Supplementary Information for: Electric-field driven stability control of skyrmions in an ultrathin transition-metal film

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Electric field	B_1	Y_1	K_1	$\Delta E^{uudd}_{3\overline{\mathrm{K}}/4}$	$\Delta E^{uudd}_{\overline{\mathrm{M}}/2}$	$\Delta E^{3Q}_{\overline{\mathrm{M}}}$
+0.5 V/Å	−0.34	1.05	−1.23	−4.23	−12.65	−20.54
0.0 V/Å	−0.39	1.00	−1.36	−5.31	−13.33	−21.88
−0.5 V/Å	−0.33	1.00	−1.53	−6.91	−14.86	−23.38

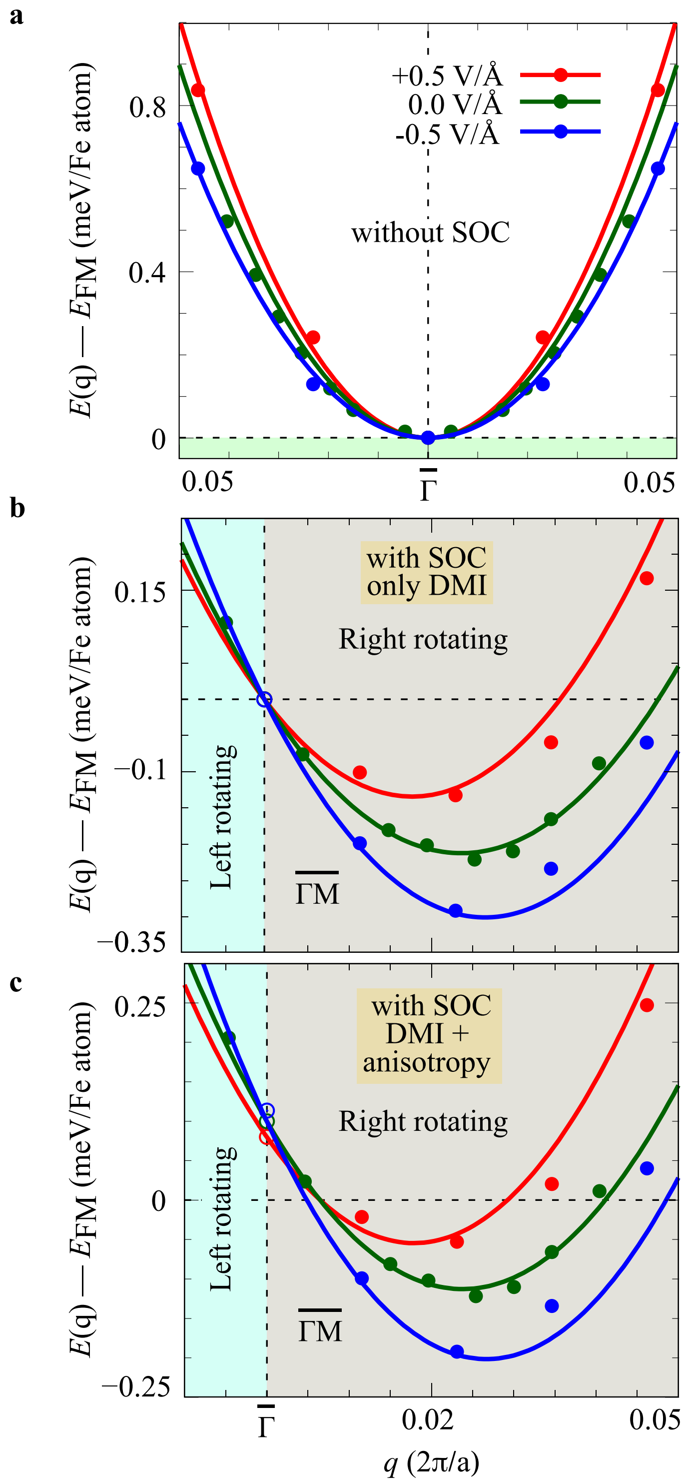
Supplementary Table 1 | Higher-order exchange constants from DFT without and with electric fields. Energy difference between the multi-Q and single-Q states and higher-order constants from DFT. Biquadratic (B_1), three-site four spin (Y_1) and four-site four spin (K_1) constants for Fe/Rh/Re(0001) at $\mathcal{E}= \pm0.5$ V/Å and $\mathcal{E}= 0.0$ V/Å. ΔE is the energy difference between the multi-Q and spin spiral states (single-Q) defined in Eqs. (5-7) in the method section of the main text. Higher-order constants are calculated using Eqs. (2-4) in the method section of the main text. The data of $\mathcal{E}= 0.0$ V/Å are taken from Ref. [1]. All values are given in meV.

