ≈ 2 -3 nm. In Fig. 19c data of 19 comparable kicking events is shown. The same micro-tip was used for inducing motion using +200 mV and –200 mV of the two atoms shown in Fig. 19a.

After every kicking event, the atoms were imaged to record their new positions and subsequently manipulated back to their original position using atom manipulation with typical tunneling parameters of 4-5 mV and 60 nA. We estimate an accuracy of the atom position to be ± 1 lattice site. Kicking the Co atoms from identical starting positions, with the same distribution of the surrounding defects, improves the comparability between kicking events and decreases the variability in the kicking distance. This is because the defects can affect the motion of the atoms, as they act as pinning centres. Generally a Co atom on top of such a defect is more difficult to move, with the strength of the pinning depending on the defect type. While the atoms can usually still be moved using atom manipulation, atom kicking does often not result in any jumping of the adatoms. The need for atom manipulation after every event limits the number of possible events performed with the same micro-tip as tip changes have a certain probability to occur.

We observe a broad distribution of travel distances after atom kicking at +200 mV and –200 mV with an average distance of ≈ 2 -3 nm or ≈ 10 lattice sites and a maximum distance of up to 8 nm or 26-28 lattice sites, see Fig. 19c. This is likely limited by the amount of defects on the surfaces. In Fig. 19a the red arrows indicate the movement direction of the Co atoms. The atoms were positioned in such a way as to allow the largest movement distance along these directions without hitting defects. However, the atoms will inevitably encounter defects for large-enough kicking distances, which limits the maximum travel distance. For a defect-free surface, an overall shift of the distribution towards larger kicking distances is expected. Increasing the bias voltage above 200 mV did not appear to increase the average movement distance significantly. The maximum observed movement distance was ≈ 10 nm for +500 mV.

Comparing the travel distances for both polarities in Fig. 19c the distribution is quite similar, with both polarities resulting in atoms being moved across many lattice sites. However, in both cases the atoms move away from the tip. If the motion was driven by an inhomogeneous electric field between tip and sample, one would expect two electric fields pointing in opposite directions to result in a force on an electric dipole towards lower electric field (repulsion) in one case and towards higher electric fields (attraction) in the other, see Fig. 19b, as has been observed for Au and Ag atoms on Ni(111) [122]. The direc-

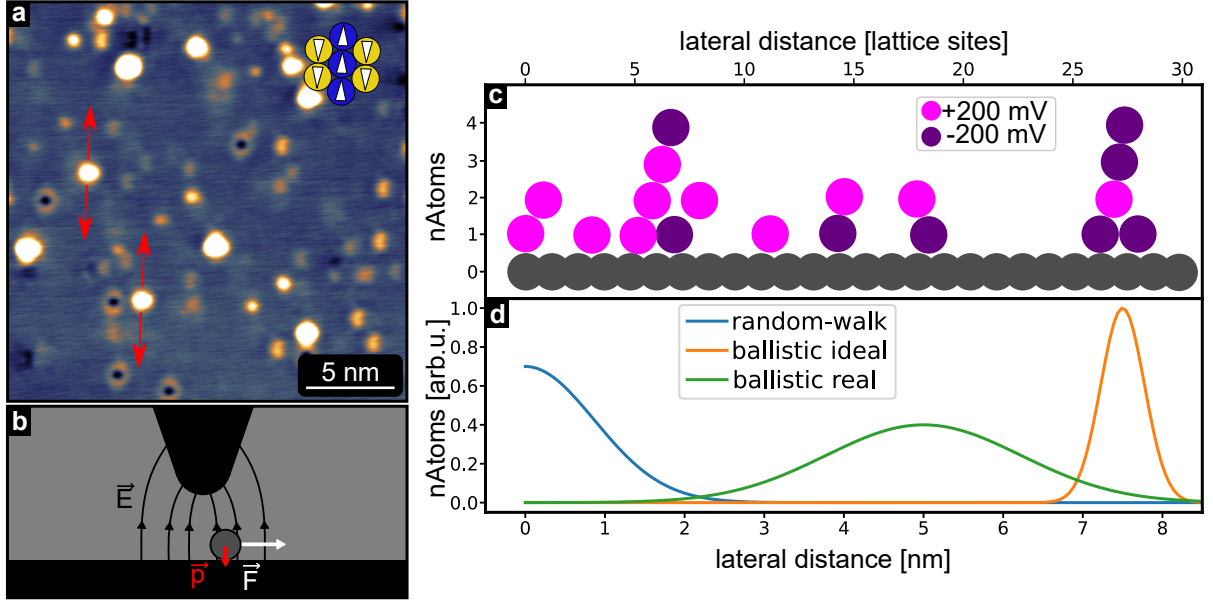


Fig. 19 **a**, STM overview image of single Co atoms on Mn/Re(0001). The red arrows represent the movement direction of two Co atoms, for which motion was repeatedly initiated using a voltage pulse. $U = +10$ mV, $I = 300$ pA. **b**, Sketch of an inhomogeneous electric field between tip and sample for $U > 0$ and the resulting force (white arrow) on an electric dipole (red arrow) pointing down. **c**, Travel distances for the two atoms in **a** initiated with voltage pulses with +200 mV and -200 mV. Compared to our paper [117] the distribution has been corrected, as one of the distances was actually 1.8 nm, not 0.5 nm. **d**, Expected travel distance distribution for a random-walk, ideal-ballistic motion and real-ballistic motion.

tion of the electric field depends on the bias voltage as well as the work function of tip and sample. We can roughly estimate the work function difference between tip and sample using the work function of Chromium ($\phi_{\text{Cr}} = 4.5$ eV) and Rhenium ($\phi_{\text{Rh}} = 4.72$ eV) to be ≈ 200 meV. So, while we cannot be sure that the direction of the electric field inverts between +200 mV and -200 mV, we repeated the tip-assisted diffusion experiments using +500 mV and -500 mV as well and observed similarly repulsive behaviour for both polarities indicating that the electric field plays no major role in this process.

The average travel distance does appear slightly larger for -200 mV. This effect could simply be the result of too few data points. Assuming this effect is real, however, one could imagine the electric field contributing only in a minor way to the kicking process, slightly reducing the average travel distance for positive polarity while slightly boosting it for the negative polarities. Calculations of the Co electric dipole moment by Soumyajyoti Halder, indicate an electron transfer from the Mn layer to the Co atom, resulting in an electric dipole pointing down. This should result in an attractive/repulsive force for an electric field pointing down($U < 0$)/up($U > 0$) respectively, see Fig. 19b, exactly the opposite effect of what we observe. Thus, even though the value of the calculated dipole moment is sizeable, at $p = 2.9$ D, we can conclude that the electric field does not play a role in the kicking process.

Calculations show a difference in the DOS above and below the Fermi level [56], which

may be responsible for the difference in the average travel distance between +200 mV and -200 mV. While the tip is stabilized at the same tip-sample distance in both cases, the voltage pulse at -200 mV would result in a higher current flowing through the Co atom, leading to a larger average travel distance.

The distribution of travel distances also reveals information about the possible mechanism behind the tip-induced motion process. For a random-walk-like process one would expect a distribution centred around zero (i.e. under the tip) and therefore a maximum of travel distance around zero and an exponential tail towards higher travel distances, see the blue curve Fig. 19d. Such a mechanism could be caused by Joule heating where the tunneling electrons locally increase the sample temperature. Anisotropic diffusion barriers could result in hopping occurring only across $\uparrow\downarrow$ -bridge sites and not across $\uparrow\uparrow$ -bridge sites. The uncorrelated hopping processes in both directions along the $\uparrow\uparrow$ -rows would be equally likely, which would mean that on average no net-movement occurs and the result would be a Gaussian distribution around zero travel distance. Such a random-walk type process can therefore be excluded as it does not fit the experimental data.

The clustering at higher travel distances indicates a ballistic process with low energy barriers along the $\uparrow\uparrow$ -rows and low damping, see orange curve in Fig. 19d. For molecules, lateral motion on surfaces induced by STM has been investigated more extensively than for single atoms. The excitation of vibrational modes in molecules using inelastic tunneling electrons was identified as an important mechanism on Pd(110) [113, 123]. This raises the possibility for the excitation of vibrational modes in the tip - Co atom - sample junction playing a role in the ballistic motion we observe. For such a ballistic process, the presence of many surface- and sub-surface-defects, as well as any small variations in atom position could lead to general broadening of the travel distance distribution and also a reduction of the average travel distance, see the green curve in Fig. 19d.

4.2.4 Atom kicking of Rh and Ir

We have seen that the magnetic Co atoms exhibit a strong anisotropy in their motion when initiated by local voltage pulses at 4.2 K. This raises the question, if a sizeable magnetic moment is required or if non-magnetic atoms show the same anisotropy. As we have seen in Fig. 16e, the $\uparrow\downarrow$ -bridge and $\uparrow\uparrow$ -bridge are also electronically different due to the underlying magnetic state.

We deposit Rh atoms onto the RW-AFM state in the same way as we did for Co. Rh is an element in the same group as Co in the periodic table and therefore electronically similar, but it is typically non-magnetic on metallic surfaces. We once again perform kicking experiments in a region with two rotational domains of the magnetic state present, see Fig. 20a. We observe no magnetic contrast on either domain and their orientation is once again derived from the asymmetry in the defect LDOS.

The Rh atoms move in the vertical direction on the left domain and diagonally on the bottom left domain, as can be seen in Fig. 20b-e. In both cases, the atoms move exclusively along the $\uparrow\uparrow$ -rows in the same way as Co. However, the observed travel distances are much smaller at $\approx 1 - 2$ nm even when initiating the motion with bias voltages of +1 V, indicating that the Rh atoms are much less mobile. Even for +2 V bias pulses a maximum travel distance of 3 nm was observed. The threshold voltage needed for

initiating single site hopping for Rh atoms was generally also much higher at 200–250 mV. Similar to Co atoms and some of the native defects, Rh atoms show an asymmetry in the LDOS. However, compared to Co this asymmetry is more apparent and located closer to the atom.

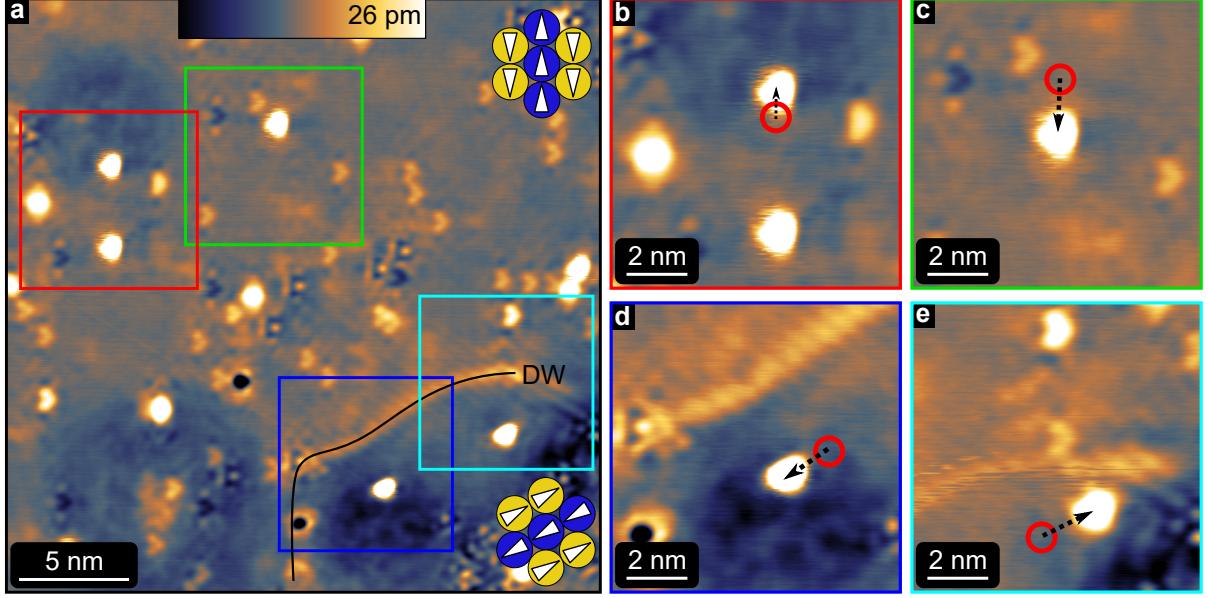


Fig. 20 **a**, STM overview image of single Rh atoms on Mn/Re(0001). Two rotational domains are visible and marked by the blue-yellow sketch. The domain wall is marked by a black line. Native defects and Rh atoms both exhibit an asymmetry in the LDOS. **b-e**, The Rh atoms move along the $\uparrow\uparrow$ -rows when motion is initiated with $U_p = +1$ V, $\Delta t = 0.4$ s voltage pulses. All images: $U = +25$ mV, $I = 2$ nA, $T = 4.2$ K.

We have found that the atom mobility decreases when changing the adatom species from Co to Rh. To see if this trend continues, we investigate single Ir atoms. We observe that even when pulsing with +2 V, Ir atoms were stationary or moved a single lattice site at most, clearly indicating that Ir adatoms are even less mobile than Rh adatoms. However, in some cases we observe the Ir atom moving with the STM tip during scanning at high bias, similar to the tip-assisted diffusion of Co, compare Fig. 17 and Fig. 21. The Ir atom moves exclusively along the $\uparrow\uparrow$ -rows during this dragging process, indicating that the same anisotropic movement direction as for Co and Rh can be expected.

As we have seen, the trend of decreasing mobility going from Co to Rh to Ir can be confirmed. Several different mechanisms that could contribute to this decrease come to mind: This effect could be due the increasing size of the d -orbitals going from Co to Ir which may lead to increasing adatom-surface interaction and therefore increased damping during the movement process. The increasingly higher atom mass could also have an effect on a ballistic transport process. For instance, the faster moving Co atom could move larger distances on the surfaces in case of damping processes scaling with time.

The DFT calculations show that the binding energy of Co is 1.5 eV lower than for Rh, because during kicking some of the energy of the tunneling electrons is needed for the excitation of the atom, the excess energy would be smaller in the case of Rh. This

is supported by the lower threshold voltage for Co atom motion compared to Rh atom motion. Lastly, one could imagine the spin-degree of freedom of the Co atom playing a role, similar to how molecules are able to store energy in their vibrations. Spin excitations of the Co atom or possibly also in the Mn layer might contribute to the excitation process and the travel distance.

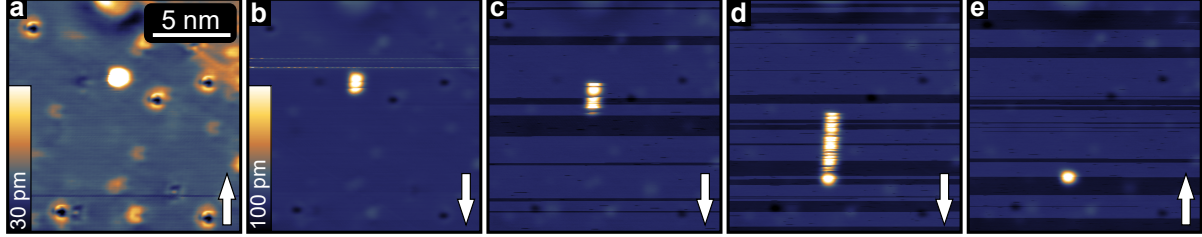


Fig. 21 **a**, Single Ir atom on Mn/Re(0001). The $\uparrow\uparrow$ -rows run vertically. The white arrow indicates the slow scan direction. $U = +100$ mV. **b-d**, The Ir atom is moving downwards with increasing distances. **b**: $U = +1$ V, **c**: $U = +2$ V, **d**: $U = +2.5$ V. **e**, The Iridium is pinned at a native defect. $U = +2.5$ V. All images: $I = 50$ nA, $T = 4.2$ K.

4.2.5 DFT calculations

Soumyajyoti Haldar (Univ. of Kiel) has done minimum energy path calculations for Co, Rh and Cu adatoms on Mn/Re(0001) using the climbing image nudged-elastic band method [124, 125]. Fig. 22a shows the atom positions of the calculated paths from the initial hcp hollow-site (I) along the $\uparrow\uparrow$ -rows (blue path) and along the $\uparrow\downarrow$ -rows (red path) to the next/final hcp site (F). The hcp-site is the preferred adsorption site with a lower energy compared to the fcc-site. Both paths cross over a bridge-site, an fcc-site then a second bridge site. Atoms on the blue path along the $\uparrow\uparrow$ -rows cross two identical $\uparrow\downarrow$ -bridge sites, while atoms on the red path first cross a $\uparrow\uparrow$ -bridge site followed by a $\uparrow\downarrow$ -bridge site.

For the Co atom in the hollow-site positions, where it aligns its intrinsic magnetic moment with the majority of nearest-neighbour Mn atoms, the calculations show a magnetic moment of $1.6 \mu_B$. This spin alignment means that along the blue path the Co spin in the final hollow-site (F1) is the same as for the initial hollow-site (I1). Along the red path, the Co magnetic moment has to flip along the path as the final hollow-site (F2) prefers alignment opposite to the initial hollow-site (I2). The minimum energy paths for the Co atom show a higher overall energy barrier for the red path compared to the blue path, in agreement with the experimental findings, see Fig. 22b. On the blue path, the Co atom avoids the perfect fcc-hollow-site position and its magnetic moment points in the same direction the entire time. Along the red path, the Co atom moves over the perfect fcc-hollow-site as its magnetic moment is aligned correctly.

Interestingly, the Co atom prefers the $\uparrow\uparrow$ -bridge site over the $\uparrow\downarrow$ -bridge site, because the height of the energy barrier is reduced, compare the first half of the red and blue path. However, across the second bridge site the Co atom is required to flip its magnetic moment, resulting in a much higher energy barrier. The magnetic moment is quenched on this unfavourable bridge site due to a collinear constraint in the calculations. An unconstrained magnetic moment could possibly result in a reduced barrier height but is

still expected to lead to a higher barrier than for the blue path overall. Additionally, calculations from the intermediate fcc-hollow-site (I3) to the final hollow-site (F3) were performed for the Co atom while no spin flip was allowed, see cyan path in Fig. 22a,b. The cyan path starts from the identical position on the red path at the fcc-hollow-site allowing for a combination of the first half of the red path with the cyan path. Prohibiting the spin-flip of the Co reduces the energy barrier at the second bridge site and the Co atom ends in the metastable final state (F3), with a possible relaxation to the perfect hollow-site (F2). The blue path along the $\uparrow\uparrow$ -rows remains the lowest energy path.

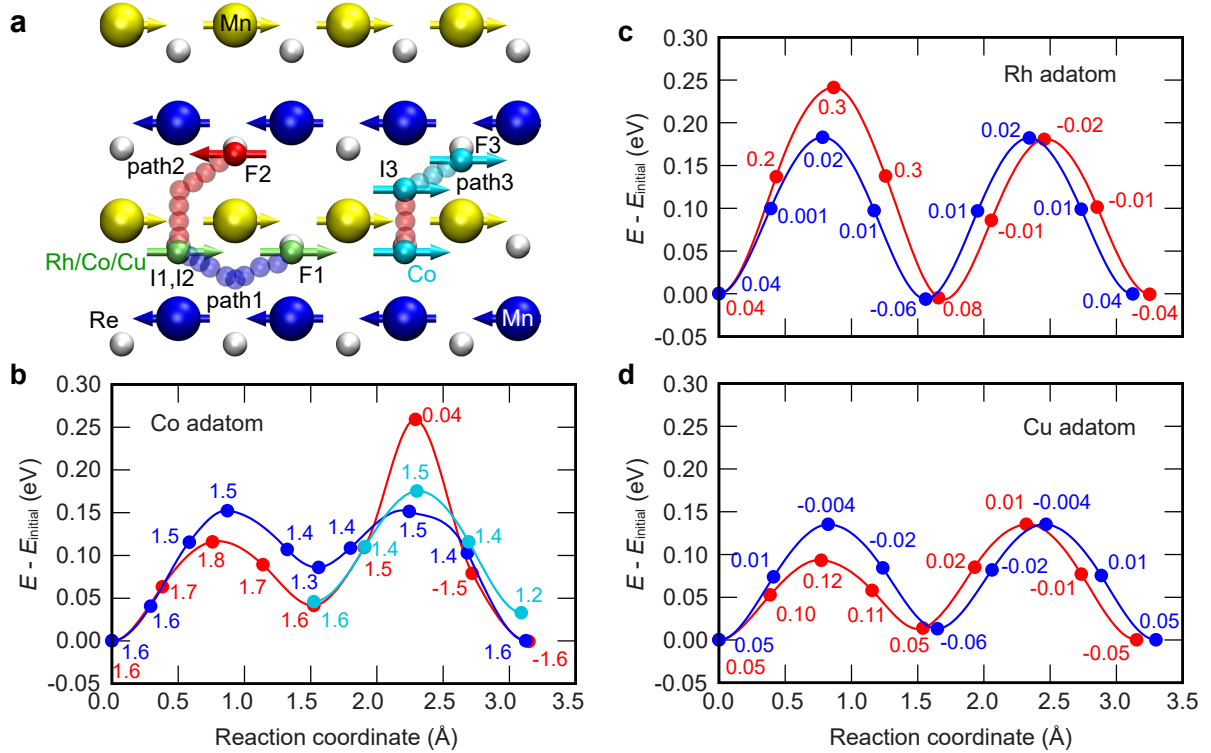


Fig. 22 a, DFT minimum energy paths for the anisotropic movement of Co, Rh and Cu atoms on the RW-AFM state of Mn/Re(0001), obtained via nudged-elastic band calculations. The yellow and blue spheres represent Mn atoms, while the grey spheres correspond to the top-most Re layer. The large coloured spheres represent initial/final adatom positions, while the small transparent coloured spheres indicate intermediate positions on the minimum energy paths. **b-d**, Energy along the minimum energy paths 1 and 2 for Co, Rh and Cu from the initial state I1 and I2 to the final states F1 and F2, for the blue and red path as indicated in **a**, respectively. For the Co atom the energy is also given for the minimum energy path 3 from I3 to F3 (cyan symbols and line in **a**). The numbers along the path indicate the magnetic moment of the adatoms in μ_B . The DFT calculation and Figure were done by Soumyajyoti Halder [117].

For the Rh atom, the DFT calculations are in agreement with the experiments as well, see Fig. 22c. The red path along the $\uparrow\downarrow$ -rows shows an increased energy barrier compared to the blue path along the $\uparrow\uparrow$ -rows. In the hollow-site positions, the induced magnetic moment is quite small, and on the order of 0.04–0.08 μ_B . The Rh atom exhibits an induced magnetic moment 0.3 μ_B at the unfavourable $\uparrow\uparrow$ -bridge site on the red path, which is much

larger than the induced moment at the more favourable $\uparrow\downarrow$ -bridge sites, indicating that the polarization of the Rh atom is leading to an increased energy barrier. This runs contrary to the behaviour of the Co atoms, where a reduction of the magnetic moment results in an increase in energy. We attribute this effect to the different spin-dependent hybridization of the Co and Rh atoms with the Mn surface layer. Nevertheless, the DFT calculations predict the same preferential movement direction of the surface for Co and Rh, in agreement with the experiments. Interestingly, for the Rh atom the magnetism induced 15 pm shift is vital, as the DFT calculations for the unshifted layer show minimum energy paths that are almost isotropic.

For the Cu atom we find the opposite behaviour. Here, the polarization of the Cu atom on the $\uparrow\uparrow$ -bridge site results in a lower energy barrier on the red path compared to the blue path. The induced magnetic moments are of a similar size compared to Rh; however, the polarization of the Cu atom on the $\uparrow\uparrow$ -bridge site is energetically favourable. This opposite effect can be attributed once again to the different hybridization environment because of the filled 3*d*-bands. The motion of the Cu atoms is predicted to still be anisotropic but, unlike Co and Rh, no longer just one-dimensional. In kicking experiments, this behaviour may be visible as an avoidance of the $\uparrow\uparrow$ -row direction. For a more in-depth discussion of the DFT results, see [117].

4.2.6 Atom tracking

The DFT calculations calculated the minimum energy path for the Co and Rh atoms and, while they show agreement with experimental data, the actual kicking process may in-fact be quite different to a typical diffusion process. The potential landscape an excited adatom sees during a ballistic trajectory is likely different compared to the potential landscape calculated in the DFT data. The observed motion of Co atoms further away from the tip during kicking, see black circles in Fig. 18b-e, is perhaps more closely comparable to adatom surface diffusion. There, the Co atoms occasionally move one or two lattice sites, and appear to hop almost exclusively along the $\uparrow\uparrow$ -rows. As a further experiment, we induce the motion of the adatoms slightly above the voltage threshold for single site hopping, and by letting the STM tip continuously follow the hopping adatoms, using the atom tracking feature as described earlier, we can record the x, y and z coordinates of the STM tip over time.

We perform these atom tracking measurements on the same sample area as for the Co kicking in Fig. 18a, see Fig. 23a. The lateral position of the tip following a hopping Co atom over a time frame of 251 s is shown in Fig. 23b. The hopping was induced with tunneling parameters of $U = +110$ mV, $I = 500$ pA. The motion of the tip and therefore the Co atom is highly anisotropic, moving a distance of ≈ 7.3 nm along the $\uparrow\uparrow$ -rows until reversing direction at the defect left of the domain wall, see Fig. 23a,b. For the last part of the x - y tip coordinates, the z -positions were also measured, see Fig. 23c. After every hop of the Co atom, which corresponds to a sharp drop in the z -position of the tip, it takes from 1 – 20 s before another hop occurs. This is because after the Co hops further away from the tip some time is needed for the tip to track closer towards the centre/top of the atom again, see corresponding slow increase in the z -position of the tip over time. As the tip gets closer to the centre of the Co atom, the fraction of the tunnel current

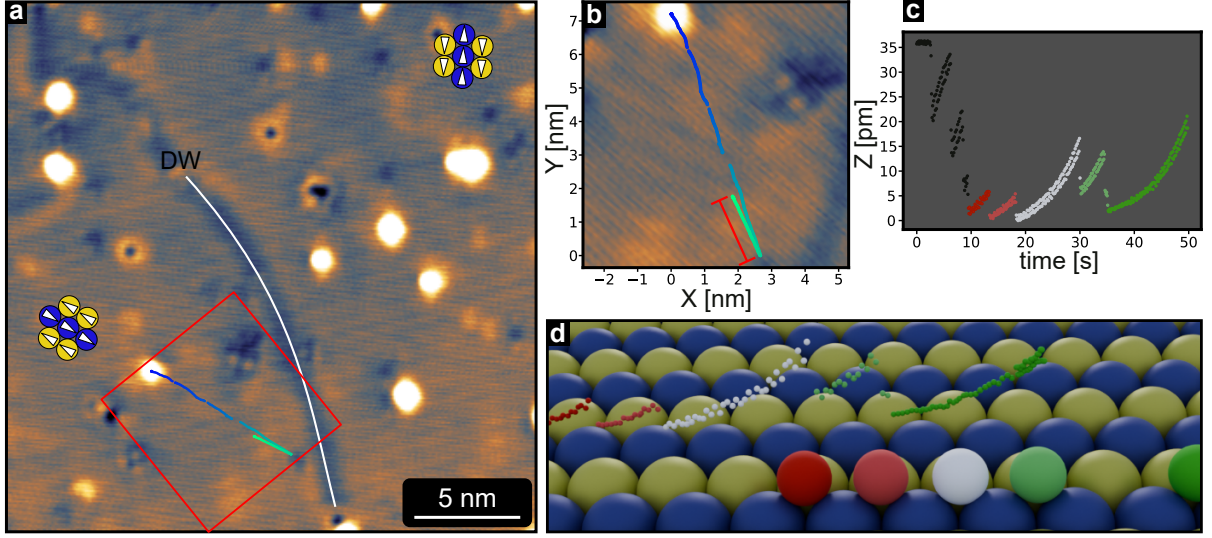


Fig. 23 **a**, STM overview image of two rotational domains on Mn/Re(0001). The domain wall is marked by the white line and the domain orientations are marked by the blue-yellow sketches. $U = +8$ mV, $I = 500$ pA, $T = 4.2$ K. The image was blurred with a 2-pixel Gaussian to remove high frequency noise. **b**, x and y -coordinate of the tip plotted vs. time. Time is increasing from blue to turquoise. The tip position can be correlated to the previous STM image, see red square and path in **a**. The Co motion is initiated by the tip using parameters $U = +110$ mV, $I = 500$ pA. **c**, Measured z -coordinate of the STM tip over time. The z -coordinates correspond to the x - y -coordinates in **b** after the inversion of the direction, see red line. **d**, 3D render of the time-correlated x - y and z -coordinates, shown as the small coloured spheres. The colour of the spheres can be correlated to the Co atom in a certain hollow-site position, see colour of corresponding larger spheres and in **c**. The blue and yellow atoms represent the RW-AFM state.

flowing through the atom increases until another hop is initiated.

In most cases, the Co atom jumps when the tip is still moving up the side of the atom, akin to the pushing mode in atom manipulation experiments [19]. The recording of the z -position of the tip was started right at the point of direction inversion, when the STM was able to track to the centre of the Co atom without initiating a hop, due to the increased pinning near the defect, see flat z -position line near zero in Fig. 23c. The Co atom is not performing a true random walk under the tip, as once again the presence of the tip breaks the equivalence of the two directions along the $\uparrow\uparrow$ -rows. Once the atom starts moving in one direction a change in direction is less probable.

Combining the x - y data with the z -data, allows for plotting the position of the STM tip in 3D, see Fig. 23d. By taking into account the tip height before and after the Co atom hop as well as the general extent of the Co atom on the surface one can assign a likely hcp-hollow-site position for each section of the z -position trace where the tip was moving up the side of the atom, see the colouring of the small and large spheres in Fig. 23c,d. The z -coordinates were scaled up by a factor of 20 relative to x - y to improve the visualization. The trace indicates that in one of the hops the Co atom jumped a distance of two lattice constants instead of the typical single site hop.

In this section, we have shown that adatoms such as Co, Rh and Ir move anisotropically at 4.2 K when their motion is initiated by the STM tip either via electrons tunneling at a particular bias or via additional voltage pulses. The atoms move in an anisotropic potential landscape due to the symmetry-breaking RW-AFM state. For the Rh atoms the DFT calculations suggest, that the magnetism induced lateral shift of the Mn layer plays a major role. For the Co atoms we speculate, that this anisotropy is related to the avoidance of flipping the magnetic moments. This anisotropic motion raises implications regarding the growth of nanostructures and thin films on such a magnetic surface, which we will investigate in the following section.

4.3 Anisotropic growth of Co on Mn/Re(0001)

The focus in this section is the anisotropic growth of Co on the RW-AFM state of Mn/Re(0001), which is also discussed in our manuscript titled "Reciprocity of magnetism and nanostructure growth", see [Publications](#).

The RW-AFM state of Mn/Re(0001) leads to a symmetry-breaking in the electronic DOS as well as a 15 pm lateral structural shift in the Mn layer from the hollow-sites towards the bridge sites. We have found that Co and Rh adatoms move anisotropically on this uniaxial magnetic state at 4 K, see Sec. 4.2. It follows, that, in principle, the magnetic state should be able to directly or indirectly affect the growth of nanostructures.

The contribution of diffusion to this growth can be separated into two categories: Adatoms freely diffusing on a terrace and atoms diffusing while attached to edges. For a more detailed introduction to different diffusion processes and their effect on thin film growth, see Sec. 2.5.

Anisotropic diffusion of adatoms on terraces, as observed for Co and Rh atoms on Mn/Re(0001) at 4 K, can lead to anisotropic or even 1D growth in cases where free surface diffusion dominates over diffusion at edges. An example of this is the formation of bi-atomic Fe atom chains on the (1×5) reconstruction of Ir(001) [114], although the corrugation of the surface plays a significant role.

If the temperature is high enough that atoms remain mobile while attached to edges, diffusion processes along these edges can start to play a role. Here, edges that are structurally and electronically different can show different adatom mobility and therefore different growth speed; the result is that the slowest growing edges are dominant on the sample.

In the previous section, the Co and Rh atoms were not mobile at the measurement temperature and the motion was initiated by local voltage pulses. For nanostructure growth, higher temperatures, where single atoms are mobile on the surface, are necessary.

To investigate the possible effect of magnetism on the growth of nanostructures, we need to deposit the material we want to grow above and below the magnetic ordering temperature. Using DFT parameters and Monte-Carlo simulations, the Néel temperature has previously been estimated to be ≈ 160 K [57, 126]. Therefore, we deposit Co at 300 K and at 80 K and can directly compare the two cases. In Fig. 24a we can see that at 300 K the Co forms roughly hexagonal and triangular islands on the Mn and Re, respectively, following the symmetry of the surface. The island edges are oriented along the close-packed row directions.

Below the Néel temperature the growth is very different, see Fig. 24b. On the Mn areas, we find elongated Co islands growing with a large aspect ratio, while on the Re substrate only small clusters form; this indicates that the Co adatom mobility is much lower on the Re areas than on the Mn areas. We observe that these elongated Co nanostructures are usually oriented in the same direction as other nanostructures in their vicinity. However, across the whole Mn area, we generally find three elongation directions at angles of 120° to each other. This large difference in the island shape for growth above and below the magnetic ordering temperature and the clear symmetry-breaking of the Co islands grown at 80 K, can be seen as the first clear indication that the magnetic state has some effect on the nanostructure growth.

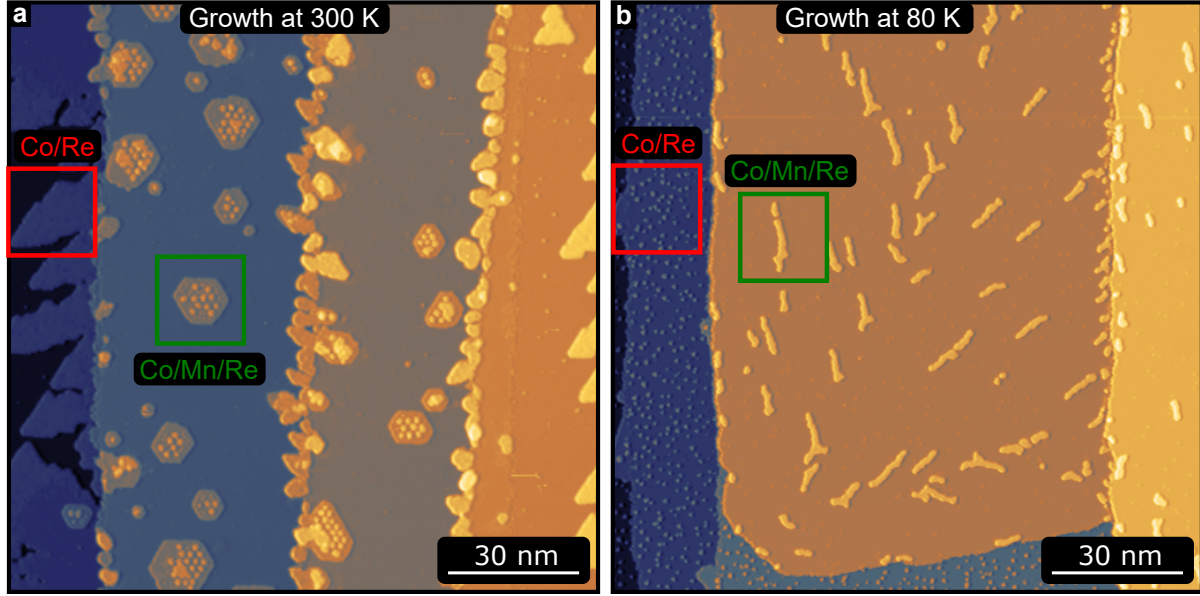


Fig. 24 **a**, Partially differentiated STM image of Co deposited on Mn/Re(0001) at room temperature. Hexagonal and triangular Cobalt islands are visible on the Mn monolayer and Re substrate, respectively. $U = 200$ mV, $I = 500$ pA. **b**, Partially differentiated STM image of Co deposited at 80 K. Islands grow with a high aspect ratio on the Mn ML. The orientation depends on the underlying rotational magnetic domain of the RW-AFM state. Co forms small clusters on the bare Re surface. $U = 500$ mV, $I = 500$ pA. Both images were measured at 4.2 K.

4.3.1 Growth above the Néel temperature

When measuring the hexagonal Co islands, grown at 300 K, at a temperature of 4.2 K, the antiferromagnetic order in the Mn layer is established and we find the presence of multiple rotational domains, as indicated by the bright orange domain walls in the dI/dU -maps in Fig. 25a,c, around the islands. These small domains are typically not present for a Mn layer without Co on top and the domain walls are all connected to the Co islands, a clear indication that the domains nucleate during sample cool-down due to the presence of the hexagonal Co islands.

Previous investigations of the systems of Mn/Re(0001), have shown that edges found at step edges or vacancy islands can select for particular rotational domains of the RW-AFM state [57]. Spethmann *et al.* observed that at these edges the missing atoms necessarily result in the $\uparrow\downarrow$ -edge being energetically more favourable, when considering only Heisenberg exchange with the assumption that magnetic parameters are identical to the interior of the Mn monolayer region and that the edge is oriented along a close-packed row direction and atomically perfect. A similar behaviour at the boundary between the RW-AFM state and the Co island may be able to explain our experimental observations.

The orientation of the magnetic domains can be derived using the asymmetry in the native defects or using SP-STM, see Fig. 25b. We observe that the local domain structure always results in one of the $\uparrow\downarrow$ -rows being parallel to the island edges ($\uparrow\downarrow$ -boundary), resulting in an alternating spin pattern of Mn ML at these edges. However, two types of boundary configurations are possible in principle and can be seen in the sketch of a

hexagonal Co island on a single magnetic domain, see Fig. 25d.

