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Supplementary Materials for

Atomic-scale spin-polarization maps using functionalized superconducting probes

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Supplementary Text
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Supplementary Text

Lorentzian fits to the YSR spectra

We fit a sum of two Lorentzian distributions to the spectra obtained with the YSR tip using only the voltage range $V \in \pm\, 0.56$ mV, i.e. inside the gap of the substrate:

$$\mathrm{d}I/\mathrm{d}V(V) \propto \sum_{\mathrm{i}\in\{\mathrm{e},\mathrm{h}\}} \frac{a_{\mathrm{i}}\sigma_{\mathrm{i}}^{2}}{\pi((\mathrm{e}V - E_{\mathrm{i}})^2 + \sigma_{\mathrm{i}}^{2})} \qquad (S1)$$

The maximum of each Lorentzian is directly given by a_e and a_h in this form, while E_{i} and σ_i represent the energies and the widths of the peaks, respectively. All fits to the spectra used in Fig. 3D and E of the main text are shown in Figure S2. In the region of interest, the experimental data, especially the peak heights, are well reproduced by the fit. Adding a broadened Dynes-DOS would maybe give better agreement for the voltage region above ±0.56 mV. However, this would introduce additional free parameters to the fit without having influence on the asymmetry, which is the only desired property we want to obtain.

Suppression of superconductivity in external magnetic fields

In Figure S3, we present tunneling spectroscopy data obtained with a clean Nb tip without in-gap states on the bare Nb(110) substrate in different external magnetic fields B_z. In the range of $0 < B_z < 0.1$T, we observe no visible change in the dI/dV spectra, indicating that we measure below the lower critical field $B_{\mathrm{c},1}$ of the Nb substrate. For $0.125\,\mathrm{T} < B_z < B_{\mathrm{c},2} = 0.4\,\mathrm{T}$, which is the upper critical field of Nb known from literature (*31*), one of the gaps gradually closes until the spectra show a BCS-type gap of $\Delta \approx 1.40$ meV. The second gap is only barely affected by the external field up to $B_z \approx 1.4$ T. We point out that this gap has to stem from the superconducting cluster on the tip apex, since we are able to induce YSR states by picked up magnetic atoms at fields above $B_{\mathrm{c},2}$. When applying even higher magnetic fields, superconductivity in the tip is finally quenched at $B_z = 3$ T (Fig. S3B).

Measurements with different YSR tips

In Figure S4, we show additional examples of YSR-SP-STM on ferromagnetic $1a - [001]$ Mn chains with different YSR tips. In both examples, a single Mn atom is attached to the tip apex. Note that at least the tip in Fig. S4A obviously features a Yu-Shiba-Rusinov multiplet. In addition, the hole-like YSR peak is intrinsically stronger in intensity than the particle-like peak for both tips shown in this Figure. When tunneling into the ferromagnetic chain, the hole-like peak shows increased intensity in this case. This is the opposite behavior to the YSR tip shown in the main text. A possible interpretation of this effect is that the YSR impurities are on different sides of the quantum phase transition where the YSR peaks cross E_{F} (*21, 24*). Note that the tip used for Fig. S4A/B exhibits a relatively strong double feature, as it can be seen in the chain's STM topography. We argue that this is probably leading to the strong overshooting of the asymmetry signal on the left chain end, since we saw less of this stray signal with better imaging capability of the tip. Tunneling into multiple areas of the sample at the same time might lead to additional contributions to the measured spin-polarization in this case. Interestingly, a larger average YSR asymmetry is observed with these two tips when compared to Fig. 4B of the main manuscript, although a similar structure is investigated. This might be interpreted as a sign for an unresolved YSR multiplet on the tip in the main text, reducing the total tip spin-polarization below 100%.