Electric field	J_1/J_1'	J_2/J_2'	J_3/J_3'	J_4/J_4'	J_5/J_5'	J_6/J_6'	J_7/J_7'	B_1	Y_1	K_1
+0.5 V/Å	10.42	0.02	−0.10	−0.24	0.27	−0.01	−0.12	0.00	0.00	0.00
	9.37	−1.03	0.07	−0.24	0.27	−0.01	−0.12	−0.34	1.05	−1.23
0.0 V/Å	9.85	0.23	−0.15	−0.22	0.27	0.05	−0.16	0.00	0.00	0.00
	8.85	−0.77	0.05	−0.22	0.27	0.05	−0.16	−0.39	1.00	−1.36
−0.5 V/Å	9.08	0.45	−0.14	−0.20	0.23	0.01	−0.12	0.00	0.00	0.00
	8.08	−0.55	0.03	−0.20	0.23	0.01	−0.12	−0.33	1.00	−1.53

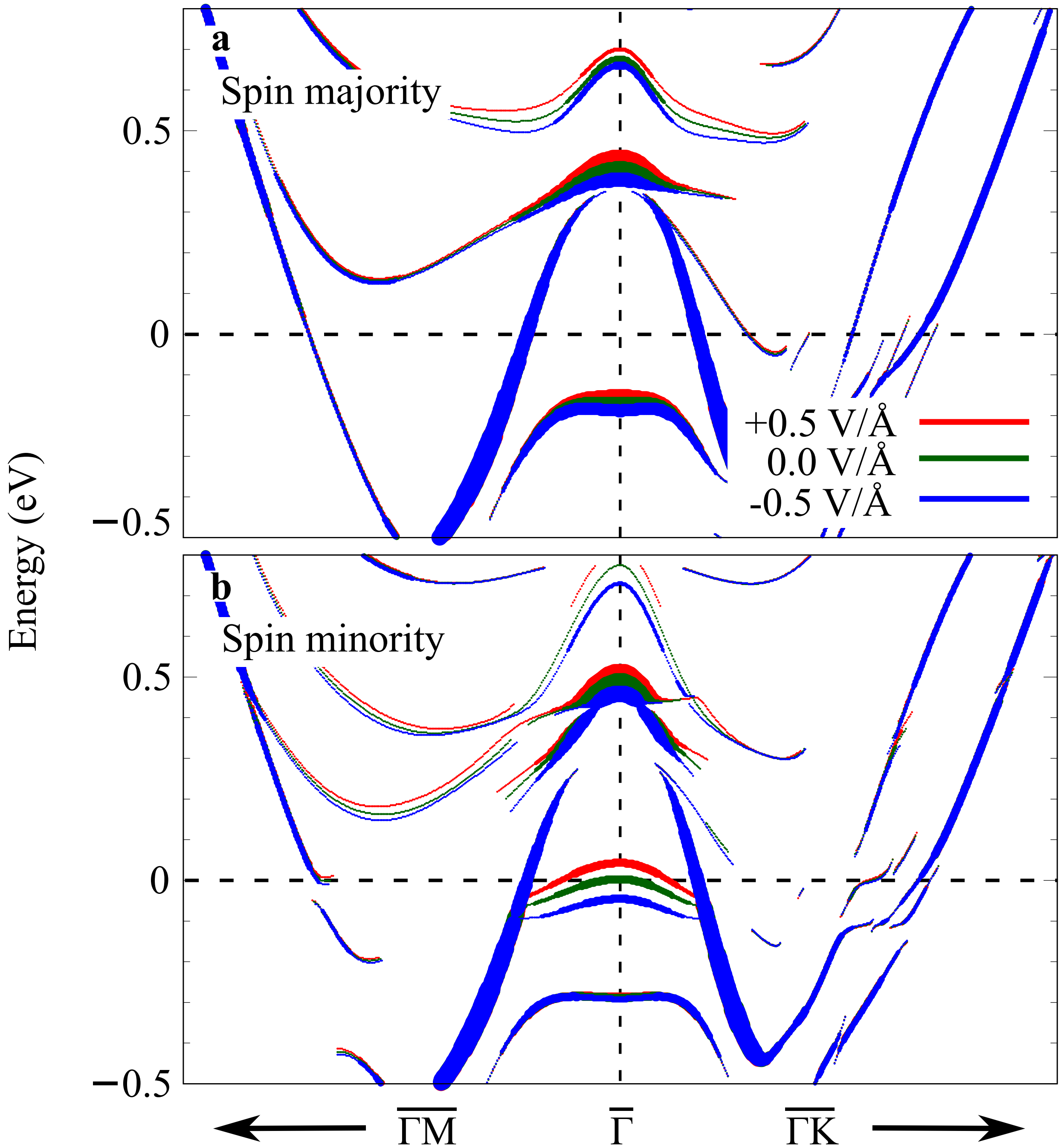
Supplementary Table 2 | Interaction parameters from DFT without and with electric fields. The Heisenberg pair-wise exchange parameters (J_i/J_i'), biquadratic (B_1), three-site four spin (Y_1) and four-site four spin (K_1) constants for Fe/Rh/Re(0001) at $\mathcal{E}= \pm0.5$ V/Å and $\mathcal{E}= 0$ V/Å. The modified and unmodified Heisenberg pair-wise exchange parameters are denoted as J_i' and J_i, respectively. The exchange constants are modified upon including the higher-order interactions according to Eqs. (8-10) in the method section of the main text. The data of $\mathcal{E}= 0$ V/Å are taken from Ref. [1]. All values are given in meV.