The second type of configuration at the boundary is found when the $\uparrow\uparrow$ -rows run parallel to the Co island edge leading to spins oriented in the same direction along the boundary ($\uparrow\uparrow$ -boundary), see white rectangles in Fig. 25d. This boundary configuration is never observed in our samples, which indicates that it is energetically unfavourable. This configuration is avoided by nucleating the observed additional small domains, clearly demonstrating that the hexagonal Co islands disturb the magnetic state.

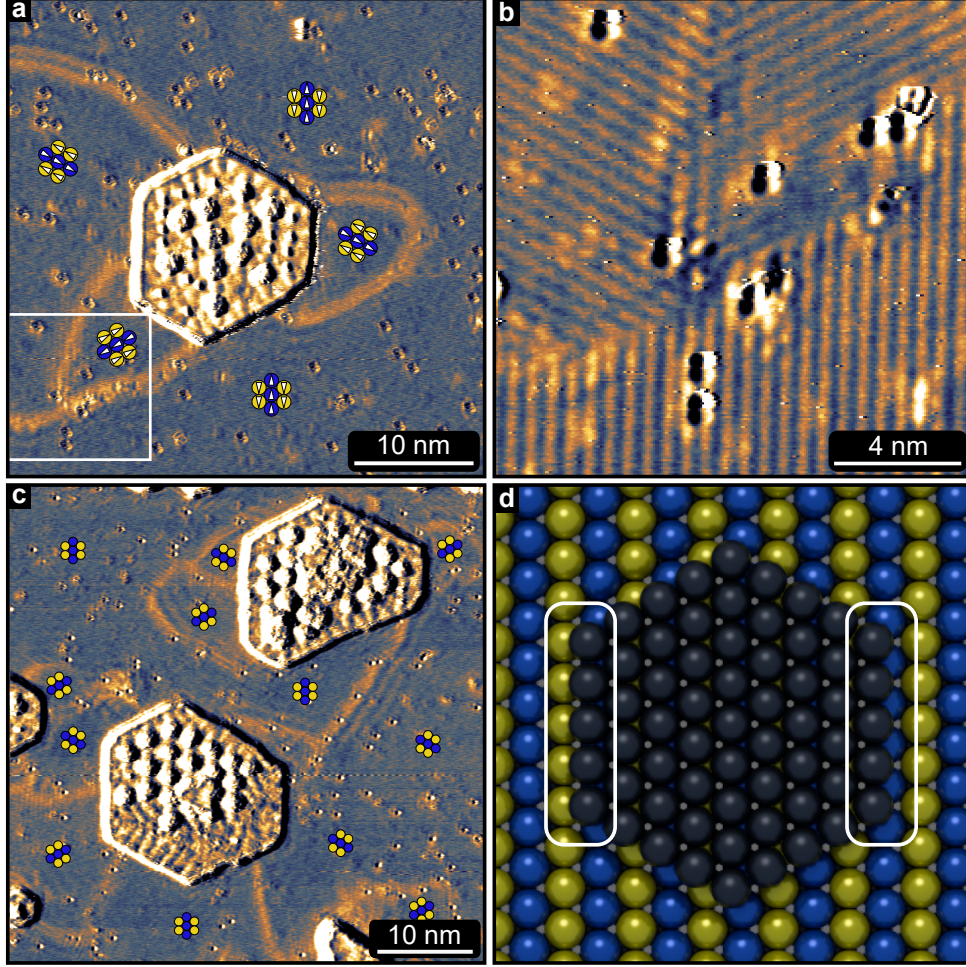


Fig. 25 **a**, dI/dU -map of a hexagonal Co island on the Mn ML grown at 300 K. The orange lines are the domain walls. Blue-yellow sketches mark domain orientation. $U = 10$ mV, $I = 3$ nA. **b**, SP-STM image of the region marked by the white rectangle in **a**. $U = 10$ mV, $I = 5$ nA. **c**, dI/dU -map of multiple hexagonal Co islands grown at 300 K on the Mn ML. $U = 10$ mV, $I = 3$ nA. **d**, 3D-sketch of a hexagonal island on a single domain of the RW-AFM state. The two inequivalent $\uparrow\uparrow$ -edges are marked by the white rectangles. **a-c**: $T = 4.2$ K.

In our case, the preference for the $\uparrow\downarrow$ -boundary configuration may be related to the magnetic state of the Co islands. While we were unable to observe any magnetic contrast on the Co/Mn/Re(0001) regions and therefore cannot determine their magnetic state, we expect a ferromagnetic configuration to be most likely. For an OOP ferromagnetic Co island, one might expect very similar behaviour to a vacancy island, as the Co spins

are perpendicular to both ferromagnetic sublattices. In addition, a simple Monte Carlo simulation of the formation of an IP ferromagnetic island, done by André Kubetzka using Monte Crystal [18], indicate that for weak coupling at the interface between a ferromagnetic and RW-AFM region, $\uparrow\uparrow$ -edges are avoided as well.

The room temperature growth of Co islands can be used to increase the density of domain walls and form a domain wall network compared to the pristine magnetic layer. In the system of a Mn-DL on Ir(111), which hosts the same magnetic state, strain induced by argon bubbles can be used in a similar way to nucleate domain walls, forming a network [58].

On top of the Co ML island we find Co clusters as the start of the second layer, sometimes even growing in a hexagonal arrangement, see Fig. 25a,c. Looking more closely at topographic images, we observe a contrast of bright and dark spots on the Co monolayer regions which can be identified as a reconstruction, see Fig. 26a. The arrangement of second layer Co clusters is directly related to this underlying reconstruction. This reconstruction is not unexpected, as the ideal Co lattice constant is only $\approx 90\%$ of the Re lattice constant. Instead of adhering to the Re lattice constant, which induces significant strain, the Cobalt relaxes to an intralayer distance closer to its desired lattice constant.

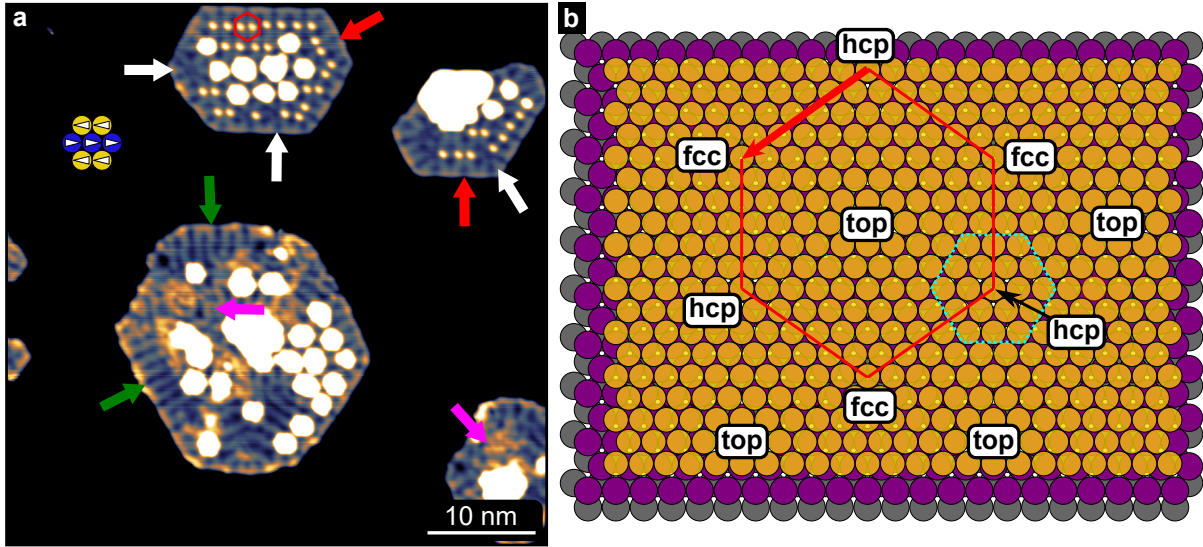


Fig. 26 a, STM image of multiple hexagonal Co islands grown on Mn/Re(0001) at 300 K. The colour-map is adjusted to provide contrast only on top of the Co layer. The direction of the close-packed rows is marked by the blue-yellow sketch. Coloured arrows mark different contrast of dark spots in the reconstruction: Honeycomb (red), hexagonal (white), uniaxial (green), unordered (pink). $U = 50$ mV, $I = 2$ nA, $T = 4.2$ K. **b**, Sketch of a Co layer with 90% of the lattice constant of the Rh and Mn layers, giving rise to a Moiré pattern. Red hexagon connects different regions where the Co atoms are located in the hollow-site positions. Cyan dotted hexagon represents a 1.5 nm wide region around a Co atom in the hcp hollow-site position.

We observe some Co ML regions, which host a honeycomb like pattern of dark spots, see red hexagon in Fig. 26a, with a distance between spots of ≈ 1.4 nm and variations of ± 0.1 nm across the sample. This type of pattern resembles a Moiré pattern where the dark spots correspond to regions of atoms in both fcc and hcp hollow-site positions. We

can model this Moiré pattern by placing a Co layer at 90% of the Re lattice constant on top of the normal Mn layer, with the Re lattice constant as depicted in Fig. 26b. The distances between neighbouring hollow-site areas, see red hexagon and red arrows, are 1.5 nm and are in decent agreement with the areas hosting the honeycomb pattern on the Co ML, see red hexagon in Fig. 26a.

However, in other Co regions the dark spots of the reconstruction appear instead in a hexagonal pattern or as uniaxial dark stripes. Even somewhat unordered regions remain, marked by white, green, and pink coloured arrows, respectively, highlighting that no clear long-range order exists for this reconstruction. The hexagonal pattern shows approximately the same distance between dark spots as the honeycomb pattern. For the uniaxial pattern we observe a distance between stripes of ≈ 1.2 nm, which fits well with a Moiré with reduced intralayer Co distance only along one direction, assuming that both the hcp and fcc regions appear as dark stripes.

Overall, we expect the Co reconstruction to be negligible to the observed effect on the surrounding magnetic state and therefore expect the same behaviour for a pseudomorphic hexagonal island.

4.3.2 Growth below the Néel temperature

Depositing Co below the Néel temperature at 80 K results in Co 1D-nanostructures with their long axis pointing approximately perpendicular to the $\uparrow\uparrow$ -rows, see Fig. 27a. We find that the orientation is directly tied to the underlying rotational domain of the RW-AFM state as Co nanostructures on different rotational domains grow at approximately 120° to each other, see Fig. 27b.

If the growth of the nanostructures was mainly driven by the anisotropic diffusion observed at 4 K in the previous section, we would indeed expect the elongation direction to be tied to the underlying magnetic domains, however, it should be oriented along the $\uparrow\uparrow$ -rows instead of perpendicular to them. Although the anisotropy of the diffusion may have decreased significantly at 80 K, we find it unlikely that the anisotropy inverts. Additionally, the elongation direction is not along the close-packed row direction, further indicating that the anisotropy in the free diffusion of Co atoms on the Mn monolayer terraces cannot explain the observed 1D growth.

The density of Co nanostructures appears to be on average higher at or near the domain walls, see Fig. 24b, potentially related to the fact that adatoms were observed to often be pinned at the domain walls at 4.2 K [57]. If this effect is present at 80 K as well, it may explain the greater number of nucleation centres, and therefore nanostructures, located there. Near the domain walls, the shape of the Co islands deviates from being elongated strictly in one direction, and instead islands with bends can be observed. It appears, that two different effects contribute to this: On one hand, the islands grow preferentially along the domain walls. On the other hand, because one domain favours one growth direction and the other domain favours one rotated by 120° , parts of the Co nanostructures that do not grow perfectly along the domain wall show two different elongation directions. Interestingly, this also means that one can relatively easily estimate the position of domain walls by just looking at regions where nanostructures grow, that are not strictly elongated along just one direction.

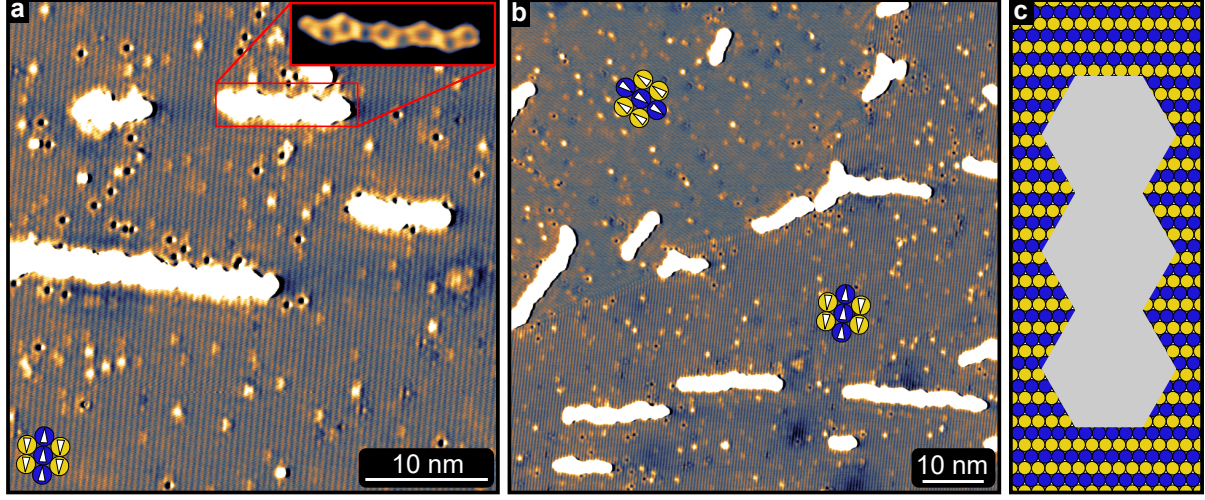


Fig. 27 **a**, Partially differentiated SP-STM image of elongated Co islands grown at 80 K. The rows of the RW-AFM state are oriented perpendicular to the long axis of the islands. Inset shows the topography of such a 1D-nanostructure. $U = 10$ mV, $I = 7$ nA. **b**, Partially differentiated SP-STM image of Co nanostructures on two different rotational domains of the magnetic state, growing preferentially along different directions. $U = 10$ mV, $I = 4$ nA. **c**, Sketch of the orientation of the $\uparrow\uparrow$ -rows at the edges of such a 1D-nanostructure. All images were measured at 4.2 K.

The long edge of the Co nanostructures shows a zig-zag structure, see inset in Fig. 27a. For the hexagonal Co islands, we have found that the edges running along the close-packed row directions are energetically favourable. As the long axis of the 1D Co nanostructures is not pointing along a close-packed row direction, the zig-zag edge naturally emerges as the optimal configuration. This means that the $\uparrow\uparrow$ -rows that are oriented perpendicular to the long axis of the Co 1D-nanostructures intersect the edges at $120^\circ/60^\circ$ giving rise to $\uparrow\downarrow$ -edges, which as we have seen for the hexagonal Co islands in Fig. 25 are energetically more favourable. Only at the ends of the 1D nanostructures the unfavourable configuration remains, see sketch in Fig. 27c. In contrast to the hexagonal Co islands grown at 300 K that disturb the RW-AFM state when cooled below the magnetic ordering temperature, the Co 1D-nanostructures grow in a way that minimizes the disturbance of the magnetic state and typically does not result in any additional domains in the vicinity of the structures.

We observe a periodic pattern of dark spots on top of the 1D-nanostructures indicating that the island is reconstructed, similar to the hexagonal islands grown at 300 K, see inset in Fig. 27a. The Co nanostructures appear to be made from of roughly hexagonal building blocks, see rough sketch in Fig. 27c. The size of these building blocks is likely related to the Moiré pattern discussed previously and can possibly be identified as a small approximately 1.5 nm wide hexagonal region around a Co atom in one of the hollow-site positions, see cyan dotted hexagon in Fig. 26b. One could imagine that adding Co atoms to an already existing hexagonal unit is energetically less favourable, as atoms would be placed at top sites or bridge sites. Preferential nucleation of the next unit at the $\uparrow\uparrow$ -edge could then lead to the observed anisotropic growth.

The 1D-nanostructures are oriented optimally for the particular rotational domain of

the RW-AFM state that they grow on. However, if the magnetic state changes after they are grown, we find that they pin their favoured rotational domain locally, resulting in the observed magnetic structure in Fig. 28. The observed configuration suggests that a domain wall swept over the terrace, changing the main domain of the terrace after the 1D-nanostructures were already grown. The estimated domain structure present during growth and during measurement is marked in Fig. 28a-b, respectively. The exact previous domain wall position during the growth can no longer be reconstructed, nevertheless, an estimate of the previous overall domain structure can be derived from the orientation of 1D nanostructures. The current domain wall positions and domain structure can be measured directly, as the domain wall positions are visible in maps of differential tunneling conductance and even in the topography to some extent, although the colour-map contrast is not optimized for that in Fig. 28a,b.

Taking a closer look, we can observe that each small group of 1D-nanostructures is surrounded by a domain wall, with the domain orientation inside corresponding to the orientation present during growth, which is therefore also energetically favourable, see blue-yellow sketches indicating the domain orientation in Fig. 28c. This indicates that the energy cost of the domain walls is lower compared to a configuration where a different rotational domain of the RW-AFM is in direct vicinity to the Co 1D structures, because the unfavourable $\uparrow\uparrow$ -edges are introduced for parts of the long zig-zag edge. This is likely further supported by the fact that the domain walls go right through the tip of Co 1D-nanostructures, allowing for both the long edges and short edges to have a $\uparrow\downarrow$ -row configuration.

This is the only sample area where we have observed such a change in the magnetic structure after growth. While this data was not measured in the virgin state, but after a sweep to 9 T and back to 0 T, we expect that this change in domain structure was not induced by the magnetic field, as this antiferromagnetic state is typically not susceptible to change even for higher magnetic fields. However, one could imagine the magnetic field affecting the Co islands and therefore indirectly the magnetic state of the Mn layer.

4.3.3 Investigation of the growth mechanism

When considering diffusion along the edges, the observed anisotropic growth requires the Co atoms arriving at the long edge, which would be an increasing fraction of all incoming Co atoms as the aspect ratio grows, to be quite mobile. The attached Co atoms need to be able to move to the short edges and remain there, which means that the Co atoms need to be less mobile at these short edges. It is likely that the Co reconstruction plays a significant role in allowing for the large aspect-ratios of the 1D-nanostructures, still magnetism is a necessary requirement as we have seen by comparing growth above and below the magnetic ordering temperature.

The question arises, what one would expect if the Co island grew pseudomorphically. If the size of the nanostructures is not limited in some direction by a reconstruction, we speculate that Co islands would likely grow in a diamond shape, where the unfavourable $\uparrow\uparrow$ -edges do not exist. In this case, the nanostructure growth would still be anisotropic and uniaxial but the aspect ratio of the structures would be smaller.

Another growth mechanism may involve Co atoms that attach at the $\uparrow\downarrow$ -boundary of

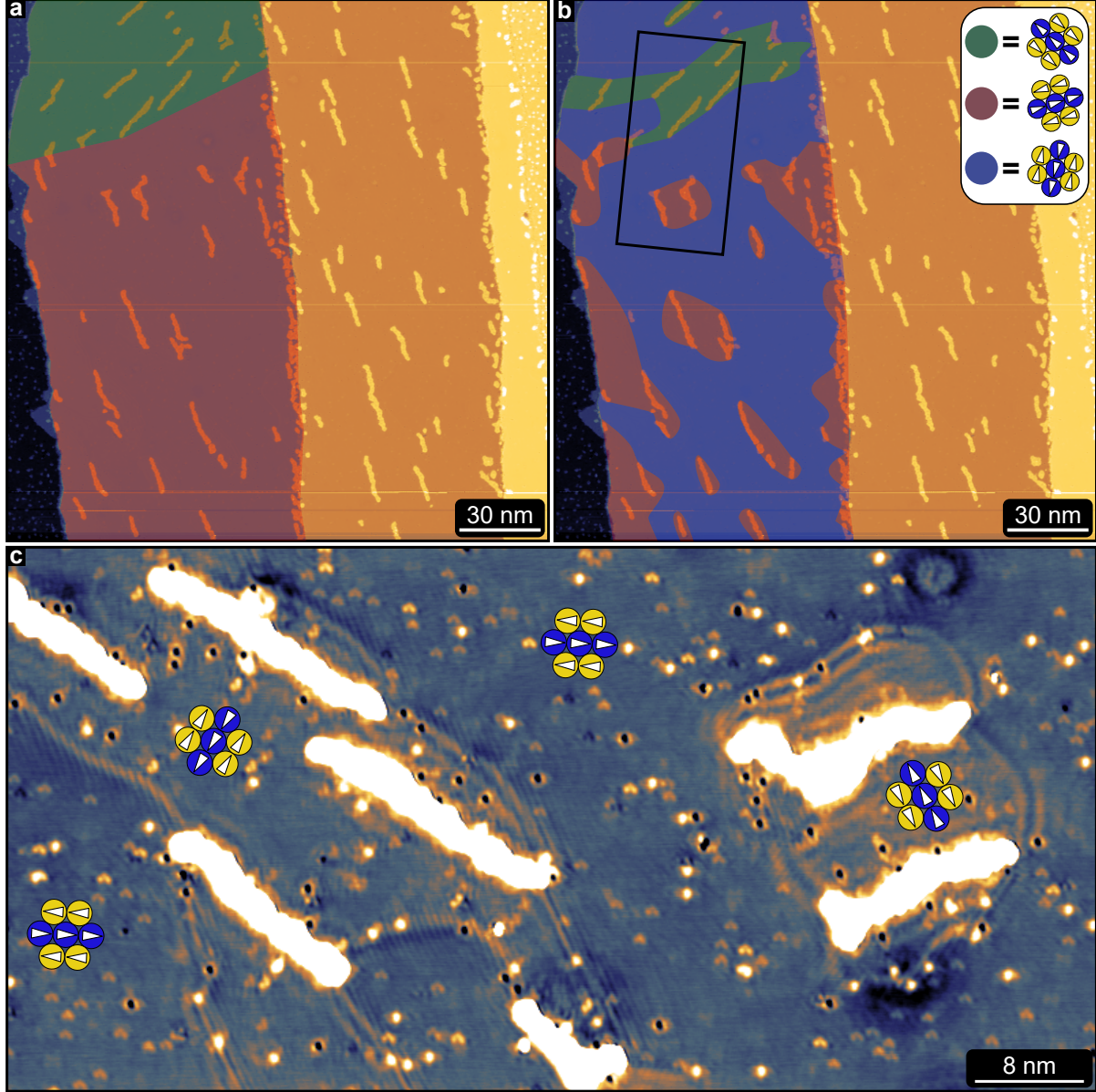


Fig. 28 a-b, STM overview image of a sample with Co deposited on Mn/Re(0001) at 80 K. On the large Mn terrace on the left the rotational domains are shaded according to the rotational domain beneath. In **a** the domain structure is estimated through the growth direction of the 1D-nanostructure, while for **b** the measured domain layout is represented. **c**, STM image of the area marked in **b**. Domain walls appear as bright orange lines. Domain orientation is marked by blue-yellow sketches. All images: $U = 5$ mV, $I = 5$ nA, $T = 4.2$ K.

the Co nanostructures to "evaporate" back onto the terraces to continue diffusion freely. The energy barrier for this evaporation process at the unfavourable $\uparrow\uparrow$ -boundary would be required to be much higher. The mechanism cannot be experimentally differentiated from the edge diffusion mechanism and may even coexist with it. However, one would expect the energy barriers for diffusion along the edges to be significantly smaller than for adatoms evaporating back onto the Mn ML terraces and therefore for the edge diffusion mechanism to dominate overall.

For the proposed growth mechanism, which is likely mainly driven by diffusion processes of Co atoms/clusters at the island edges, the Co atoms preferentially nucleate at the short edges due to lower adatom mobility there. Another possible explanation for this preferential nucleation exists. So far, a change in the magnetic state in the vicinity of the Co nanostructures has not been considered. Although we do not know the magnetic state of the Co islands, it is likely that the RW-AFM state may be modified near this boundary.

We have seen that for the hexagonal islands grown at 300 K the edges, hosting the unfavourable $\uparrow\uparrow$ -boundary, nucleate additional rotational domains. For the elongated Co islands this edge appears only at the ends of the 1D nanostructure, but a distortion of the magnetic state away from the perfect RW-AFM state towards the 2Q state may be energetically favourable. We have seen that Co appears to preferentially grow on domain walls, which host a 2Q state in their centre, see also Sec. 2.3.7. A distortion towards a 2Q state at the short edges might therefore lead to the pinning of additional Co atoms and clusters and could be an alternative mechanism for preferential Co growth. Close up spin-polarized STM measurements in the direct vicinity of these islands show no visible distortion of the RW-AFM state near the short edges of the 1D nanostructures, see Fig. 29e,f; however, we cannot exclude a small distortion of the RW-AFM state.

4.3.4 The magnetic state at the short edge

One can see that the elongated islands are typically surrounded by a single domain of the magnetic state; however, in some rare cases an additional domain can be observed at a $\uparrow\uparrow$ -edge, see Fig. 29d,h. It appears that for particular configurations, the overall energy can be lowered further by nucleating a small domain at the short edge, because the energy cost of a domain wall, which is already quite small in this system in general, can be lowered further due to the surrounding defect configuration until it becomes smaller than the energy gained by introducing the $\uparrow\uparrow$ -edge. An alternative explanation could be that the state with the small domains at the short edges is actually the ground state, but the system is able to remain in a meta-stable state due to a large enough energy barrier.

A more common way to reduce the overall energy of the 1D nanostructures is to capture a domain wall and attach it to the end of a 1D Co nanostructure. This cannot be realized for all nanostructures, but an example is displayed in Fig. 29a,c, where the shape of the domain wall is marked by the white line at the location of the red square in the bottom left. For the pristine Mn layer, the domain wall would run mostly straight with a smooth profile, not the zig-zag-like shape observed here. This pinning at the short edge eliminates the unfavourable $\uparrow\uparrow$ -edge and transforms it into a $\uparrow\downarrow$ -edge. This is supported by the fact that domain walls can move with relatively low energy in this system [57].

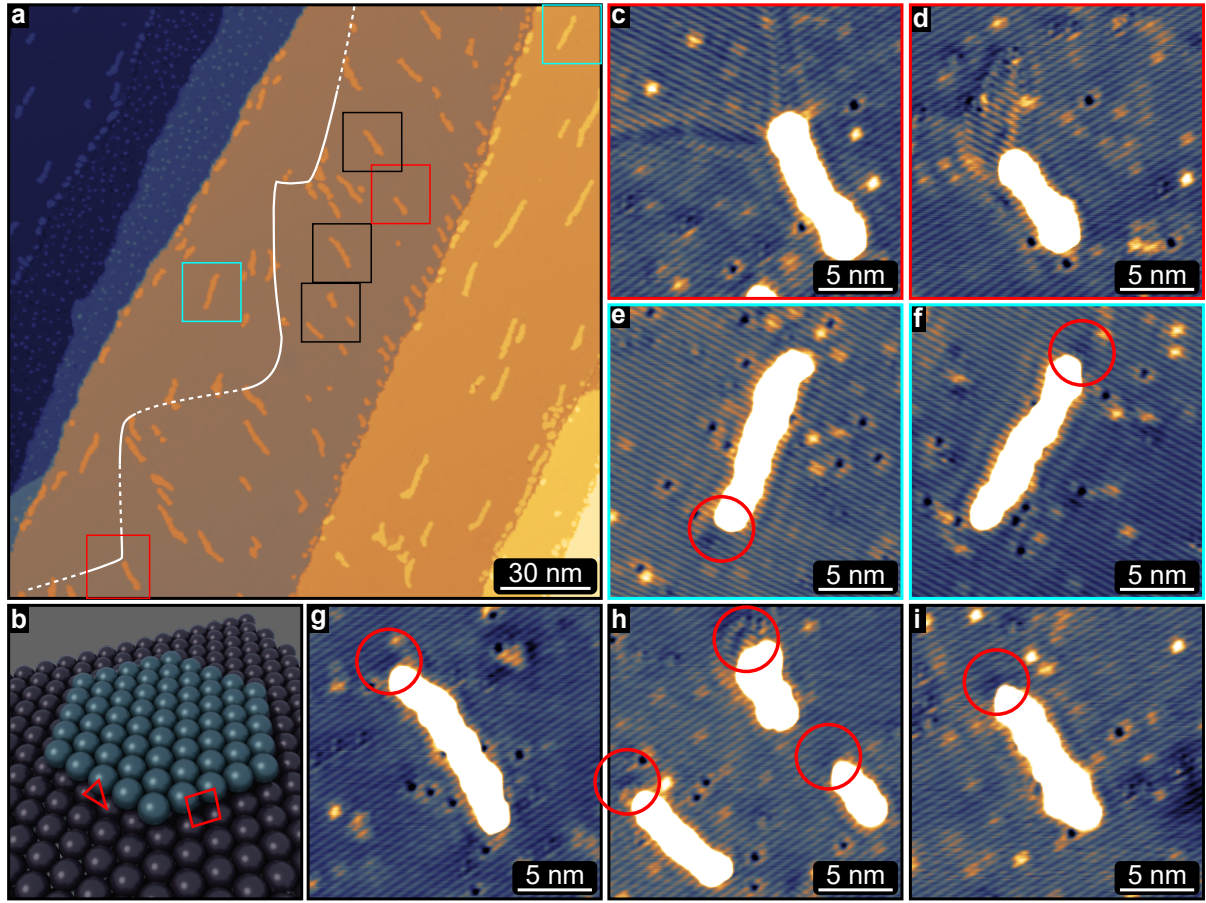


Fig. 29 **a**, STM overview image of Co deposited at 80 K on top of Mn/Re(0001). The white line indicates the position of a domain wall on the centre terrace; here, a solid line indicates confirmed measured positions, and a dotted line indicates inferred positions. **b**, 3D-sketch of a hexagonal Co island on a hexagonal substrate. The edges along the close-packed row directions come in two variations with different coordination environment. Edges going in the same direction are of one type on one side of the island and of the second type on the opposite side. **c**, SP-STM image of the magnetic state around a Co island. The domain wall is positioned in such a way, that the short edge of the Co islands is exposed to the other rotational domain. **d**, SP-STM image of the magnetic state around a Co island. A domain at the short edge of the island is visible. **e-f**, SP-STM image of the magnetic state for two islands growing on a rotational domain with the same orientation but on different terraces. **e** is marked by the cyan square in **a** and located on the left, while **f** is located one terrace higher (right). The red circles indicate the short edge with slightly darker contrast on the Mn layer compared to the opposite short edge. **g-i**, SP-STM images of the magnetic state for multiple nanostructures. The red circles highlight the in-equivalence of the two short edges, as the contrast on the indicated top edges is slightly darker than on the bottom short edge. All images: $U = 5$ mV, $I = 5$ nA, $T = 4.2$ K.

The edges of the Co islands are oriented along the close-packed row directions for Co structures grown at 300 K as well as at 80 K. For each edge direction, two types of edges with different coordination to the Mn layer below are possible, see red triangle and square in Fig. 29b. These edges appear at opposite ends of the hexagonal islands and can be

energetically different. This is the case in this system, as can be seen by the hexagonal Co island in Fig. 25a. It is technically not a perfect hexagon but truncated with opposite edges being of different length.

For the Co 1D-nanostructures grown at 80 K, the two short edges are electronically different to the long edges, due to the symmetry-breaking of the magnetic state. In addition, the two short edges at opposite ends appear different. One edge reproducibly shows a dark spot on the nearby Mn ML, while the other edge does not, see red circles in Fig. 29e-i. This dark spot always appears for the same type of edge, as can be seen by comparing Fig. 29e and Fig. 29g-i, located on the same terrace but on different rotational domains. The edge with the dark spot rotates by 120° when comparing 1D-nanostructures on different rotational domains on the same terrace. For structures on the same rotational domain but located on a terrace one step edge up, the edge with the dark spot rotates by 180° , compare Fig. 29e and Fig. 29f. The scan areas for the imaged islands are marked in the overview STM image in Fig. 29a.

This dark spot appears to be of electronic origin as it has also been observed with a tip showing no magnetic contrast. We speculate that this dark spot might be standing-electron-wave contrast, although no spatial oscillation is visible, perhaps due to a fast lateral decay. Nevertheless, this clearly illustrates the fact that the two short edges are not equivalent. No similar electronic contrast is observed for the hexagonal islands grown at 300 K. A possible explanation could be that such an electronic contrast is only visible at the unfavourable $\uparrow\uparrow$ -boundaries, which are avoided for the hexagonal islands by the nucleation of additional domains.

Additionally, we observed ≈ 10 of the uncommon cases where a small domain was nucleated at a short edge, see Fig. 29d,h. We found these domains always at the short edge with the dark spot and they have not been observed on the other edge. Although the sample count is quite limited, the chance to coincidentally find all of them at the same edge, assuming equal probability, is ≈ 1 permille. This means, that the short edge hosting the dark spots is energetically even more unfavourable than the opposing short edge, which in some rare cases allows for an overall reduction in energy for the former but not the latter when the energy cost of a domain wall is small enough.

This inequality between the short edges could in principle lead to a difference in the diffusion barrier for atoms/clusters attached to these edges and therefore in a difference in growth speed. To investigate if the growth happens preferentially at one of the short edges, we grow Co 1D-nanostructures in a two-step process. Fig. 30a,b shows STM images of the same area with two different Co coverages (i.e. ≈ 0.12 ML & ≈ 0.20 ML), this is possible because Co is deposited with the sample remaining in the STM. Comparing the two coverages, see also the red mask derived from the Co structures in Fig. 30a overlaid on top of the STM image in Fig. 30b, we find no clear indication that the Co 1D-nanostructures grow preferentially on one of the short edges.

4.3.5 AFM Bitter technique

An overview STM image of the same sample with an even higher coverage of ≈ 0.3 ML is shown in Fig. 30c. This image clearly illustrates that the deposition of Co can map the large scale antiferromagnetic domain structures, including the orientation of the antifer-

romagnetic domains, domain wall positions, and therefore also the domain size.

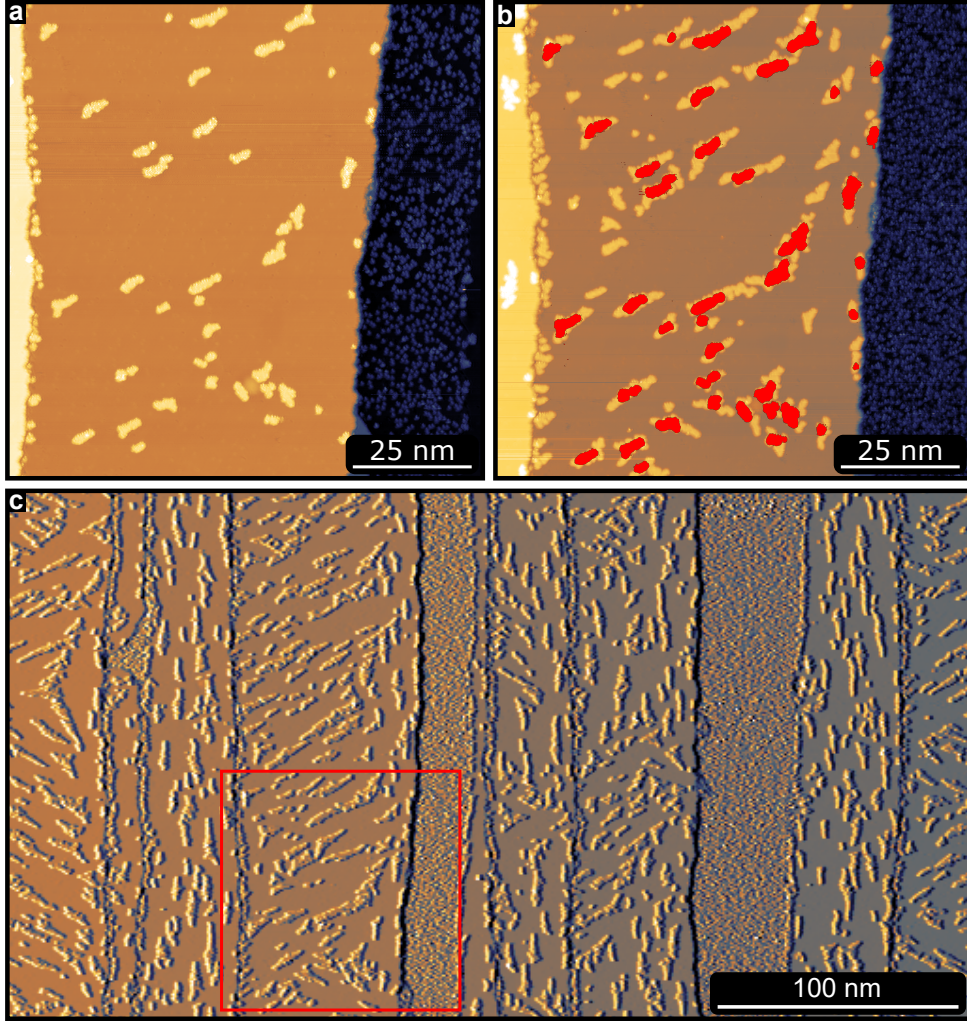


Fig. 30 **a**, STM image of ≈ 0.12 ML of Co on Mn/Re(0001) deposited at 80 K. $U = 100$ mV, $I = 500$ pA. **b**, STM image of ≈ 0.20 ML of Co on Mn/Re(0001) deposited at 80 K. The Co island shapes from **a** are overlaid as a red mask. No clear preference for growth at one short edge over the other is visible. $U = 100$ mV, $I = 500$ pA. **c**, Overview STM image of ≈ 0.3 ML of Co on Mn/Re(0001) deposited at 80 K. The orientation and shape of the nanostructures can be used to derive the orientation of the magnetic domains and the domain wall positions. The red square marks the scan area in **a-b**. $U = 500$ mV, $I = 500$ pA. All: 80 K.

