Supplementary Material

Guiding spin spirals by local uniaxial strain relief

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S1. Tunneling spectroscopy measurement of the Fe-DL on Ir(111)

Fig. S1(a) shows the corresponding d*I*/d*U* map to the topography shown in Fig. 1(a) of the main text. A spatially resolved spectroscopy measurement has been performed in the area of the indicated square and selected bias voltage cuts are shown as insets to Fig. S1(a). The conductance maps of the reconstructed surface are strongly bias dependent, in line with the strong bias dependence of the topography images shown in Fig. 1(b),(c) of the main text. The tunneling spectra displayed in Fig. S1(b) are taken at h_1, bcc, and h_2 sites and reveal the electronic differences between these different stackings.

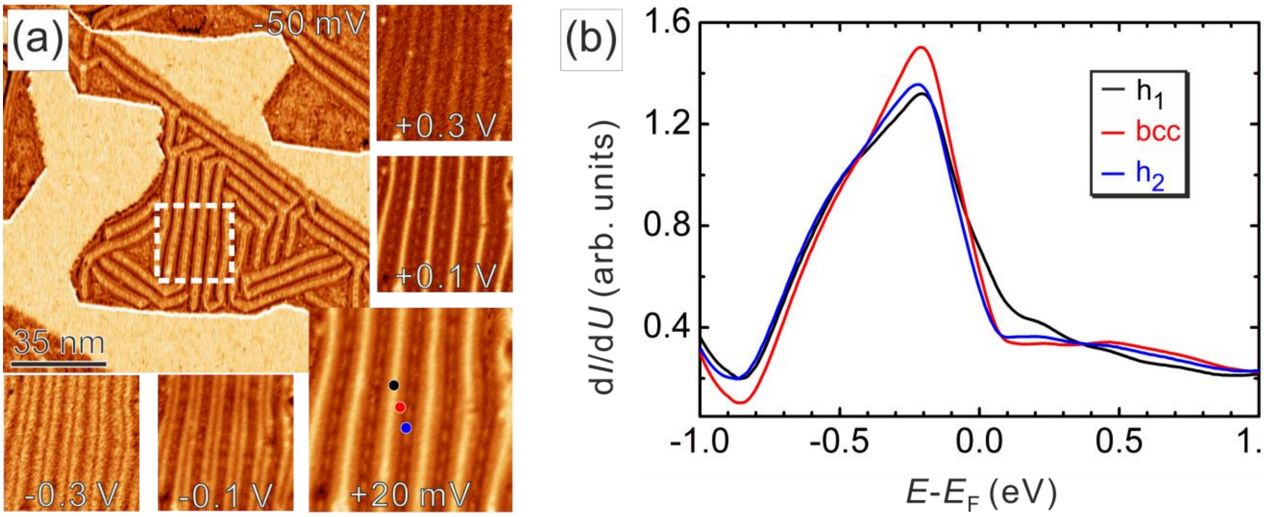

Figure S1. (a) Corresponding d*I*/d*U* map to the topography shown in Fig. 1(a) of the main text. Insets: bias dependent differential conductance maps on the reconstructed Fe-DL on Ir(111). (b) The corresponding tunneling spectra taken at h_1, bcc, and h_2 atomic sites (see colored dots in inset of (a)) reveal the electronic differences, which also lead to the strong bias dependence of topography images (spectroscopy set-point parameters: U = +1.0 V, I = 1.0 nA).

S2. Spin-averaged and spin-polarized measurements

Spin-averaged STM measurements are typically performed with non-magnetic metallic tips. Typical topography images of the reconstructed Fe-DL on Ir(111) obtained with a bare W tip are shown in Fig. S2. Direct comparison of the data acquired at zero magnetic field (a) and under magnetic field (b) of the same sample area shows no detectable change.

