Supplementary: Correlation of Magnetism and Disordered Shiba Bands in Fe Monolayer Islands on Nb(110)

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Supplementary Note 1

Thin Fe films grown on oxygen-reconstructed Nb(110)

Figure S1a shows an STM image of a thin film of Fe grown on Nb(110) after the crystal was heated with a reduced power of only 350 W. After such a preparation of the Nb crystal, STM images taken before the Fe growth reveal that the surface is still covered by an oxygen-reconstruction. The 1st ML is not yet fully closed, and 2nd ML and 3rd ML islands are elongated along the [001]-direction. The Fe film has a characteristic reconstruction which is very reminiscent of the reconstruction that was observed in previous work using STM of room temperature deposited Fe on the (110) surface of Nb layers grown on Al₂O₃ or W(110).¹ This reconstruction is very different from all three reconstructions that are reported in the main manuscript where monolayers of Fe have been grown on the oxygen-free Nb(110) surface achieved by annealing the Nb crystal to higher temperatures with a power of at least 510 W. We, therefore, conclude that the previous work reports Fe grown on oxygen-reconstructed Nb(110), while we report in the main manuscript on Fe films grown on oxygen-free clean Nb(110).
Supplementary Note 2

Local spin-resolved spectra and hysteresis loops

In order to determine the bias voltage ranges with a considerable spin contrast for the SP-STS measurements, point spectra (Figures S2c,d and e,f) have been taken in the range of ±1 V at different positions of the islands (Figures S2a,b) and for different magnetic fields during a hysteresis loop (Figures S2g,h). The voltages subsequently selected for dI/dV maps were −400 mV, 200 mV, and 400 mV for the type I, 320 mV for the type II, and 375 mV for the type III MLs (see vertical dashed gray lines in the point spectra).

For the type I ML, there is a contrast reversal between $V = −400$ mV and $V = +200$ mV, as well as for both of these bias voltages between the stripe (Pos. 1) and flat (Pos. 2) areas on the reconstruction (c.f. Figures S2c,d). Furthermore, the hysteresis loops extracted from these two different areas also reveal an inverted behaviour (c.f. Figures S2g, upper
and lower panels). We accordingly assign these inversions to an inversion of the vacuum spin-polarization above the stripes with respect to the flat regions, but not to any kind of alternating domain structure (see also Supplementary Note 3). For the type III ML, both, the point spectra (Figures S2e,f), as well as the hysteresis loop (Figure S2h) hardly show any magnetic-field dependent contrast in the entire bias and magnetic field range, except for a region in the small magnetic field range \( B_z < 0.5 \, \text{T} \). We assign this to a very small coercive field of this type of reconstruction on the order of the coercive field of the used tip.

Figure S2: (a), (b) STM images of a type I and a type III ML island, respectively, indicating the locations where point spectra have been taken \((V = 400 \, \text{mV} \) (a), \( V = 375 \, \text{mV} \) (b) and \( I = 1 \, \text{nA} \)). (c), (d) and (e), (f) Point spectra taken at the different magnetic fields indicated in the legend in the order shown by the accordingly colored points in the hysteresis loops in (g,h) on the locations marked in (a,b) \(( I_{\text{stab}} = 500 \, \text{pA}, \, V_{\text{stab}} = 1 \, \text{V} \) and \( V_{\text{mod}} = 10 \, \text{mV} \)). (g), (h) Hysteresis loops extracted from the \( \frac{dI}{dV} \) signal averaged over the areas of the type I and type III islands shown in the insets. The colored points are the magnetic field values for which the point spectra in (c) to (f) have been taken.
Supplementary Note 3

Magnetic field dependent spin-resolved line spectra

In order to further investigate the lateral change in the spin-resolved contrast across the flat and stripe regions of the type I reconstruction, which is apparent from Figure 2a of the main manuscript as well as from Figures S2 c,d,g, we present here dI/dV line profiles extracted across several flat and stripe areas of a type I island while looping the external magnetic field and, thereby, reversing the island magnetization, see Figure S3.