$\mathcal{E}= +0.5$ V/Å	$4s$	$4p$	$3d$
spin-majority	$+2.1\times10^{-4}$	-4.4×10^{-4}	$+3.6\times10^{-3}$
spin-minority	-3.3×10^{-4}	-9.3×10^{-4}	-3.4×10^{-3}
$\mathcal{E}= -0.5$ V/Å	$4s$	$4p$	$3d$
spin-majority	-3.6×10^{-4}	$+2.0\times10^{-4}$	-3.0×10^{-3}
spin-minority	$+2.0\times10^{-4}$	$+9.2\times10^{-4}$	$+2.6\times10^{-3}$

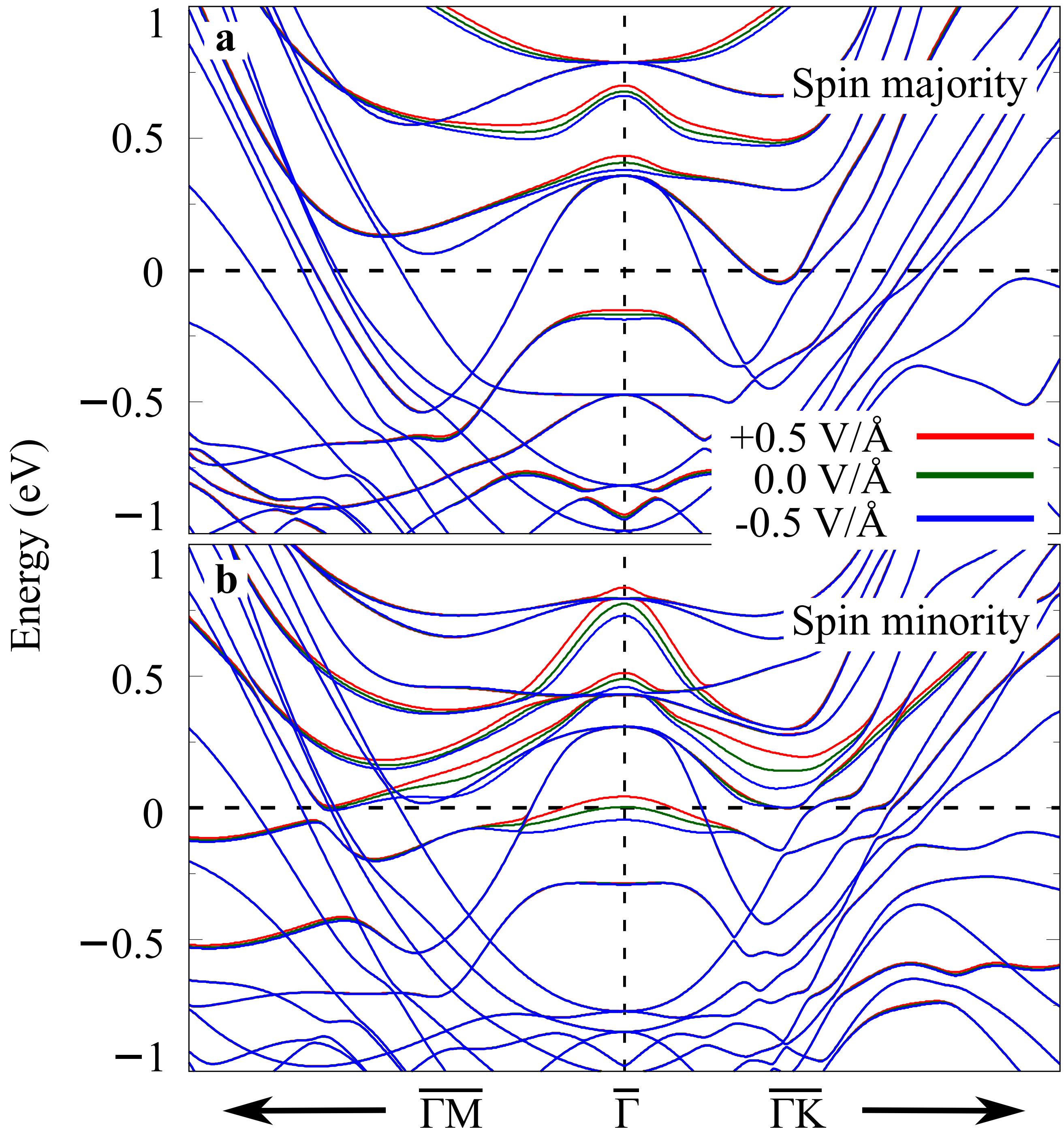
Supplementary Table 3 | Change in electron occupation of Fe with electric fields. Difference in electron occupation of Fe $4s$, $4p$ and $3d$ valence orbitals for $\mathcal{E}= \pm0.5$ V/Å with respect to $\mathcal{E}= 0.0$ V/Å. The values are shown for majority (spin up) and minority (spin down) spin channels and obtained from integration of the charge within the muffin-tin spheres.