Firstly, the elongation directions directly encode the orientation of the underlying magnetic state. Secondly, the domain wall position can be estimated by observing the transition from one elongation direction to another and also by following the lines of Co nanostructures, that are not strictly elongated along just one direction. However, the position of the domain walls of the pristine film with no Co can only be approximated within a range of 10-20 nm, as we have seen that the domain wall position can be locally affected by the Co nanostructures. Nevertheless, the anisotropic growth of Co in this system serves as an antiferromagnetic analog of the Bitter technique. The Bitter technique is typically

used to image domain walls in ferromagnets using magnetic nanoparticles [127, 128]. The stray-field near the domain walls leads to preferential aggregation of the nanoparticles there.

This AFM Bitter technique analog, should in principle be expandable to other antiferromagnetic systems, with the only requirements being a high enough mobility of adatoms at a temperature below the Néel temperature and an anisotropy in the growth of a nanostructure due to the antiferromagnetic state. One could also imagine a more broadly applicable, more limited technique, not requiring anisotropic nanostructure growth, where the domain walls could be visualized due to an increased nanostructure density compared to the domains but no information about the orientation of the rotational domains could be derived. Because there is no need for magnetic resolution or even spatial resolution in the atomic range, these methods are usable for other scanning probe techniques as well as other imaging techniques with a spatial resolution on the order of the size of the magnetic domains.

We observed the anisotropic growth of the Co thin films up to a temperature of 100 K. This behaviour is expected due to the predicted Néel temperature of 160 K by Monte-Carlo-Simulation, based on parameters calculated by DFT. However, Elmers *et al.* estimate a Néel temperature of 75 ± 5 K from the temperature-dependent band-splitting measurements using momentum microscopy [129]. They also measured a constant, non-zero band-splitting above the measured transition temperature, which was interpreted as short range magnetic order persisting up to 110 K. However, this is in contradiction to our experimental observation of magnetically-driven anisotropic growth at 100 K. This discrepancy could be caused by differing thin film quality (i.e. different levels of contamination). Another possible explanation could be that small clusters of Co atoms on the magnetic state of Mn/Re(0001) lead to an increase of the magnetic ordering temperature locally.

4.4 Summary

We have shown the effect of magnetism on adatom diffusion and nanostructure growth in the system of Mn/Re(0001), hosting a uniaxial antiferromagnetic state. The magnetic state breaks the symmetry of the hexagonal substrate, resulting also in symmetry-breaking of the local density of states and a lateral shift of the Mn layer.

Single Co adatoms were found to move preferentially along the $\uparrow\uparrow$ -rows at 4.2 K in this system, when motion was initiated during scanning, using local voltage pulses or even when hovering the tip above the adatom at medium bias of 100 mV. Similar behaviour was found for Rh and Ir atoms as well, although the movement distance and adatom mobility decreased from Co via Rh to Ir. DFT calculations for Co and Rh atoms are in good agreement with experiments, as they find a moderately anisotropic potential landscape, leading to the same anisotropic motion.

When depositing Co in larger amounts on the same Mn/Re(0001) system at 80 K, we observe the growth of 1D-nanostructures with the orientation of their long axis being coupled to the underlying rotational domain of the antiferromagnetic state. The nanostructure morphology completely changes when depositing Co above the magnetic ordering temperature, where Co grows in typical hexagonal and triangular islands. The

Co 1D-nanostructures are oriented with their long axis approximately perpendicular to the $\uparrow\uparrow$ -rows, which indicates that the anisotropic growth is not driven by the anisotropic motion of Co atoms observed at 4.2 K. Instead, the diffusion of Co atoms/clusters along the edges of the Co nanostructures likely plays a crucial role. The Co atoms/clusters must preferentially nucleate at the short edges of the nanostructures, likely due to higher diffusion barriers at the energetically unfavourable short edges.

We have shown that magnetism can be used as a means to control adatom diffusion and nanostructure growth on AFM surfaces with low magnetic symmetry.

5 The System of Mn/Ta(110)

5.1 Introduction

Localized magnetic quasi-particles, such as merons and skyrmions, have been at the forefront of spintronic research in the last two decades due to the potential improvements they offer compared to current information-storage technologies [130–133]. Antiferromagnetic (AFM) states hosting topological magnetic spin textures have been of increasing interest recently because they exhibit certain advantages over their ferromagnetic counterparts [13, 14]. This includes a net zero magnetization, high resilience against stray fields, ultra-fast dynamics, and no transversal motion when the quasi-particles are driven by a current (i.e. skyrmion-Hall-effect). So far AFM skyrmions/merons, which host oppositely magnetized ferromagnetic skyrmions/merons on different sublattices, see Fig. 1, have been mainly investigated experimentally using synthetic antiferromagnets where ferromagnetic skyrmions in spatially separated layers are coupled antiferromagnetically [65, 66]. Jani *et al.* have observed AFM merons and anti-merons at the interface between an Fe₂O₃-film and a Pt overlayer [67]. In both cases the ferromagnetic sub-lattices are located in spatially-separated layers, and no experimental observation of an AFM localized topological spin texture in a single atomic layer has been reported yet. For bcc(110) surfaces, such as Ta(110), non-collinear DMI-stabilized antiferromagnetic spin textures and spin-spirals, have been observed for Mn and Cr-layers on W(110) [54, 134]. In Sec. 5.3 we investigate a system of the same symmetry, Mn/Ta(110), using SP-STM and make use of frustrations arising at the boundary between the Mn DL and ML to stabilize topological AFM spin textures in a single magnetic layer. This discussion is partially based on our manuscript "Topological spin textures in an antiferromagnetic monolayer" available as a preprint, see [Publications](#).

As we have seen in Sec. 2.4.4, interesting physics emerges when magnetic material is brought in the vicinity of a superconductor. The interactions in such a magnet-superconductor-hybrid (MSH) system can lead to the emergence of topological superconductivity. At the boundary between trivial and topological superconductors or between two distinct topological superconducting phases, topologically protected states such as Majorana modes (MM) can form. These states are good candidates for topologically protected quantum computing [22, 135]. While extensive experimental and theoretical work has been done for 1D magnetic chains on *s*-wave superconductors [99, 136], the work on two dimensional MSH-systems is more limited. Ferromagnetic systems have been theoretically predicted and experimentally found to host gapped topological superconducting phases [29]. However, for collinear antiferromagnets or a ferromagnetic spin-spiral, topological nodal point superconducting (TNPSC) phases have been observed [30, 100]. For non-coplanar magnetic states, gapped superconductivity has been predicted theoretically, but no experimental observations have been able to confirm this yet [103–105].

The focus so far has been on investigating the emergence of edge modes between a 2D-MSH system in a topological superconducting phase and the trivial substrate. For future spintronic applications an ability to tune the emergence of such modes and their properties is desirable. In Sec. 5.4 we look at the interaction of magnetism and super-

conductivity in the system of Mn/Ta(110) using STS on both Mn layers and investigate the boundary between the two MSH-systems (Mn ML and DL) as well as between them and the trivial substrate. We show that in MSH systems hosting TNPSC phases, the emergence of edge modes at specific edges and a control over some of their properties may be achievable by locally perturbing these MSH systems or by interfacing them with other MSH systems hosting similar TNPSC phases. The discussion in this section is partially based on the manuscript "Spin-polarized edge modes between different magnet-superconductor-hybrids" released as a preprint, see [Publications](#).

5.2 Preparation of ultra-thin films of Mn/Ta(110)

The Ta single-crystal surface was cleaned by heating up to temperatures > 2300 K. At these temperatures the Oxygen reconstruction is removed and the unreconstructed bcc(110) surface remains. The cleaning process is explained in more detail by Eelbo *et al.* [137]. Mn was deposited on the Ta surface using a PBN Knudsen cell at a temperature of 690°C with an evaporation rate of ≈ 0.2 AL/s. For Mn coverages above 1 AL the deposition started $\approx 8 - 10$ min after a final flash annealing with the crystal still being at elevated temperatures of approximately $100 - 200^\circ\text{C}$. For samples with sub-monolayer coverage, which will be shown in Sec. 5.4, the crystal was cooled to room temperature before evaporation started.

A typical sample of 1.25 AL is shown in Fig. 31a. The Mn almost fully wets the Ta surface, and the DL regions grow from the step edges. Both layers grow pseudomorphically. During growth, the residual Oxygen does not accumulate at the interface between Mn and Ta; it instead gets displaced by the Mn atoms resulting in small patches on the surface where Mn does not grow.

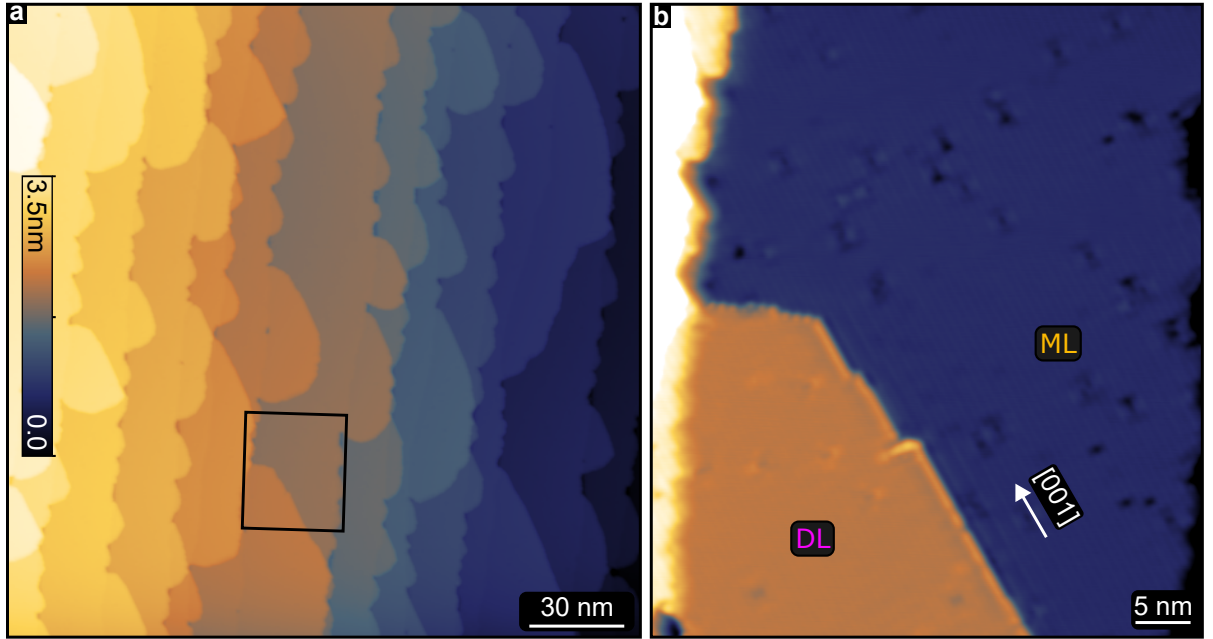


Fig. 31 **a**, STM topography image of ≈ 1.2 AL of Mn on Ta(110). $U = 600$ mV, $I = 500$ pA. **b**, STM topography image of the sample area marked in **a**. A DL region can be seen on the bottom left growing from the Ta step edge. $U = -15$ mV, $I = 2$ nA. Both: $T = 4.2$ K.

5.3 Magnetic properties of the Mn ML and DL

In this section, we will first discuss the magnetic states of the Mn ML and Mn DL, followed by investigating the interaction between the two regions and the spin textures emerging due to frustration at the boundaries.

5.3.1 The magnetic state of the Mn ML and Mn DL

We start investigating the magnetic state of the Mn ML and DL using a Cr bulk tip. In a typical constant-current topography image measured in a sample area, where both ML and DL areas are present, the magnetic contrast is not immediately visible, see Fig. 31b. Adjusting the colour-map contrast to only the DL or ML does reveal magnetic contrast. We have found that in this system the magnetic signal, which is typically relatively weak, can better be seen in the current channel. Even by adjusting the feedback-loop parameters and the scan speed, this does not significantly change.

Current maps of the same sample area as in Fig. 31b reveal a stripe-like contrast on both the Mn ML and DL, see Fig. 32a,b. These stripes are oriented along the $[001]$ direction with spacings between bright lines along the $[1\bar{1}0]$ of two atomic rows (i.e. 466 pm).

Interestingly, on the Mn ML in Fig. 32a this stripe-like contrast vanishes in two areas near the top of the image. The regions of no contrast separate the stripe contrast in three areas, with the middle area showing an inverted contrast with respect to the other two. This indicates a coherent rotation of the spins in the $[001]$ direction, where the magnetic contrast vanishes at points where the antiferromagnetic quantization axis is perpendicular to the tip-magnetization. The rotation of spins in the Mn ML will be discussed in detail in Sec. 5.3.2. Note that in the ML area at the bottom of Fig. 32a, next to the DL island, the stripe contrast is present for a larger area, indicating a suppression of this rotation, see discussion in Sec. 5.3.3 for details.

Comparing Fig. 32a and Fig. 32b, which were measured with different micro-tips, we find that the regions that previously showed the stripe contrast now do not, and vice versa. This means that the tip-magnetization must have rotated by approximately 90° between the two images. The magnetic contrast on the DL and the nearby ML region is strongly visible in Fig. 32a, while nearly vanishing in both regions in Fig. 32b, indicating that their quantization axes are aligned.

Note that the stripe contrast at the bottom of the ML region near the $[001]$ DL edge in Fig. 32b is not of magnetic origin, but represents standing-electron-waves (or QPI). The period of the standing-waves is ≈ 900 pm, which is approximately twice the distance between bright stripes of the magnetic contrast, resulting in the superposition pattern visible in Fig. 32a, where the bright stripes near the step edge to the DL alternate in intensity.

Tracing the magnetic contrast from the DL to ML by following the dark lines across an edge reveals that they land on top of bright lines, indicating that the top DL atoms sit in the fourfold hollow-sites and couple antiferromagnetically to the bottom Mn layer, see Fig. 32c. One can sketch a corresponding lattice of atoms coloured red and blue to indicate the two sublattices with opposing magnetization, see Fig. 32d.

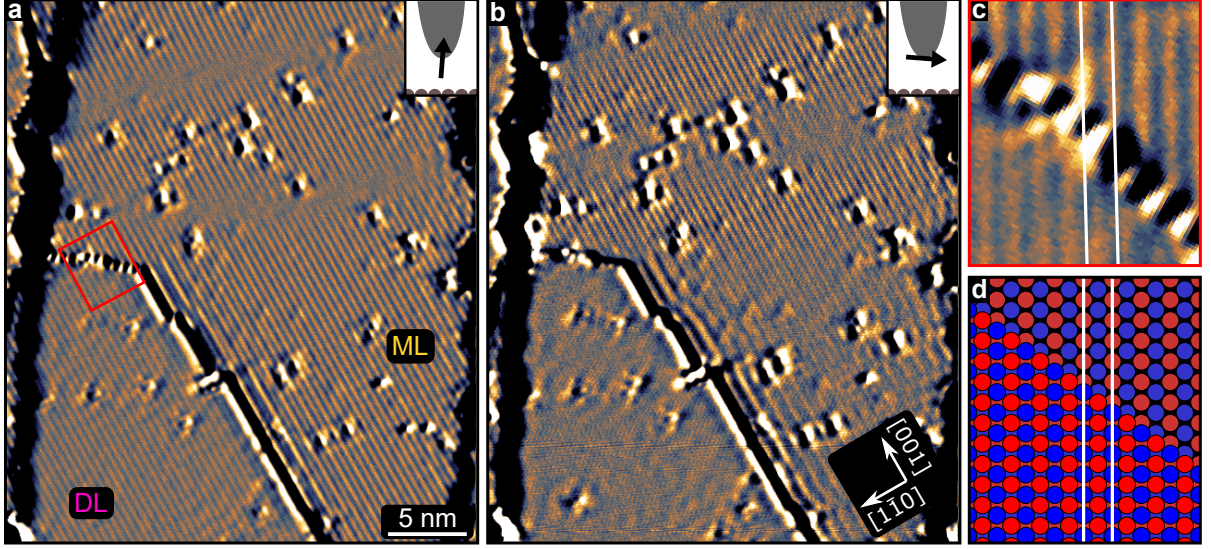


Fig. 32 a-b, SP-STM current maps of the sample area in Fig. 31b, measured with different tip magnetization. **a**: $U = -15$ mV, $I = 2$ nA, **b**: $U = -15$ mV, $I = 5$ nA. **c**, Rotated and scaled cut-out from the area marked by the red rectangle in **a**. **d**, Sketch of the atomic lattice measured in **c**. The red and blue colour of the atoms represent the opposite magnetization. Atoms in the ML are desaturated compared to the DL for enhanced visibility.

Although we know that the tip-magnetization is rotated by $\approx 90^\circ$, see insets in Fig. 32a,b, the orientation of the magnetization is completely unknown. It can be determined by using field-dependent measurements with a soft magnetic tip, as the tip rotates its magnetization axis in the external magnetic field, while the antiferromagnetic state remains unaffected. In Fig. 33 a series of images is shown, measured with a soft-magnetic Fe/W-tip, with corresponding OOP magnetic fields of $0\text{T} \rightarrow +2.5\text{T} \rightarrow 0\text{T} \rightarrow -2.5\text{T}$.

At ± 2.5 T the tip magnetization rotates OOP and the stripe-like magnetic contrast appears on both the Mn ML and DL. This clearly indicates that the Mn DL hosts a collinear OOP antiferromagnetic state and the adjacent ML regions are oriented out-of-plane as well. Additionally, a domain wall is visible, marked by the dotted line in Fig. 33b,d. The magnetic contrast on the DL area is much weaker, which is a general trend, and can also be seen in Fig. 32a. The measurements at ± 2.5 T show the inversion of the OOP component of the tip magnetization, which is particularly evident for the appearance of the interference pattern of standing-electron-waves with the magnetic contrast on the Mn ML. The resulting pattern shows an enhanced intensity for every second bright stripe or every second dark stripe in Fig. 33b and Fig. 33d, respectively.

At zero field the tip magnetization is aligned in-plane and reveals no magnetic contrast, which is only possible if the magnetization is oriented in the $[1\bar{1}0]$ direction, perpendicular to the rotation plane of the spins in the ML, as we will show in Sec. 5.3.2.

5.3.2 The ground state of the Mn ML

We have seen that the spins rotate coherently along the $[001]$ direction in some areas of the Mn ML; however, this rotation appears to be suppressed next to DL regions, which

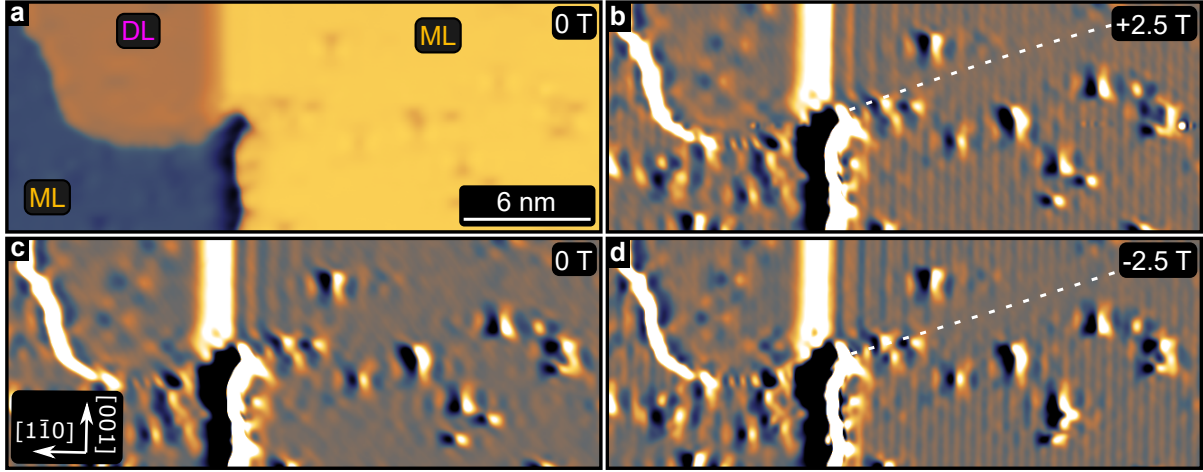


Fig. 33 **a**, STM topography image of a sample area with a DL region at the top left. **b-d**, SP-STM current maps of the same area measured with a soft magnetic tip and at the magnetic field indicated at the top right corner of each image. Dashed white line indicates the position of in-plane spins. Parameters for all images: $U = 10$ mV, $I = 1$ nA, $T = 8.7$ K, with the magnetic field going from $0\text{ T} \rightarrow +2.5\text{ T} \rightarrow 0\text{ T} \rightarrow -2.5\text{ T}$.

	$[1\bar{1}0]$	$[001]$	$[110]$
K_{ML}	0.00	-1.71	-2.23
K_{BL}	0.00	-0.35	-0.96

Table 1 Magneto-crystalline anisotropy constants along the different crystallographic directions for the ML/BL calculated by Tim Drevelow using DFT.

host an OOP collinear antiferromagnetic state. To investigate the ground state of the Mn ML we need to investigate the undisturbed regions far away from any DL areas. Fig. 34b shows a sample area on a large Mn ML terrace. The vanishing of the stripe-like magnetic contrast is highly periodic and the regions of no-contrast are completely straight and oriented along the $[1\bar{1}0]$ direction, in contrast to the previous sample areas where these regions were slightly curved. This periodic modulation of the magnetic contrast clearly indicates a rotation of the quantization axis in the $[001]$ direction, although we cannot derive the plane of rotation of this non-collinear AFM spin-spiral state purely based on this measurement. However, from the field-dependent measurements shown in Fig. 33 we know that the spin-spiral has an OOP component and an IP component.

Differentiating between a helical and cycloidal spin-spiral is not possible experimentally without control/knowledge of the in-plane magnetization, nevertheless one would expect a significant DMI contribution in this high SOC Ta system, which should result in a cycloidal rotation of the spins (i.e. in the plane opened up by the $[001]$ direction and surface normal ($[110]$)), see Fig. 34a. The spins could rotate, in principle, also in any plane between perfectly cycloidal and perfectly helical, but then we should be able to observe two different rotational domains of the magnetic state, which we do not.

DFT calculations by Tim Drevelow (Kiel Univ.) find a cycloidal spin-spiral with a $\mathbf{q}$ -vector along the $[001]$ direction. The factor determining $\mathbf{q}$ is the crystalline anisotropy,

which favours the $[001]$ direction over the $[1\bar{1}0]$ direction, see Table 1. The DMI contribution is smaller, nevertheless lowering the energy for coherent cycloidal rotation in either direction, but would actually prefer the spin-spiral to be oriented in the $[1\bar{1}0]$ direction. The cycloidal nature of the spin-spiral is also in agreement with the similar systems of Cr and Mn on W(110), both systems hosting a cycloidal AFM spin-spiral propagating along the $[001]$ direction and the $[1\bar{1}0]$ direction, respectively [54, 134].

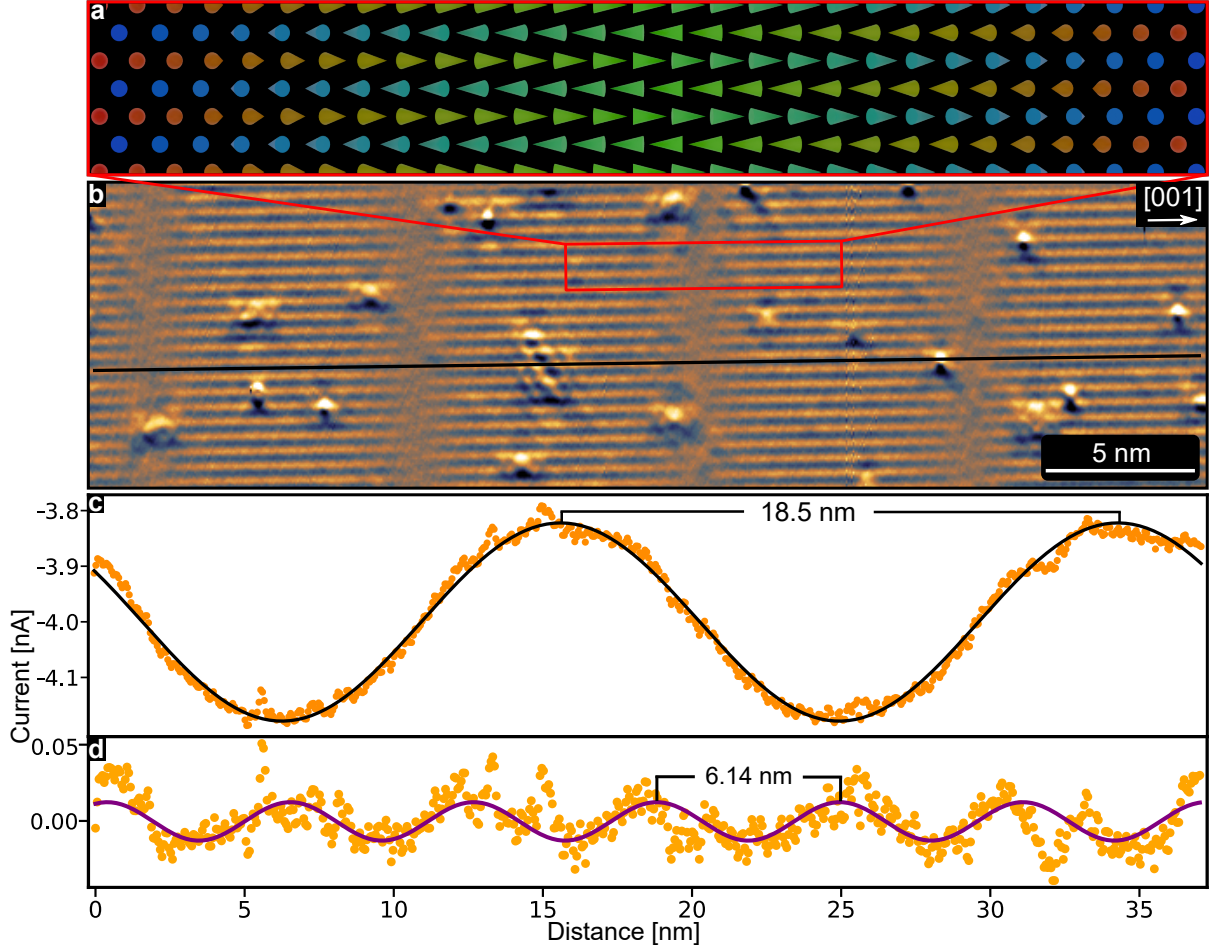


Fig. 34 **a**, Sketch of half a period of a cycloidal antiferromagnetic spin-spiral. The atomic magnetic moments are represented by cones with the colour encoding the direction. Red (up), blue (down), green (in-plane). **b**, SP-STM current map of a Mn ML area hosting an antiferromagnetic spin-spiral, indicated by the periodically vanishing stripe contrast. The region indicated by the red rectangle corresponds to the sketch in **a** for a tip magnetization pointing up. $U = -15$ mV, $I = 4$ nA, $T = 4.2$ K. **c**, The orange dots represent the experimental line-profile created by averaging 17 individual line-profiles taken with the direction and length indicated by the black line in **b**, but spaced by a two-atomic-row distance apart and located on top of the atomic rows. The black line shows a sinusoidal fit. **d**, The orange dots indicate the difference between the experimental data and the fit in **c**. The purple line is a sinusoidal fit.

The AFM spin-spiral appears homogeneous, which means that the angle between neighbouring spins is constant over the spin-spiral period. Due to this homogeneity and

because the tip magnetization during the measurement in Fig. 34b is unknown, we cannot say which regions of the image correspond to in-plane magnetization and which correspond to out-of-plane magnetization. The appearance of the magnetic spin-spiral contrast is independent of a rotation of the tip magnetization in the plane created by the [001] axis and the surface normal ([110]), only a lateral shift of the regions of no magnetic contrast in the [001] direction would be observable.

Averaging multiple line-profiles taken on every second atomic row, results in the combined line-profile in Fig. 34c, see also black line in Fig. 34b. One can see that the magnetic signal is highly periodic and follows a sinusoidal curve closely, as one would expect for a homogeneous spin-spiral. The overall 360° rotation of the spins occurs with a period of ≈ 18.5 nm and OOP/IP regions appear every 9.25 nm. We note, that OOP/IP regions spaced half a period apart (i.e. 9.25 nm) show an inverted magnetic structure, one could say that they are perfectly out-of-phase, see Fig. 34a. Slight deviations from a perfect sinusoidal curve, appear with a period which is three times smaller, see Fig. 34d, and accordingly can be attributed to the difference between the anisotropy energy in the [001] and out-of-plane ([110]) directions, see Table 1.

We have not observed any electronic contrast between IP and OOP regions or across the spin-spiral period in general, for a range of bias voltages from +1 V to -1 V. Two electronic contrast mechanisms need to be considered, see Sec. 2.1.4. NCMR contrast emerges due to changes in the angle between neighbouring spins. These changes are quite small in the spin-spiral of the Mn ML, because it is nearly perfectly homogeneous. Therefore, NCMR contrast is not expected. The second mechanism is the TAMR effect, where spins oriented in different crystallographic directions are electronically different. This contrast mechanism could possibly exist in this system, for instance as an electronic contrast between IP and OOP regions, but has not been observed.

5.3.3 Magnetic frustration at the ML/DL boundary

The ground state of the Mn ML is an AFM spin-spiral, but as we have also seen, in the vicinity of the DL the rotation can be completely suppressed, see Fig. 32a. The top and bottom layers in the DL regions are aligned antiferromagnetically and this collinear AFM state in the bottom layer imposes a certain OOP configuration on the nearby Mn ML regions. The ML spin-spiral is no longer nearly homogeneous in the vicinity of the DL regions. As the OOP regions are favoured, we will refer to the regions where the spins are aligned in the plane as "domain walls" going forward.

To better understand why the spin rotation in the ML is suppressed, Fig. 35 shows a sketch with part of a spin-spiral in the ML and the collinear OOP AFM state in the DL. The presence of the spin-spiral on the ML necessitates that both possible OOP regions interface with the collinear DL AFM state at the boundary. The OOP configuration at the top of the image allows for a continuous anti-parallel alignment across the boundary. In contrast, at the front of the sketch the inverted configuration, found after rotating the spins 180° , clearly leads to a ferromagnetic alignment of two atomic rows in the ML at the boundary and therefore magnetic frustration.

The question arises, what happens to a magnetic state at the boundary between the Mn ML and DL to avoid these configurations that are likely energetically unfavourable?

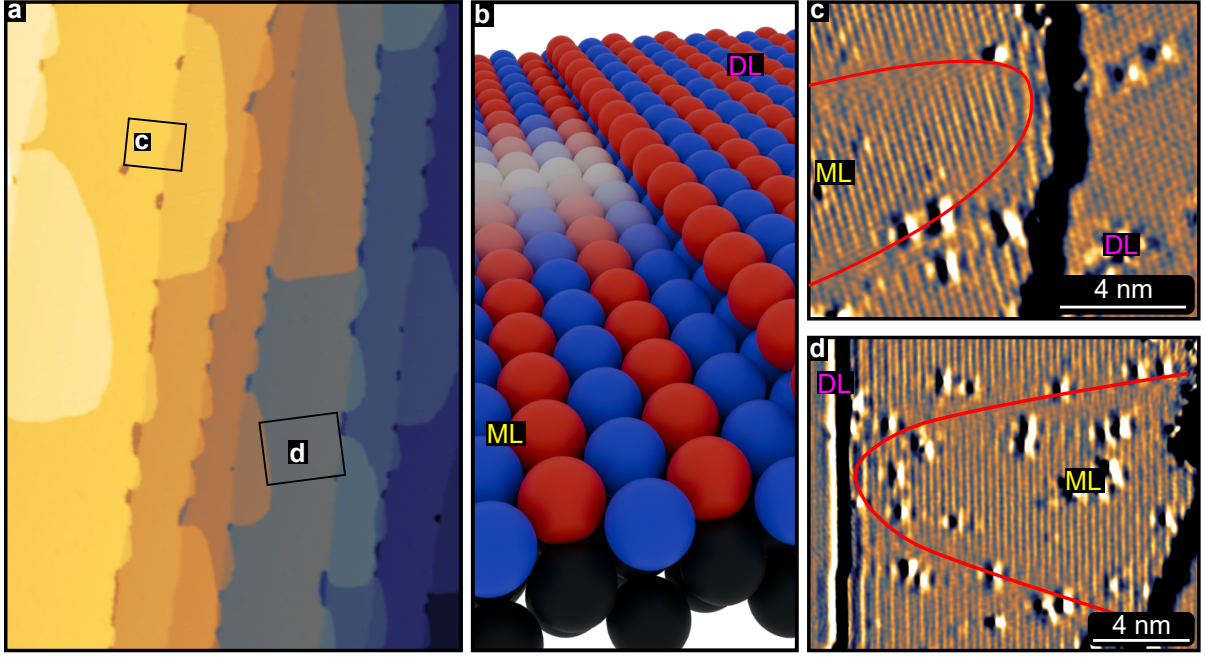


Fig. 35 **a**, Constant-current STM image of a sample with 1.25 ML of Mn on Ta(110). $U = 200$ mV, $I = 300$ pA. **b**, Sketch of a hypothetical magnetic configuration at an edge between a region of ML hosting an AFM spin-spiral (left) and a region of DL hosting a collinear OOP AFM state. The red and blue colours encode the OOP component of the magnetization. **c**, SP-STM current image of a region of Mn ML (left) bordering to Mn DL one step edge below (right). Scan region marked in **a**. $U = -15$ mV, $I = 1$ nA. **d**, SP-STM current image of a region of Mn ML (right) bordering a Mn DL region growing on top (left). Scan region marked in **a**. $U = -15$ mV, $I = 3$ nA. For **c-d** high frequency noise was removed.

One possibility appears to be the complete suppression of the spin-spiral, imposing a purely collinear state on the ML. While this does avoid the unfavourable configurations at the boundary, for larger terrace sizes the energy gained by rotating the spins increases and we expect to find the presence of an AFM spin-spiral for large enough terrace widths.

The overview STM image in Fig. 35a shows almost fully closed Mn ML and DL regions growing from the step edges. In Fig. 35c, near the boundary between ML and DL at a buried step edge we find the energetically unfavourable OOP region to be cut off, with the domain walls connecting to form a half-loop structure, see red line in Fig. 35c. We find these half-loops located all along the boundary. Clearly, the terrace to the left of the DL region and therefore the width of the ML region is large enough to favour the AFM spin-spiral.

Interestingly, we find the same half-loop structure also at a boundary between a ML region and a DL region growing on top (i.e. not across a buried step edge), see Fig. 35d. In both cases the exact magnetic configuration in close vicinity to the boundary is hard to observe because of standing-electron-waves and defects. However, we can speculate that at the end of the half-loop, spins oriented with a component along $[1\bar{1}0]$ likely exist, because one OOP domain transitions to the other along this direction and DMI favours cycloidal rotations.

Fig. 36 shows the spin texture of such a half-loop simulated using micro-magnetic modelling by Tim Drevelow. These simulations will be discussed in more detail in Sec. 5.3.6.

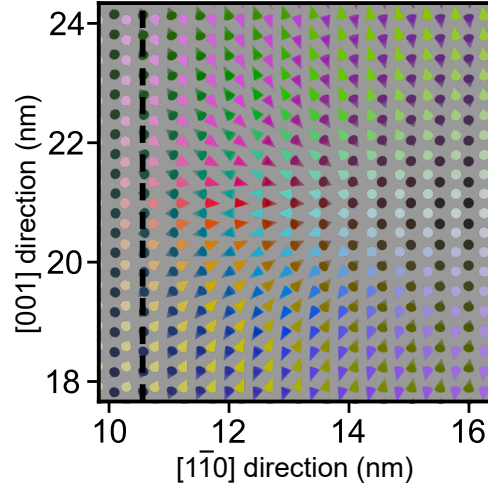


Fig. 36 Sketch of the spin texture of a half-loop simulated using micro-magnetic modelling by Tim Drevelow, Univ. Kiel. Spins are represented as cones with the following colouring: Black (down), white (up), colour (in-plane). The dashed black line marks the boundary between the ML (right) and DL (left).

5.3.4 The phase-relation between DL regions

Naturally we have to ask what happens in the areas where the ML is sandwiched between two DL regions, see for example Fig. 35a around the black rectangle on the bottom right. Here, the ML connects to the DL growing on top of it on the left and also borders to the DL growing on the terrace one step edge down on the right. We have seen that the DL regions impose their collinear AFM state on the Mn ML. What happens when the two DL regions are in-phase or out-of-phase (i.e. impose the same/opposite OOP configuration on the Mn ML)?

It seems likely that during the cool-down of the sample the DL regions establish their magnetic order first. At this point, the coupling between different DL regions should be minimal, allowing the AFM state to arise with two different phase relations to the underlying lattice. Fig. 37a shows a 3D sketch of a ML region sandwiched between two DL regions that are in-phase. The antiferromagnetic alignment is continuous between the bottom layer of the DL on the left, across the ML, to the top layer of the DL on the right. In contrast, Fig. 37b shows the same sketch for an out-of-phase alignment of the DL regions. Here, magnetic frustration arises, highlighted as two rows in the ML being aligned ferromagnetically, which is energetically unfavourable. One can expect that for the in-phase DL alignment a complete suppression of the AFM spin-spiral is favourable at least for ML areas below a certain width.

