Supporting Information:

Sensing Noncollinear Magnetism at the Atomic Scale Combining Magnetic Exchange and Spin-Polarized Imaging

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S1. Experimental details

All measurements were performed with an Fe bulk tip attached to the free prong of the tuning fork. Two different types of tuning forks were used: (a) an asymmetric tuning fork with the Fe tip contacted via an additional electrode printed on the prong; (b) a symmetric tuning fork where the Fe tip is contacted by a 12 µm thin wire. The latter solution is suggested to avoid cross talk between the tunneling current and the deflection of the tuning-fork prong.\(^1\)\(^2\) The tuning-fork oscillation amplitude was calibrated using the tunneling current-controlled amplitude determination\(^3\). Owing to the calibration procedure, an error of the

Figure S1: Characterization of the tip magnetization: SP-STM constant-current topography (raw data) of different domains on a bilayer Fe island (25 x 25 nm\(^2\), \(V_m=50\) mV, \(I_t=291\) pA) for (a) a tip with a dominating out-of-plane magnetization, (b) and (c) tips with out-of-plane as well as in-plane magnetization, (d) a non-magnetic tip. The three boxes in every image indicate the apparent-height modulation along all three rotational domains of the spin spiral. The scale bar is 6 nm in all images. Color scales: (a) 0.19-0.27 nm, (b) 0.16-0.23 nm, (c) 0.10-0.19 nm, (d) 0.17-0.28 nm.
oscillation amplitude of 10% is assumed.

The constant frequency shift images were acquired in the attractive force regime. While for larger tip-surface separations \( d \gtrsim 500 \text{ pm} \) long-range van der Waals, electrostatic and magnetic dipolar forces dominate the total tip-sample force, short-range forces become dominant at smaller separations \( d \approx 100 \text{ pm} \). Therefore, the oscillation amplitude was chosen such that it is in the order of the range of the tip-surface interaction, minimizing the amplitude noise and enhancing the sensitivity to short-range interactions. Thus, we used oscillation amplitudes \( z_{\text{mod}} \leq 110 \text{ pm} \).

**S2. Tip preparation and characterization**

In order obtain spin contrast, the Fe tip attached to the tuning fork is dipped into regions where the Fe islands on Ir(111) are 3-4 monolayers thick. To check the orientation of the tip spin polarization, we image bilayer Fe islands and compare the spin contrast within each of the spin-spiral domains that are rotated by 120° with respect to each other (highlighted by boxes in Figs. S1a-d). The modulations in the apparent height along the spin spirals have a spacing of about 1.4 nm. For tips with a dominating out-of-plane magnetization, the SP-STM contrast along the spin spiral is identical on all three rotational domains (Fig. S1a), while a tip with significant in-plane magnetization results in different amplitudes along the three different directions or smaller total spin contrast (Fig. S1b and c). Tips with no spin polarization lead to images where the spin spirals appear blurred (Fig. S1d).

In order to account for small long-range van der Waals interactions, we mainly use tips that exhibit a frequency shift \( |\Delta f| \sim 7 \text{ Hz} \) at typical tunneling parameters (50 mV, 100 pA).
Figure S2: Image processing of Fig. 1: (a) and (b) shows raw-data images of the SP-STM constant-current topography shown in Fig. 1 of the main text with two differently adjusted color scales in order to see (a) the contrast of the Fe bilayer spin spirals and (b) the defects and nano-skyrmion lattice in the Fe monolayer. (c) shows the images merged with a Laplace-filtered image (as shown in the manuscript), thus enabling visualization of all the structures within a single image (55 x 45 nm$^2$, $V_s$ =50 mV, $I_t$ = 100 pA). The scale bar is 10 nm. Color scales: (a) 0.25-0.82 nm, (b) 0.30-0.38 nm, (c) 0.33-1.1 nm.