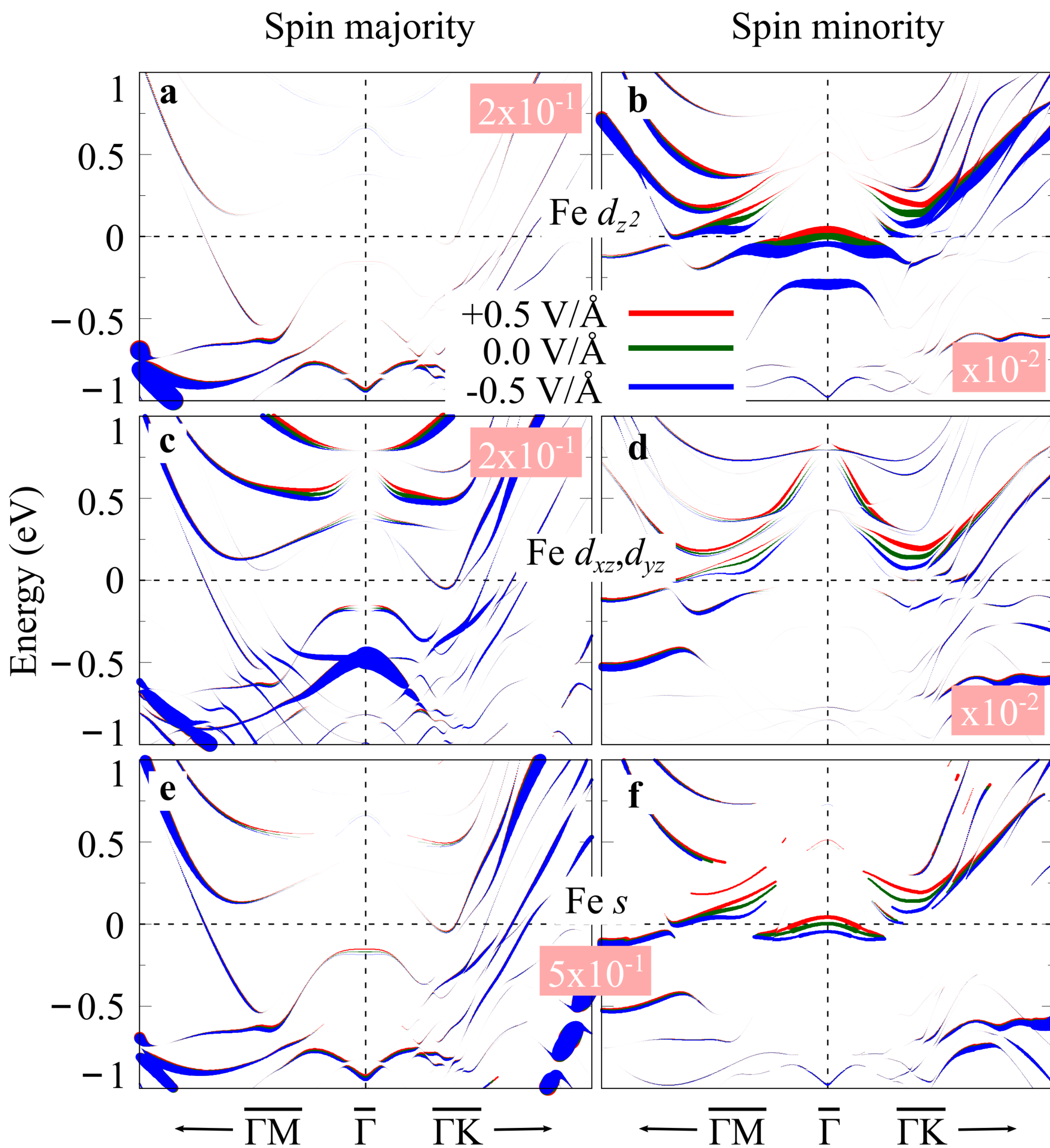
Supplementary Figure 1 | Highlighting the effect of SOC on the ground state by adding DMI and MAE successively. **a** Energy dispersion $E(\mathbf{q})$ of flat spin spirals close to the $\overline{\Gamma}$ point along $\overline{\Gamma\mathrm{M}}$ without SOC for Fe/Rh/Re(0001) at $\mathcal{E}= +0.5$ V/Å (red), $\mathcal{E}= 0.0$ V/Å (green), and $\mathcal{E}= -0.5$ V/Å (blue). The filled circles represent DFT data and the solid lines are the fit to the Heisenberg model. $E(\mathbf{q})$ of flat cycloidal spin spirals **b** including DMI and **c** DMI plus MAE along $\overline{\Gamma\mathrm{M}}$ for zero and two electric fields. The filled circles represent DFT data and the solid lines are fit to the Heisenberg plus DMI spin models. Grey (cyan) shaded region indicate right (left) rotating spin spirals. MAE shifts the energy by $K/2$ with respect to the FM state.