Fig. 38a shows the sample region around the black rectangle in Fig. 35a, where the domain walls of the spin-spiral, marked by the red lines, form a continuous snake-like structure with half-loops forming at/near the boundary to the DL areas. One can clearly

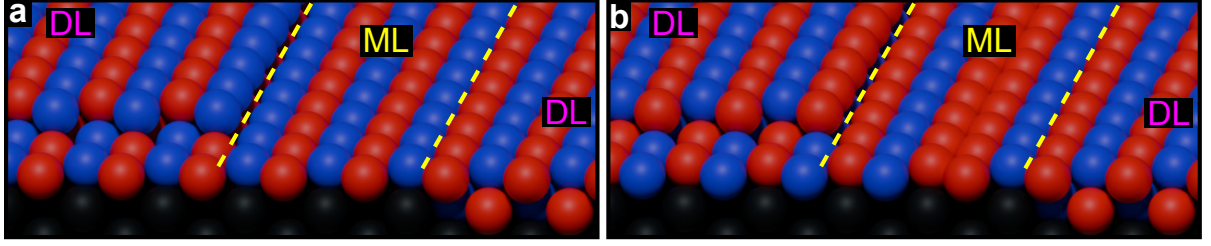


Fig. 37 a-b, Sketches of a ML region sandwiched between two DL regions. The DL region on the left grows on top of the ML, while the DL region on the right grows on the terrace one step edge down (i.e. the ML grows across a buried step edge). The yellow dashed lines indicate the boundary between the ML and DL. The DL regions are in-phase/out-of-phase in **a/b**, respectively. The magnetic state in **b** is purely hypothetical.

see that the DL regions on the left and right favour different OOP regions of the spin-spiral, indicated by the green circles in Fig. 38a, and the unfavourable regions are expelled and cut-off, forming the half-loops, see red circles. This demonstrates that the magnetic states of the DL regions are out-of-phase.

Fig. 38b shows a different sample area, hosting a ML area of larger width. The domain walls, once again indicated by the red lines, connect to form loops. The OOP regions outside the loops are favoured by both of the DL areas on either side, with the unfavourable one being present inside the loops. The much wider terrace allows the domain walls to run exactly along the $[1\bar{1}0]$ direction, unlike the out-of-phase case displayed in Fig. 38a, likely due to the terrace being too narrow.

Additionally, for this much wider terrace the half-loops appear to be located further away from the DL edge, as the favourable domain is visible continuously near the DL edge, see pink line in Fig. 38b,c. The line running along a single atomic row stays on top the bright line in the magnetic contrast; in contrast, in the middle of the terrace a similar line misses the bright lines of the magnetic contrast on the opposite OOP region, clearly indicating the presence of the spin-spiral state in the centre of the terrace, see green line in Fig. 38b,d. The previously observed half-loops were located more closely to the DL edge, possibly because the ML areas were too narrow, making them harder to resolve. For example, in Fig. 38a moving the ends of the half-loops further away from the DL edge would result in the domain walls being oriented less in the $[1\bar{1}0]$ direction, which is energetically unfavourable.

In Fig. 38a-b one can see that the Mn ML is not fully closed and the domain walls preferentially terminate at holes. For domain walls terminating at holes located at a buried step edge between a ML and DL regions, see cyan arrows in Fig. 38a-b, this is favoured because it allows for the avoidance of half-loops, which are energetically more expensive. For the formation of half-loops, wide ML terraces with hole-free boundaries between DL and ML are optimal, as this maximizes magnetic frustration at the ML/DL boundary. As mentioned in Sec. 5.2 the holes in the Mn ML are mostly caused by residual Oxygen impurities on the Ta surface and therefore a cleaner Ta(110) surface could improve the Mn growth and reduce the number of holes.

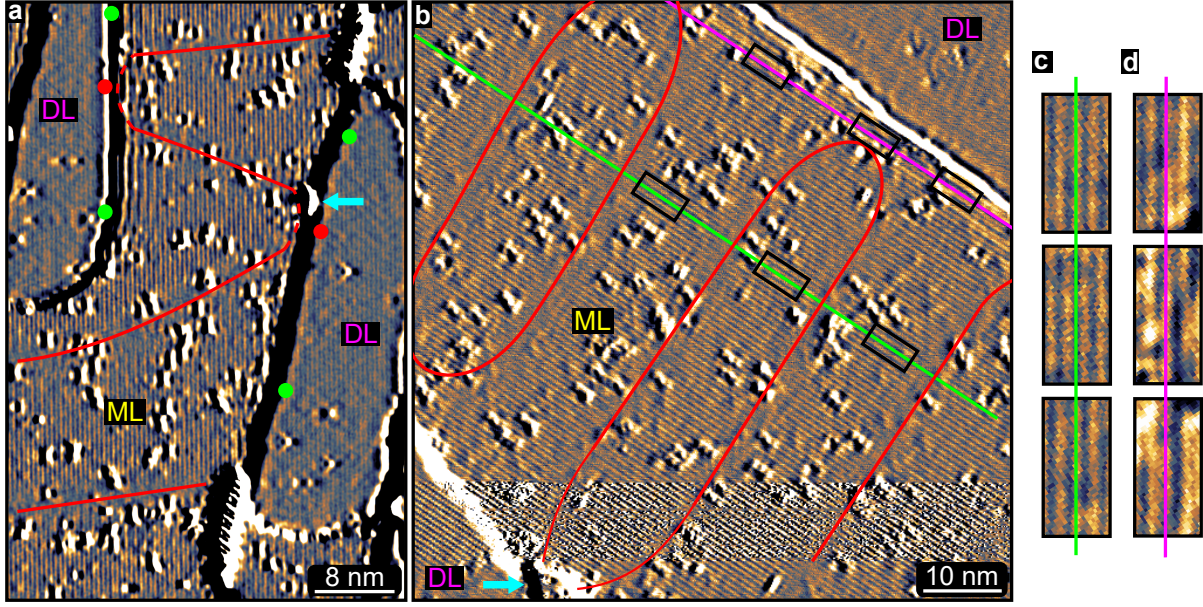


Fig. 38 **a**, SP-STM current image of a sample with 1.25 ML of Mn. The red lines indicate the domain walls. The green/red dots indicate favourable/unfavourable regions at the ML-DL interface. $U = -15$ mV, $I = 1$ nA. High frequency noise was removed. **b**, SP-STM current image of a large Mn ML area with regions of Mn DL at top right and bottom left. The red lines indicate the domain walls. $U = -10$ mV, $I = 1$ nA. **c-d**, Zoom-ins of the black rectangles on the green/pink line in **b**. All images were measured at 4.2 K.

5.3.5 The relation of the Mn ML width and the magnetic state

We will now investigate the effect of the width of the Mn ML areas on the magnetic state. The sketches in Fig. 39a-c show the magnetic state for a Mn ML sandwiched between two in-phase DL regions, with the width of the Mn ML decreasing from a to c. As the width becomes smaller, the loops transform from approximately elliptical to approximately spherical. The experimentally observed magnetic state in Fig. 38b corresponds to Fig. 39a.

The sketches in Fig. 39d-f show the magnetic state for a Mn ML sandwiched between two out-of-phase DL regions. As the width of the Mn ML region decreases, the domain walls marked by the red lines no longer run perfectly horizontal in the middle of the ML area. The experimentally observed magnetic state shown in Fig. 38a corresponds to Fig. 39e.

We previously mentioned how during the cool-down of the sample from the preparation temperature to the measurement temperature, the OOP AFM state can arise in two phase-shifted configurations on the underlying lattice. For completely isolated DL regions, a statistical distribution of 1 : 1 for in-phase/out-of-phase configurations of DL areas would be expected. Experimentally we find no such distribution locally, instead nearby DL regions are more likely to be in-phase than out-of-phase. The two main contributing factors we can determine to be relevant are the size of the DL islands and the separation between the DL islands (i.e. the width of the ML area).

In Fig. 40a-b a thin Mn ML area is adjacent to a large region of DL at the bottom

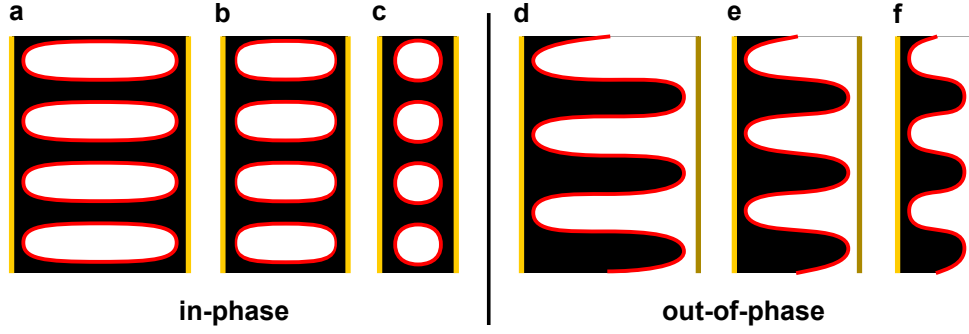


Fig. 39 **a-c**, Sketch of the magnetic state in a Mn ML sandwiched between two in-phase DL regions. **d-f**, Sketch of the magnetic state in a Mn ML sandwiched between two out-of-phase DL regions. **a-f**, The red lines mark the domain walls. White and black coloured regions indicate the two possible phase-configurations of the OOP ML areas.

and to two smaller DL islands growing from the step edges at the top. With the red line indicating the only visible domain wall, it becomes clear that the rotation of the spins in the ML is completely suppressed and the AFM state in the small DL regions is in-phase with the large DL region at the bottom. With decreasing width of the Mn ML regions, the energy gained by the spin-spiral state decreases relative to the energy cost at the boundary to the DL. The formation of magnetic states with nearly circular loops, see sketch for the in-phase configuration displayed in Fig. 39c, appears to be energetically unfavourable and is not found experimentally.

A complete suppression of the spin-spiral was almost always observed for such ML terraces with such a small width, which may indicate that at some point during the cool-down process that coupling between neighbouring DL regions becomes strong enough to still allow for an inversion in the magnetization of small islands at the given temperature.

Exceptions to this appear if domain walls can be positioned with low energy due to holes in the ML, see red line on the right side in Fig. 40c-d, which demonstrates a domain wall allowing for an out-of-phase configuration of the bottom DL and top right DL. The overall energy of the domain walls depends on multiple factors, mainly the length and orientation of the domain wall. However, both the length and orientation depend on the width of the terraces, the angle of the step edges, and the position of ML edges not bordering DL areas (i.e. step edges to the Ta).

The domain wall in Fig. 40d is oriented mainly along the $[1\bar{1}0]$ direction, which is the energetically optimal direction. If the direction of the step edges was different, the same domain wall would be oriented less along the $[1\bar{1}0]$ direction and would therefore be less favourable. We expect that such a difference may force the bottom and top right DL regions in Fig. 40d to be in-phase during sample cool-down.

A similar effect could be seen if the edges between the Mn ML and the Ta, where the domain wall can attach, are spaced further apart (horizontally), because the angle of the domain wall would be less favourable and overall longer. This can be seen in Fig. 40a-b, where the holes in the Mn ML, marked by white arrows, are spaced far apart and a domain wall running between the two holes would not be oriented along the $[1\bar{1}0]$ direction, but more along the $[001]$ direction.

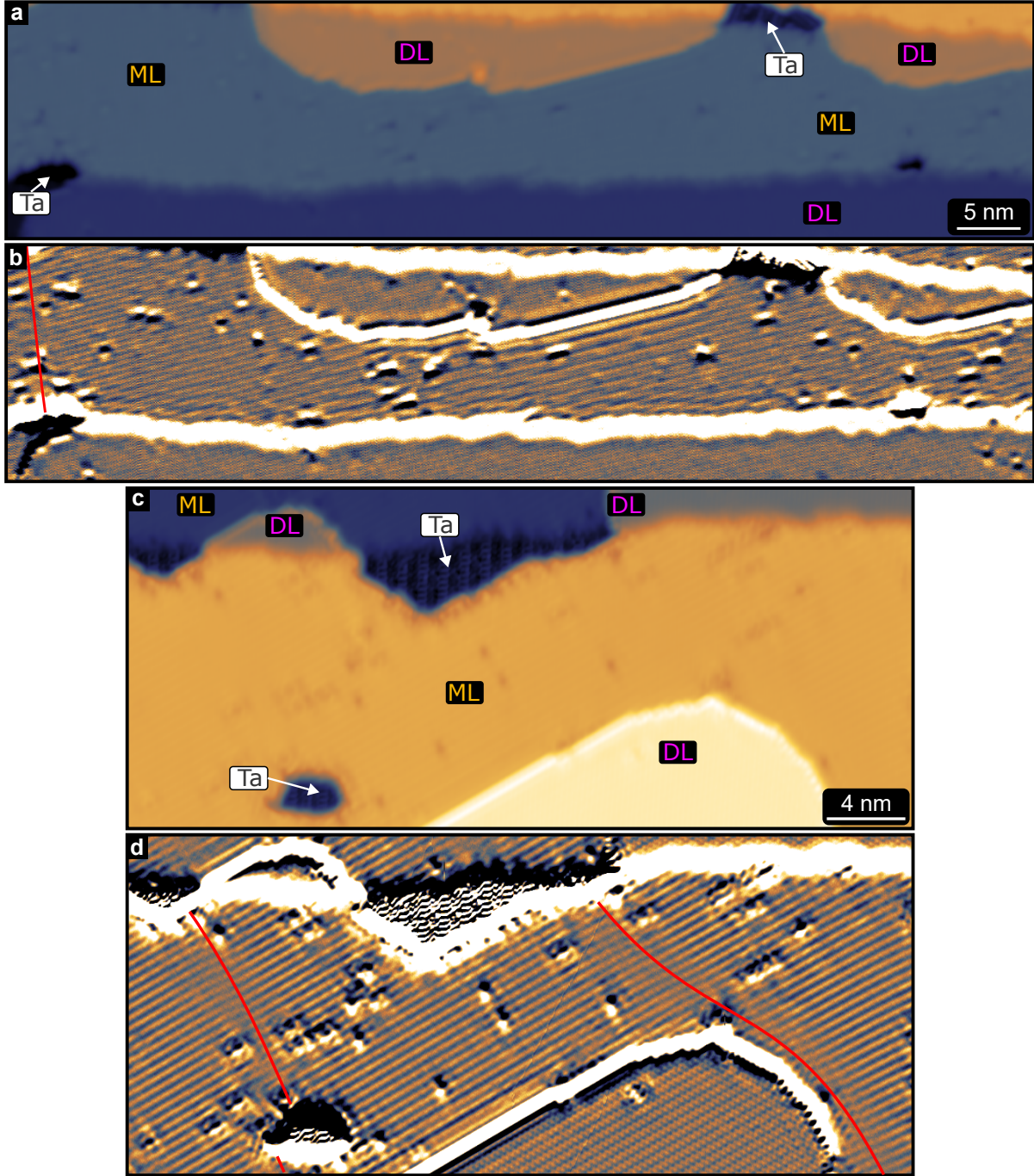


Fig. 40 **a-b**, Topography image and spin-resolved current map of a ML region sandwiched between a large DL region at the bottom and two smaller DL regions at the top. The red line marks a domain wall. $U = -15$ mV, $I = 1$ nA. **c-d**, Topography image and spin-resolved current map of a ML area with one DL region at the bottom and two DL regions at the top. The red lines mark domain walls. $U = -15$ mV, $I = 990$ pA. all: $T = 4.2$ K.

Even between larger DL regions, where the width of the ML stripe between them is < 20 nm, we find a complete suppression of the spin-spiral. However, this means, that during cool-down, the coupling between these islands becomes large enough to prevent

out-of-phase alignment. Only for larger separations between large DL islands (i.e. wider ML terraces) can an out-of-phase configuration be found.

Our experimental findings indicate that the magnetic state sketched in Fig. 39f does not exist because an out-of-phase configuration of two large DL areas with such a small Mn ML area between them is not possible. Furthermore, the magnetic state sketched in Fig. 39c also does not exist for such small Mn areas because it is too energetically unfavourable

5.3.6 Micro-magnetic modelling of the loops

To better understand the structure of the magnetic state around the half-loops, micro-magnetic simulations were performed for one of the sublattices, with parameters derived from the DFT calculations. Simulating the large number of discrete atomistic spins necessary to represent this spin-spiral state is computationally too expensive. Micro-magnetic simulations model a continuous magnetization density, which requires a separation of the AFM spin-spiral into two ferromagnetic sublattices. The separation can be applied here because the antiferromagnetic alignment of the sublattice and the rotation of the spin-spiral occur on two different length scales. The in-phase/out-of-phase behaviour of the DL regions can be modelled by two ferromagnetic regions on the left and right aligned parallel/anti-parallel, respectively.

The result of the simulation is shown in Fig. 41a-b. For both cases the AFM spin-spiral develops in the centre of the ML region, with the DMI providing a rotational sense and the magnetic anisotropy selecting the plane of rotation.

Near the boundaries to the DL regions, half-loop structures emerge, where the unfavourable OOP region of the spin-spiral vanishes and the domain walls (in-plane regions) connect to form half-loops, see also Fig. 41d-e. Here, the former figure shows the moments at the atom position of one sublattice, derived from the continuous magnetization density simulated using the micro-magnetic model. The spin texture in the latter figure is recovered by inverting the magnetic moments of the other sublattice. The magnetization at the ends of the half-loop points along the $[1\bar{1}0]$ direction avoiding any discontinuities in the magnetization, with the rotational sense and direction determined by the sign of the DMI. The magnetic state at the position of the half-loop looks similar to half a magnetic skyrmion on one sublattice, see Fig. 41d.

We find that for in-phase DL arrangement the half-loops appear to form elongated skyrmions, see Fig. 41a. Calculations of the topological charge density indicate that each half-loop holds a topological charge of $-\frac{1}{2}$, resulting in a combined topological charge of -1 for each loop, while a conventional circular skyrmion typically holds the same topological charge but it is located in the centre of the skyrmion instead. For the out-of-phase arrangement, we find a continuous winding domain wall instead of individual loops, see Fig. 41b, with the topological charge on the left and right being of opposing sign, resulting in a total topological charge of 0.

When considering two antiferromagnetically aligned sublattices instead of a single ferromagnetic sublattice, the resulting total topological charge of the system always cancels, as inverting the magnetization also inverts the sign of the topological charge. Overall, the micromagnetic simulation can reproduce the two cases that we observe experimentally.

The half-loops emerge due to frustration at the boundary between the OOP collinear antiferromagnetic state in the DL and the non-collinear AFM state in the ML. Depending on the phase between the DL regions, we find the formation of an array of loops, which can be understood as AFM skyrmions, or the formation of a single topologically trivial winding domain wall.

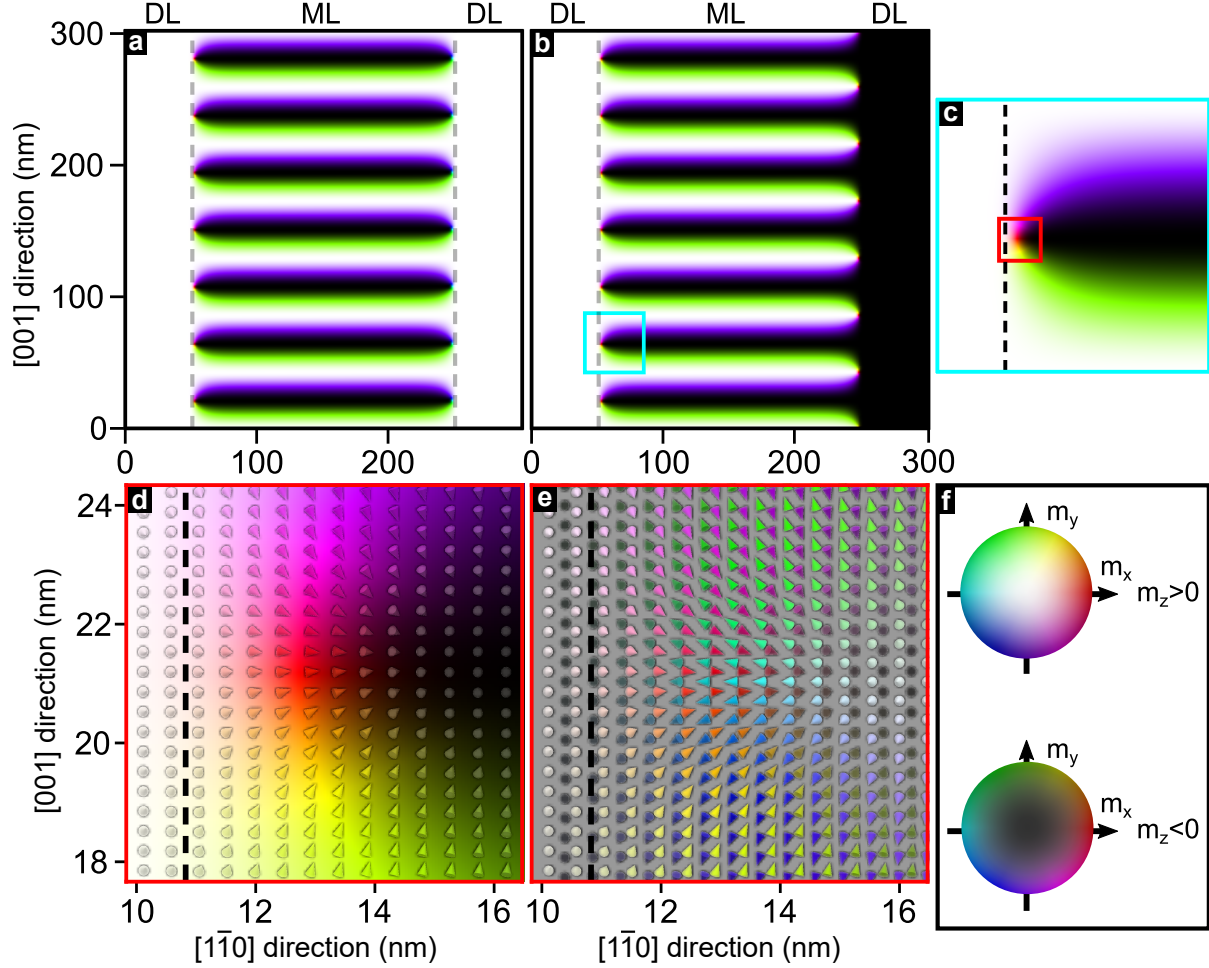


Fig. 41 a-c, Colour coded magnetization density obtained from a micro-magnetic simulation. **f**, Colour code used for the magnetization density maps. **a-b**, Simulated magnetization density for parallel/anti-parallel alignment of the magnetization in the DL regions, representing in-phase/out-of-phase DL regions, respectively. **c**, Magnetization-density of the region marked by the cyan rectangle in **b**. **d**, Magnetization density of the region marked by the red rectangle in **c**. The cones show the magnetization direction probed at the atomic positions. **e**, Atomistic magnetic moments recovered from the data in **d** by inverting the magnetization of the other sublattice. **a-e**, The dashed lines mark the boundary between ML and DL. Figure adapted from our manuscript, see [Publications](#), based on the micro-magnetic simulations by Tim Drevelow, Univ. of Kiel.

To find AFM skyrmions experimentally, both boundaries between the DL and ML regions need to be approximately oriented along the propagation axis of the AFM spin-spiral, which typically means, at least for step-flow growth, that the step edge needs to

run approximately parallel to the propagation axis or the $[001]$ direction in this system. Additionally, a two-fold rotation axis is required to define the propagation direction of the spin-spiral, here provided by the low-symmetry $\text{bcc}(110)$ surface, and a broken inversion symmetry to allow for the DMI to impose a unique rotational sense, always present at surfaces.

Lastly, let us compare an SP-STM simulation of the half loops with our experimental data. Fig. 42a-b shows two SP-STM current maps of a half-loop measured with different tip magnetization, with the estimated tip magnetization, derived via a comparison to SP-STM simulations, shown in the sketches above each image.

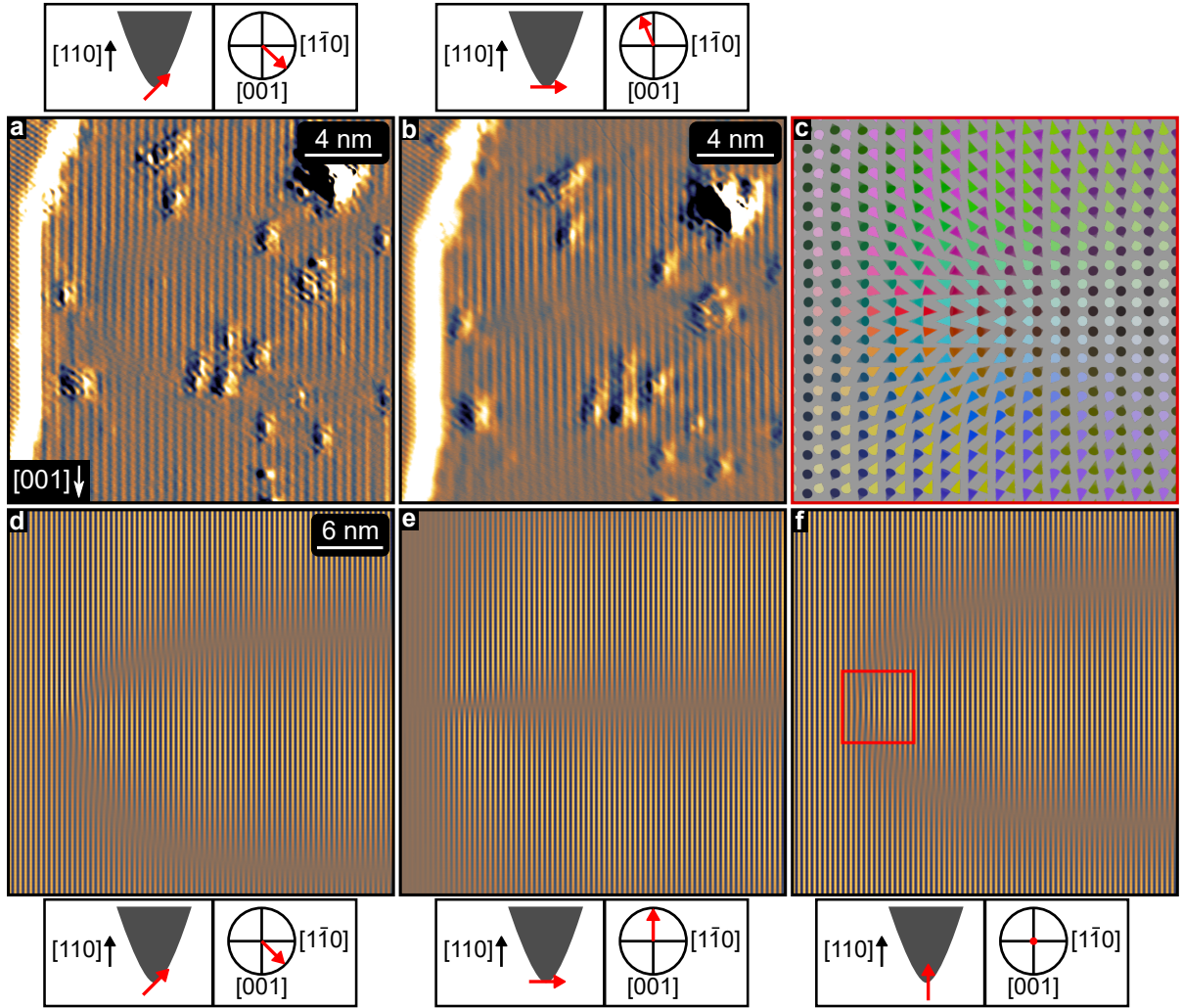


Fig. 42 a-b, SP-STM current maps of a Mn ML region bordering the Mn DL across a buried step edge on the left, measured with the estimated tip magnetization shown above the image, respectively. $U = -15$ mV, (a): $I = 2$ nA, (b): $I = 4$ nA, $T = 4.2$ K. **c**, Sketch of the spin texture of a half-loop simulated using micro-magnetic modelling. **d-f**, Simulated SP-STM images using the spin-configuration obtained from the micro-magnetic modelling. The area in red rectangle in **f** corresponds to the spin texture in **c**. Micro-magnetic simulations done by Tim Drevelow, Univ. Kiel.

In Fig. 42b the tip magnetization is oriented mostly IP, as we hardly observe any magnetic contrast away from the position of the half-loop. Additionally, the contrast on the DL (left side) is purely structural with no magnetic component. We find good agreement with a simulated SP-STM image using an IP tip with the magnetization oriented along the $[001]$ direction, see Fig. 42e. Due to a small asymmetry in the magnitude of the experimental magnetic stripe contrast near the boundary to the DL of the domain walls forming the half-loop, we can conclude that the experimental tip magnetization must have a small component in the $[1\bar{1}0]$ direction but is mainly oriented in the $[001]$ direction.

In Fig. 42a the tip magnetization has both significant out-of-plane and in-plane components because a comparison to the in-plane magnetized tip in Fig. 42b shows no complete inversion of the regions that do/don't show magnetic contrast. Comparing the simulated SP-STM images for an IP and OOP tip, see Fig. 42e-f, shows such an inversion of contrast regions. A tip magnetization with both an OOP component and an IP component in the $[001]$ direction, shows a lateral shift of the regions without magnetic contrast, see Fig. 42d. Such a shift, relative to a completely IP or completely OOP tip, is also observed experimentally, compare the position of the hole in the Mn ML relative to the position of magnetic contrast in the half-loop in Fig. 42a,b. We can conclude that the areas of vanishing magnetic contrast in the normal spin-spiral (i.e. right side of Fig. 42a) do not correspond to the domain walls (in-plane areas) but some canted quantization axis instead. Additionally, we observe a strong asymmetry in the half-loop near the boundary to the DL. In the top half, we observe magnetic contrast in the IP regions, while we do not observe any contrast in the bottom half of the half-loop, indicating a significant magnetization component in the $[1\bar{1}0]$ direction, compare Fig. 42a and Fig. 42d.

Even though the micro-magnetic model is simple, as it uses an easy-plane anisotropy in the $[110]$ - $[001]$ -plane and it does not consider holes and defects in the Mn ML, the agreement between experimentally observed half-loops and SP-STM simulations using the micro-magnetic model is good.

5.4 Properties emerging from superconductivity

In the previous section, we have investigated the magnetic states of the Mn ML and DL on Ta(110), now we turn to the interaction between these magnetic states and the superconducting substrate.

5.4.1 The superconducting gap of the Mn ML and DL

We start by investigating the density of states around the Fermi energy using tunneling spectroscopy measurements at a temperature of 1.3 K. Here, the STM tip is first positioned and stabilized at the sample region of interest, the feedback loop is disabled, and then the bias voltage is swept through the desired voltage range while the current and the dI/dU signal are recorded.

We perform these spectroscopy measurements on the bare Ta substrate, the Mn ML, and the Mn DL, see coloured crosses in Fig. 43a, and the dI/dU signal measured in the range from -4 to $+4$ mV is displayed in Fig. 43b. The modulation voltage for recording the dI/dU signal has been set to $U_{\text{mod}} = 50\mu\text{V}$ for all point spectra and zero bias dI/dU -maps that are shown in this chapter. This modulation amplitude is small enough to not wash out energetic features inside the superconducting gap while still providing a decent signal-to-noise ratio, see also methods in Sec. 3. We observe the characteristic drop in the density of states on the Ta substrate (grey curve) around the Fermi energy expected for a superconductor. Due to thermal broadening at the measurement temperature of 1.3 K and Tantalum's relatively low critical temperature of $T_c = 4.48$ K, the spectrum does not show sharp coherence peaks or a drop to zero intensity in the centre of the gap. As we are using a Cr-bulk tip (i.e. a non-superconducting tip), the position of the coherence peaks is in agreement with the expected value of $\Delta(T = 0) = 0.72$ meV found in the literature [138].

We find a similar drop in the dI/dU signal for spectra taken on the Mn ML and DL, see orange and violet curves in Fig. 43b, indicating that both of these layers are also superconducting due to the proximity effect of the Ta substrate. The increased dI/dU signal inside the superconducting gap for the magnetic layers demonstrates the presence of in-gap states which are expected to arise due to the interaction of magnetism and superconductivity and the locally reduced energies to break Cooper pairs, see Sec. 2.4.4 for more information.

Due to particle-hole symmetry, these in-gap states appear symmetrically around the Fermi energy; however, their intensities can be different, resulting in an asymmetry of the LDOS also visible in the spectra on the ML and DL. This asymmetry is more pronounced for the spectrum on the DL, here the left coherence peak seems to be shifted towards the Fermi energy due to the in-gap states close to it. The right coherence peak seems to be unaffected by the in-gap states, indicating a strong discrepancy in the intensity of the states above and below the Fermi energy. We expect that the difference between the ML and DL spectra is mostly related to differences in the material properties.

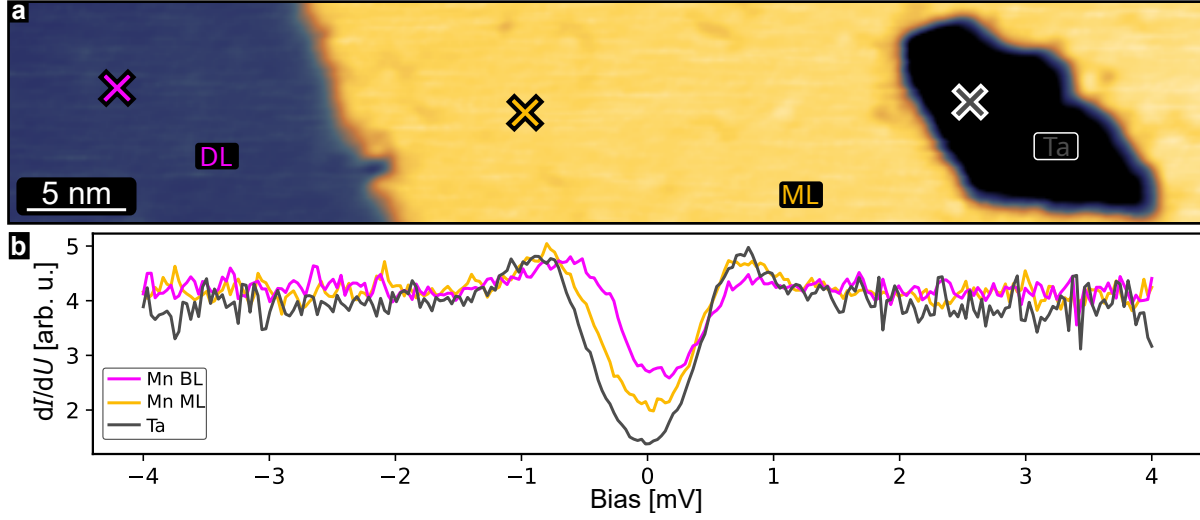


Fig. 43 **a**, STM constant-current image of 1.25 ML of Mn on Ta(110). On the left the Mn grows across a buried step edge, while a hole in the ML reveals the bare Ta substrate. $U = -15$ mV, $I = 5$ nA. **b**, Individual spectroscopy measurements at 1.3 K taken at the positions marked by the coloured crosses in **a**. Stabilization bias $U_s = 4$ mV, stabilization current $I_s = 1$ nA, bias modulation $U_{\text{mod}} = 50 \mu\text{V}$; all: Cr bulk tip.

5.4.2 The in-gap states of IP vs OOP ML regions

Previously, oscillations in the LDOS inside the superconducting gap of a MSH system hosting a ferromagnetic spin-spiral have been observed with both the full spiral period and half the period [30]. The oscillation with the spin-spiral period was attributed to the spin-polarization of the in-gap states, while the oscillation with half the period was attributed to an oscillation in the Rashba SOC caused by the non-collinear spin structure. It was demonstrated that the LDOS in the superconducting gap can be different for OOP magnetized and IP magnetized regions.

To investigate if a similar effect exists in the system of Mn/Ta(110), we measured 60 evenly spaced spectra on the Mn ML hosting the AFM spin-spiral along the 30 nm long red line in the [001] direction, see Fig. 44a. The spectra were positioned to be taken along an atomic row in the [001] direction; however, due to thermal drift over the measurement duration of 3.5 hours, the sample drifted approximately 0.9 nm, which is why the red line is not completely vertical. The energy-resolved dI/dU signal of these spectra is plotted as a waterfall plot in Fig. 44b. We observe no visible modulation in the LDOS along the line, which spans over more than one spin-spiral period and therefore crosses over multiple IP and OOP regions. The spectra marked by the green/cyan arrows deviate from the rest. This can be attributed to defects indicated by a similar green/cyan arrow in the topography image. We can conclude that the LDOS in the superconducting gap is similar in the IP and OOP regions. Lower measurement temperatures or the use of a superconducting tip can improve the energy resolution and may reveal differences. However, with a SC tip a direct spatial correlation with the magnetic state is not possible.