Naively, one might conclude, that this change in the sign of the spin-polarization could as well be due to a reversal of the local magnetization between the flat and stripe regions, e.g., due to a non-collinear spin structure or very small anti-parallel domains. However, this scenario is highly unlikely due to the following reasons. (i) In case of a non-collinear spin structure imaged by magnetic field dependent SP-STS using a soft magnetic tip as the one in the present experiment, we would expect to observe lateral shifts of the maxima and minima of the dI/dV values measured across the flat and stripe regions as a function of the magnetic field.\(^3\) However, for the entire magnetization curve, none of such lateral shifts are observed, see Figures S3b,c. (ii) For a hypothetical collinear structure of anti-parallel domains, the sizes of the domains would be unreasonably small. Moreover, such domains should as well laterally shift with variations in the magnetic field, which is not observed. Therefore, the data presented in Figure S3 corroborates the conclusion, that the type I Fe ML is out-of-plane ferromagnetic, and that the sign reversal of the spin asymmetry from the stripe to the flat regions (Figure 2a of the main manuscript) is due to an inversion of the vacuum spin-polarization measured by SP-STS.
Figure S3: (a) dI/dV map of a type I ML island showing the lines and arrows along which the dI/dV line profiles in (b,c) have been extracted. (b), (c) dI/dV line profiles along the lines in (a) plotted in normal and waterfall fashion, respectively, from the dI/dV maps recorded during a hysteresis loop at the external magnetic field values given by the color scale on the right. Each dI/dV line profile is an average of three individual profiles positioned parallel, side by side (green, blue and red lines in (a)).

Supplementary Note 4

Monte Carlo simulations

In order to rationalize the measured magnetization curves of the three different types of Fe islands we performed atomistic Monte Carlo simulations of a classical spin model using a single-spin Metropolis algorithm. The magnetic island is described by the Hamiltonian

$$ H = -\sum_{i,j} J_{i,j} (S_i \cdot S_j) + \sum_i K_z (S_i^z)^2 + \sum_i K_x (S_i^x)^2 - \mu \sum_i S_i^z \cdot B_z $$

(1)

with localized Heisenberg spins $S_i$ on a triangular lattice of 60 × 60 sites and a stripe shape, the effective exchange interaction coefficients $J_{i,j}$ which are restricted to nearest neighbors ($J_1$), on-site anisotropy energy parameters which can be chosen either out-of-plane ($K_z$) or within the plane ($K_x$), the magnetic moment $\mu$, and the external magnetic field in out-of-plane orientation $B_z$. We assume the experimental temperature of $T = 4.5$ K, a reasonable size of the magnetic moment consistent with the island material (Fe) of $\mu = 2.5 \mu_B$, and calculate the resulting spin configurations for magnetic field loops $B_z = 3 \text{T} \rightarrow -3 \text{T} \rightarrow 3 \text{T}$. We vary $J_1 = 1 \ldots 5$ meV (per bond) and $K_z, K_x = -0.05 \ldots -0.5$ meV within reason-
able constraints\(^4\) in order to reproduce the experimentally measured magnetization curves, particularly the coercive fields. The resulting \(z\)-component of the magnetization averaged over the whole lattice as a function of \(B_z\) for different selections of parameters is shown in Figure S4. For the cases of out-of-plane anisotropy \((K_z < 0, K_x = 0)\), the island reverses its magnetization by domain wall nucleation and propagation, as shown by the snap-shots of the spin-configurations given in the insets. Since the domain wall energy per unit area is proportional to \(\sqrt{J_1 \cdot K_z}\),\(^6\) while the Zeeman energy per unit area is proportional to \(B_z\), the coercivity is roughly proportional to \(\sqrt{J_1 \cdot K_z}\). Hence the coercive field is similar for \((J_1 = 5 \text{ meV}, K_z = -0.1 \text{ meV})\) and \((J_1 = 1 \text{ meV}, K_z = -0.45 \text{ meV})\), and considerably smaller for \((J_1 = 1 \text{ meV}, K_z = -0.1 \text{ meV})\). When this product is very small \((J_1 = 1 \text{ meV}, K_z = -0.05 \text{ meV})\), the island’s coercive field gets comparable to the tip coercive field \(B_c^{\text{tip}} \approx 0.1 \text{ T}\) (see Figure 2e in the main manuscript). In this case, SP-STS would be no longer sensitive to the coercive field and would yield a largely constant signal, consistent with the experimental observation for type III islands. In contrast, even for a relatively strong in-plane magnetic anisotropy \((J_1 = 1 \text{ meV}, K_z = -1 \text{ meV})\), the experimentally available maximum magnetic field of \(B_z = \pm 3 \text{ T}\) still yields an out-of-plane magnetization of 25% of the saturation magnetization (see data set with light green diamonds in Figure S4) which would be experimentally detectable, in conflict with the measurements. We thus conclude, that the type III islands most probably have a small out-of-plane magnetic anisotropy comparable to that of the tip, and that the type II and type I islands have increasingly larger out-of-plane anisotropies and/or exchange constants.
Figure S4: Monte Carlo simulation of the spin configuration of a stripe of a triangular spin lattice during an out-of-plane magnetic field loop (60 × 60 sites, $\mu = 2.5 \mu_B$, $T = 4.5$ K). The graphs show the $z$-component of the magnetization averaged over the whole lattice, $\langle M_z \rangle$, as a function of $B_z$ for different selections of parameters, given on the top. The two insets illustrate snap-shots of the calculated spin configurations close to the coercive fields $B_z = \pm 2$ T for one set of parameters ($J_1 = 5 \text{ meV}$, $K_z = -0.1 \text{ meV}$) where the gray scale corresponds to $S_i^z$ and the colored arrows indicate the orientations of each of the spins in the lattice.
Supplementary Note 5

