

Supplementary Information:

Ultrafast current and field driven domain-wall dynamics in van der Waals antiferromagnet MnPS₃

Ignacio M. Alliat¹, Richard F. L. Evans², Kostya S. Novoselov^{3,4}, & Elton J. G. Santos^{5,†}

¹*School of Mathematics and Physics, Queen's University Belfast, BT7 1NN, UK*

²*Department of Physics, The University of York, YO10 5DD, UK*

³*Department of Material Science & Engineering, National University of Singapore, Block EA, 9 Engineering Drive 1, 117575, Singapore*

⁴*Chongqing 2D Materials Institute, Liangjiang New Area, Chongqing 400714, China*

⁵*Institute for Condensed Matter Physics and Complex Systems, School of Physics and Astronomy, The University of Edinburgh, EH9 3FD, UK.*

[†]*Corresponding author: esantos@ed.ac.uk*

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30 **1 Ab initio Density Functional Theory including Hubbard– U corrections**

31 Ab-initio calculations were performed using Density Functional Theory (DFT) as implemented in
32 the VASP package^{1,2}. Within the generalized gradient approximation (GGA), the Perdew-Burke-
33 Ernzerhof (PBE) exchange-correlation functional was used. No van der Waals interactions were
34 taken into account given that the system of interest is a monolayer. A Hubbard U correction was
35 included through the Dudarev formulation³. The exchange parameter J was fixed as $J = 0.9$ eV
36 while the U energy was calculated through the linear response method as described below (Sup-
37plementary Note 2) resulting in a magnitude of $U = 2.58$ eV. These values of J and U were used in
38 all calculations. The PAW pseudopotentials were used, including GW corrections. The electronic
39 convergence criterion was set to 1×10^{-7} eV, while the structural optimizations were performed until
40 all forces were lower than 0.001 eV/Å. All calculations were spin polarized. A converged Gaussian
41 smearing with a low sigma value (0.022 eV) was used, as it resulted appropriate for semiconduc-
42tors and insulators. A vacuum space of 24 Å was introduced to avoid interactions between vertical
43 image replications of the unit cell. All calculations were performed with an energy cutoff of 600
44 eV, which is considerably higher than the maximum ENMAX in the pseudopotentials (208 eV)
45 and ensures very well converged results (Supplementary Fig. S1). For structural optimizations,
46 the $\mathbf{k}$ -space was sampled via a converged $7 \times 7 \times 1$ automatically generated Gamma-centered grid

47 (Supplementary Fig. S2). The **k**-point density was doubled for the calculations of the magnetic
48 parameters (exchange interactions, magnetic anisotropies, spin moment), where the energies of
49 different magnetic configurations are compared. This resulted in a $14 \times 7 \times 1$ grid for a $1 \times 2 \times 1$
50 supercell.