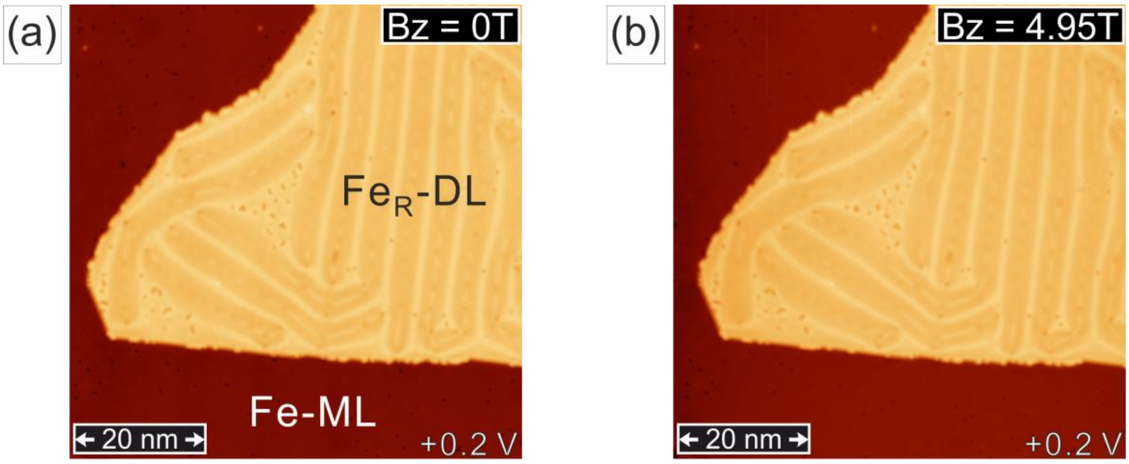

Figure S2. (a) Topography of the reconstructed Fe-DL on Ir(111) at zero magnetic field acquired with a bare W tip. (b) The same sample area measured under external applied field of B_z = 4.95 T using the same W tip. There is no detectable change observed by comparing the images shown in (a) and (b) (measurement parameters: I = 1 nA, T = 4.8 K).

Spin-polarized measurements require the use of spin-polarized tips. To disentangle the magnetic contrast from structural/electronic contributions, it is useful to perform spin-averaged and spin-polarized measurements at the same sample area. One way to do this is to use a tip that is (super-) paramagnetic without external magnetic field, but exhibits a stable spin-polarization in an applied field. This method has been demonstrated in previous studies [1,2], where a spin-polarized tip was prepared by picking up a single magnetic atom from the sample surface and applying an external magnetic field. At our measurement temperature of about 5 K also clusters of several magnetic atoms have a high switching rate of their magnetization without external magnetic field, but can be aligned in significantly smaller external magnetic fields compared to single atoms [3]. While such tips may exhibit magnetocrystalline anisotropy, in first approximation the Zeeman-energy is sufficient to align the tip magnetization with the external magnetic field.

For example, Fig. S3(a) shows a sketch of a spin-averaged STM measurement taken with a superparamagnetic cluster at the end of a non-magnetic tip. The rapid magnetization switching of the cluster in zero external field gives rise to a non-magnetic measurement due to the limited time resolution in typical STM experiments. This is equivalent to a measurement with a non-spin-polarized tungsten tip. However, when applying an external magnetic field, Fig. S3(b), the magnetization direction of a superparamagnetic cluster can be oriented along the field direction, allowing magnetic sensitivity to different components of the sample's local magnetization direction, e.g. by out-of-plane or in-plane magnetic fields, as depicted in Fig. S3(b).

The use of more magnetic material at the tip leads to spin-polarized measurements also in the absence of an external magnetic field. While a reference spin-averaged measurement is not possible in this case, it has been demonstrated that magnetic fields of 1 T are sufficient to align the magnetization of an Fe-coated W tip at 5 K [4]. The use of antiferromagnetic tip materials, e.g. Cr, leads to spin-polarized tips that are unaffected by external magnetic fields [5].

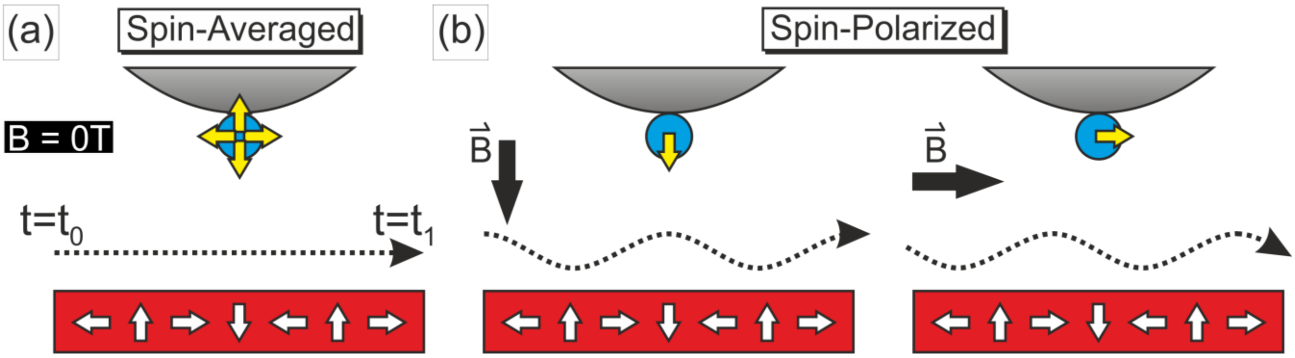

Figure S3. Schematics of SP-STM with a superparamagnetic cluster (blue) at the end of a non-magnetic tip (gray). (a) Due to rapid switching of the superparamagnetic cluster in zero field, the STM line profile measured from time t_0 to t_1 has no net contribution from the spin-polarized current. (b) In an external magnetic field, the magnetization direction of the cluster can be aligned, allowing magnetic sensitivity to different components of the sample's local magnetization direction.