S3. Raw-data image of Figure 1

The SP-STM image shown in Fig. 1a was processed in order to visualize both the square nano-skyrmion lattice in the fcc monolayer as well as the spin spirals in the bilayer Fe islands. For that purpose, the image was merged with a Laplace-filtered image. Fig. S2 shows the raw data images again with the color contrast adjusted to (a) the bilayer spin spiral, (b) the monolayer fcc nano-skyrmion structure. (c) shows the processed image again as shown in Fig. 1a.

S4. MExFM-SP-STM with different defects/ adsorbates on the fcc skyrmion lattice on the Fe monolayer on Ir(111)

Figure S3: MExFM-SP-STM with different defects: (a) SP-STM constant-current topography (8.3 x 5.9 nm$^2$, $V_s$ = 50 mV, $I_t$ = 100 pA) of a fcc Fe island showing the square skyrmion lattice and two different types of defects, labeled with 1 and 2. The inset shows the 2D FFT of the image showing the square structure of the fcc skyrmiior lattice. (b) MExFM image of the image in (a). (c) Simultaneously measured current map. (b) and (c): $\Delta f_{set} = -39$ Hz, $V_s = 0.1$ mV, $z_{mod} = 77$ pm. The scale bar is 2 nm in all images. Color scales: (a) 0.2-35 pm, (b) 0-16 pm, (c) 0.1-1.1 nA.
Fig. S3a shows a SP-STM image of a region with two defects, labeled “1” and “2.” While defect 1 appears as a depletion in the apparent height and is spatially confined, defect 2 only leads to a small height variation. The MExFM image in Fig. S3b and the simultaneously measured current map in Fig. S3c give further information. The MExFM image shows that defect 1 is indeed an adsorbate on the fcc skyrmion lattice, while defect 2 also results in a height corrugation, but is less spatially defined. We suggest that defect 2 might be a defect in the Ir or an adsorbate in between the Ir substrate and the Fe layer. The bright lines in Fig. S3c show changes of the tip magnetization that influence the spin-polarized current, but not the MExFM contrast in Fig. S3b.

**S5. Amplitude, excitation voltage, and frequency shift for the MExFM image in Fig. 2**

In order to give further information on the measurement parameters and mechanisms of Fig. 2b and c, the simultaneously measured amplitude, excitation voltage and frequency shift are shown in Fig. S4. The amplitude and the frequency shift appear without any features indicating no disturbances during the measurements. In particular, the featureless image of the excitation voltage (b) indicates a non-dissipative interaction.

![Figure S4: Additional channels for Fig. 2b and c](image)

**Figure S4: Additional channels for Fig. 2b and c:** (a) Amplitude, (b) excitation voltage of the tuning fork, and (c) frequency shift $\Delta f$ simultaneously measured with the MExFM image in Fig. 2b ($\Delta f_{\text{set}} = -14.6$ Hz, $z_{\text{mod}} = 102$ pm, $V_s = 0.2$ mV). The scale bar is 2 nm in all images. Color scales: (a) 102-102.8 pm, (b) 1.74-3.93 mV, (c) $-14.75$ to $-14.58$ Hz. A roughness analysis distribution for the images yields a Gaussian distribution with a FWHM of (a) 0.24 pm, (b) 0.65 mV, and (c) 43 mHz.

**S6. MExFM data with shifted contrast in the MExFM and current image**

The images shown in Fig. S5 were taken with a tip that had a frequency shift offset of $-10$ Hz, indicating a larger contribution from long-range van der Waals forces indicating a blunt tip. In this case, we
observed a lateral shift of the corrugation between the MExFM image (Fig. S5b) and the simultaneously measured current map (Fig. S5c), as indicated by the lines that mark the same position. However, for most of our tips (10 out of 12 tips with which we acquired MExFM images) the corrugation maxima in the MExFM image and in the spin-polarized current map are at the same positions. We believe that this shift can be caused by modifications of the spin polarization relative to the exchange force for tips that change their structure near the apex.⁷