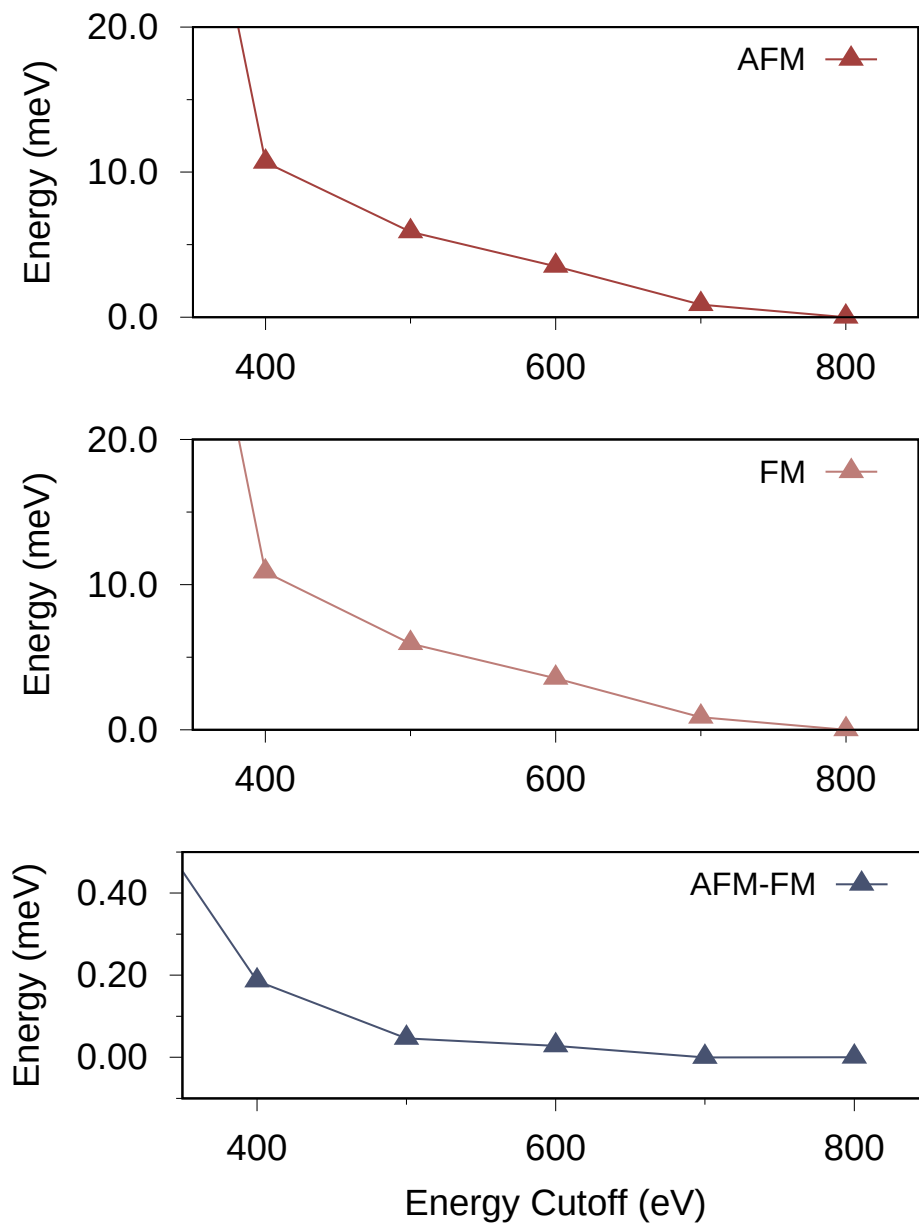


Figure S1: Convergence of the energy cutoff utilized to calculate the magnetic properties of monolayer MnPS_3 . Two spin configurations in terms of anti-ferromagnetic (AFM) and ferromagnetic (FM) were utilized. Results are shown individually for both AFM and FM, but the choice criterion was based on how well converged the energy difference between AFM and FM is rather than their absolute value. It is concluded that both differ by an accuracy better than $\sim 25 \mu\text{eV}$ when the cutoff increases from 600 eV (as used in this work) to 700 eV. Hence, 600 eV is deemed acceptable. The energies are relative to the largest cutoff utilized, e.g. 800 eV.