For the measurements shown in Fig. 2 of the main text we have picked up some Fe with a W tip, leading to spin-averaged measurements at B = 0 T due to rapid magnetization switching of the Fe cluster. Figure S4(a)-(d) shows spin-averaged measurements obtained with this tip at B = 0T at four different bias voltages. In contrast to a measurement with a bare W tip of Fig. S2, the application of an external magnetic field of B = 4 T changes the appearance of the images, see Fig. S4(e)-(h). This change is due to the alignment of the magnetization of the Fe cluster at the tip with the external magnetic field, leading to a spin-polarized contribution to the tunnel current. Comparison between the spin-averaged and spin-polarized images shows that the observation of magnetic contrast of the spin spiral state is strongly bias- and stacking dependent, as anticipated from the spectroscopy measurement shown in Fig. S1.

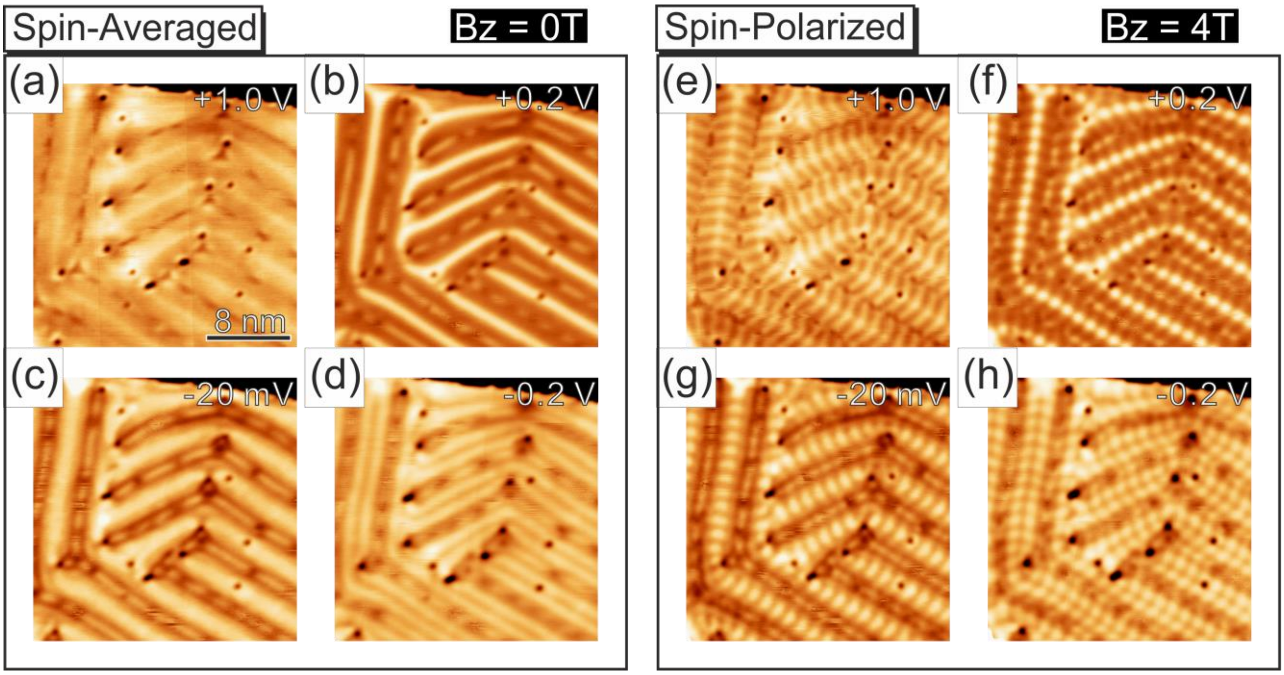

Figure S4. (a)-(d) Spin-averaged topography images taken with an Fe cluster/W tip at B = 0 T, showing also a strong bias dependence as found for the conductance maps in Fig. S1(a). (e)-(h) Spin-polarized measurements at the corresponding bias voltages performed at the same location at B = 4 T, where the Fe cluster at the tip is aligned with the applied magnetic field; a comparison of spin-averaged and spin-polarized measurements enables a disentanglement of the spin contrast from the structural/electronic contributions for the reconstructed Fe-DL.

Another way to obtain spin-averaged and spin-polarized images at the same sample position is an *in-situ* modification of the tip at a different sample location between measurements. This method was employed to obtain the images displayed in Fig. 4(a) and (b) of the main text, measured with a bare W tip and a tip that had picked up Fe from a position outside the image area. A similar procedure was performed to modify the magnetization direction of the bulk Cr tip between the images displayed in Fig. 3(c) and (d) of the main text. Such a bulk Cr tip leads to spin-polarized images also at $B = 0$ T, see Fig. 4(c) of the main text.

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