**Figure S5: MExFM for a tip leading to a shifted contrast:** (a) SP-STM constant-current topography ($V_s = 50$ mV, $I_t = 100$ pA), (b) MExFM image ($\Delta f_{\text{set}} = -60$ Hz, $V_s = 0.2$ mV, $z_{\text{mod}} = 58$ pm). The frequency shift offset at $z = 0$ is $-10$ Hz indicating strong long-range van der Waals forces caused by a blunt tip. The arrows in (b) and (c) show the shift of the corrugation between the AFM topography and the simultaneously measured current. All image sizes: 3 x 3 nm². The scale bar is 1 nm in all images. Color scales: (a) 0-20 pm, (b) 1.3-4.5 pm, (c) 852 pA-1.5 nA.

**S7. Procedure for acquisition of frequency shift vs. distance curves**

The tip prepared as described in section 6 was stabilized above the position of the skyrmion lattice with maximum spin-polarized signal (cf. red-circled area of the inset image in Fig. 4). The tuning fork oscillation was then started with oscillation amplitudes (defined as half the peak-to-peak value) between 40 pm and 80 pm. The current-feedback loop was then switched off at $V_s = 50$ mV and $I_t = 100$ pA. The voltage was decreased to values between 0.1 mV and 0.3 mV, the current-voltage preamplifier gain is set between $10^7 - 10^9$.

Next, the tip was retreated to $z = 200$ pm in order to probe the long-range forces and then approached to the surface to values down to $z = -480$ pm. We measured both the approach and retract sweeps and
recorded the frequency shift, spin-polarized current, amplitude, and excitation voltage. Two to eight sweeps were acquired at this position and averaged before going to the neighboring position of opposite spin contrast within the skyrmion lattice (cf. blue-circled area of the inset image in Fig. 4), with the current-feedback loop switched off. The same distance sweep as described above was repeated on the dark position. To ensure that no drift occurred during acquisition, follow-up curves on each of the two positions were taken. If the distance-dependent spin-polarized current curves of these two full sets of data acquisition exhibit a shift along the z-axis of more than 5 pm (which is more than the typical variation between the forward and backward sweep in the current), then the data was disregarded. Otherwise, the different curves at each of the two positions with opposite spin contrast were averaged, and the

![Figure S6: Distance-dependent measurements on the fcc Fe nano-skyrmion lattice grouped by spin polarization](image)

(a) Spin-polarized asymmetry for mainly out-of-plane magnetized tips with $\mathcal{A}(z) \geq 0.15$, and (c) with $\mathcal{A}(z) \leq 0.15$. (b) and (d) display the corresponding exchange force $F_{ex}(z)$, respectively. The color code for all sub-figures reflects the averaged spin-polarized asymmetry $\mathcal{A}$ with blue to red representing low to high.
frequency-shift difference \( \Delta f_{\text{ex}} \) was calculated. Prior to subtracting, the \( \Delta f(z) \) curves were smoothed by a Savitzky-Golay filter (polynomial order: 1, frame width: 7). The magnetic exchange force \( F_{\text{ex}} \) was calculated (Ref. 8) from \( \Delta f_{\text{ex}}(z) \) using formula 1. The shown magnetic exchange force curves in Fig. 4 and Fig. S6 were smoothed by Savitzky-Golay filter (polynomial order: 1, frame width: 5).

In order to illustrate the reproducibility and spread of our data, we show all acquired datasets in Fig. S6 for \( F_{\text{ex}}(z) \) derived from \( \Delta f_{\text{ex}}(z) \) as well as the spin-polarized current asymmetry \( \mathcal{A}(z) \). The data is split into two groups of larger \( \mathcal{A}(z) \geq 0.15 \), (a) and smaller \( \mathcal{A}(z) \leq 0.15 \), (c) spin-polarized current asymmetry, respectively, together with the respective magnetic exchange force \( F_{\text{ex}}(z) \) ((b) and (d)). The color code for all sub-figures reflects the averaged spin-polarized asymmetry \( \mathcal{A}(z) \) with blue to red representing low to high. For all shown curves, no drastic increase in the tuning-fork excitation voltage was observed, which would have indicated a non-stable tip with dissipative processes\(^9\).