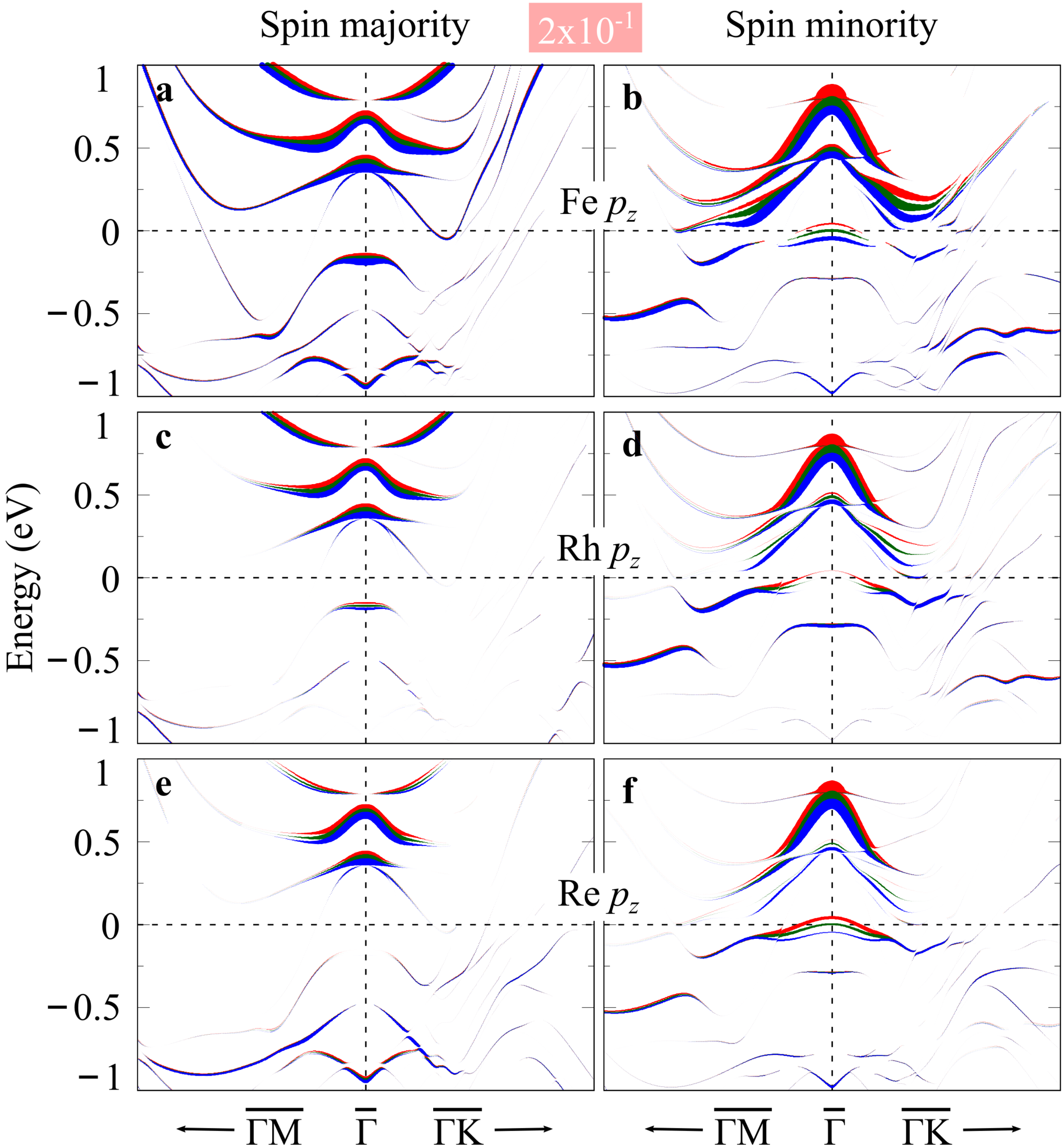
Supplementary Figure 2 | Effect of electric fields on the surface states. **a** Spin majority and **b** spin minority surface states of Fe/Rh/Re(0001) around $\overline{\Gamma}$ for $\mathcal{E}$= +0.5 V/Å (red), $\mathcal{E}$= 0.0 V/Å (green) and $\mathcal{E}$= −0.5 V/Å (blue). Only around $\overline{\Gamma}$, i.e., part of the $\overline{\Gamma\mathrm{M}}$ and $\overline{\Gamma\mathrm{K}}$ directions are visible. The Fermi energy is set to zero.