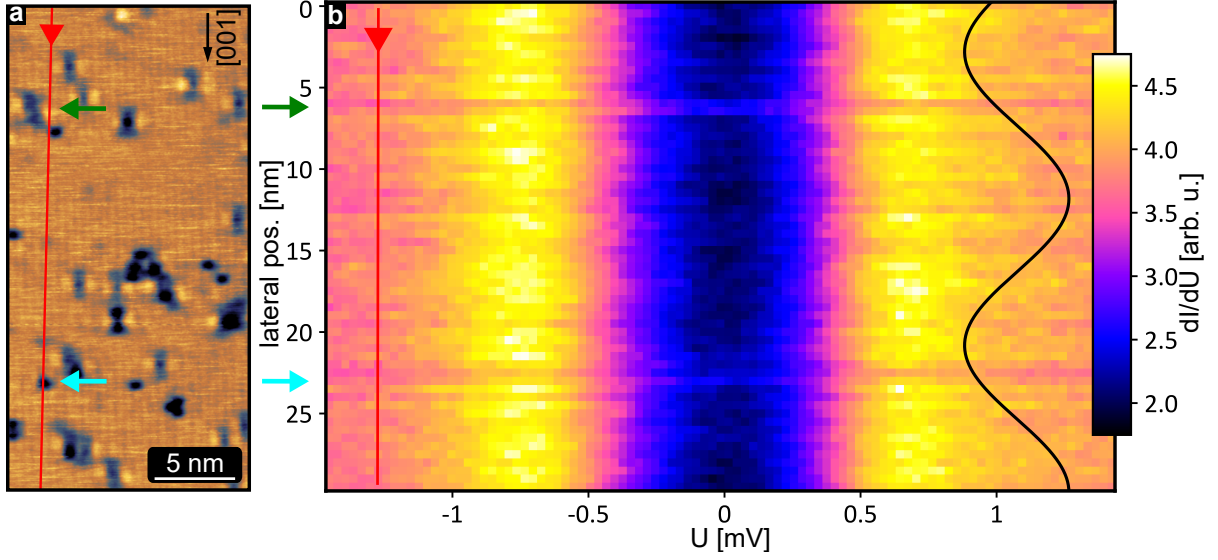


Fig. 44 **a**, SP-STM image of a Mn ML regions hosting an AFM spin-spiral with a period of ≈ 18.5 nm. $U = 4$ mV, $I = 1$ nA. **b**, Waterfall plot of 60 dI/dU spectra taken along the 30 nm long red line shown in **a**. The green and cyan arrows indicate spectra taken near defects marked by similar arrows in **a**. The black curve indicates the period of the spin-spiral. All spectra: stabilization bias $U_s = 4$ mV, stabilization current $I_s = 1$ nA, bias modulation $U_{\text{mod}} = 50 \mu\text{V}$. all: $T = 1.3$ K, Cr bulk tip.

5.4.3 The Mn ML/Ta edge mode

We have discussed that the interaction of magnetism and superconductivity can not only lead to states inside the superconducting gap, but also to topological superconductivity, see Sec. 2.4.4. Interesting physics emerge at the boundary between two topologically distinct regions, typically resulting in edge modes. In our system of Mn-layers on Ta(110) we have three distinct sample regions: the Ta substrate, the Mn ML and the Mn DL. We start by investigating the boundary between the Mn ML and the topologically trivial elemental superconducting Ta substrate. For this purpose, we grow a sub-ML coverage of Mn on Ta(110) at room temperature, which allows for the formation of isolated Mn islands.

Fig. 45a shows an STM topography image of such a Mn ML island, with some straight edges running along high-symmetry directions on the bottom right. In general, we find straight edges for the Mn ML to be relatively uncommon, with most edges appearing similar to the left side of the islands.

Due to the small size of the islands, we observe a collinear AFM state, instead of the AFM spin-spiral observed in the extended ML, see Fig. 45b. Although we have observed no significant differences in LDOS around the Fermi energy for IP and OOP regions of the spin-spiral, the simple collinear state further simplifies the investigation of possible edge modes between the Mn ML and Ta substrate and serves as a good starting point.

Fig. 45c shows a zero bias dI/dU -map of the same Mn ML island measured with a non-magnetic tip using the multipass measurement mode. For details on this measurement mode and its differences to constant-height mode, see Sec. 2.1.3. We find an increased

dI/dU signal, which indicates a higher LDOS, at the edges of the ML region and therefore the presence of an edge mode. This edge mode appears around the entire ML island, which is reminiscent of the edge mode between a fully-gapped topological superconducting system and the trivial substrate [29]. However, one would typically expect a collinear antiferromagnet to show topologically nodal point superconductivity instead, due to a collinear antiferromagnet's effective time reversal symmetry, see also Sec. 2.4.4.

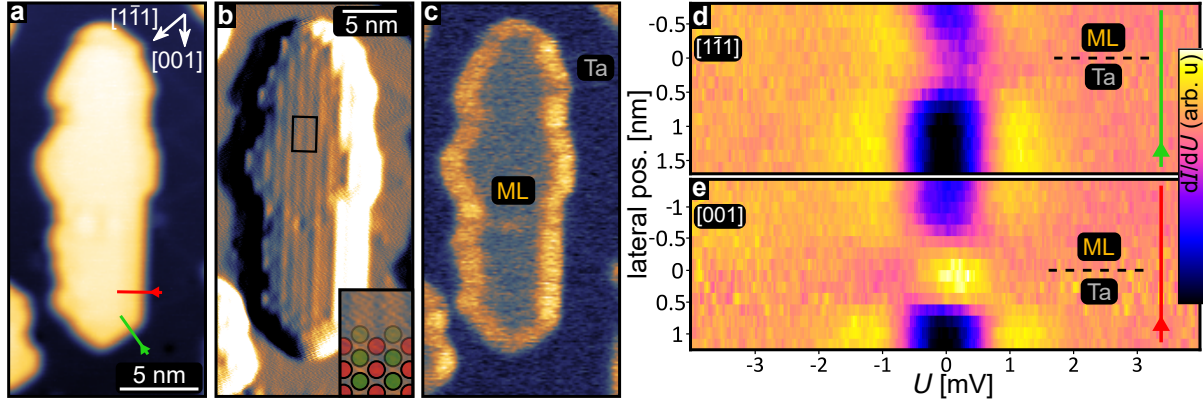


Fig. 45 **a**, STM constant-current image of an island with several straight edges running along high symmetry directions. $U = +4$ mV, $I = 1$ nA. **b**, SP-STM current map of the same island using a spin-polarized tip; inset shows the row-wise AFM state. $U = +5$ mV, $I = 1$ nA, $T = 4.2$ K. **c**, Zero bias dI/dU -map measured in multipass mode. $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$. Measured at the tip-sample distance obtained in **a**. **d-e**, Waterfall plots of dI/dU spectra taken along the green/red lines in **a**, depicting the spatially- and energy-resolved dI/dU signal across the $[1\bar{1}0]/[001]$ edges, respectively. Each spectrum was normalized to the y-intercept of a line fitted to the data outside the gap. All spectra: stabilization bias $U_s = 4$ mV, stabilization current $I_s = 1$ nA, bias modulation $U_{\text{mod}} = 50 \mu\text{V}$. **a,c-e**: $T = 1.3$ K all: Cr bulk tip.

We investigate the edge mode for the two edges oriented along the $[001]$ and $[1\bar{1}1]$ directions by measuring multiple spectra along the red and green lines in Fig. 45a, respectively. The dI/dU spectra moving from Ta to the Mn ML across the edge are plotted as two waterfall plots, see Fig. 45d-e. In each plot, the spectra taken on the bare Ta are plotted at the bottom and the ML at the top, with the edge between them roughly marked by the dotted black line. The additional in-gap states caused by the magnetism in the ML are visible as increased signal intensity inside the gap compared to Ta.

When looking at the differences between the two plots, it becomes clear that close to the Fermi energy at the position of the edge a bright yellow peak in the dI/dU signal is visible at the $[001]$ edge, while the $[1\bar{1}1]$ edge shows no such peak, instead only a slightly increased signal intensity compared to the ML island interior is visible. We see an identical behaviour of the edge mode for a different sample area where a Mn ML region has well developed edges in all three high symmetry directions, see Fig. 46a.

A similar zero bias dI/dU -map shows the same increased LDOS at the $[001]$ edge with lower LDOS at the $[1\bar{1}1]$ and $[1\bar{1}0]$ edges, see Fig. 46b. The spectra on the Ta substrate, the Mn ML, and the different edges are shown for the ML island in the previous Fig. 45a, and this ML region in Fig. 46e/f, respectively. These two ML regions are located on two different samples and were measured with two different micro-tips. One can see that the

LDOS inside the gap matches quite well, with the strongest edge mode intensity at the $[001]$ edge (red), followed by the $[1\bar{1}1]$ edge (green), and then the $[1\bar{1}0]$ edge (cyan). All three edges show increased intensity compared to the Mn ML interior (violet).

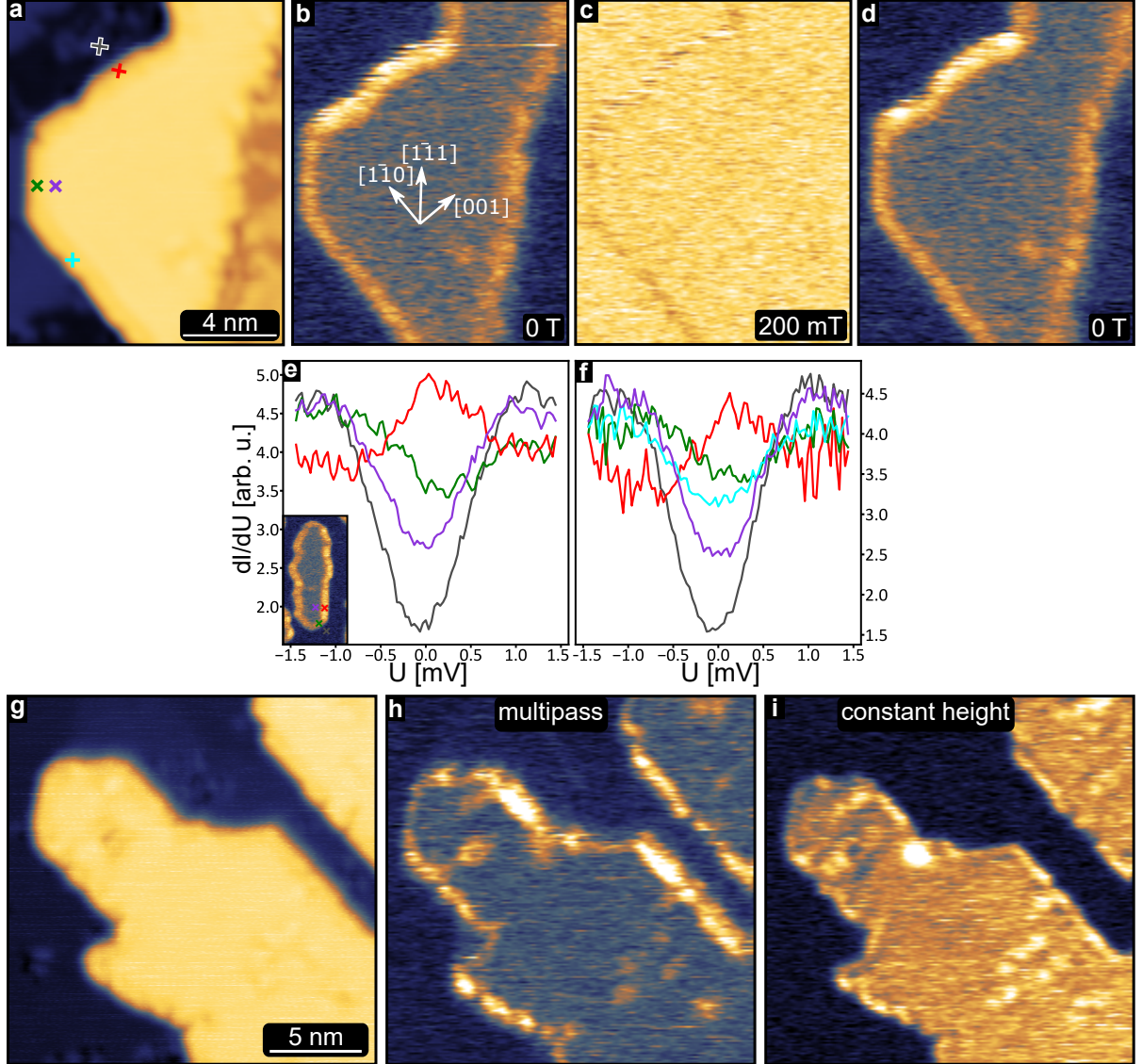


Fig. 46 **a**, STM topography image of a Mn ML island growing from the Ta step edge. Three well-developed edges running along the high symmetry directions are visible. **b-d**, A magnetic field dependent series of zero bias dI/dU -maps measured in multipass mode by tracing the topography from **a**. Field values: 0 mT $\rightarrow$ +200 mT $\rightarrow$ 0 mT. **e-f**, Point spectra measured on the Ta (grey), the Mn ML (purple), the $[001]$ edge (red), the $[1\bar{1}1]$ edge (green), and the $[1\bar{1}0]$ edge (cyan) at the positions marked by the correspondingly coloured cross in **a** or the inset in **e**. **g**, STM topography image of a Mn ML region growing from an edge. **h-i**, Zero bias dI/dU -maps measured in multipass mode by tracing the topography from **g** with an offset of -140 pm and constant-height mode, respectively. Measurement parameters: a-d: $U = +4$ mV, $I = 1$ nA, $U_{\text{mod}} = 50 \mu\text{V}$; e-f: stabilization bias $U_s = 4$ mV, stabilization current $I_s = 1$ nA, modulation bias $U_{\text{mod}} = 50 \mu\text{V}$; g: $U = -40$ mV, $I = 1$ nA; h-i: $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$.

At the boundary between the Mn ML and Ta across the buried step edge, we also observe an increased density of states, see right side in Fig. 46b, indicating an edge mode as one would expect for a transition between a topological superconducting state and the Ta substrate. This boundary is typically not oriented along a high symmetry direction and is never observed as perfectly straight, unlike the edges on the left side in Fig. 46a.

When quenching the superconductivity using an external magnetic field of 200 mT, the edge contrast in the dI/dU -map vanishes and returns upon switching off the magnetic field, see Fig. 46b-d, which demonstrates that the observed edge modes are indeed connected to the superconductivity and not a feedback artefact.

Due to a possibility of feedback artefacts in multipass mode, one typically measures dI/dU -maps in constant-height mode; however, interestingly for the ML, the intensity of the edge mode is strongly diminished compared to the multipass mode, see Fig. 46g-h, only the edge mode at the brightest [001] is barely visible. Two explanations seem possible for this effect: Firstly, the edge mode may be located more in the Ta below, which the multipass mode can resolve but the constant-height mode cannot. This is because the tip follows the contour of the surface in multipass mode and comes closer to the Ta substrate. The same considerations apply to the edge mode at the buried step edge because we find the Mn ML to be structurally higher. The Mn ML shows a larger apparent height than a step edge on the Ta substrate for large range of different bias voltages of $\approx 70 - 120$ pm, see Fig. 46a. Secondly, the decay of the states into the vacuum may be different vertically and laterally and similarly in multipass mode the tip may be able to resolve such a signal more strongly.

Overall, the edge mode of the Mn ML to the Ta has the characteristics of a nodal point superconductor, as the edge modes at different crystallographic edges appear with different intensity. From the fact that edge modes are present at all three edges, we can constrain the nodal point configuration. This is further supported by tight-binding calculations predicting a large phase space of nodal-point superconductivity for antiferromagnets [102], as well as by the similar experimental system of Mn/Nb(110), which hosts the same magnetic state, being identified as a nodal point superconductor [100]. As will be discussed in more detail in Sec. 5.4.8, tight-binding calculations performed in the context of this work can nicely reproduce our experimental findings assuming TNPSC in the Mn ML.

5.4.4 Mn ML/Ta edge mode for non-collinear magnetic textures

Next, we investigate the edge mode at locations where the spin-spiral is not completely suppressed. We have already seen that in-gap states in the centre of the ML island are similar between IP and OOP regions, however, the edge mode may behave differently. In Fig. 47a, a Mn ML island is shown, which is large enough to allow for half a period of the spin-spiral to exist, see the SP-STM current map in Fig. 47b.

Without the presence of the OOP collinear AFM state of the Mn DL as a reference, a determination of the tip-magnetization is more challenging. However, we know that the tip magnetization has a significant OOP component, because we observe magnetic contrast on smaller DL islands, where the spin-spiral rotation is suppressed and a collinear OOP AFM state is present. The regions where we lose the magnetic contrast, see red line in Fig. 47b, correspond to a quantization axis pointing perpendicular to the tip magnetization vector

and do not necessarily correspond to the in-plane regions. Nevertheless, the discrepancy between the region marked by the red line and the IP region is expected to be small.

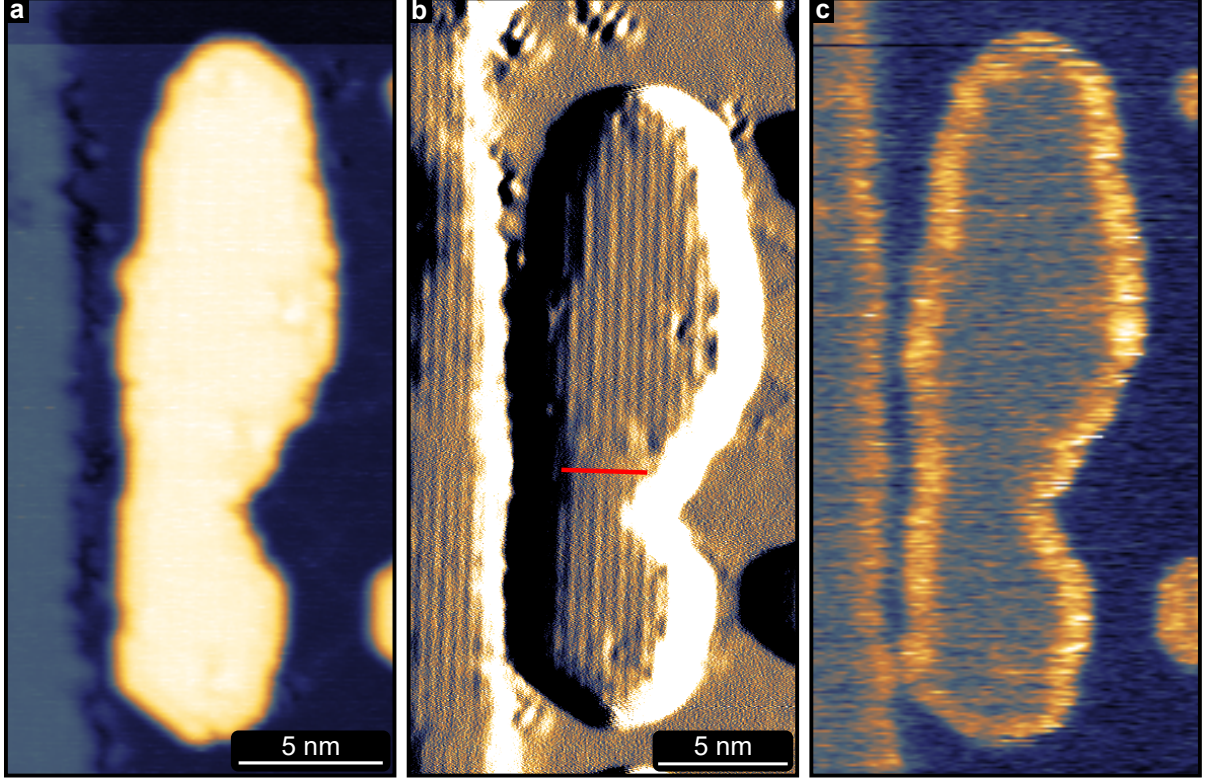


Fig. 47 **a**, STM topography image of a Mn ML island. **b**, SP-STM current map of the same island as in **a**, measured with a spin-polarized tip. The red line marks a region of non-contrast corresponding approximately to an in-plane region. **c**, Zero bias dI/dU -map of the same island as in **a**, measured using multipass mode by tracing the topography in **a**. Parameters: **a**: $U = +4$ mV, $I = 1$ nA; **b**: $U = +5$ mV, $I = 1$ nA, $T = 4.2$ K; **c**: $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$; **a,c**: $T = 1.3$ K, Cr-bulk tip.

The zero bias dI/dU -map of the same island is shown in Fig. 47c and we once again observe the edge mode at all the edges; however, there is no evidence of any additional modulation in the intensity of the edge mode at the edges near the IP region, even for some of the straight [001] edges on the left side of the island. The variations in the edge mode due to the structure of the edge appear to be significantly larger than any variations induced by the difference of the IP and OOP regions.

There are various factors possibly contributing to the explanation as to why we do not observe any differences in the edge mode for IP and OOP regions: Firstly, the energy resolution of our measurement might not be sufficient to resolve any small changes between IP and OOP regions. Secondly, the variations in the edge mode due the structure/orientation of the edges may be significantly larger than the one induced by the IP and OOP regions; thirdly, a spin-polarized tip may be required to resolve these changes. However, even though we have measured multiple other zero bias dI/dU -maps of other ML islands hosting IP regions, some even with weakly spin-polarized tips, we have not

observed any modulation in the edge mode intensity that could clearly be attributed to the change from OOP to IP spins.

A way to disentangle structural contributions to the edge mode from the impact of the quantization axis, would be the investigation of longer straight edges along the [001] direction. However, we have not found IP regions next to long straight ML edges, possibly because these edges are already very rare by themselves and the IP regions mostly appear at the points where the ML islands are at their smallest width, which are also the points where straight edges appear even less often. Such a sample area would allow for the elimination of the intensity variations due to structural or orientation reasons and isolate the magnetic signal. Perhaps by growing larger Mn ML islands both the probability for IP regions and longer ML edges could be increased at the same time.

5.4.5 Investigating the in-gap states of a non-coplanar spin texture

Theory predicts non-coplanar antiferromagnetic spin textures to host fully gapped topological superconducting systems, unlike their collinear counterparts, see [103, 104]. In the previous section (Sec. 5.3) we have seen, that for samples with > 1 ML Mn coverage, the DL, which hosts a collinear OOP AFM state, leads to frustration of the AFM spin-spiral state in the Mn ML resulting in the formation of half-loop magnetic structures near the boundary between the two, see Fig. 48a-b. From the theory side these structures were simulated to be non-coplanar and hold a half-integer topological charge on each of the ferromagnetic sublattices.

Here, we investigate if these structures show any indication of changes to the in-gap states in their surroundings by measuring zero bias dI/dU -maps in both multipass and constant-height mode, see Fig. 48c-d. However, we do not observe any changes to the LDOS at the Fermi energy in the area around the half-loop that cannot be attributed to defects in the Mn ML. We repeated this experiment for two other half-loops at two different locations on the same sample with similar terrace widths and observe the same results.

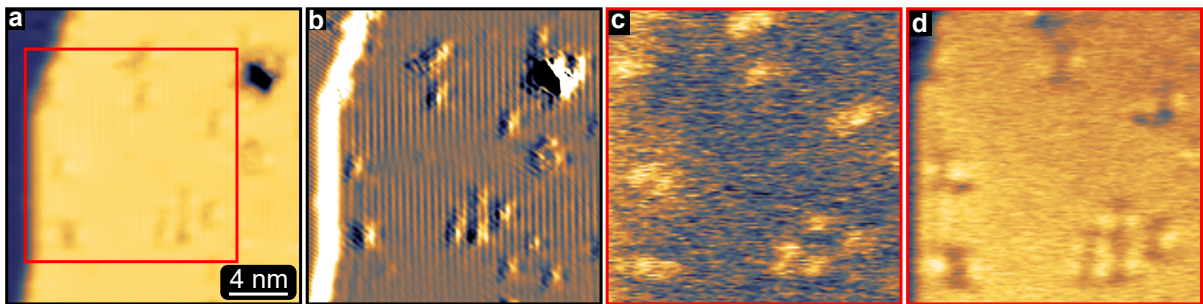


Fig. 48 **a**, STM topography image of a Mn ML region bordering the Mn DL to the left. **b**, SP-STC current map of the same region as in **a**, measured using an in-plane spin-polarized tip. **c-d**, Zero bias dI/dU -maps of the region marked by the red rectangle in **a**, measured using multipass/constant-height mode, respectively. For the multipass measurement the topography measured at $U = +4$ mV, $I = 1$ nA was retraced. Measurement parameters: a-b: $U = -15$ mV, $I = 2$ nA, $T = 4.2$ K; c-d: $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$, $T = 1.3$ K.

The fact that we do not find any variation in the LDOS inside the superconducting gap could be due to our limited energy resolution and may be resolved by measuring at lower temperatures. Using a SC tip is not possible, as magnetic contrast is necessary to locate the half-loop structures. Furthermore, the half-loop structures investigated here form closer to the DL boundary compared to the simulation. Experimentally we have also observed half-loops further from the DL-ML edge, see Fig. 38b, due to much larger terrace widths, and it may be interesting to reassess them to investigate the resulting in-gap states.

Compared to the non-coplanar spin textures investigated theoretically [103, 104], mainly the 3Q state and small-scale multi-Q textures, the half loops in this system are larger. While they still host a non-zero scalar spin chirality, the canting angles between nearest-neighbour moments are smaller. Smaller canting angles in general lead to smaller topological gaps [104], which likely leads to smaller changes in the LDOS when comparing the coplanar and non-coplanar regions. Tight-binding calculations of the spin texture in this system may provide further insight.

5.4.6 The Mn DL/Ta edge mode

We have seen that the edge mode observed at the boundary between the Mn ML and the Ta substrate is consistent with a nodal point superconducting phase. Let us now turn our attention to the Mn DL. We first investigate the boundary between the Mn DL and the Ta substrate. In Fig. 49a a sample of 1.25 ML of Mn on Ta(110) is shown, with a DL region growing from the step edge and perfectly covering the full width of ML below. Due to Oxygen impurities the ML does not cover the entire Ta substrate, here such an exposed Ta region is bordering to the DL, resulting in a boundary between the DL and Ta substrate. Several regions of the edge are atomically straight and are oriented along the [001] direction. In general, the DL grows with many atomically straight edges along the [001] direction, see also the previous section Sec. 5.3.

Fig. 49b-c show zero bias dI/dU -maps of these edges measured in constant-height mode. Along the straight [001] edge, an increased dI/dU signal, compared to the interior of the DL island, is visible, indicating the presence of an edge mode. Deviations from the atomically straight edges along the [001] direction show reduced contrast similar to the behaviour of the Mn ML. The straight horizontal edge in Fig. 49b indicates an $[1\bar{1}0]$ edge between the Mn DL and the Mn ML which we will discuss later.

The presence of an edge mode between the Mn DL and the Ta substrate is an indication that the two systems are in different topological superconducting phases, and as the Ta is necessarily in a topologically trivial superconducting state, the Mn DL cannot. Experimentally, only the [001] edge between the Mn DL and Ta substrate is accessible, while both of the $[1\bar{1}0]$ and $[1\bar{1}1]$ edges are not. This removes the possibility of investigating the edge mode intensity levels for the different edges and if an edge mode is present for all edge directions, making it hard to distinguish between a nodal point superconducting phase and a gapped superconducting phase. However, given the ML appears to host a nodal point superconducting phase and the theoretical predictions for collinear AFM systems in general, we can conclude that the DL is a nodal point superconductor as well.

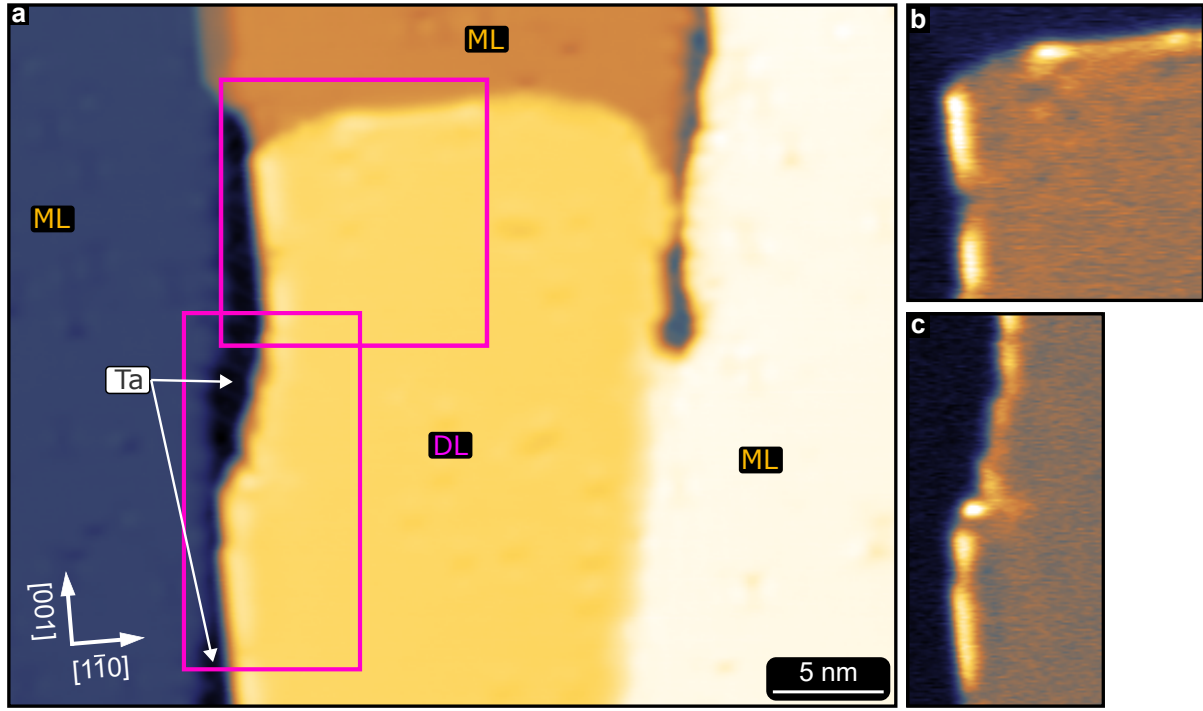


Fig. 49 **a**, STM topography image of 1.25 ML of Mn on Ta(110). The regions within the pink rectangles show vertical edges from the Mn DL directly to the Ta substrate and are running along the [001] direction. $U = 15$ mV, $I = 300$ pA. **b-c**, Zero bias dI/dU -maps of the regions marked by the pink rectangles in **a** measured in constant-height mode. The tip was stabilized at $U = +4$ mV, $I = 1$ nA on the DL. $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$; all: Cr bulk tip, $T = 1.3$ K.

5.4.7 The Mn DL/ML edge mode

We will now investigate the boundary between the two magnet-superconductor-hybrid systems, the Mn ML and Mn DL. In Fig. 50b, a sample of 1.25 AL of Mn on Ta(110) is shown, with the DL island growing from the step edge and forming very straight edges along the [001] direction. At the measurement bias of +5 mV the edges of the Mn DL appear higher, which indicates an increased LDOS at the edge. This can be understood as an edge mode independent of the superconducting state, because it appears identical when the superconductivity is quenched. We will discuss this in detail when looking at the magnetic field dependent measurements.

The corresponding SP-STM current map shows a weak magnetic contrast on the Mn ML and no magnetic contrast on the DL. The spin-spiral is suppressed everywhere in this sample area, as we observe no regions where the magnetic contrast vanishes. This means that the magnetic state of the Mn ML is OOP everywhere, which indicates a tip magnetization with a significant OOP component. However, the magnetic contrast on the DL is too weak to resolve, see Fig. 50a. Interestingly, we observe two different contrast levels at the edges on the right side of the DL islands. For a current map we can only compare edges on the same side of islands, as the STM tip will move up the island edge on one side and down the island edge on the other, as can be seen from the generally brighter contrast on the right edges and the generally darker contrast on the left edges.

Additionally, by just comparing right edges, we remove any effects of tip asymmetry.

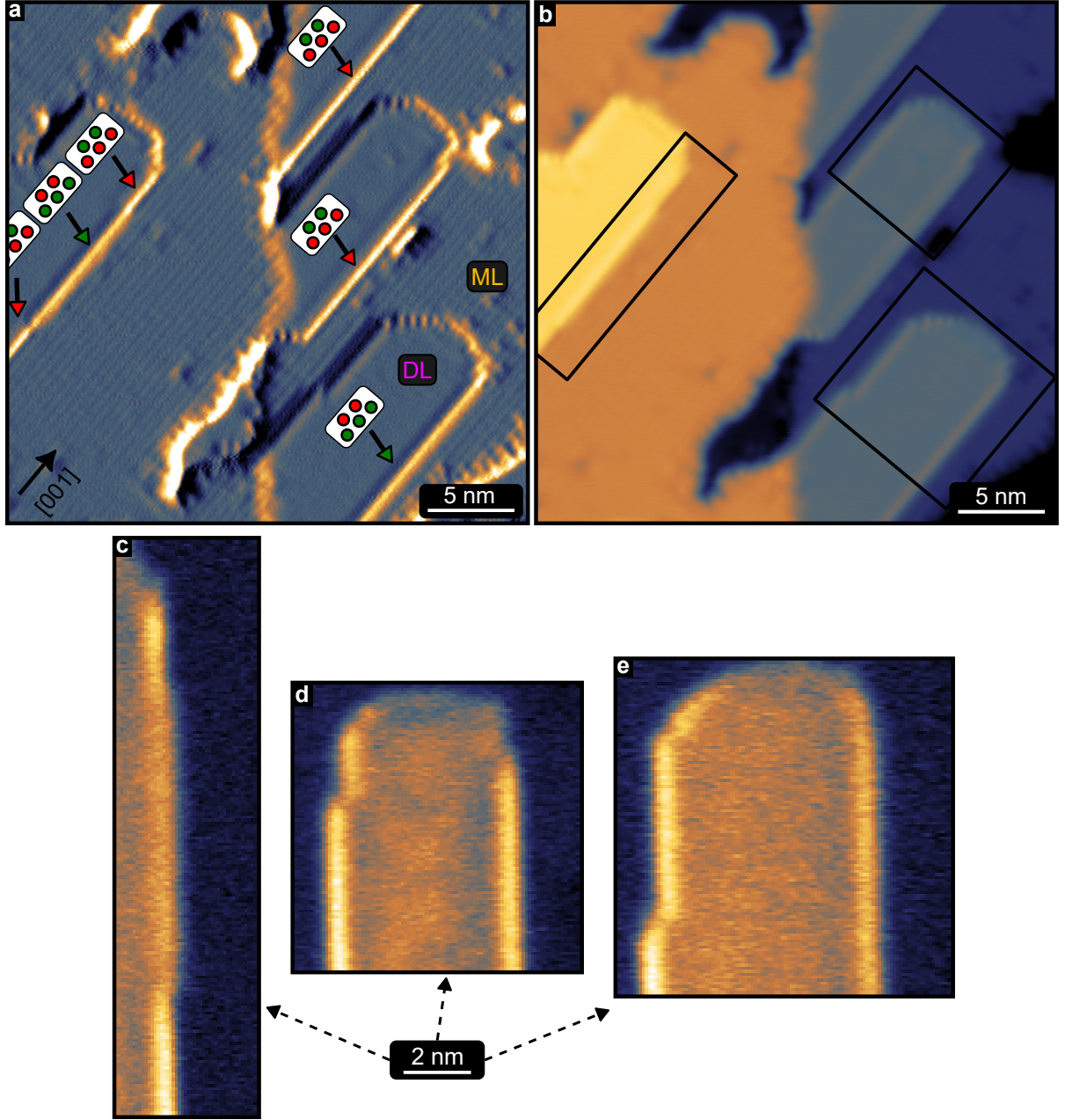


Fig. 50 **a-b**, SP-STM current map/topography image of 1.25 ML of Mn on Ta(110). The DL islands growing from the step edges exhibit atomically straight edges in the [001] direction. The right-side island edges show a two-stage contrast as indicated by the red/green spheres and arrows. $U = 5$ mV, $I = 1$ nA. **c-e**, Zero bias dI/dU -maps of the regions marked by the black rectangles in **b** measured in constant-height mode. The tip was stabilized at $U = +4$ mV, $I = 1$ nA on the DL. $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$; all: Cr bulk tip, $T = 1.3$ K.

The edge on the left in Fig. 50a is of special interest; here, two different contrast levels are present at the same edge, in this case slightly brighter at the top and bottom

and slightly darker in the middle. The reason for this two-stage contrast is the edge termination of the AFM state, resulting in a row of either spin-up or spin-down atoms at the edge. This also indicates that this edge exhibits two kinks, each a single atomic row wide. By comparing the contrast levels at the right edges of the different DL islands, we can assign the two possible terminations of the AFM state, as indicated by the red/green sketches and arrows in Fig. 50a.

We measured zero bias dI/dU -maps in the areas marked by the black rectangles in Fig. 50b. We find a strong signal intensity at the $[001]$ edges, for the DL islands shown in Fig. 50c-e, indicating the presence of an edge mode. In Fig. 50c, we observe two contrast levels of the edge mode at the edge hosting the two kinks. These two contrast levels can be attributed to the two opposite terminations of the AFM state, indicating that the edge mode is spin-polarized. As discussed before, only the intensity of edge modes at edges on the right side of the islands can be compared, not between left edges and right edges.

We also find evidence for an edge mode at the $[1\bar{1}1]$ edges, see Fig. 50e. The edges along the $[1\bar{1}0]$ direction show no increase in the dI/dU signal compared to the island interior. However, to exclude the presence of an edge mode, we need to investigate edges that are perfectly straight, like the ones in the other two directions. We have already observed a perfectly straight edge in the $[1\bar{1}0]$ direction in Fig. 49b, and there we observe an increased signal intensity. Even though it is weaker than the contrast at the other edges, this does indicate the presence of an edge mode as well.

In Fig. 51, multipass and constant-height images of the edge with the two single atomic kinks, see Fig. 50a, are displayed, measured as a magnetic field-dependent series with the same micro-tip. Note that for all images the forward and backward channels are qualitatively the same. In addition to the topography images and zero bias dI/dU -maps, the zero bias current maps are also shown for both measurement modes, using a blue-red colour scale with blue indicating negative currents, red indicating positive currents, and white indicating zero current. At zero bias there should be no current flowing in either direction; however, due to the bias modulation which was typically set to $U_{\text{mod}} = 50\mu\text{V}$ (RMS), a net current can arise if the LDOS in the range of $-71\mu\text{V}$ to $+71\mu\text{V}$ exhibits an effective non-zero slope. A positive slope would result in a positive current and a negative slope similarly in a negative current. We find that the current maps for constant-height and multipass generally show the same features.