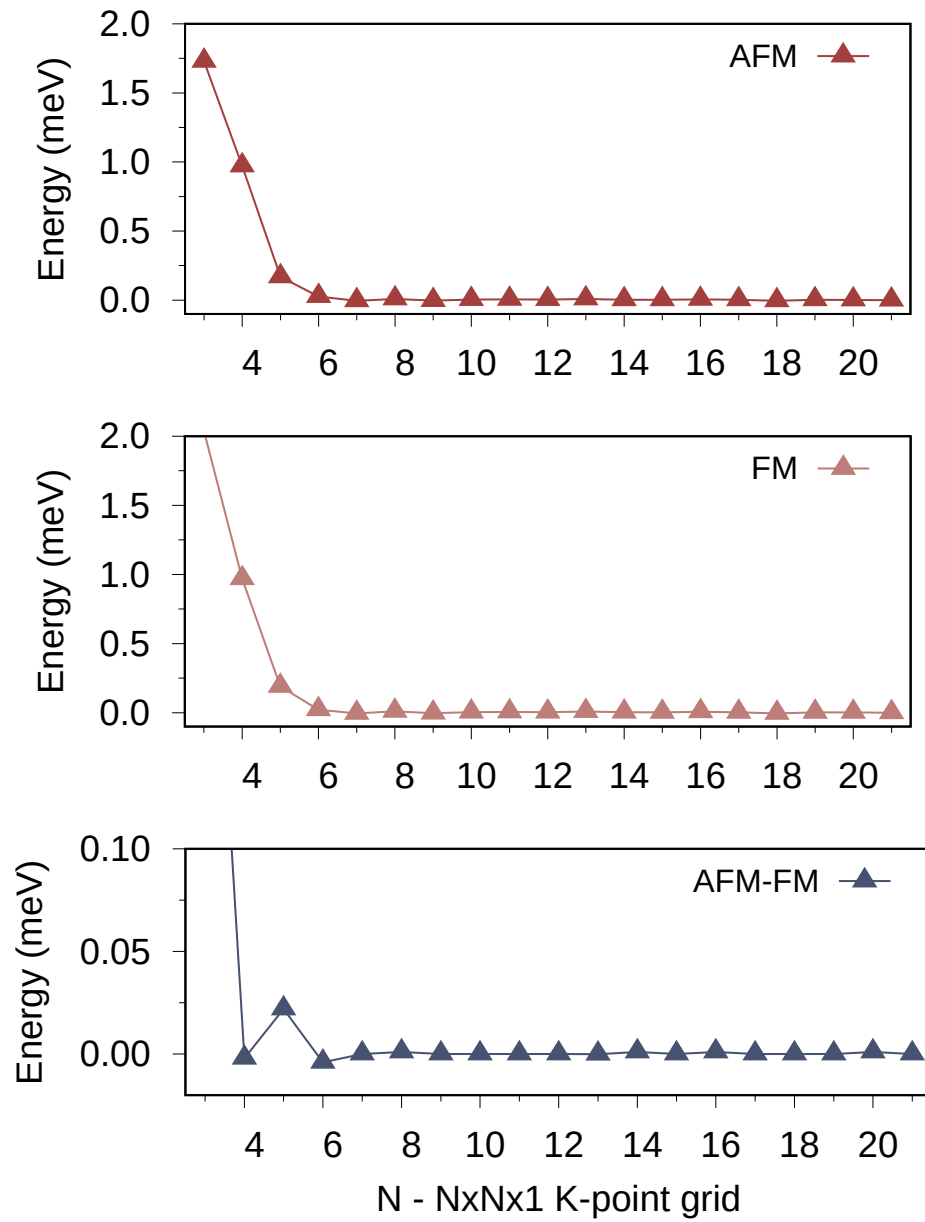


Figure S2: Convergence of the k-sampling grid utilized to calculate the magnetic properties of monolayer MnPS₃. Two spin configurations in terms of anti-ferromagnetic (AFM) and ferromagnetic (FM) were utilized. The difference between both spin configurations is showed on the bottom panel with negligible energy difference at $7 \times 7 \times 1$ grid. The energies are relative to the densest grid utilized.

2 Linear response calculations for evaluating the Hubbard U term

The GGA ground state was initially determined at $U = 0$ eV. This involved optimizing the lattice parameter as described in Supplementary Note 3. The lattice parameter obtained was 6.057 Å. Then, once the structure was determined, a single point calculation with a denser $\mathbf{k}$ -point grid ($14 \times 14 \times 1$) was done in order to obtain a reliable WAVECAR and CHGCAR files to be used in the linear response (LR) calculations. The LR simulations were done with a hidden VASP functionality (LDAUTYPE = 3) and they consist on perturbatively increasing the Hubbard U parameter of only one Mn atom in the unit cell with respect to a reference value of 0 eV. We did this in steps of 0.1 eV, up to ± 0.5 eV. Then, we obtain the density response function (χ) of this Mn atom as the derivative of its charge density with respect to the perturbation. We do this self-consistently (ICHARG = 1) and non-self-consistently (ICHARG = 11) to obtain χ and χ_0 , respectively. Then, we calculate the Hubbard parameter⁴ as $U = \chi_0^{-1} - \chi^{-1}$. The same procedure is done for a $1 \times$ unit cell (1 out of 2 Mn atoms is perturbed), a 2×2 (1 out of 8) and a 3×3 supercell (1 out of 18). The resulting value of U for the 3×3 supercell is $U = 2.5807$ eV, and it is used for all calculations in this work. The results can be found in Supplementary Figures S3-S4.

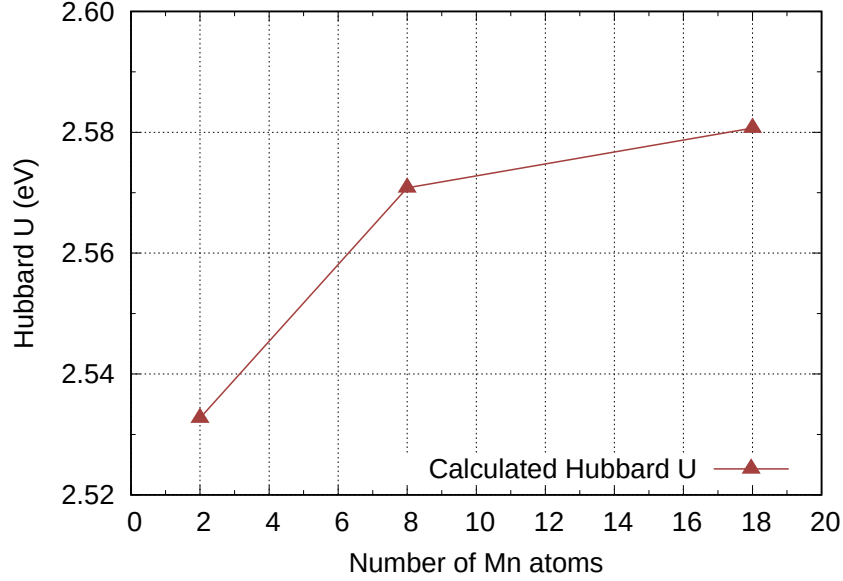


Figure S3: Converged value of the Hubbard U parameter.