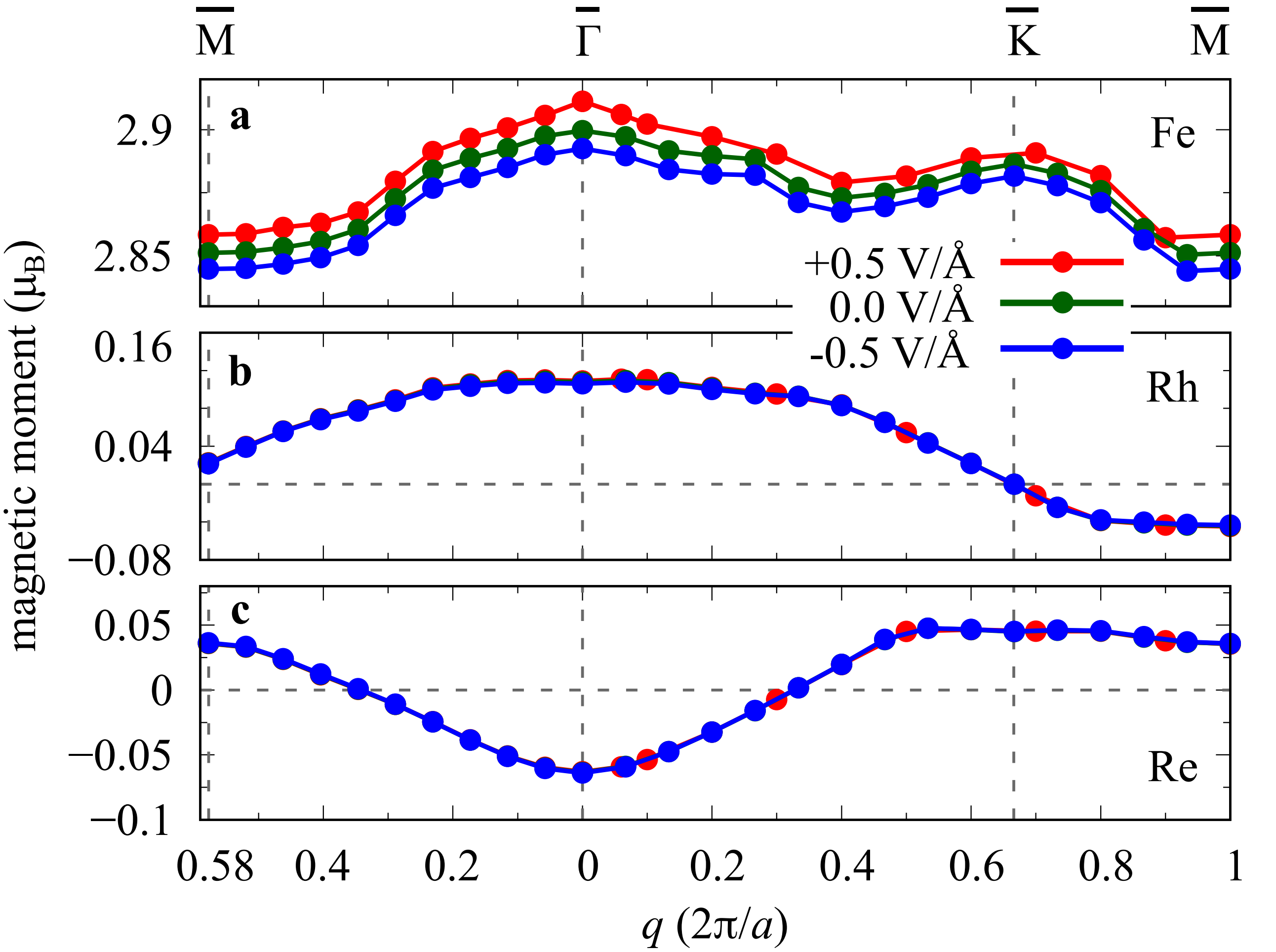
Supplementary Figure 3 | Effect of electric fields on the band structure. **a** Spin majority and **b** spin minority band structures of Fe/Rh/Re(0001) around $\overline{\Gamma}$ or $\mathcal{E}= +0.5$ V/Å (red), $\mathcal{E}= 0.0$ V/Å (green) and $\mathcal{E}= -0.5$ V/Å (blue). Only around $\overline{\Gamma}$, i.e., part of the $\overline{\Gamma\mathrm{M}}$ and $\overline{\Gamma\mathrm{K}}$ directions are visible. The Fermi energy is set to zero.