As we have already seen in Fig. 50a, the $[001]$ edge with two kinks shows two contrast levels, unrelated to the superconducting state, due to the termination of the AFM state. The zero bias conductance maps similarly show two contrast levels indicating that the edge mode is spin-polarized, see Fig. 51b,d. Interestingly, we also see an impact of the different spin-polarization on the appearance of the edges in the zero bias current maps, see Fig. 51c,e. For the middle edge, we observe a strong positive current followed by a negative current when crossing the edge from left to right. For the bottom and top edges, we observe a much weaker positive current followed by a similar or stronger negative current. This indicates that the different intensity of the features in the LDOS at near zero bias at the oppositely magnetized edges, results in a distinct lateral distribution of the slope in their vicinity.

When quenching the superconductivity using a $+200\text{ mT}$ magnetic field, we find a complete vanishing of the dI/dU signal at the edge in multipass mode, while some contrast

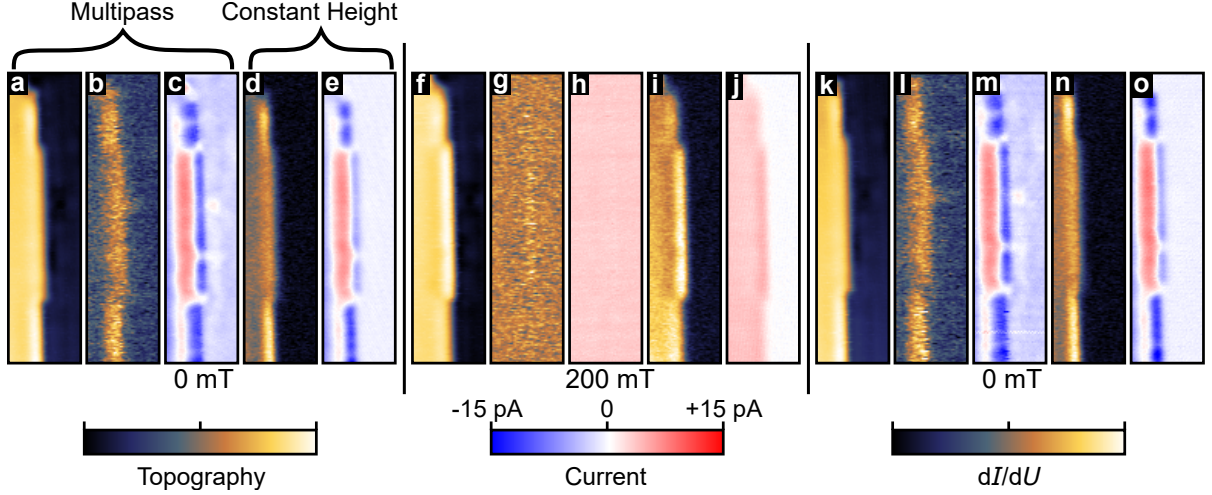


Fig. 51 **a,f,k**, STM constant-current topography images of an edge between the Mn DL (left) and Mn ML (right). $U = +4$ mV, $I = 1$ nA. **b,g,l**, Zero bias dI/dU -maps of the same region measured in multipass mode. $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$. **c,h,m**, Zero bias current maps of the same region measured in multipass mode. $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$. **d,i,n**, Zero bias dI/dU -maps of the same region measured in constant-height mode. $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$. **e,j,o**, Zero bias current maps of the same region measured in constant-height mode. $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$. All images were measured with the same tip at $T = 1.3$ K as a magnetic field dependent series at the following field values: a-e: 0 mT $\rightarrow$ f-j: +200 mT $\rightarrow$ k-o: 0 mT; all: Cr bulk tip.

remains in the constant-height data, see Fig. 51g,i. The contrast at the edge inverts between the constant-current topography at 0 mT and +200 mT, see Fig. 51a,f, respectively, appearing like the z-component of the tip magnetization inverted. However, this seems unlikely to happen at such low magnetic fields. It is more likely that the spin-polarization of the sample states around the Fermi energy changes between the superconducting and normal-conducting state. Perhaps the spin-polarization of the edge mode in the superconducting state is opposite to the spin-polarization of the normal-conducting state. The spin-polarized dI/dU contrast in the constant-height data, see Fig. 51i, is due to this spin-polarization at the Fermi energy in the normal-conducting state and appears therefore very similar to the constant-current multipass topography image, see Fig. 51f, which measures the spin-polarization of the states in the range of 0 mV and +4 mV.

The clear indication of the vanishing of the edge mode due to the quenching of superconductivity can actually be observed in the zero bias current maps, see Fig. 51h,j, which appear both mostly featureless and only show small positive currents because the LDOS around the Fermi energy of a metal is similarly featureless and monotone and in this case shows a positive slope.

The reason that the contrast in the multipass mode completely vanishes is related to the feedback loop. From the constant-current topography image, we know that the tip is further away at the bright edge, which results in a decreased dI/dU signal at this edge. Overall, the differences in tip-sample distance at 4 mV, see Fig. 51f, and the zero bias dI/dU signal, see constant-height data in Fig. 51i, almost perfectly compensate. This loss

of information due to the feedback loop clearly demonstrates the potential problems of multipass mode measurements and that, if the topography is featureless, the multipass mode and the constant-height modes are identical on atomically flat surfaces. For the edge mode between the Mn DL and ML, the constant-height measuring mode not only avoids feedback artefacts, due to the bright edge, but also generally provides a better signal to noise ratio. Constant-height mode and multipass mode are best employed together if possible, as the constant-height mode can indicate feedback artefacts in the multipass mode. Additionally, the multipass mode can show details about the dI/dU and current signal, which cannot be revealed by constant-height mode, at sample positions where the apparent height changes (i.e. edges).

In Fig. 52 multiple spectra were taken from the Mn ML across two oppositely terminated [001] edges onto the DL. The example spectra displayed in Fig. 52c were taken at the similarly-coloured locations marked in the current map, see Fig. 51a, with the entire series of spectra plotted as a waterfall plot for the black and dashed black line in Fig. 51d/e, respectively. For the waterfall plots, a line was fitted to the region outside the superconducting gap of each spectrum and then each spectrum was normalized using the y-intercept.

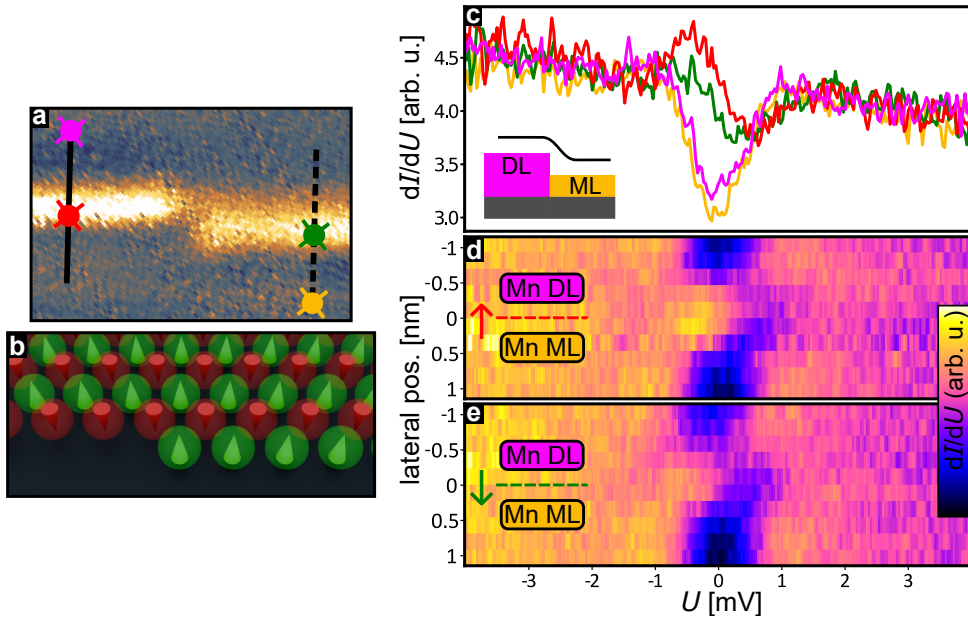


Fig. 52 **a**, SP-STM current map of a kink in the atomically straight [001] edge in Fig. 50c. $U = 5$ mV, $I = 1$ mA. **b**, 3D sketch of the kink in **a**. **c**, dI/dU spectroscopy measurements taken at the locations marked by the coloured circles in **a**, with one on the Mn ML (orange), one on the Mn DL (violet), and two on the oppositely magnetized edges (red and green). **d-e**, Waterfall plots of a series of spectra taken across the oppositely magnetized DL edges along the black and dashed black line in **a**, respectively. Each individual spectrum was normalized to the y-intercept of a line fitted to the data outside the gap. Measurement parameters c-e: The tip was stabilized at $U = +4$ mV, $I = 1$ nA; $U_{\text{mod}} = 50 \mu\text{V}$; all: Cr bulk tip, $T = 1.3$ K.

The spectra on the Mn ML and DL are similar, with slightly higher LDOS inside the

gap for the DL, in accordance to what we observed before in Fig. 43b. The red and green spectra were taken on the oppositely magnetized edges and both show an increase in the LDOS inside the gap compared to the bulk ML or DL as expected for an edge mode. The differences between the two curves can be attributed to the spin-polarization of the edge mode and the spin-polarization of the tip. When comparing the waterfall plots for the whole series of spectra taken across the edges, we observe a similar difference in the intensity at the edge. The estimated edge position is marked with the red and green dashed lines.

Estimating the position of the edge mode in relation to the edge (i.e. the last row of DL atoms) is not trivial. One has to look at the trace of the z-position of the tip for the series of spectra or a topography line-profile for the multipass measurements and compare them with the dI/dU signal that has been adjusted for the delay of the lock-in amplifier. First, we need to establish the position of the last row of DL atoms for our constant-current topography line-profiles.

Fig. 53a-b show a topography image and an SP-STM current map of a [001] edge between a DL region on the left and a ML region on the right. A line-profile was extracted from the topography channel along the white line. We can use the bright lines of the magnetic contrast to find the centre of the atomic rows, represented by the red and green dashed lines. Due to the antiferromagnetic alignment between DL and ML, the maxima on the DL are marked by red dashed lines, while the maxima on the ML are marked by green dashed lines. Following this periodicity to the edge, we can establish the possible positions of the last row of DL atoms.

To identify the most likely position of this last row of DL atoms, we need the assistance of simulations where this position is well known. In Fig. 53d a line-profile across a hexagonal island, see inset, was simulated using an *s*-wave single atom tip in constant-current mode for different tip heights (i.e. different current set points) and then shifted. The last row of DL atoms is marked by the dashed grey line. We find that even before the tip reaches the last row of DL atoms, the tip needs to approach the sample to keep the current constant because the missing atoms decrease the overall tunnel current. This holds true for all three tip-sample distances, but is least pronounced for a tip very close to the sample providing atomic contrast, see grey line in Fig. 53d. Typical imaging parameters are associated with the tip-sample distances of the red and blue curves. Here, an approximate 5% drop in apparent height is seen at the position of the last row of DL atoms.

The simulations allow us to make a better estimation of the position of the last atomic row in the experimental line-profile. The best estimate is given by the solid red line in Fig. 53c, however, the strong LDOS at the edge of the DL makes this more difficult. The differences of a real STM tip, which is not always terminated by a single atom at the tip apex, compared to the simulations are expected to be minor and to generally show a similar trend in the falloff of the apparent height.

To find the position of the edge in relation to the zero bias dI/dU -maps, we need to compare the topography profiles, for which we can now make a reasonable estimate of the position of the last row of DL atoms, to the profiles extracted from the dI/dU -maps. Fig. 53e shows line-profiles for the topography and the dI/dU signal, one for each of the oppositely magnetized edges, extracted from Fig. 51a,b, respectively. To align them

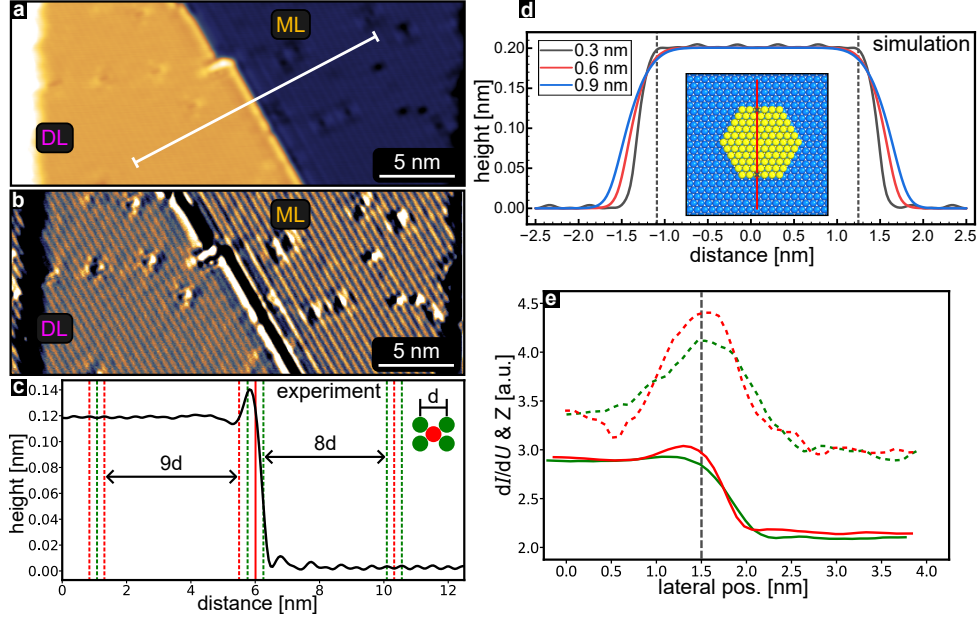


Fig. 53 a-b, STM topography image and current map of a $[001]$ edge between a Mn DL region (left) and a Mn ML region (right). $U = -15$ mV, $I = 5$ nA, $T = 4.2$ K. **c**, Line-profile across the same edge, as indicated by the white line in **a**. The vertical red and green dashed lines mark the position of the atomic rows, as indicated by the magnetic contrast. The solid red line indicates the best estimate for the position of the centre of the last row of DL atoms. **d**, Simulated constant-current line-profile across a hexagonal island, see inset, at different tip-sample distances, courtesy of André Kubetzka. **e**, Topography (solid lines) and dI/dU (dashed lines) line-profiles taken across the two oppositely magnetized (red and green) DL edges in Fig. 51a,b, respectively. The red and green curves were shifted relative to each other by a single atomic row to account for the kink. The dI/dU profiles were shifted relative to the topography profiles to account for the delay by the lock-in amplifier.

correctly, the dI/dU profiles are first shifted with regard to the topography profiles to account for the lock-in delay. Subsequently, the profiles of oppositely magnetized edges (red and green lines) are shifted in relation to each other by a single atomic row to account for the kink in the edge.

The dashed black line represents our estimate for the DL edge position and we find that it aligns very closely with the maximum of the dI/dU signal, which indicates that the edge mode is located on the last row of DL atoms, which is expected because the edge mode is strongly visible in constant-height mode. If the edge mode was located on the ML, constant-height mode measurements would not be able to resolve it.

As a second method to estimate the position of the edge mode in relation to the edge, the zero-bias dI/dU -maps for both the multipass and constant-height mode were overlaid onto a 3d representation of the topography of the DL island, see Fig. 54a-b, respectively. The constant-height measurement was performed immediately after the multipass measurement; therefore, a deviation between the multipass topography signal and the constant-height dI/dU signal is expected to be negligible. However, the dI/dU -maps for both modes needed to be adjusted for the delay of the lock-in amplifier, which

was calculated to be a two-pixel shift for this scan speed and pixel density. The exact methods to calculate the lock-in delay and how to adjust for it, are discussed in detail in Sec. 3.

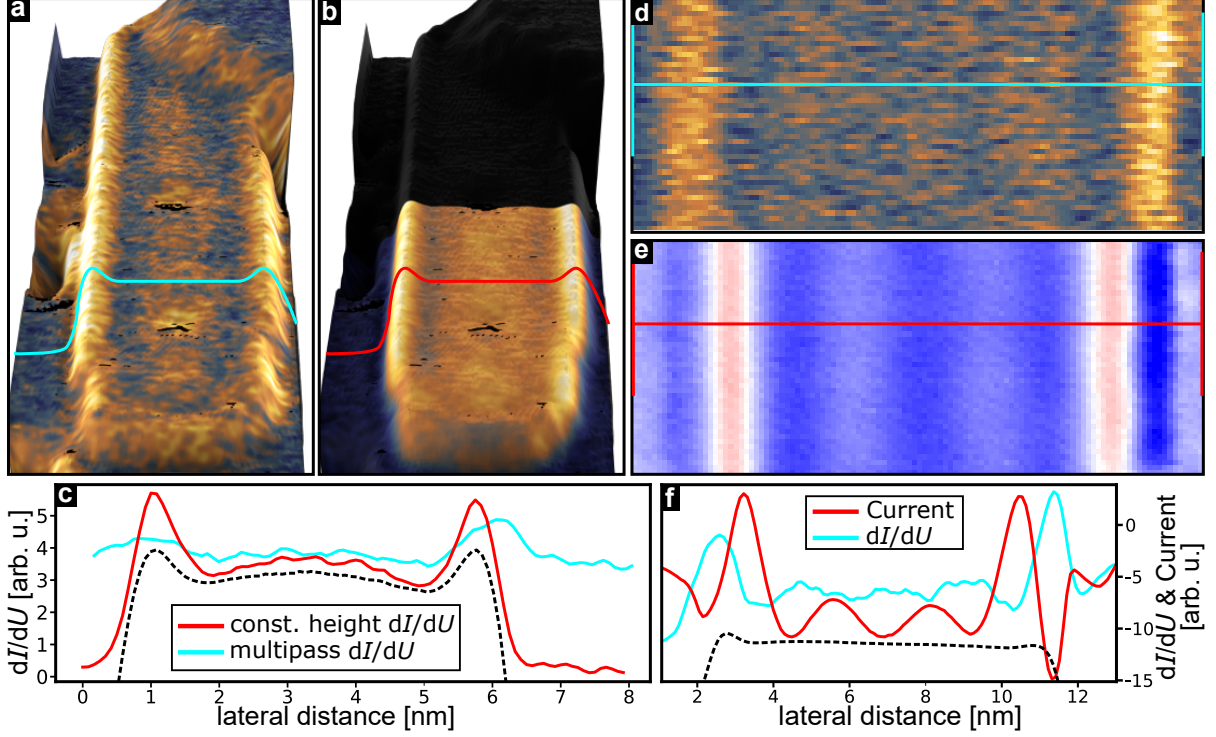


Fig. 54 **a-b**, 3D plots of a DL island with the colouring given by the multipass and constant-height dI/dU signal, respectively, measured at $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$. The dI/dU -maps were adjusted for the delay of the lock-in amplifier. The topography signal was measured at $U = +4$ mV, $I = 1$ nA. **c**, Line-profiles across the entire DL island for the multipass (cyan) and constant-height (red) dI/dU -maps, respectively. For each line-profile 5 pixels were averaged to improve the signal-to-noise ratio. The dotted black line shows the multipass topography at $U = +4$ mV. **d-e**, Zero bias dI/dU and current map, respectively, measured in multipass mode with the topography profile measured at $U = +4$ mV, $I = 1$ nA. $U = 0$ mV, $U_{\text{mod}} = 50 \mu\text{V}$. **f**, Line-profiles across the width of a Mn DL island extracted from the dI/dU -map (cyan) and current map (red), respectively. For each line-profile 32 pixels were averaged to improve the signal-to-noise ratio. The dotted black line shows the multipass topography at $U = +4$ mV; all: Cr bulk tip, $T = 1.3$ K.

The resulting, coloured, 3D projections show the position of the edge mode right at the edge of the DL, in good agreement with our estimation using the topography line-profiles and comparing those to the adjusted dI/dU line-profiles. The multipass and constant-height maps show a very similar position of the edge mode. We have seen that the edge mode appears wider for the multipass mode, see Fig. 51b,e. This appears to be mainly due to the fact that the tip still measures the increased signal when tracing down the side of the DL island, where the signal in the constant-height has a sharp drop off, see Fig. 54a-b.

We observe an interesting standing-wave pattern in the centre of the DL islands, more

strongly visible in the multipass maps, but generally also visible in the constant-height data, see Fig. 54c. After the strong peak in the dI/dU signal at the edge we first observe a dip in the signal followed by a broad peak in the centre of the island.

This standing-wave pattern appears differently depending on the width of the island. The zero bias multipass data of a wider DL island is shown in Fig. 54d, and we observe three peaks in the island interior. Interestingly, the corresponding current map shows only two maxima in the island interior with a better overall signal-to-noise ratio and signal amplitude, see Fig. 54e. Comparing two line-profiles from the two maps, we find that the peaks in the current signal appear roughly at the position of the minima of the dI/dU signal.

This standing-wave pattern is likely caused by the decaying oscillating edge modes from both island edges interfering with each other. Alternatively, the standing-wave pattern could be caused by states in the island interior reflecting from the edges. For larger island widths, which are relatively uncommon, we find not just one maximum in the island interior but two or even three; however, we were limited by the sizes of the DL islands on our sample. The zero bias current signal encodes information of the slope of the LDOS in a small region around the Fermi energy, as mentioned before. Here, a minimum in the dI/dU profile indicates a minimal increase of the in-gap LDOS due to the edge mode, reducing the overall slope around the Fermi energy and therefore the magnitude of the current. In contrast, at the position of the maxima in the dI/dU profile, the LDOS in the gap is changed strongly due to the edge mode, and we see an asymmetric peak in the LDOS, see also Fig. 52c, resulting in an increasingly negative slope around the Fermi energy and therefore an increasingly negative current.

Both the constant-height measurement mode and the multipass measurement mode show the presence of a low-energy spin-polarized edge mode at the boundary between the Mn ML and DL.

5.4.8 The tight-binding model

Tight-binding calculations to model the system of Mn layers on Ta were done by Felix Nickel for our manuscript on this system, see [Publications](#). The results will be briefly discussed here, but for more details refer to the corresponding manuscript.

From the experimental side alone, we cannot definitively say if the Mn ML and DL are in nodal point superconducting states, only that the experimental data for both appear consistent with this assumption. Similarly, from the theory side, we assume topological nodal point superconductivity for the Mn ML and DL and see if we can qualitatively reproduce the experimental data. It is important to note that the goal was not to adjust the tight-binding parameters until they perfectly reproduce the experimentally observed edge modes and their intensity levels relative to each other. Instead, the focus was on deriving a qualitative model capturing the essential effects, such as the experimentally observed edge mode between the Mn ML and DL. An edge mode between two different MSH systems has not been investigated experimentally or theoretically before.

However, the tight-binding parameters were not completely randomly chosen either, as the starting parameters are based on previous theoretical investigations [100]. Adjustments for the two MSH systems were made based on the ratio of the induced magnetic

moments at the interface as calculated by DFT, with the resulting parameter set confirmed to be in a nodal-point superconducting phase. The boundaries along different crystallographic directions between different systems were modelled using a ribbon geometry, where the ribbons of the systems alternate in one direction and are infinite in the other direction.

First, using the chosen tight-binding parameters, the bulk properties of the Mn ML and DL are characterized, and we find that the ML exhibits six nodal points while the DL exhibits 4, see the sketches of the 2D Brillouin zone in Fig. 55a,c, respectively. Both systems host 2 pairs of nodal points on the edge of the Brillouin zone (BZ), with the Mn ML hosting another pair near the centre.

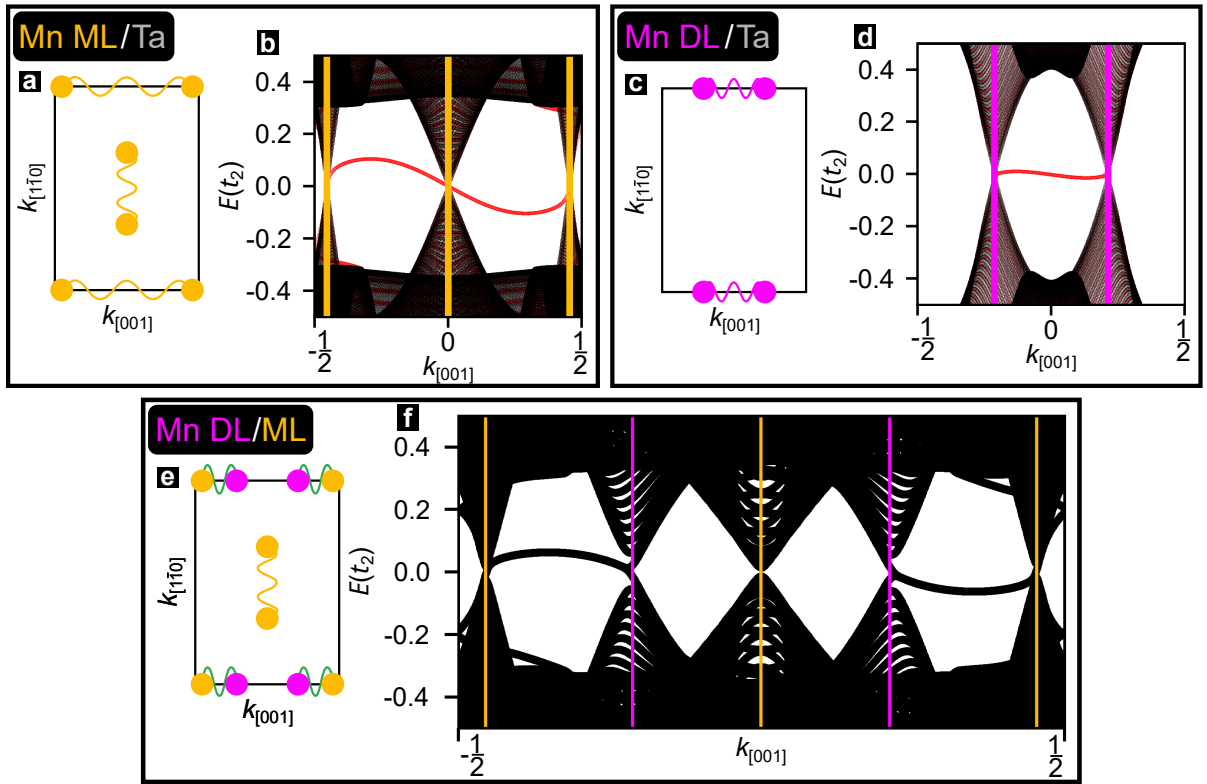


Fig. 55 **a**, 2D-Brillouin zone (BZ) of the Mn-ML/Ta(110) MSH system with the positions of the nodal points marked as modelled using tight-binding formalism. Nodal points found to be connected by an edge mode, when the Mn-ML/Ta(110) system borders the trivial Ta, are connected by yellow lines. **b**, Projected band-structure along the [001] direction from an infinite sized Mn ML sample (black) and the dispersion of a stripe geometry between the Mn ML and Ta (red). **c**, 2D-BZ of the Mn-DL/Ta(110) MSH system with the positions of the nodal points marked as modelled, similar to **a**. **d**, Projected band-structure along the [001] direction for the Mn DL and Ta edge, similar to **b**. **e**, Sketch of the 2D-BZ with the bulk nodal points of the Mn ML and DL marked. Nodal points which are connected by edge modes, for a boundary between the ML and DL, are indicated by the green lines. **f**, Band structure along the [001] direction for a ribbon geometry of alternating stripes of Mn ML and DL. Coloured vertical lines mark the position of the bulk nodal points in the ML (orange) and DL (violet). Figure based on Figure by Felix Nickel in our manuscript, see [Publications](#).

To investigate the presence of edge modes along the $[001]$ direction for the Mn ML, we use a ribbon geometry with alternating regions of Mn ML and Ta in the $[1\bar{1}0]$ direction and infinite extent in the $[001]$ direction. The resulting band-structure is shown in red in Fig. 55b, with the black points indicating the bulk band-structure calculated for a Mn ML of infinite extent in both directions. We find an edge mode, see red line in Fig. 55b, propagating between the nodal points near the edges of the BZ, see yellow line in the BZ sketch of Fig. 55a. Investigating the $[1\bar{1}0]$ edge by using the same stripe geometry rotated by 90° , we also find an edge mode at this edge connecting the two nodal points near the centre of the Brillouin zone, see yellow line in the BZ sketch of Fig. 55a.

Similar calculations for the Mn DL reveal an edge mode at the $[001]$ edge, due to nodal points at the edge of the BZ, similar to the Mn ML, see purple and red lines in Fig. 55c-d. However, no edge mode exists at the $[1\bar{1}0]$ edge, see Fig. 55c, because the DL does not have nodal points near the centre of the BZ and the pair of nodal points at the edges of the Brillouin zone fall on top of each other when projected along this direction.

In both cases our tight-binding model reproduces the experimentally observed edge states. For the Mn ML, the edge state observed along the $[1\bar{1}1]$ follows from the calculated nodal-point configuration as well. For the Mn DL, the $[1\bar{1}0]$ edge to the Ta substrate is experimentally not accessible, the presence/absence of an edge mode cannot be determined.

Let us turn to the main focus of the tight-binding investigation, the boundary between the Mn DL and the Mn ML. The result of the calculations for a stripe geometry of alternating ML and DL regions with infinite extent of the stripes in the $[001]$ direction is shown in Fig. 55f. We find edge modes propagating between nodal points of the Mn ML and DL, which means between nodal points of different nodal-point superconducting phases. This is qualitatively different from previously investigated systems, where the boundary between nodal point superconducting phases and a trivial substrate were investigated [30, 100], see also sketch of the 2D-Brillouin zone in Fig. 55e. Note that in the $[1\bar{1}0]$ direction the edge mode between the nodal points of the Mn ML remains, which would result in an increased LDOS also at the $[1\bar{1}0]$ edge and the $[1\bar{1}1]$ edges consistent with the experimental results. These nodal points are independent of the Mn DL properties as they only result from the nodal points of the ML. However, we see that if the nodal points in the centre of the Brillouin zone were not present (i.e. even if the Mn ML and DL had the same number of nodal points), an edge mode would still emerge at the $[001]$ edge. This also demonstrates that the presence or absence of the edge mode at the $[1\bar{1}0]$ edge can, in principle, be controlled by choosing an MSH system with a specific nodal point layout for the Mn DL system to interface with.

The tight-binding parameters used for ML and DL differ only in the coupling strength of the SC to the magnetic layer, and the two MSH systems can be smoothly transformed into each other. During this transformation from ML to DL, the nodal points in the centre of the Brillouin zone merge and detach from the Fermi surface, while the nodal points on the $[001]$ edge of the Brillouin zone shift closer together. These shifted nodal points at the edge of the Brillouin zone are then connected to their unshifted counterparts at boundaries between the shifted (DL) and unshifted (ML) systems. The fact that an edge mode between a nodal point and its slightly shifted nodal point can emerge, raises quite interesting possibilities of edge mode engineering. A local perturbation, for instance

an electric field, can lead to small changes in material parameters resulting in differences in the nodal point layout and the emergence of an edge mode between perturbed and unperturbed regions.

5.5 Summary

In this section, we have first investigated the antiferromagnetic states hosted in the Mn ML and DL. While an AFM spin-spiral emerges as the ground state in the extended ML, the presence of the DL and its collinear OOP AFM state leads to frustration in the ML around it. We find a complete suppression of the spin-spiral in many cases, especially for narrow ML stripes. For wider ML regions the energy gained by the spin-spiral in the centre of the terrace can win over the energy loss at the boundary to the Mn DL, where we find the presence of half-loops (i.e. connected IP regions), which host a non-coplanar spin texture at their turning point and can be understood as AFM half-skyrmions. In cases where the Mn ML is bordered by one DL region on each side, we can observe the formation of a topologically trivial winding domain wall (in-plane region), when the magnetic states of the DL regions are out-of-phase. In the in-phase case we find topologically non-trivial loops, which can be understood as elongated AFM skyrmions.

The interaction of the magnetic states in the Mn ML and DL with the superconducting substrate results in edge modes forming at the boundaries between these MSH systems and the trivial Ta, consistent with both MSH systems being in a topological nodal point superconducting phase. We do not observe changes of the in-gap states for in-plane regions and half-loop structures in the Mn ML, although the effects may be too small to be resolved with our limited energy resolution. At the boundary between what appears to be two topological nodal point superconducting states, we find the presence of edge modes and for the edge mode in the [001] direction we find a strong spin-polarization.

Our experimental findings are in good agreement with a tight-binding model assuming nodal-point superconductivity in both MSH systems. We have shown that interfacing different MSH systems hosting TNPSC phases allows for some control over the emergence of edge modes at certain edges. A further exciting possibility is the engineering of edge modes for nodal-point superconductors through local perturbations such as local electric fields. Overall, we demonstrate MSH systems hosting TNPSC phases can in principle provide certain controllability of the emergence and the properties of edge modes and are therefore potential candidates for future spintronic applications.

6 Conclusion and Outlook

In this thesis, antiferromagnetic model type systems were investigated using SP-STM. For future application in spintronic devices a good understanding of the interactions governing the behaviour of these ultra-thin AFM films is required.

By inducing adatom motion using local voltage pulse and by growing Co nanostructures on the uniaxial RW-AFM state of fcc-stacked Mn/Re(0001), which breaks the symmetry of the hexagonal surface, we have shown that magnetism is able to not only affect the motion of adatoms but also the growth of nanostructures. This finding has potential implications for processes such as catalysis, molecular self-assembly, and the self-assembly of nanostructures. Further investigations into the effect of magnetism on growth are required to assess the effects of temperature and material choice. Additionally, investigations into different symmetry-breaking magnetic states and into the effects of on the growth of multilayer films are necessary to broaden the scope of possible applications.

Mn ML and DL on the low-symmetry Ta(110) surface host a non-collinear spin-spiral and an OOP collinear AFM state as their magnetic ground state, respectively. We have shown that the frustration at the interface between the two layers leads to the formation of AFM half-skyrmion states near the boundary and that two such boundaries on either side of a Mn ML area can lead to the formation of elongated topological AFM skyrmions. This is to our knowledge the first experimental observation of AFM skyrmions in a single magnetic layer. We expect that in this system a further refinement of the Mn thin film growth to eliminate vacancies in the Mn ML and to maximize the interface between ML and DL regions, while ensuring a sufficient width of the Mn ML areas, may vastly increase the density of AFM skyrmions. This system may serve as a model to further investigate the creation and annihilation, as well as the motion of AFM skyrmions. Control over these processes is necessary for future applications such as in racetrack memory.

We have also investigated the interaction of the magnetism in the Mn ML and DL with the superconducting state of the Ta substrate. We observe zero energy edge modes between the two MSH systems and the substrate, as well as between the two MSH systems. In the latter case, we observe a strong spin-polarization of the edge mode at the [001] edge between the ML and DL. Our experimental findings are in good agreement with tight-binding calculations, assuming TNPSC in both. We demonstrate that MSH systems hosting TNPSC phases can provide certain controllability over the emergence and properties of edge modes, by either interfacing different MSH systems or via local perturbations. We conclude that these MSH systems may provide a platform for the controlled engineering of edge modes, with potential applications in spintronic devices.

Appendix A Ultra-thin oxide films on Ir(111)

In the first part of this chapter, transition metal oxides (i.e. Fe-, Co-, and Mn-oxides) on the Ir(111) surface are investigated. Although the main focus of this investigation was the magnetism of these oxides, as they are typically antiferromagnetically ordered in bulk, the thin film morphology and oxide structure will also be discussed. Transition metal oxides may be able to serve as a platform for antiferromagnetic topological quasi-particles like AFM-skyrmions or AFM-merons. While their ferromagnetic counterparts can often be stabilized by driving a spin-spiral phase with a magnetic field, they cannot due to their net zero magnetization. Oxides are of interest because they are typically polar, hosting OOP electric dipole moments. Their polar nature increases the degree to which their properties can be tuned via an electric field, allowing for manipulations of the AFM state.

The Ir(111) single crystal was first cleaned by cycles of Ar ion sputtering at room temperature and annealing up to $\approx 1000^\circ\text{C}$ on an electron-beam (e-beam) heating stage in an Oxygen pressure of 5×10^{-8} - 2×10^{-7} mbar. Before any metal deposition, the crystal was flash annealed to $\approx 1500^\circ\text{C}$. The metal films were then grown on a sample still at an elevated temperature approximately 10 – 20 minutes later. This was followed by heating at a very low power in the range of 1 – 8 W for 2 minutes on the electron-beam heating stage with an additional 2 – 3 minutes in an Oxygen atmosphere of 5×10^{-7} mbar at the same temperature. A rough estimate for the sample temperature in the range of 1 – 8 W on the e-beam stage is $300 - 800^\circ\text{C}$. The exact power for each sample preparation is mentioned in the specific sample sections. The samples were then cooled to the respective measurement temperature in the STMs.

A.1 FeO/Ir(111)

For the Fe-oxide (FeO) thin films ≈ 0.65 ML of Fe were deposited on the Ir(111) surface. The approximate deposition rate was derived from prior Fe preparations. Fig. 56b shows an FeO sample after oxidation at ≈ 1 W on the e-beam heating stage.

The sample shows large regions of the FeO ML covering most of the Ir(111) surface, see blue areas in Fig. 56b. In these regions a hexagonal pattern of dark spots is visible, reminiscent of a Moiré pattern, which appears for layers with a lattice mismatch. Such a Moiré pattern is expected for FeO on transition metal substrates and has been observed for Pt(111) [139, 140], Au(111) [141] and also Ir(111) [142] before, with the Fe-oxide layer expanding with respect to the underlying transition metal substrate. The lattice of dark spots across the FeO area shows small changes in orientation, suggesting that the coupling of the FeO layer to the Ir(111) substrate is not very strict and small variations in the angle with respect to the substrate are possible.