**Figure S7:** MExFM image of the fcc skyrmion lattice on first layer Fe on Ir with changed voltage: (a) SP-STM constant-current topography of the skyrmion lattice on the fcc Fe monolayer (sizes all images: 5 x 2.8 nm\(^2\), \( V_s = 50 \) mV, \( I_t = 100 \) pA). (b) MExFM image of the lower part of (a), \( \Delta f_{\text{ex}}: -36 \) Hz, \( V_s = 0.0 \) mV (lower part), \( V_s = 0.4 \) mV (upper part), \( z_{\text{mod}} = 180 \) pm)). (c) same image as (b), but smoothed by a 2-point Gaussian filter. (c) Simultaneously measured current map. At the scan line marked by the black arrow the voltage was increased from 0 to 0.4 mV. The arrows in the images mark the same defect. A plane has been subtracted from all images. The scale bar is 1 nm. Color scales: (a) 3.7-49 pm, (b) 18.6-22.1 pm, (c) 12.7-18.4 pm, (d) 0.0-4.4 nA.
S8. Discussion of cross-talk

In combined STM/AFM imaging with a tuning fork, it is important to rule out potential cross-talk between the current and the frequency shift, where the tunneling current may introduce an interference with the deflection of the tuning fork. It has been suggested before to use a separate wire for the tunneling current to minimize this unwanted signal.\textsuperscript{1,2} For our measurements we used two different types of tuning forks, as described above. For both forks, we checked for cross-talk by changing the applied bias voltage, and thus the current, while acquiring a MExFM image. Fig. S7 shows an SP-STM image (a) and an MExFM image (b,c) together with the simultaneously measured current (d) for the fcc skyrmion lattice on the Fe monolayer on Ir(111). At the horizontal position marked by the black arrow in (d), the voltage was changed from $V_{S} = 0.0$ mV (lower part) to $V_{S} = 0.4$ mV (upper part). While there is a clear signal change in the current image, no change in the MExFM corrugation is observed. Fig. S8 shows a constant-height image of the frequency shift ($\Delta f$) (a,b) and the simultaneously acquired current map. At the line indicated by the arrow, the bias voltage was changed from $V_{S} = 0.1$ mV (upper part) to 0 mV (lower part). Again, while there is a clear signal change in the current image, no change in the MExFM corrugation is observed.

**Figure S8:** Constant-height imaging of the fcc skyrmion lattice on the Fe monolayer on Ir(111): (a) Frequency shift $\Delta f$ (raw data), (b) smoothed by a Gaussian filter, and (c) simultaneously acquired current (raw data). Size: 4.0 x 4.0 nm$^2$ for all images, $z_{\text{mod}} = 51$ pm, $z = -400$ pm with the current-feedback loop opened above the bright position of the skyrmion lattice at $V_{s} = 50$ mV and $I_{t} = 100$ pA. At the line indicated by the arrow, the bias voltage was changed from $V_{S} = 0.1$ mV (upper part) to 0 mV (lower part). There is a small voltage offset ($\approx |0.05|$ mV to |0.1| mV) which is not constant in time and which could not be compensated entirely. While there is a clear change in the current, the $\Delta f$ images (a) and (b) show no change. This shows that the corrugation in the $\Delta f$ image is not influenced by any potential cross-talk with the current. The scale bar is 1 nm in all images. Color scales: (a) and (b) –34.9 to –33.6 Hz, (c) –135 pA to 290 pA.
observed. This further reveals that the AFM image at close tip-surface distances corresponds to a MExFM image where the signal is attributed to the local variations in exchange force between the magnetic tip and the surface nano-skyrmion lattice. We note that the tip used to acquire the images in Fig. S8 was one out of two tips for which the maxima in the corrugation in $\Delta f$ and in the spin-polarized current are not at the same positions (see section S6).

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