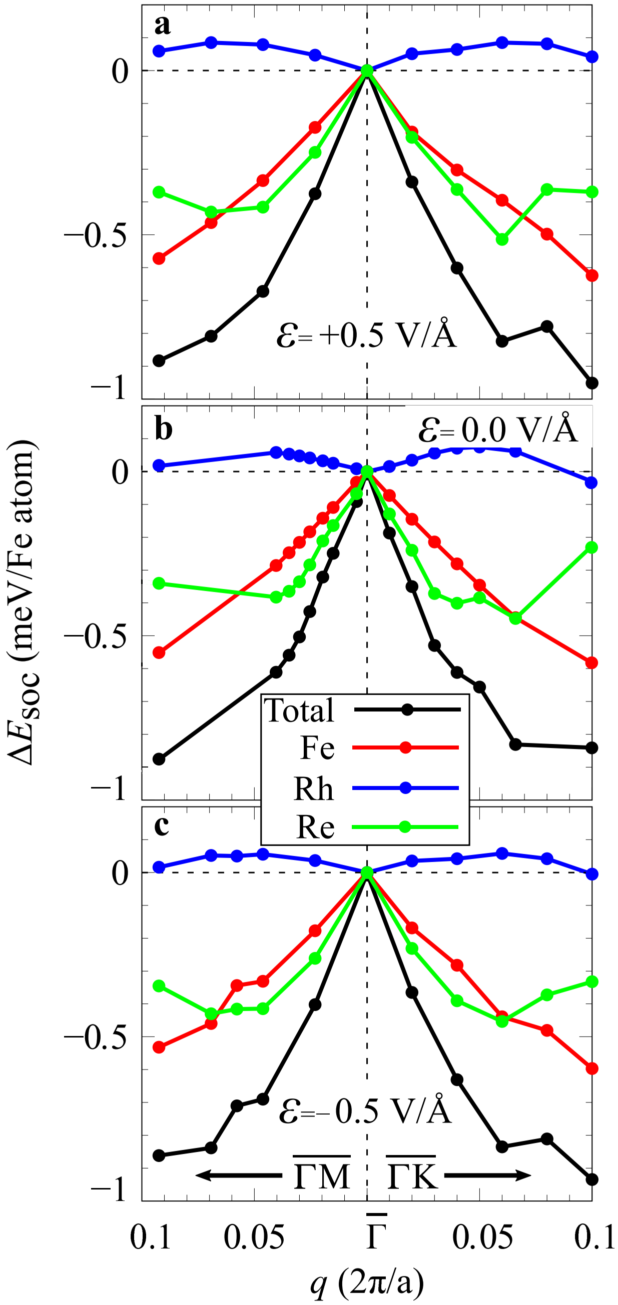
Supplementary Figure 4 | Fat band representation of the projected band structure of Fe for zero and finite electric fields. Spin majority and spin minority bands of Fe are projected onto **a,b** d_{z^2}, **c,d** d_{xy} and **e,f** d_{yz} orbitals. Weight of the orbital character is indicated by thickness (fatness). Only around $\overline{\Gamma}$, i.e., part of the $\overline{\Gamma \mathrm{M}}$ and $\overline{\Gamma \mathrm{K}}$ directions are shown for $\mathcal{E}= +0.5$ V/Å (red), $\mathcal{E}= 0.0$ V/Å (green) and $\mathcal{E}= -0.5$ V/Å (blue). Weight of spin majority and spin minority d_{z^2} and $d_{xz,yz}$ states are multiplied by a factor of 0.2 and 0.01, respectively and s states by a factor of 0.5 for better visualization. The Fermi energy is set to zero.