The apparent height at the bias voltage of 500 mV is only ≈ 120 pm, see Fig. 56a, much lower than the height of the unoxidized Fe ML, which is ≈ 280 pm. With the additional Oxygen likely integrated as an additional layer on top of the Fe, as also indicated in previous investigations [142], the height of a FeO ML is physically larger than the Fe ML; a reduction in conductivity could explain the lower apparent height as a smaller tip-sample distance would be necessary to achieve the same current as on the Fe ML.

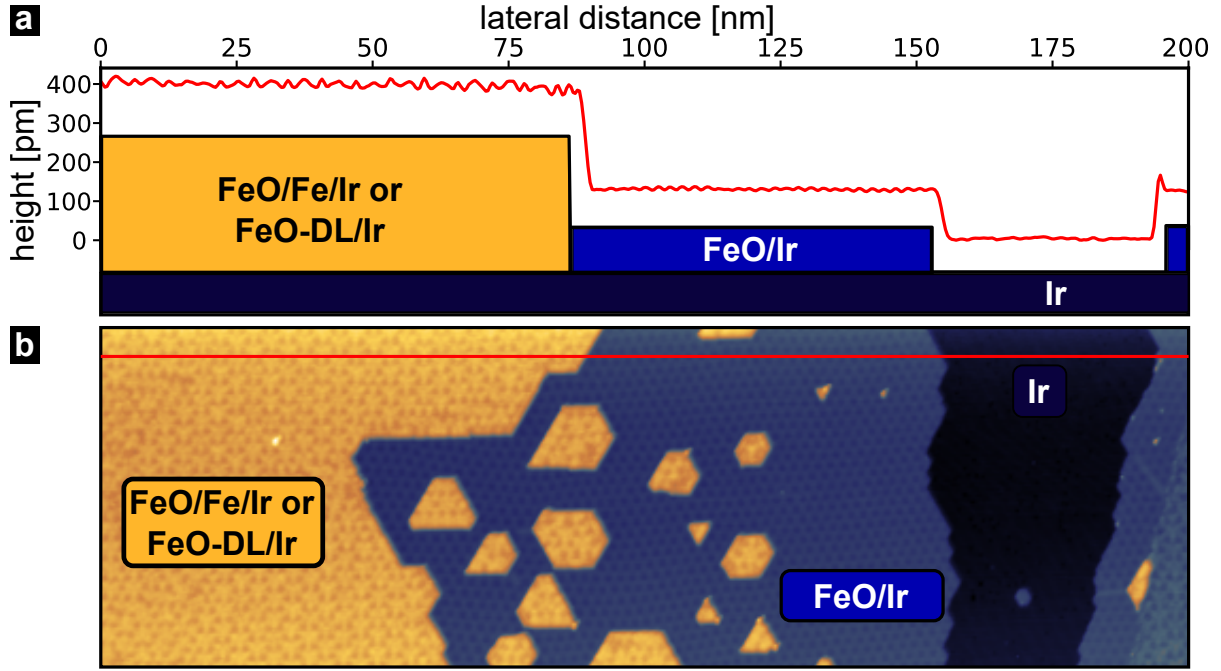


Fig. 56 **a**, Line-profile extracted from the image in **b**. **b**, STM overview topography image of an oxidized Fe film on Ir(111). $U = 500$ mV, $I = 500$ pA, $T = 4.2$ K.

In addition to regions of FeO, a higher layer configuration can be observed as well, see the left side of Fig. 56a-b, respectively. Here, a honeycomb pattern of dark spots is visible on top, but on other sample regions areas with hexagonal spots of different sizes can be found, see Fig. 57, suggesting that multiple different structural phases exist. This higher layer could correspond to regions of an Fe DL before oxidation. During oxidation an expansion of the FeO lattice is expected, possibly resulting in additional Fe atoms from the Fe ML being incorporated into this higher layer oxide structure. It is unclear if the oxidation results in the formation of an Oxygen layer only on top of the Fe DL, similar to the Fe ML, or if an Oxygen layer also forms between Fe layers.

The focus of this investigation is on the FeO ML, which only hosts one structural phase and will be easier to investigate regarding magnetism. A close-up atomic resolution image of the Moiré pattern in the FeO ML is shown in Fig. 58a. Previous investigations of FeO on transition-metal substrates suggest that the Oxygen atoms are located in the top layer, therefore the atoms visible here are likely the Oxygen atoms as one is less likely to see the Fe atoms in the layer below. However, atomic resolution of the Fe layer below is possible as well [143]. For the purpose of analysing the relationship of the FeO with respect to the Ir substrate, it does not matter which atomic layer is visible but these atoms will be referred to as Oxygen atoms in the following.

From the FFT of the atomic resolution image in Fig. 58a, see Fig. 58c, as well as real-space line-profiles, one finds an Oxygen-Oxygen distance of ≈ 310 pm, which is larger than the Ir(111) lattice constant of 272 pm. As sketched in Fig. 58b, this larger FeO lattice results in a Moiré superstructure, where the stacking between the FeO and Ir sub-

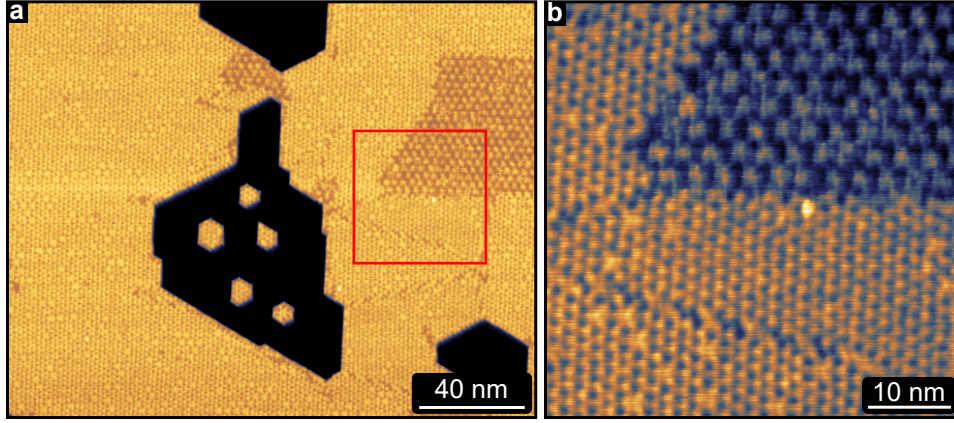


Fig. 57 **a**, STM overview topography image of the second layer in an oxidized Fe film on Ir(111). $U = 500$ mV, $I = 300$ pA. **b**, STM topography image in the area indicated by the red rectangle in **a**. $U = 500$ mV, $I = 500$ pA. Both: $T = 4.2$ K.

strate varies periodically from top site, to hcp site, to bridge site, to fcc site, back to top site. However, correlating the observed regions in the STM image to the different stacking positions is not trivial, because while the top-site regions may be visible with the largest apparent height at one bias, at a different bias they can appear the lowest. Such a correlation has been done for FeO/Pt(111) by Nilius *et al.* [144] using field-emission resonances or apparent-barrier-height measurements to determine the relative surface dipole that changes over the unit cell and stacking sequence, but for our purposes this is of minor importance.

For the atomic resolution image in Fig. 58a the Moiré lattice can be approximately divided into three regions: The dark blue regions, the roughly equally sized orange regions, and the smaller bright yellow regions. The fact that the bright yellow regions are the smallest lets us mark these as top site regions as a best guess. The other two roughly equally-sized regions would then correspond to fcc and hcp stacking, respectively. This is compatible with the fact that Fe grows in both fcc and hcp stacking on the Ir(111) substrate with only a relatively small energy difference of 7.6 meV per atom [145].

From the 2D-FFT, a Moiré lattice constant of ≈ 2.05 nm can be determined. By comparing the ratio of the distance between atomic spots and Moiré superstructure spots one finds the Moiré lattice constant to be ≈ 6.5 times larger than the Oxygen-Oxygen distance. For the LEED pattern measured for an FeO sample, see Fig. 58d, a ratio between atomic spots and superstructure spots of ≈ 7.5 is found. The atomic LEED spots likely correspond to the Ir(111) lattice constant, not the FeO lattice constant as in the 2D-FFT image; this suggests a Moiré superstructure caused by 13 Oxygen atoms placed over a distance of 15 Ir atoms, resulting in the unit cell marked by the white parallelograms in Fig. 58a-b. These measurements are in good agreement with the results obtained by Ma *et al.* [142].

The LEED pattern was taken on a sample with lower Fe coverage before oxidation and therefore much less higher-layer FeO coverage, which explains why only spots expected from the FeO ML are visible. Secondly, the LEED pattern does not show any broadening of the LEED superstructure spots that one would expect to arise due to the different

orientations of the FeO with respect to the Ir lattice. However, the sample was oxidized at a $\approx 15\%$ higher power compared to the ones shown with STM here, which may result in less variation in the angle of the Moiré lattice across the sample.

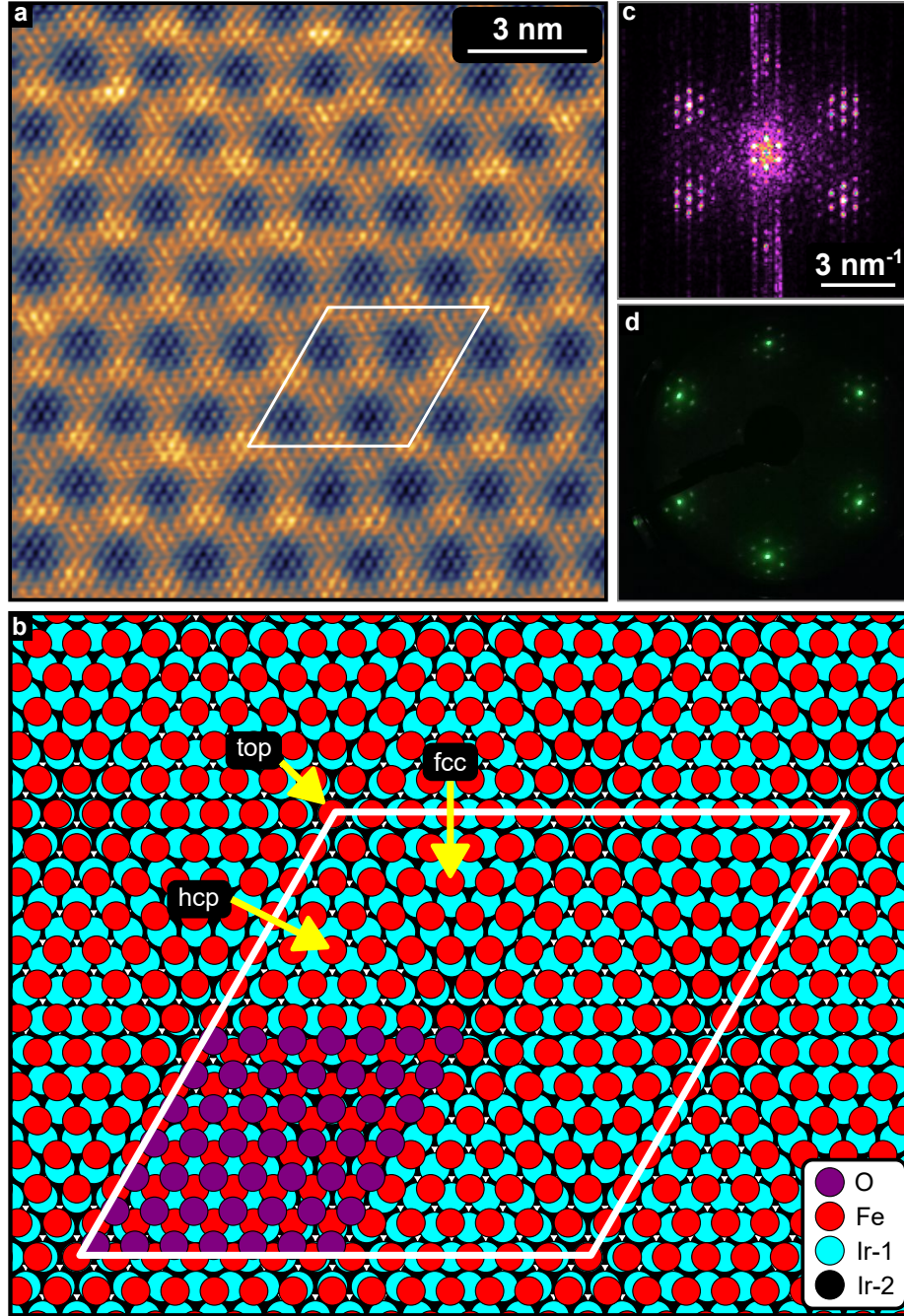


Fig. 58 **a**, STM topography image of a single ML of FeO on Ir(111) hosting a Moiré pattern, caused by the expanded FeO lattice. The white lines indicate the Moiré unit cell. $U = 40 \text{ mV}$, $I = 2 \text{ nA}$, $T = 4.2 \text{ K}$. **b**, Sketch of a Moiré pattern caused by a $\approx 16\%$ expanded top layer representing the Fe-oxide ML, with 13 Oxygen or Fe atoms for every 15 Ir atoms. Part of the top layer of Oxygen was removed for better visibility. **c**, FFT of the STM image in **a**. **d**, LEED pattern of an FeO sample. Centre spot covered by the electron gun.

The sample occasionally shows bright lines separating different regions of the FeO ML, see Fig. 59a. A close-up view is shown in Fig. 59b, where regions, separated by a domain wall, are labelled as domains A, B, and C. For domain A and C, the Moiré lattices show the same unit cell orientation and scale and therefore atomic resolution images show the FeO lattices are oriented in the same direction, although there is some distortion of the Moiré lattices in the region around the domain boundary. The bright lines therefore appear to correspond to boundaries between two out-of-phase domains, perhaps caused by nucleating the FeO lattice in multiple regions at the same time or due to growth with different stacking of the Oxygen layer. One could also imagine that two previously separated Fe islands merged during oxidation, as the Fe regions expand during the FeO growth.

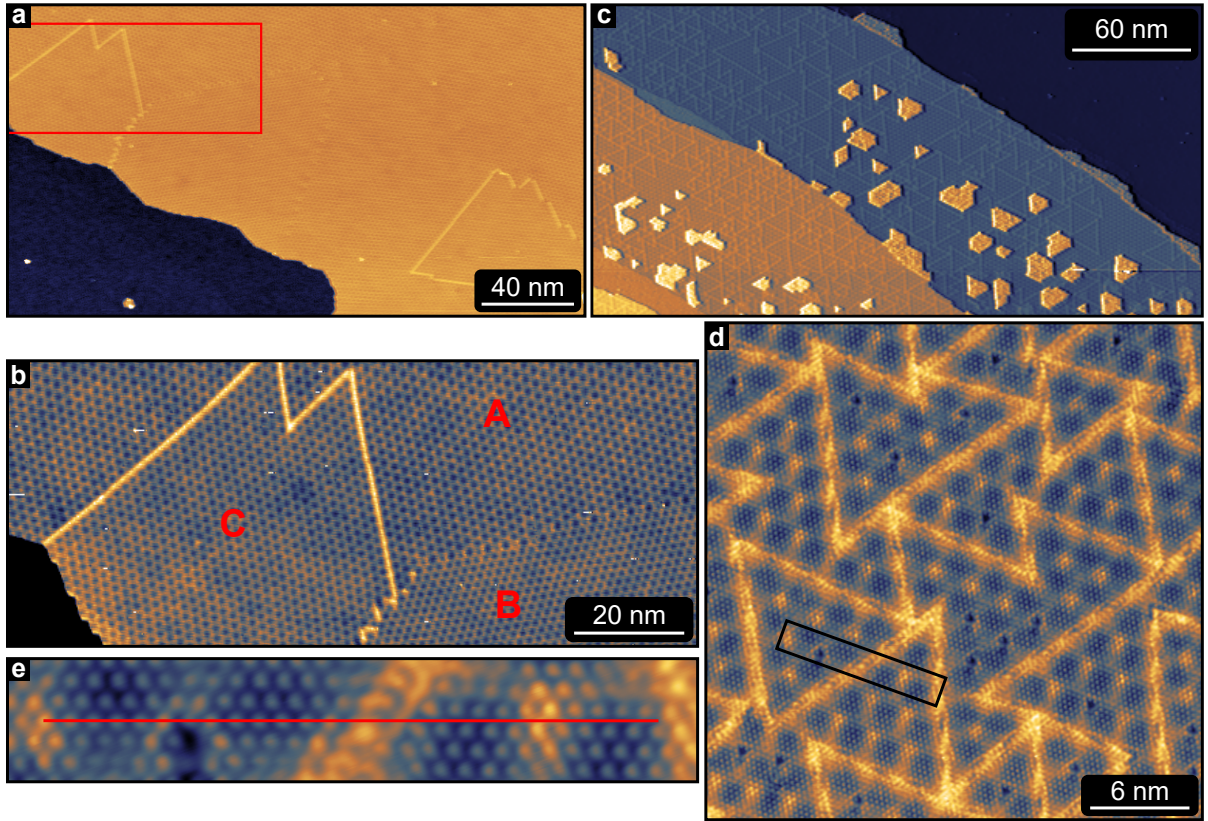


Fig. 59 **a**, STM overview image of a large FeO ML island. $U = 500$ mV, $I = 300$ pA. **b**, STM image of the area marked by the red rectangle in **a**. Three distinct domains are visible, separated by domain walls. $U = 500$ mV, $I = 500$ pA. **c**, Overview STM image of a different sample prepared in the same way as the one in **a** but showing many more domain walls forming triangular structures. $U = 800$ mV, $I = 200$ pA. **d**, Close-up atomic resolution image of a FeO region in the same sample as **c**. $U = -60$ mV, $I = 4$ nA. **e**, Close-up image of the area marked by the black rectangle in **d**. The red line marks a row of Oxygen atoms on the left domain. $U = -60$ mV, $I = 4$ nA. Measurement temperature: a-b: $T = 80$ K; c-d: $T = 8.4$ K.

The second type of domain wall between domain A and B appears to be much rarer, as it was only observed once and can be seen in the middle right of Fig. 59b. Here domains A and B exhibit differently sized and oriented Moiré unit cells. The lower domain B shows

a $\approx 13\%$ smaller Moiré lattice constant caused by a larger angle between the FeO and Ir unit cells than typical. The top domain A and bottom domain B exhibit an angle of $\approx 4.8^\circ$ between the FeO unit cells, with the Moiré unit cells rotated $\approx 30^\circ$ relative to each other. Modelling the Moiré structure, one finds an angle between the FeO and Ir lattice of $\approx 4.5^\circ$ in domain B, much larger than the $\approx 0.5^\circ$ angle observed in domain A, which falls within the typical range of $\approx \pm 0.5^\circ$.

The domain wall between domain A and B is much less bright at this bias voltage than the domain wall between domain A and C and the Moiré pattern appears to be less disturbed. Interestingly, the domain wall from domain B to domain C does show some bright line sections, which suggests that even for the domain boundary between strongly misaligned domains the phase-relation between the Moiré lattices on either side can result in both bright domain walls, where a smooth transition of the Moiré structure is not possible (B to C), and the much less visible domain walls, where such a smooth transition is possible (A to B). In general, across the FeO ML regions, small changes in the orientation of the Moiré unit cell can be observed, caused by small changes in the angle between of the FeO and Ir unit cells, in the range of $\pm 0.5^\circ$. However, no clear domain walls separating these regions are visible. Therefore, the domain wall on the right in Fig. 59b between domain A and B is likely only visible because of the uncommonly high rotation angle between FeO and Ir in domain B.

Another sample prepared in the same way as the one in Fig. 59a is shown in Fig. 59c. Here, one finds many triangles of bright lines on the FeO terraces. In a close-up atomic resolution image it becomes clear that different domains broadly have the same orientation of the rows of Oxygen atoms and Moiré spots, similar to the bright lines in the previous sample.

What caused two samples prepared in the same way to show such differences in the FeO ML growth? It appears that the FeO started growing from many more nucleation centres in the second sample, perhaps caused by a change in the oxidation temperature, although the magnitude of such a temperature change due to unavoidable small variations in the preparation process should be relatively small, which would indicate a large sensitivity to temperature.

The first possibility would be a difference in the Fe thin film morphology before the oxidation, where many small Fe islands could grow together into a closed FeO film during the oxidation process. For the Fe ML to grow as many small islands instead of fewer larger islands as is typical, would require significant contamination of the Ir surface; such contamination is unlikely unless something went wrong during the preparation, for which there are no indications.

A second possibility arises by comparison with the system of FeO on Pt(111), which has been studied more extensively. There, it has been shown that reducing the perfect FeO Moiré structure with atomic Hydrogen can result in triangles formed from Oxygen stacking-faults with Oxygen vacancies at the triangle edges [143, 146]. Inside these triangles, the Oxygen stacking changes, while the Fe lattice remains unperturbed. A closer look at the atomic rows across two of the domains reveals that a line located on an atomic row on the left domain misses the atomic row on the right domain, see Fig. 59e. Assuming that the observed atoms are indeed Oxygen, the experimental data is compatible with an Oxygen stacking change. However, in a close-up of the domain wall region, the exact atom

positions remain unclear. Perhaps, instead of a line of Oxygen vacancies, an additional row of atoms has been inserted into the lattice.

Measurements of the same FeO/Ir(111) sample in a different region with a different bias voltage are shown in Fig. 60a. At this bias voltage the domain walls appear as dark lines instead of the bright lines observed before. As the Oxygen atoms are still clearly visible in the rest of the image, it appears as if two rows of Oxygen atoms are spaced slightly further apart, as one would expect for an Oxygen dislocation line.

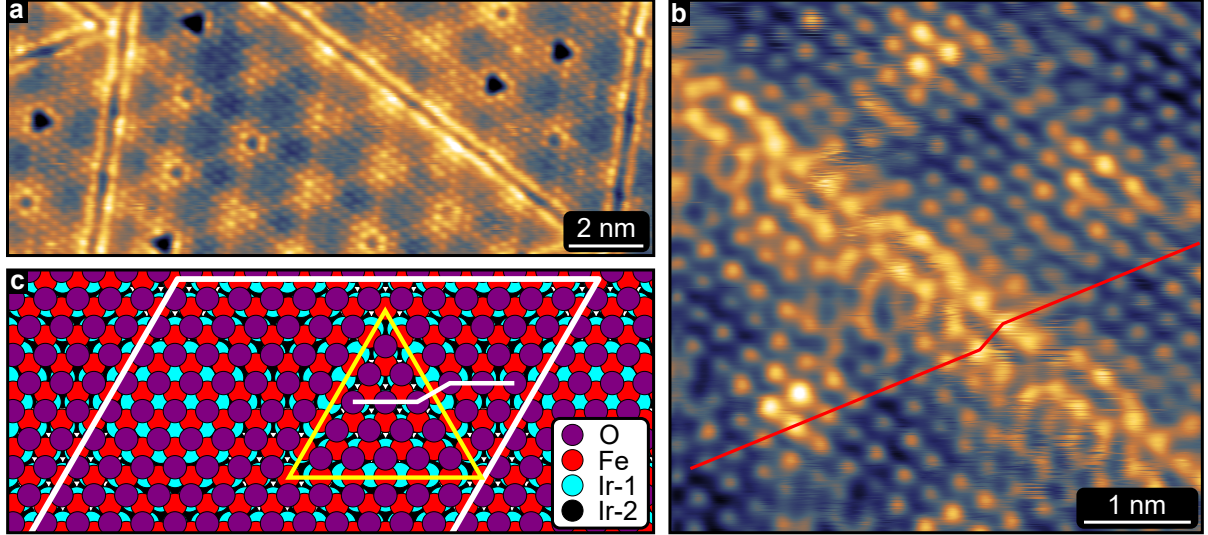


Fig. 60 **a**, STM image of a FeO/Ir(111) region hosting the triangular structures. $U = -1$ V, $I = 1$ nA, $T = 8.4$ K. **b**, Close-up STM image of a domain wall at the edge of a triangle. The red line follows the atomic row from one domain to the other. $U = -40$ mV, $I = 3$ nA, $T = 8.4$ K. **c**, Sketch of an Oxygen dislocation line as described by Merte and co-workers [146].

A closer view with atomic resolution and a different micro-tip of the STM reveals more information, as the atoms are more clearly visible at the domain walls, see Fig. 60b. If one follows one atomic row from one domain to another, one finds the atomic row offset by approximately half a row distance, as indicated by the red line. This is close to the $\approx 2/3$ of a row distance one would expect for such a dislocation line (compare to the white line in the 2D sketch of such an Oxygen stacking change in Fig. 60c).

The smallest possible dislocation configuration, enclosing only a single Fe fcc or Fe hcp region, is shown in the sketch of Fig. 60c and marked by a yellow triangle. This configuration is not observed. Instead the smallest observed triangle region appears to enclose one region of top-site Fe in the centre together with 3 regions of either fcc or hcp hollow-site Fe stacking. This configuration was labelled as a large dislocation loop by Knudsen *et al.* [143].

Although Merte and co-workers caused the formation of the dislocation loops by reducing FeO on Pt(111), it may be possible to achieve the same result by not providing enough Oxygen during the oxidation stage. Looking at larger domains and the fewer domain walls in the previous sample with atomic resolution, see Fig. 59a-b, evidence of the Oxygen lattice being shifted by approximately half an atomic row is found. Therefore, the difference in the density of these domain walls in these two samples could be directly

related to the Oxygen coverage and could be explained by small changes in the Oxygen partial-pressure during preparation.

Perhaps additional evidence for the lack of Oxygen is the presence of what are likely isolated Oxygen vacancies in the sample with the many dislocation lines, see Fig. 60a. One can observe both single Oxygen vacancies and three Oxygen atom vacancies, both are positioned at the same location of the Moiré pattern every time, which likely correspond to the top site positions.

These dislocations may also be caused by a change in the temperature during oxidation, as perhaps a small variation in temperature results in both Oxygen stackings growing, instead of mainly one. Small temperature changes can also not be excluded during our preparation as they can be induced by changes in the sample position or changes in pressure, which affects electron-beam heater performance when operated at a constant filament current and at a constant electron acceleration voltage.

One last option to consider is the possibility that the atomic resolution in Fig. 59d-e or Fig. 60a-b depicts not Oxygen atoms but Fe atoms instead, with the stacking fault occurring in the Fe layer and the Oxygen layer either remaining undisturbed or exhibiting the same change in stacking. Perhaps such a change is caused by an attempt to avoid Fe atoms on top-sites. While this possibility cannot be excluded, the more likely explanation is provided by the Oxygen dislocation lines and stacking changes, as these have been observed before for similar systems and can be more easily explained regarding our sample preparations.

The magnetism in the FeO ML has been investigated using SP-STM with antiferromagnetic Cr-bulk tips. As the Oxygen layer is located on top, magnetic contrast needs to emerge from the Fe layer below, which should be achievable in principle as atomic resolution of the Fe layers is possible. Measurements of the FeO with many different micro-tips, that were changed by poking in to the substrate or by voltage pulses, however, have not been able to reveal a magnetic contrast visible to the naked eye. An antiferromagnetic state with a small unit cell like a row-wise AFM state, a Néel state or a 3Q state should be distinguishable from the modulation of the Moiré pattern; however, the intrinsic intensity modulations in the FeO layer, see for instance Fig. 60b, make it more challenging and a weak magnetic signal may not be noticeable by eye.

To investigate this, 2D-FFTs of STM images taken on different samples with different micro-tips and at different bias voltages were calculated. For some of these images, additional spots in the FFT image are revealed, corresponding to a $(\sqrt{3} \times \sqrt{3}) - R30^\circ$ -superstructure, see the STM image and the corresponding calculated 2D-FFT image in Fig. 61a-b. The fact that the presence of the spots was tip dependent is a good indication of its magnetic origin. A magnetic $(\sqrt{3} \times \sqrt{3}) - R30^\circ$ -superstructure would correspond to an antiferromagnetic Néel state with 120° angles between neighbouring spins, see Fig. 7. This result would be consistent with theoretical investigations of magnetism in the system of FeO on Pt(111), which have shown a preference for antiferromagnetic states [147].

A reconstructed real-space image using only the frequencies in and around the superstructure spots in the FFT was calculated and overlaid on top of the original STM data. One finds that the maxima of this superstructure contrast are located not on the centre

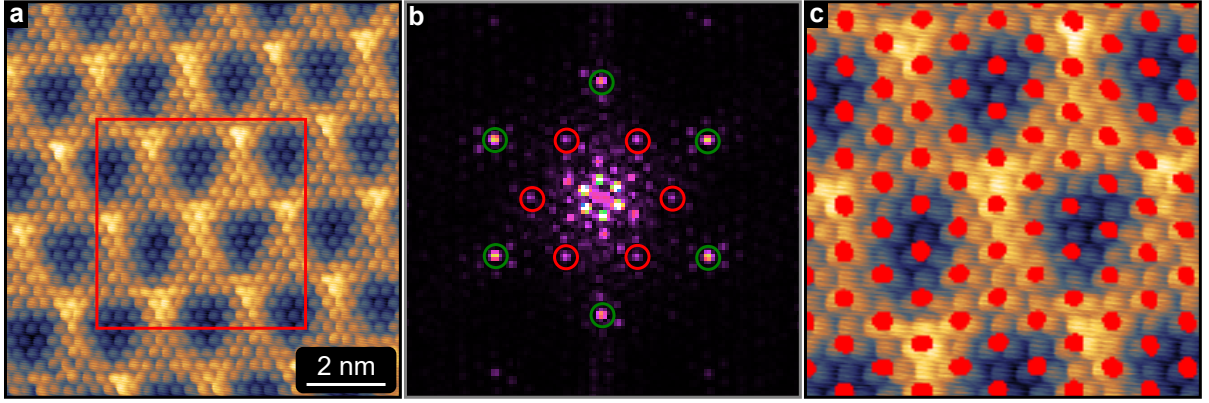


Fig. 61 **a**, Atomic resolution STM image of a FeO ML region. $U = 20$ mV, $I = 4$ nA, $T = 8.4$ K. **b**, FFT of the image in **a**. The green circles indicate the spots of the atomic lattice while the red spots indicate a $(\sqrt{3} \times \sqrt{3}) - R30^\circ$ -superstructure. **c**, Zoom-in of the region marked by the red rectangle in **a**. A reconstructed real-space image of the six superstructure spots indicated by the red circles in **b** is overlaid onto the image.

of the atoms but near the hollow-site positions, suggesting this contrast originates from the layer below, see Fig. 61c. This is consistent with our previous assumption that the observed atoms correspond to Oxygen atoms and provides additional evidence that our interpretation of triangular structures corresponding to Oxygen dislocation lines is valid.

While in principle the FeO ML does show signs of magnetism in the Fe layer, the intensity of the magnetic signal is very weak. However, perhaps by chance, the in-plane component of the tip-magnetization was small in all cases and a stronger magnetic contrast is possible for in-plane tips. Nevertheless, the observed magnetic contrast is too weak to allow for visual feedback during measurement, making the experimental investigation of changes, caused for instance by an electric field, quite difficult. This magnetic contrast was observed for different micro-tips both at 4.2 K and 8.4 K.

A.2 CoO/Ir(111)

Approximately 0.25 ML of Co were deposited on Ir(111) and then oxidized at the same temperature used for the Fe film. The resulting sample shows Co islands that appear to be only partially oxidized, see Fig. 62a. On the Ir substrate and unoxidized Co areas, dark contaminations, which are attributed to residual Oxygen, can be seen. At the edges of the Co island, one finds regions with larger apparent height which host a hexagonal superstructure, see dark spots in Fig. 62b. However, this Moiré superstructure does not appear well ordered.

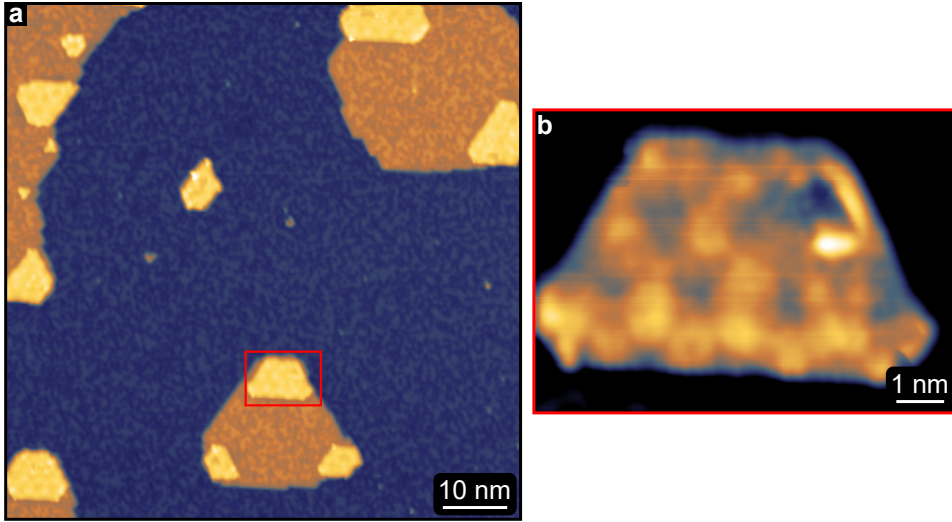


Fig. 62 **a**, STM overview image of ≈ 0.25 ML of Co on Ir(111) after oxidation at ≈ 1 W on the e-beam stage. $U = 405$ mV, $I = 400$ pA. **b**, STM image of the region marked by the red rectangle in **a**. $U = 15$ mV, $I = 500$ pA. Both: $T = 4.2$ K.

To increase the quality of the oxide, the temperature during the oxidization step is raised by increasing the power from ≈ 1 W to ≈ 4 W on the e-beam stage. The resulting sample shows large flat terraces with a small strip of oxide, which hosts a periodic pattern of dark spots, see Fig. 63a-b. Assuming this strip is the CoO, the coverage is much lower than expected and may indicate desorption of the Co or intermixing with the Ir.

The dark spots are arranged in a hexagonal lattice with a lattice constant of ≈ 555 pm, which is approximately two times the Ir lattice constant. This would correspond to a $p(2 \times 2)$ -superstructure and is also confirmed by LEED, see Fig. 63e. A closer look at the atomic structure reveals a honeycomb lattice of bright atoms, see Fig. 63c, with a distance between atoms of ≈ 325 pm, corresponding to $\approx 1.2 \times a_{\text{Ir}}$, which can only represent one atom sitting in one stacking and the other atom sitting in the second-nearest-neighbour hollow-site of the other stacking. This is also in agreement with the difference in apparent height between neighbouring atoms; one appears slightly brighter than the other, see Fig. 63c.

It remains unclear if the observed atoms are Oxygen or Co here, however, as one Co stacking is dominantly found for Co ML islands on Ir(111) [148], one can construct a structure model that fits the experimental data with the assumption that the Oxygen

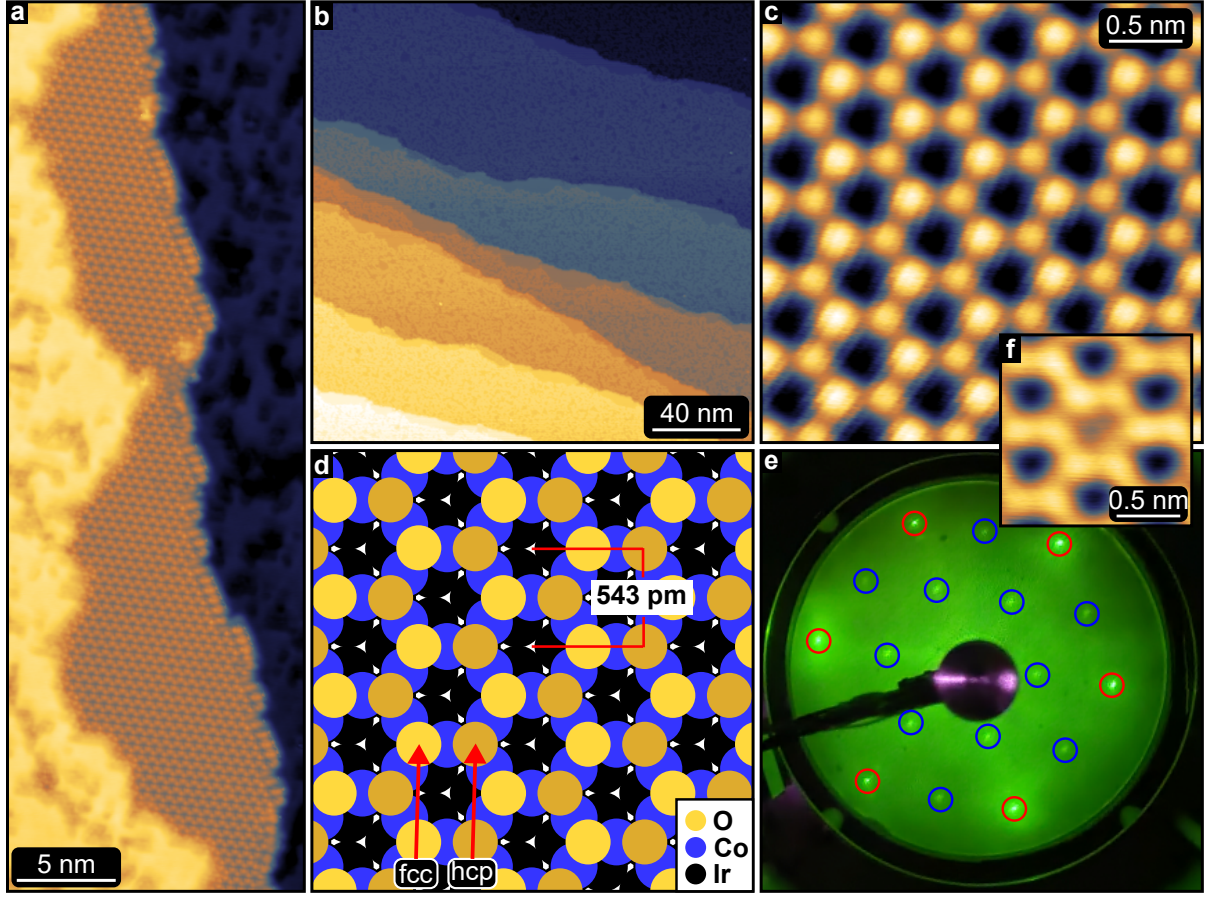


Fig. 63 **a**, STM image of a Co oxide strip, after oxidation at ≈ 4 W on the e-beam stage, showing a periodic pattern of dark spots. $U = 15$ mV, $I = 300$ pA. **b**, STM overview image of the region surrounding the scan area in **a**. $U = 605$ mV, $I = 200$ pA. **c**, Close-up image of the periodic pattern seen in **a**. $U = 55$ mV, $I = 300$ pA. **d**, Sketch of the proposed structure model. **e**, LEED image of the sample shown in Fig. 64. The red/blue circles indicate the LEED spots corresponding to the atomic lattice of Ir and the two rotational domains of the $p(2 \times 2)$ -superstructure, respectively. **f**, Close-up image of a hexagon of dark spots, with the middle spot showing a different contrast to the others. $U = 12$ mV, $I = 600$ pA. All: $T = 4.2$ K.

atoms are visible, see Fig. 63d. The Co atoms stay in the same hollow-site positions as the unoxidized film with a Co atom missing in the centre of the Oxygen honeycomb structure. This fits the experimental data because two different contrast levels are observed inside the Oxygen honeycomb, see Fig. 63f, one of which is quite rare and is interpreted as a Co atom filling the hole in the Co lattice. The Oxygen atoms sit in hollow-sites of both stackings and form the honeycomb lattice.