Statistics of the coercive fields

Table 1 summarizes the coercive fields $B_c$ extracted from magnetization curves similar to those shown in the main manuscript (Figure 2), that have been measured using different spin-polarized tips on Fe islands of different ML types, sizes and shapes shown in Figure S5. Apparently, $B_c$ is largely determined by the type of the reconstruction, and only insignificantly by the island’s size or shape. Overall, the coercive field considerably decreases from the type I reconstruction ($B_{cI} = 2.3 \pm 0.3$ T) over the type II reconstruction ($B_{cII} = 0.7 \pm 0.3$ T) to the type III reconstruction ($B_{cIII} < 0.4$ T). Note, that for the latter type of ML, there is no spin-dependent contrast beyond the range where the tip magnetization is reversing ($B_z > 0.5$ T). Therefore, we can only give upper borders for the corresponding coercive fields of the type III Fe ML.

Table 1: Coercive fields $B_c$ of the islands of different ML types with numbers shown in Figure S5, extracted from the hysteresis loops measured by different tips as given. The error in $B_c$ is defined by the last (first) magnetic field value before (behind) the magnetization reversal of the island. For the type III islands, no clear contrast reversal is observed outside of the coercive field of the tip. Therefore, this magnetic field range can only be used to determine upper borders for $B_c$.

| ML type | island no. | $B_c$ [T]               |
|---------|------------|-------------------------|
|         |            | tip 1 | tip 2 | tip 3 |<|<|<|<|
| I       | 1          |        | 2.2 ± 0.2 | 2.2 ± 0.2 |<|<|<|<|
| I       | 2          |        | 2.2 ± 0.2 | 2.2 ± 0.2 |<|<|<|<|
| I       | 3          |        | 2.2 ± 0.2 | 2.2 ± 0.2 |<|<|<|<|
| I       | 4          |        | 2.5 ± 0.1 | 2.5 ± 0.1 |<|<|<|<|
| I       | 8          |        |          | 2.2 ± 0.2 |<|<|<|<|
| II      | 5          | 0.75 ± 0.25 | 0.6 ± 0.1 |<|<|<|<|
| II      | 6          |        | 0.6 ± 0.1 | 0.6 ± 0.1 |<|<|<|<|
| II      | 9          | 0.45 ± 0.05 | <0.4 |<|<|<|<|
| III     | 7          |        | <0.4 |<|<|<|<|
| III     | 10         |        | <0.4 |<|<|<|<|
Figure S5: (a), (b) Overview STM images ($V = 450 \text{ mV}$ (a), $V = 400 \text{ mV}$ (b) and $I = 1 \text{ nA}$) of the different islands for which hysteresis loops have been measured. The numbers refer to the island number. The according Fe ML types and measured coercive fields $B_c$, which have been extracted from the hysteresis loops, are given in Supplementary Table 1.