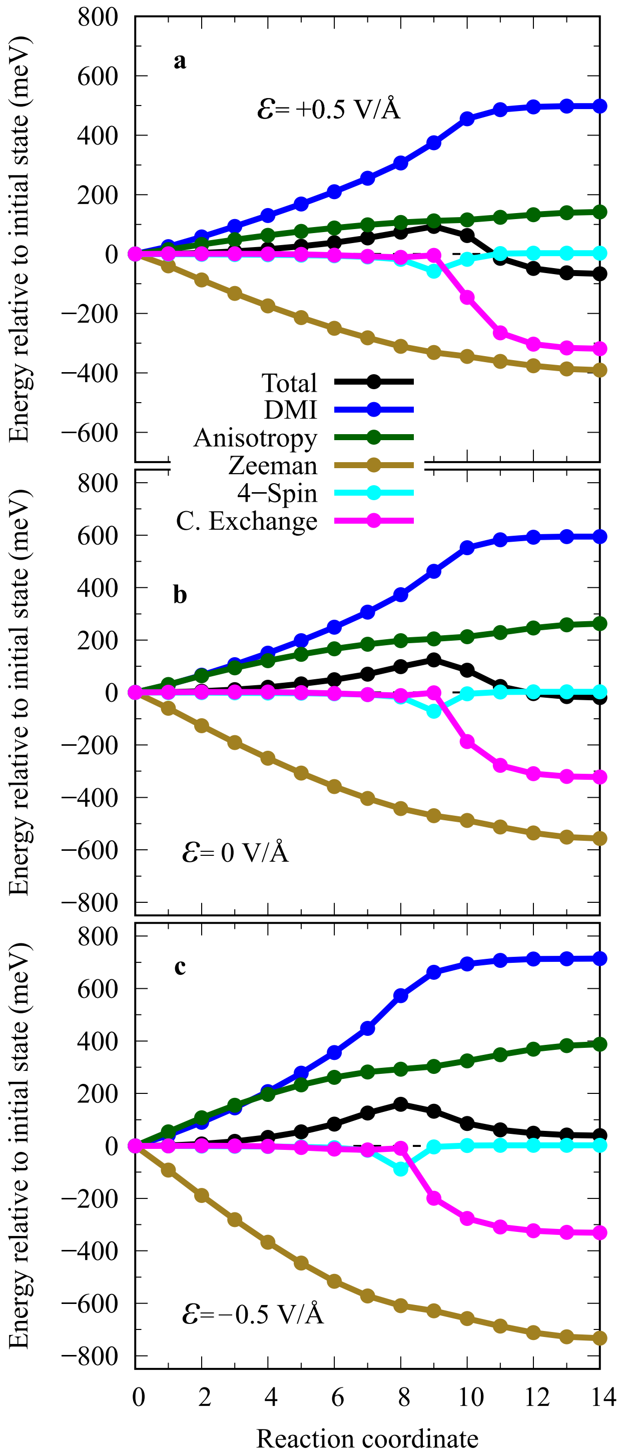
Supplementary Figure 5 | Fat band representation of the projected band structure of Fe, Rh and Re for zero and finite electric fields. Spin majority and spin minority bands of **a,b** Fe, **c,d** Rh and Re **e,f** are projected onto p_z orbitals. Weight of the orbital character is indicated by thickness (fatness). Only around $\overline{\Gamma}$, i.e., part of the $\overline{\Gamma M}$ and $\overline{\Gamma K}$ directions are shown for $\mathcal{E}= +0.5$ V/Å (red), $\mathcal{E}= 0.0$ V/Å (green) and $\mathcal{E}= -0.5$ V/Å (blue). Weight of spin majority and spin minority states are multiplied by a factor of 0.2 for better visualization. The Fermi energy is set to zero.



Supplementary Figure 6 | Effect of the electric fields on the magnetic moment. The magnetic moment of **a** Fe, **b** Rh and **c** Re (first layer) of Fe/Rh/Re(0001) as a function of spin spiral wave vector **q** at $\mathcal{E}$= +0.5 V/Å (red), $\mathcal{E}$= 0 V/Å (green) and $\mathcal{E}$= −0.5 V/Å (blue). The data of $\mathcal{E}$= 0 V/Å are taken from Ref. [2].



Supplementary Figure 7 | Atom resolved DMI for zero and finite electric fields. Total and atom resolved (Fe, Rh and nine Re layers) DMI energy (ΔE_{soc}) of flat cycloidal spin spirals for Fe/Rh/Re(0001) close to the $\overline{\Gamma}$ point along two high symmetry directions ($\overline{\Gamma\text{K}}$ and $\overline{\Gamma\text{M}}$) at **a** $\mathcal{E}= +0.5$ V/Å **b** $\mathcal{E}= 0$ V/Å and **c** $\mathcal{E}= -0.5$ V/Å.



Supplementary Figure 8 | Energy decomposition of minimum energy path (MEP) for an isolated skyrmion collapse. Total and individual energy contributions of Fe/Rh/Re(0001) along MEP for **a** $\mathcal{E}$= +0.5 V/Å **b** $\mathcal{E}$= 0.0 V/Å and **c** $\mathcal{E}$= −0.5 V/Å at B= 3 T. The energies are plotted with respect to the skyrmion (initial) state. Combined exchange (C. exchange) indicates a sum of the contribution from Heisenberg pair-wise exchange, biquadratic and three-site four spin interactions. For brevity, the four-site four spin interaction is denoted as 4-Spin.

[1] Paul, S., Haldar, S., von Malottki, S. & Heinze, S. Role of higher-order exchange interactions for skyrmion stability. *Nat. Commun.* **11**, 4756 (2020).

[2] Paul, S. & Heinze, S. Tailoring magnetic interactions in atomic bilayers of Rh and Fe on Re(0001). *Phys. Rev. B* **101**, 104408 (2020).