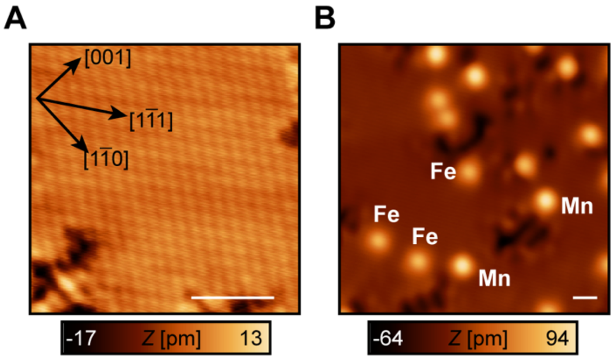

Fig. S1. Preparation of the Nb(110) sample. (**A**) Atomically resolved constant-current STM image of the clean Nb(110) surface before the deposition of adatoms ($V = -10$ mV, $I = 1$ nA). (**B**) Constant-current STM image of the sample with randomly distributed Fe and Mn adatoms ($V = -20$ mV, $I = 0.2$ nA). The white bars correspond to 1 nm.

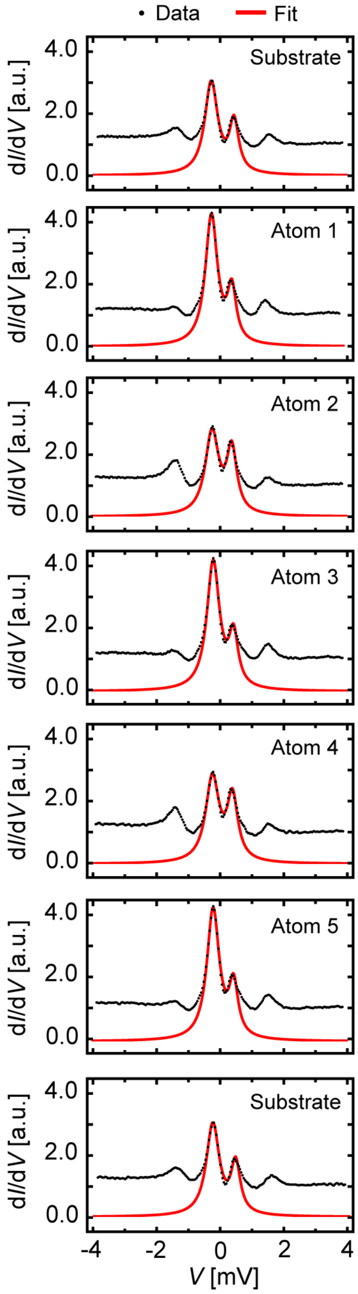

Fig. S2. Lorentzian fits to the spectra shown in Figure 3D of the main text. The black dots are the measured d*I*/d*V* curves also presented in Fig. 3D of the main manuscript. The red lines are fits to the Lorentzians defined in Eq. S1.

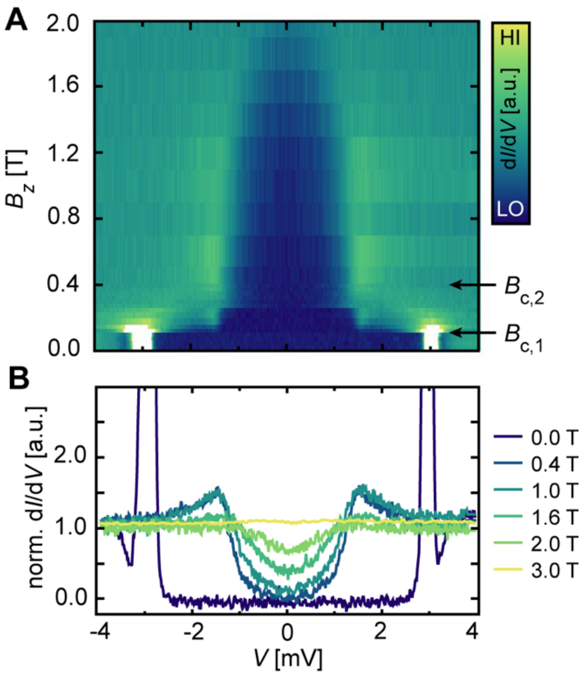

Fig. S3. Evolution of tip and sample gap in different external magnetic fields. (**A**) Stacked dI/dV spectra in the range of $0 < B_z < 2$ T ($V_{stab} = -6$ mV, $I_{stab} = 1$ nA, $V_{mod} = 20$ µV). The lower and upper critical fields $B_{c,1}$ and $B_{c,2}$ of the Nb substrate, respectively, are indicated by arrows. (**B**) Selected single dI/dV spectra in the range of $0 < B_z < 3$ T.