Supplementary Note 6

Autocorrelations of the spectroscopic line profiles

In order to quantify the disorder of the Shiba bands in the different types of Fe MLs, the bias-voltage and lateral displacement dependent autocorrelation functions of the spectroscopic line profiles shown in Figures 3a-c of the main manuscript text were calculated using

$$\gamma(V, \Delta x) = \frac{1}{N} \sum_{i}^{N-n} \left( (dI/dV)_{i+n} - \overline{(dI/dV)} \right) \left( (dI/dV)_{i} - \overline{(dI/dV)} \right).$$  \hspace{1cm} (2)

Here, $(dI/dV)_i$ is the differential tunneling conductance value at point $i$ of the spectroscopic line profile, $\overline{(dI/dV)}$ is the differential tunneling conductance averaged along the spectroscopic line profile, $N$ is the total number of analyzed points along the spectroscopic line profile that have been restricted to positions on top of the islands (red dots in Figs. S6a-c), and $n$ is number of points between the two positions separated by the lateral displacement $\Delta x$. In Fig. S6d-f, these autocorrelation functions are shown for the three types of Fe MLs. Since the autocorrelation functions are symmetric with respect to $\Delta x = 0$, only the parts for $\Delta x > 0$ are shown.
There are two noteworthy observations in the region of the Shiba bands, i.e. between the dashed horizontal lines. First, it is noticeable that the overall strongest anticorrelation values are found for the type I ML ($\gamma_{\text{min}} = -0.05$), followed by the type II ML ($\gamma_{\text{min}} = -0.013$) and the type III ML ($\gamma_{\text{min}} = -0.007$). We conclude that the lateral variation in the spectral weight of the Shiba bands is strongest for type I and weakest for type III ML. Second, the first autocorrelation minima appear at displacements of $\Delta x \approx 6 \text{ nm}$ for the type I ML, at $\Delta x \approx 1 \text{ nm}$ for the type II ML, and at $\Delta x \approx 2 \text{ nm}$ for the type III ML. These length scales of the variations in the spectral weight of the Shiba bands scale roughly with the typical periodicities of the different reconstructions in the direction of the lines used for the spectral line profiles (c.p. Figs. S6a-c), which is also largest for the type I, and smallest for the type II ML.
Figure S6: (a-c) STM-images of the three Fe ML islands of each type of reconstruction from Figures 3a-c of the main manuscript text. (d-f) Plots of the bias-voltage dependent autocorrelation $\gamma$ as a function of the displacement $\Delta x$ calculated from the spectroscopic line profiles of Figures 3a-c in the main manuscript using only the points marked by the dotted lines of the corresponding ML island shown in the panels a-c above. Black dashed horizontal lines are at $e \cdot V = \pm \Delta t$ and $e \cdot V = \pm (\Delta t + \Delta s)$. Note that the scales in $\gamma$ and $\Delta x$ are decreasing going from the autocorrelation of the type I Fe ML (d) over type II (e) to type III (f).
References

(1) Wolf, C.; Köhler, U. Growth and intermixing of Nb on Fe(110) and Fe on Nb(110). *Thin Solid Films* **2006**, *500*, 347–355.

(2) Zhou, L.; Meier, F.; Wiebe, J.; Wiesendanger, R. Inversion of spin polarization above individual magnetic adatoms. *Phys. Rev. B* **2010**, *82*, 012409.

(3) Bode, M.; Heide, M.; von Bergmann, K.; Ferriani, P.; Heinze, S.; Bihlmayer, G.; Kubetzka, A.; Pietzsch, O.; Blügel, S.; Wiesendanger, R. Chiral magnetic order at surfaces driven by inversion asymmetry. *Nature* **2007**, *447*, 190–193.

(4) Hagemeister, J.; Iaia, D.; Vedmedenko, E. Y.; von Bergmann, K.; Kubetzka, A.; Wiesendanger, R. Skyrmions at the Edge: Confinement Effects in Fe/Ir(111). *Phys. Rev. Lett.* **2016**, *117*, 207202.

(5) Hagemeister, J. MONTECRYSTAL. https://github.com/JHagemeister/MonteCrystal, accessed 2020-01-12.

(6) Chikazumi, S. *Physics of Ferromagnetism*; Oxford University Press Inc.: New York, 1997.