This structure is similar to the one proposed by De Santis *et al.* [149] for the CoPt-surface-alloy-oxide, which they call the quasi- (3×3) -phase, with the difference that the superstructure in our system is exactly (2×2) as no spots other than the (2×2) -superstructure and atomic spots are visible in the LEED images, while theirs is expanded by $\approx 10\%$ in the $[0\bar{1}1]$ direction.

An additional sample was prepared with an increased expected Co coverage of $\approx$

0.4 ML and a higher oxidation temperature at a power of ≈ 8 W on the e-beam stage, see Fig. 64a. Here, a stepped surface with a low coverage of islands growing from the step edges can be observed. The CoO coverage once again appears lower than expected. A close-up image of one of the hexagonal islands reveals straight edges running along the close-packed row directions. The straight boundaries to the Ir step edges, see red arrows in Fig. 64b, are a clear indication of intermixing, as a step edge of a Ir(111) surface does not naturally look like that.

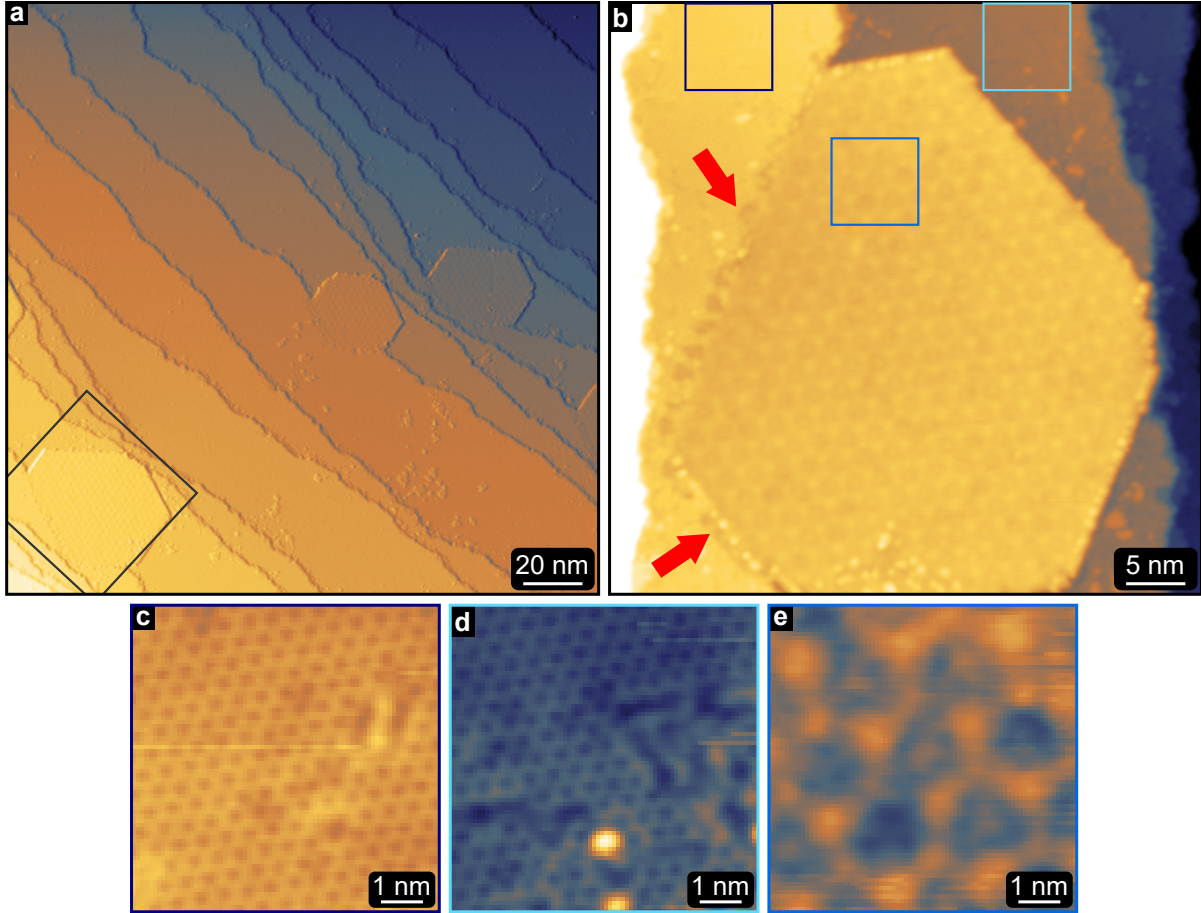


Fig. 64 **a**, Partially differentiated STM overview image of Co/Ir(111) after oxidation at ≈ 8 W on the e-beam stage. $U = 605$ mV, $I = 150$ pA. **b**, STM image of the region marked by the black rectangle in **a**. $U = 605$ mV, $I = 150$ pA. **c-e**, Cropped images of the areas marked by the respectively coloured rectangles with adjusted colour contrast. All: $T = 4.2$ K.

The hexagonal islands show a hexagonal Moiré superstructure with a distance between dark spots of ≈ 2 nm, see Fig. 64e. Interestingly, one finds the same $p(2 \times 2)$ superstructure on all measured terraces surrounding the hexagonal islands, see Fig. 64c-d, indicating that this oxide system indeed forms from a CoIr-surface-alloy. It appears that the entire Ir crystal surface hosts this $p(2 \times 2)$ structure and this is interpreted as the entire substrate surface forming a CoIr-oxide. The hexagonal islands should therefore correspond to a higher-layer CoIr-oxide configuration. This strong evidence for alloy formation requires a reconsideration of the $p(2 \times 2)$ superstructure. The two contrast levels in Fig. 63f could

now also be explained by a closed lattice of CoIr-alloy without holes where in some cases the wrong atom species is inserted as a fault. The differentiation from the structure model in Fig. 63d may not be possible via STM.

Another possibility to consider is that the observed structure in Fig. 63a and Fig. 64c-d is not a CoIr-alloy-oxide but just the CoIr-alloy, however, this seems less likely, as the apparent height compared to the substrate terraces is strongly reduced due to a decrease in conductance, similar to what was observed for FeO.

Looking back at the first preparation with the lowest oxidation temperature, where a Moiré pattern is observed on the regions growing near the island edges, see Fig. 62, the Moiré pattern appears very similar to the one in the high temperature preparation, see Fig. 64b,e. No contrast differences between the islands and the substrate can be observed for any of the bias voltages used, and one can therefore conclude that the intermixing was likely already occurring for this sample and the temperature was insufficient to form the $p(2 \times 2)$ -oxide structure.

The CoIr-alloy-oxide was investigated with regards to magnetism using a Cr bulk tip. Many tip changes were made using bias pulses and tip-surface collisions, but they did not reveal any contrast that could be assigned a magnetic origin. The CoO monolayer was not investigated due to intermixing, however, such a sample may in principle be possible to create by oxidizing at a temperature where no intermixing occurs and post-annealing to a higher-temperature if necessary, similar to the procedure for CoO on Pt(111) [149].

A.3 MnO/Ir(111)

To investigate the MnO monolayer on Ir(111) ≈ 0.75 ML of Mn was deposited, followed by annealing at the same temperature used for the FeO films (i.e. ≈ 1 W on the e-beam stage) in an Oxygen atmosphere. The resulting sample is shown in Fig. 65a, where at first glance only small islands can be found, indicating a much lower coverage than expected.

The surface between the islands on the large terraces reveals a stripe-like structure and three rotational domains, see Fig. 65c. The formation of a uniaxial superstructure is reminiscent of MnO on Pt(111), which forms a uniaxial structure as well [150], and therefore this structure likely also corresponds to MnO in our system. Some regions without this uniaxial superstructure show a larger apparent height, which can be interpreted as remaining unoxidized Mn, see red arrows in Fig. 65c.

Atomic resolution imaging reveals an expanded MnO lattice resulting in a periodic superstructure with the unit cell marked by the white lines, see Fig. 65b. The unit cell deviates from a perfectly hexagonal Moiré unit cell as seen for FeO on Ir, which indicates different MnO expansion rates in different crystallographic directions. Atomic distances vary in the range of $\approx 315 - 335$ pm. Additionally, the atomic distances are not the same throughout the unit cell, instead regions with larger atom separation near the black zig-zag lines, and smaller atom separation in the zig-zag regions between dark lines can be observed. This leads to the speculation that the dark lines represent the FeO lattice near unfavourable top and bridge sites. Interestingly, there is no evidence of a stacking difference between regions separated by dark lines.

If one assumes that this structure is the MnO ML then the observed coverage is > 1 ML as the MnO fully wets the Ir substrate and no regions exposing the bare Ir surface were observable, except for a single small hole in the MnO layer. If one accounts for the observed expansion of the MnO by assuming an average atom distance of 325 pm (19.5% expansion) our expected coverage was ≈ 0.9 ML. Three possible explanations for this discrepancy exist:

Firstly, the Mn could desorb from the Ir substrate at the oxidation temperatures. Such behaviour was found at elevated temperatures for the system of Mn/Re(0001). However, this would mean that the observed superstructure formed on the bare Ir substrate and this was not observed for the FeO preparation at the same temperature. Therefore, this explanation is considered unlikely. The second possibility involves the intermixing of the Mn and Ir substrate and the formation of a surface-alloy similar to what was found for Co on Ir. This appears to be the most likely option. Thirdly one must also consider the possibility of an unintended change occurring during the sample preparation, like a mistake or a sudden change in the evaporation rate. While this cannot be excluded, there is no evidence for such an event and the evaporation rate in preparations after this date on other samples was as expected.

The magnetism of this oxide was also investigated using an antiferromagnetic Cr bulk tip and no evidence of magnetism was found for any micro-tips produced with bias pulses or tip-sample collisions. Here, one has to consider the fact that this structure is more complex than the ones observed for FeO and CoIrO and even analysis using a 2D-FFT of the STM data did not provide any more insights. Additionally, the measurements did likely not reveal the pure MnO ML film, which could have different magnetic properties.

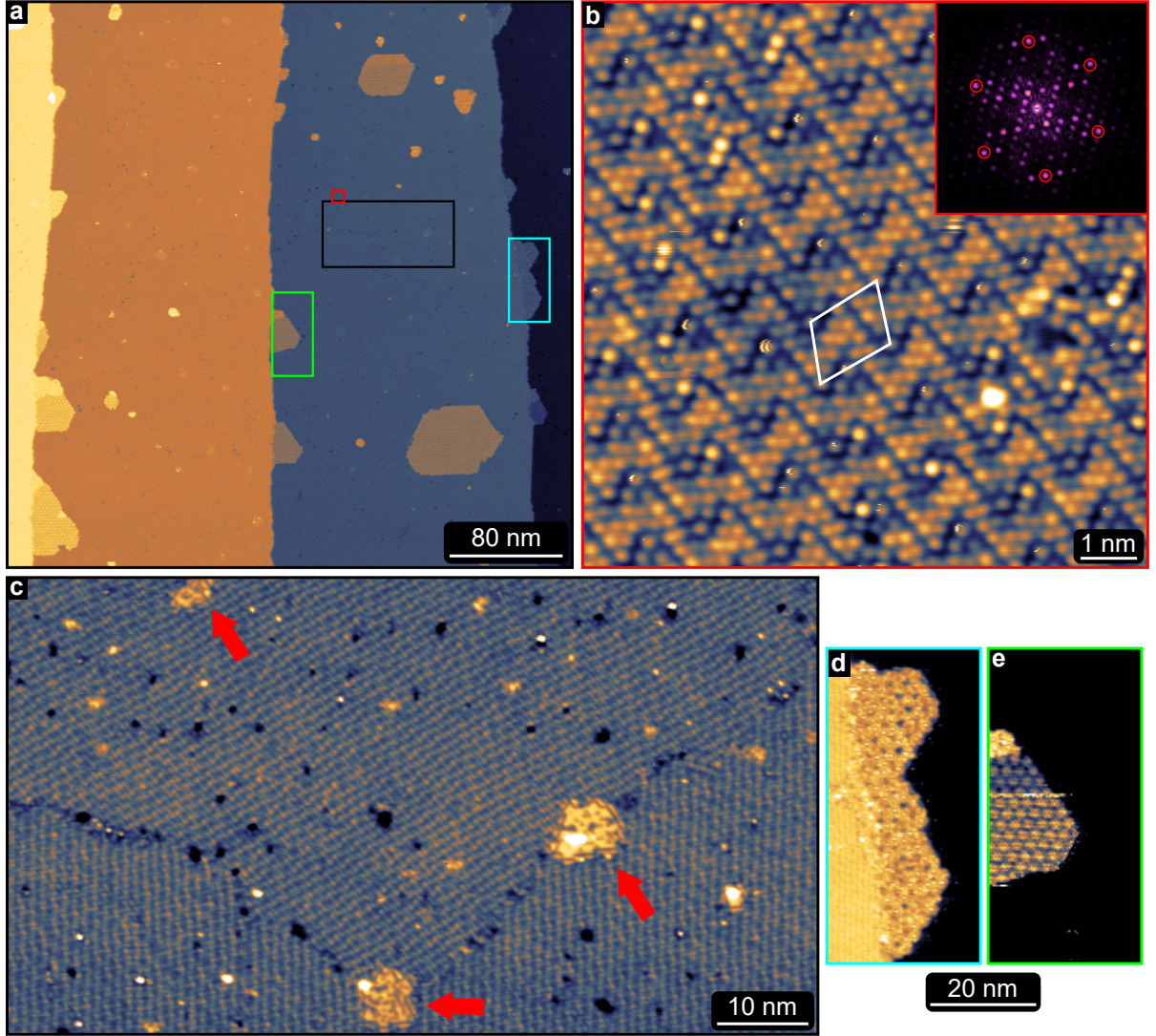


Fig. 65 **a**, STM overview image of ≈ 0.75 ML Mn deposited on Ir(111) and then oxidized at ≈ 1 W on the e-beam stage. $U = 600$ mV, $I = 500$ pA. **b**, Atomic resolution image of the region marked by the red square in **a**. The white lines indicate the unit cell of this superstructure. Inset shows FFT with red circles marking atomic spots. $U = 50$ mV, $I = 1$ nA. **c**, STM image of the region marked by the black rectangle in **a**. Red arrows indicate unoxidized Mn regions. $U = 500$ mV, $I = 800$ pA. **d-e**, Close-up image of two higher oxide layer structures marked by the respectively coloured rectangle in **a**. $U = 500$ mV, $I = 500$ pA. all: $T = 4.2$ K.

Appendix B Fe-, Co-, Mn-thin films on Ir(001)

Magnetic ultra-thin films like Fe, Co and Mn have been investigated extensively on the Ir(111) surface. They grow pseudomorphically and their magnetic states are known. For similar magnetic ultra-thin films on the square Ir(001) surface experimental results regarding magnetism are still lacking. Theoretically calculations predict interesting non-coplanar magnetic states for a Fe ML on Ir(001) [151, 152].

B.1 Lifting the (1x5)-reconstruction on Ir(001)

After a general cleaning procedure including Ar ion sputtering, Oxygen annealing, and a final flash to $\approx 1500^\circ\text{C}$, the Ir(001) surface is found to be reconstructed in a (1×5) configuration, with a periodic buckling of the Ir surface atoms, see Fig. 66a. A clean and unreconstructed (1×1) -surface can be achieved by first lifting the reconstruction by annealing and cooling in an Oxygen atmosphere followed by the removal of Oxygen using Hydrogen [153–155].

As lifting the reconstruction results in Ir atoms being expelled from the surface layer, a lower annealing temperature results in Ir islands, while a higher temperature is needed to create large flat Ir terraces. The upper temperature limit is given by the onset of Oxygen desorption which starts at $T > 750\text{ K}$ [156]. This is lower than the onset of the transition from $(1 \times 1) \rightarrow (1 \times 5)$ which starts at $T > 800\text{ K}$ [154]. After annealing the Ir(001) crystal at $\approx 4\text{ W}$ on the e-beam stage, then increasing the Oxygen partial pressure to $5e-7\text{ mbar}$, followed by cooling down the sample in the Oxygen atmosphere, a $p(1 \times 2)$ -Oxygen reconstruction is observed on the Ir terraces. On the sample, lines of Oxygen separated by two lattice constants can be seen, resulting in two rotational domains on the surface, see Fig. 66b. The bright defects are interpreted as vacancies in the Oxygen reconstruction.

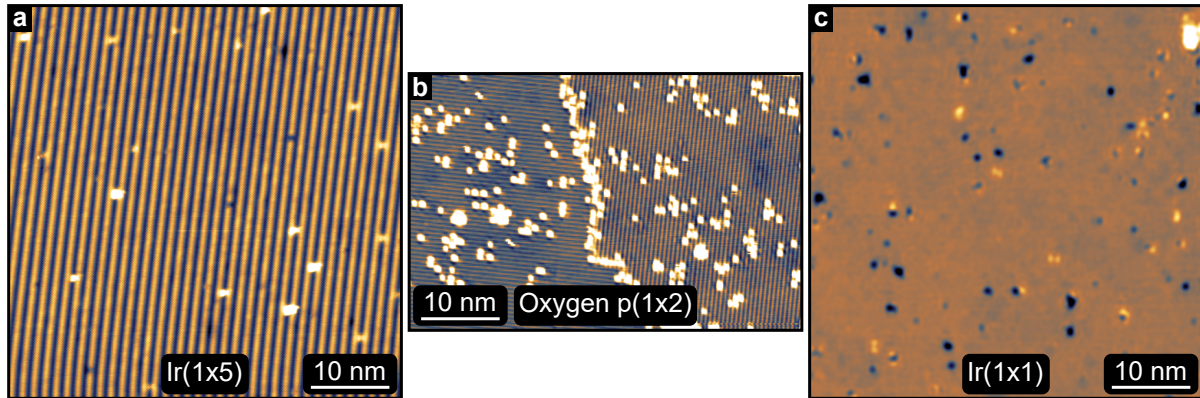


Fig. 66 **a**, STM image of the (1×5) reconstructed Ir(001) surface. $U = 40\text{ mV}$, $I = 1\text{ nA}$. **b**, STM image of the $p(1 \times 2)$ Oxygen reconstruction of the Ir(001) after annealing in Oxygen. $U = 200\text{ mV}$, $I = 800\text{ pA}$. **c**, STM image of the unreconstructed Ir(001) surface after dosing Hydrogen at elevated temperature. High frequency noise was removed using a low pass filter. $U = 15\text{ mV}$, $I = 1\text{ nA}$. all: $T = 4.2\text{ K}$.

To remove this Oxygen reconstruction the crystal is heated to 360°C on a resistive

heater, with the actual sample temperature expected to be $50 - 100^\circ$ lower, and then molecular Hydrogen is dosed at $5 \cdot 10^{-7}$ mbar for 2 minutes. The result is an unreconstructed Ir(001) surface depicted in Fig. 66c. The Fe, Co, and Mn were then deposited 5 – 10 minutes later when the sample was still at an elevated temperature of $100 - 200^\circ\text{C}$.

B.2 Fe/Ir(001)

Approximately 0.6 ML of Fe are deposited on the unreconstructed Ir(001) surface resulting in the Fe film shown in Fig. 67a. Large, sometimes interconnected Fe islands are separated by regions of the Ir substrate covered with what is expected to be Fe clusters as well as some areas showing dark patches which could be remaining Oxygen or Hydrogen. A closer look reveals flat unreconstructed pseudomorphic Fe islands, see Fig. 67b and an atomically resolved (1×1) -structure in Fig. 67d. The Ir- (1×1) is visible in Fig. 67c with some clusters on top.

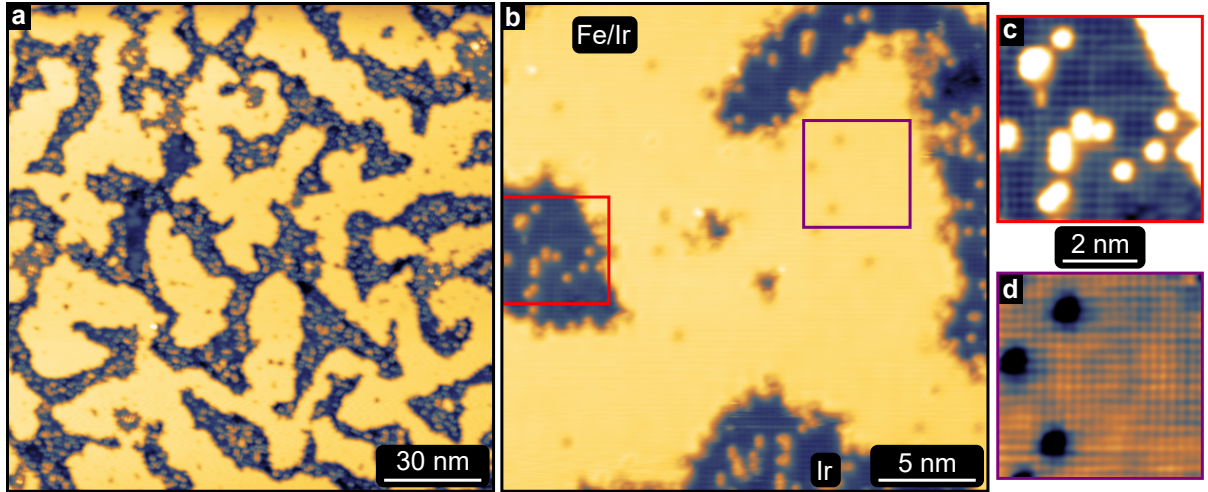


Fig. 67 **a**, STM overview image of ≈ 0.6 ML of Fe deposited on the unreconstructed Ir(001) surface. $U = 600$ mV, $I = 300$ pA. **b**, STM image of a Fe island showing a flat surface. $U = 5$ mV, $I = 1$ nA. **c-d**, Cropped regions of the image in **b** at the location marked by the coloured rectangles showing the atomic lattice. High frequency noise was removed. The colour scale was adjusted to improve contrast; all: $T = 4.2$ K.

The Fe ML was investigated regarding magnetism using a Cr bulk tip, but no evidence of any periodic magnetic pattern was found for any of our many different micro-tips created by bias pulses and tip-sample collisions. Additionally, dI/dU -maps obtained with a range of different bias voltages ($U = 200, 400, 600, 800, 1000, -200, -400, -600, -800$ mV) did not show any indications of ferromagnetism.

However, this is no proof of a non-magnetic Fe layer, as the particular STM tip used could for instance host a large amount of non-magnetic material near the tip apex. Perhaps at the bias voltages used no contrast between different ferromagnetic domains is visible, or due to the interconnected nature of the film the observed Fe regions are all hosting the same ferromagnetic domain. Additional measurements with different magnetic tips

are needed, potentially including measurements with a spin-polarized tip characterized on a different, well-known sample.

B.3 Co/Ir(001)

The growth and magnetism of Co was also investigated on this substrate. In Fig. 68a a sample of ≈ 0.6 ML of Co on Ir(001) is shown and a network of connected Co islands can be seen, similar to the growth of the Fe film. However, in between the islands one finds the bare Ir substrate with no additional clusters on top. The circular patches of larger apparent height are likely Carbon impurities, as due to bad thermal contact between the Ir(001) single crystal and the sample holder, the desired temperatures during Oxygen annealing and the final annealing step was not achieved. The increased amount of Carbon impurities aggregated into large, roughly circular patches, see cyan circles in Fig. 68a, and appears to not have affected the Co growth process.

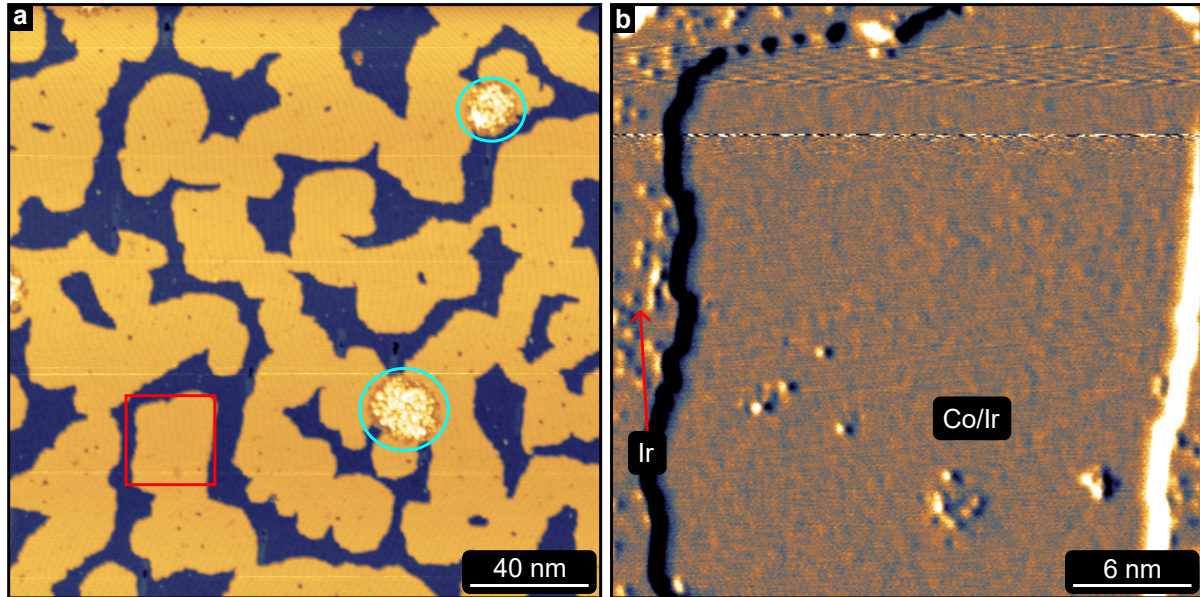


Fig. 68 **a**, Overview STM image of ≈ 0.6 ML of Co deposited on the unreconstructed Ir(001) surface. Cyan circles mark Carbon impurities. $U = 805$ mV, $I = 200$ pA. **b**, STM current map of the region marked by the red rectangle in **a**. $U = -15$ mV, $I = 1$ nA; all: $T = 4.2$ K.

A close look at one of the Co islands reveals a flat unreconstructed surface, see current map in Fig. 68b. However, similar to the Fe film no indication of a periodic magnetic signal was found, which would indicate a non-collinear or antiferromagnetic state. Additionally, no evidence for ferromagnetism was observed for dI/dU -maps at bias voltages ($U = 200, 800, -100, -200, -300, -400$ mV). Please refer to the end of the previous section on Fe as to what conclusions can and cannot be drawn from this.

B.4 Mn/Ir(001)

As a next step, ≈ 0.55 ML of Mn were deposited on the Ir(001) substrate, resulting in a thin film of interconnected Mn islands similar to Fe and Co with a higher than expected coverage of ≈ 0.8 ML, see Fig. 69a. On the surface of the Mn areas, dark lines can be observed in some regions, appearing in two different orientations due to the broken four-fold symmetry. A close-up STM image of a Mn region is shown in Fig. 69b and it becomes clear that the dark lines in Fig. 69a correspond to regions with no superstructure, while the bright areas correspond to regions with a uniaxial superstructure. The bright lines are spaced ≈ 580 pm apart, which corresponds two $\approx 1.5a_{\text{Ir}}$.

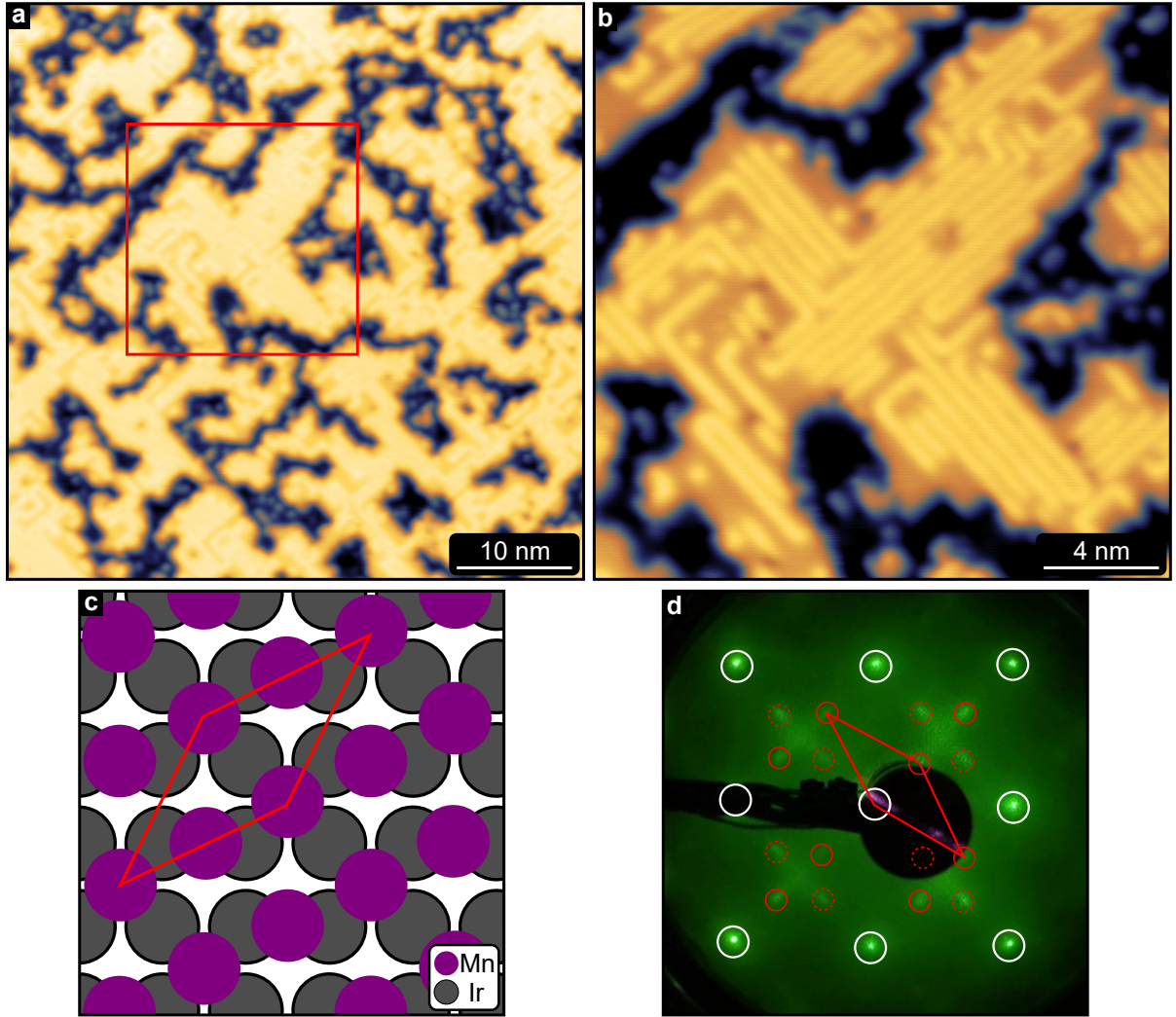


Fig. 69 **a**, Overview STM image of ≈ 0.8 ML of Mn deposited on the unreconstructed Ir(001) surface. $U = 205$ mV, $I = 400$ pA. **b**, STM image of the region marked by the red rectangle in **a**. $U = -15$ mV, $I = 300$ pA. **c**, Potential structure model with 33% lower Mn density with the unit cell marked by red lines. **d**, LEED image of a Mn/Ir(001) sample. White circles mark the atomic spots, and red/dashed-red circles mark the spots of the two rotational domains of the $c(3\sqrt{2} \times \sqrt{2}) - R45$ reconstruction; all: $T = 4.2$ K.

With the assistance of LEED measurements, see Fig. 69d, one can conclude that this superstructure is a $c(3\sqrt{2}\times\sqrt{2})-R45$ reconstruction of the Mn layer. If the superstructure was of magnetic origin, one would expect it not to appear in LEED measurements. Our higher than expected coverage could be explained by a $\approx 33\%$ lower Mn density. A tentative structure model with this density and the correct unit cell size and symmetry is shown in Fig. 69c. No additional contrast on the Mn islands, that could be attributed to a magnetic superstructure, was observed in any of our measurements.

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Publications

Anisotropic atom motion on a row-wise antiferromagnetic surface

Felix Zahner, Soumyajyoti Haldar, Roland Wiesendanger, Stefan Heinze, Kirsten von Bergmann, and André Kubetzka
Nature Communications **16**, 4942 (2025).

Reciprocity of Magnetism and Nanostructure Growth

Felix Zahner, Kirsten von Bergmann, Roland Wiesendanger, and André Kubetzka
submitted for peer-review. Preprint available: [arXiv:2509.03202](#).

Spin-polarized edge modes between different magnet-superconductor-hybrids

Felix Zahner, Felix Nickel, Roberto Lo Conte, Tim Drevelow, Roland Wiesendanger, Stefan Heinze, and Kirsten von Bergmann
Nature Communications **17**, 3457 (2026).

Topological spin textures in an antiferromagnetic monolayer

Felix Zahner, Tim Drevelow, Roberto Lo Conte, Roland Wiesendanger, Stefan Heinze, and Kirsten von Bergmann
submitted for peer-review. Preprint available: [arXiv:2512.24186](#).

Further publications not part of this thesis:

Strain-driven domain wall network with chiral junctions in an antiferromagnet

Vishesh Saxena, Mara Gutzeit, Arturo Rodríguez-Sota, Soumyajyoti Haldar, Felix Zahner, Roland Wiesendanger, André Kubetzka, Stefan Heinze, and Kirsten von Bergmann
Nature Communications **16**, 10808 (2025).

Contributed Talks

Kicking Co and Rh atoms on the row-wise antiferromagnet Mn/Re(0001)

Felix Zahner, Soumyajyoti Halder, Roland Wiesendanger, Stefan Heinze, Kirsten von Bergmann, and André Kubetzka

DPG Spring meeting, Berlin, March 2024

Spin-polarized chiral edge modes in the topological nodal-point superconductor Mn/Ta(110)

Felix Zahner, Felix Nickel, Roberto Lo Conte, Tim Drevelow, Roland Wiesendanger, Stefan Heinze, and Kirsten von Bergmann

DPG Spring meeting, Regensburg, March 2025

Spin-polarized low-energy edge modes between different magnet-superconductor-hybrid systems in Mn/Ta(110)

Felix Zahner, Felix Nickel, Roberto Lo Conte, Tim Drevelow, Roland Wiesendanger, Stefan Heinze, and Kirsten von Bergmann

STM25 conference, Seoul, June 2025

Posters

The motion of atoms on an antiferromagnetic surface: Does the magnetic texture matter?

Felix Zahner, Soumyajyoti Halder, Roland Wiesendanger, Stefan Heinze, Kirsten von Bergmann, and André Kubetzka

International conference on Magnetism, Bologna, June-July 2024

Anisotropic motion and growth on the row-wise antiferromagnetic state on Mn/Re(0001)

Felix Zahner, Soumyajyoti Halder, Roland Wiesendanger, Stefan Heinze, Kirsten von Bergmann, and André Kubetzka

SPP skyrmionics conference, Kloster Seeon, September 2024

Acknowledgements

I want to acknowledge the help and support of many people, without whom this thesis would not have been possible.

First, I would like to thank Prof. Wiesendanger for allowing me to do my PhD in his group, and for his supervision.

A very big thank you also to Kirsten von Bergmann and André Kubetzka for their supervision and general support throughout the years, as well as for the many enlightening discussions.

I also want to mention all the former and current Lab013 members who made the lab what it is today. Thank you for the nice atmosphere, and for the many discussions in and around the lab. A special thanks to Roberto for dropping that one sample.

Further, I want to thank the entire group for the nice and generally very supportive work environment. A special mention is extended to Hajo Schröder and Michael Langer for the upkeep of the helium liquefier and the general support, as well as to the colleges from the mechanical workshop, who always did great work.

I want to thank Stefan Heinze and his group from the University of Kiel for the great collaborations.

Finally, I also want to thank my friends and family, who continuously supported me throughout the years. I especially want to mention my parents who have always believed in me and helped me in so many ways throughout my life.

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