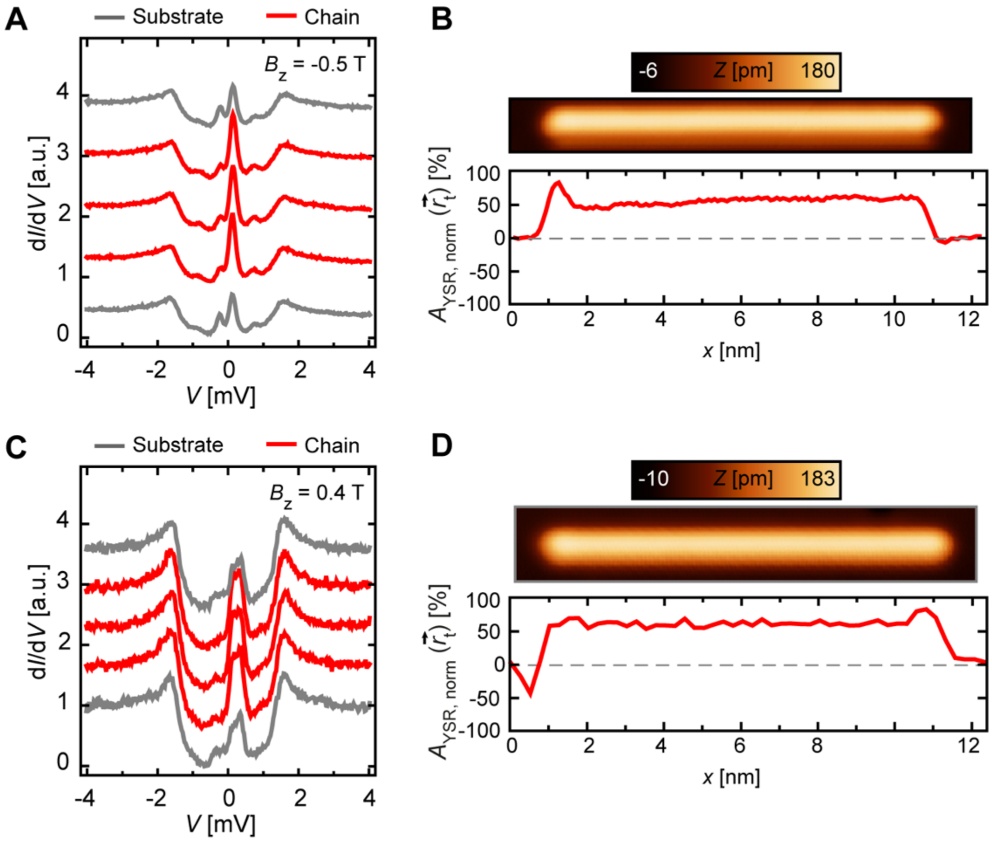

Fig. S4. Additional examples of different YSR tips. (**A**) Spectra obtained with a YSR tip along a ferromagnetic $1a - [001]$ Mn_{31} chain. (**B**) Topography of the Mn_{31} chain aligned with the YSR asymmetry along the chain. (**C**) Spectra obtained with a different YSR tip along a ferromagnetic $1a - [001]$ Mn_{32} chain. (**D**) Topography of the Mn_{32} chain aligned with the YSR asymmetry along the chain. All data is measured with $V_{stab} = -6\,mV$, $I_{stab} = 1$ nA, $V_{mod} = 40\,\mu V$.