3 Lattice parameter GGA+ U

When going from bulk to monolayer, MnPS_3 presents a transition from monoclinic (space group 12: $C2/m$) to hexagonal symmetry (space group 162: $P\bar{3}1m$)⁵. It recovers a 3-fold rotational symmetry as opposed to a monoclinic 2-fold one due to a distortion on the length of the sides of the hexagon arising from the stacking⁵. Therefore, the single-layer hexagonal structure was used to the calculations rather than a rectangular (monoclinic) cell. The structure was optimized for a Néel anti-ferromagnetic (AFM) configuration, which is the magnetic ground state for this material. As stated above, a Gamma centered $7 \times 7 \times 1$ $\mathbf{k}$ -point grid was used. The structural optimization was done by changing the magnitude of the lattice vectors in steps of 0.001 Å, relaxing the internal coordinates at every stage (not the cell, i.e. IBRION=2) and finding the minimum energy. The lattice parameter obtained was 6.109 Å (Figure S5).

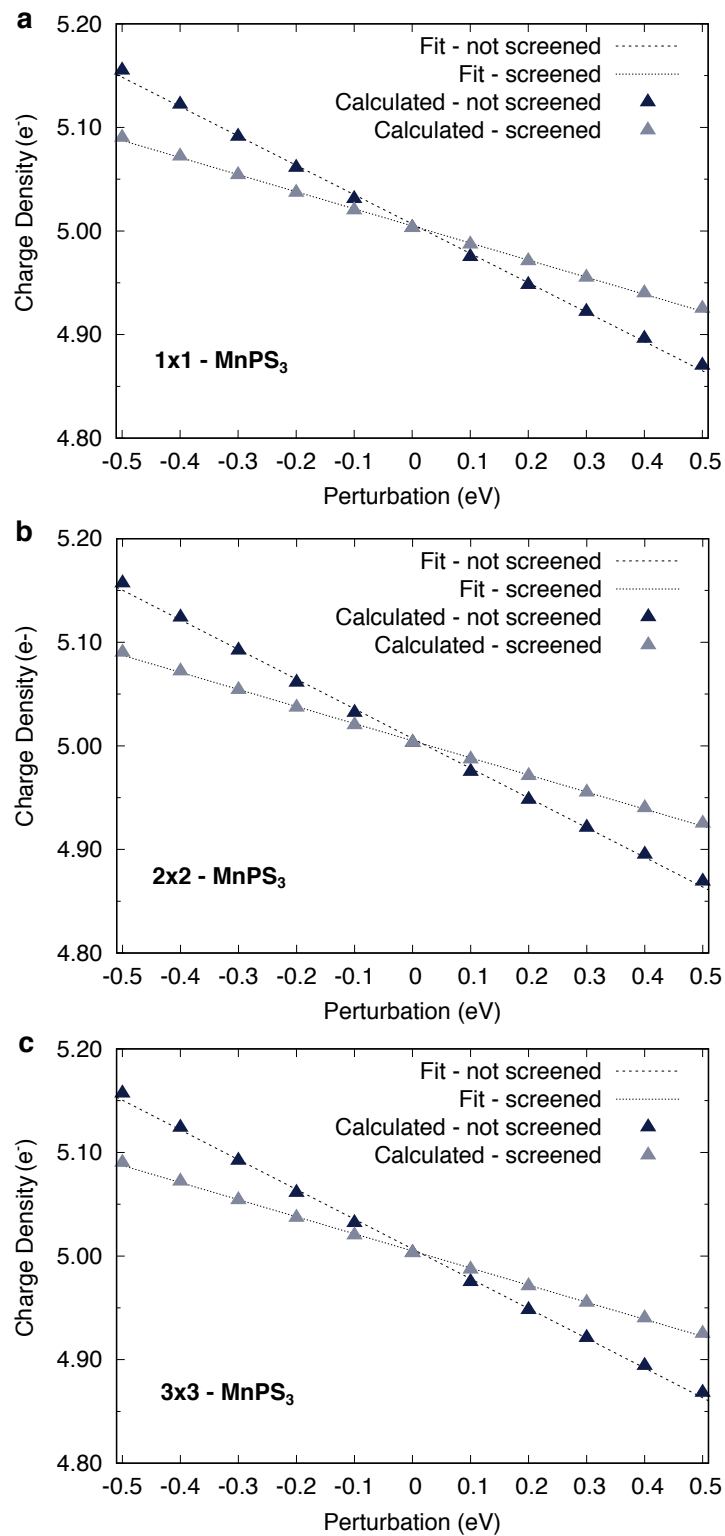


Figure S4: **a-c**, Variation of the charge density versus perturbation for screened and not screened interactions in 1×1 , 2×2 and 3×3 supercells of MnPS_3 , respectively. Minimum variations are observed as a function of the cell size.

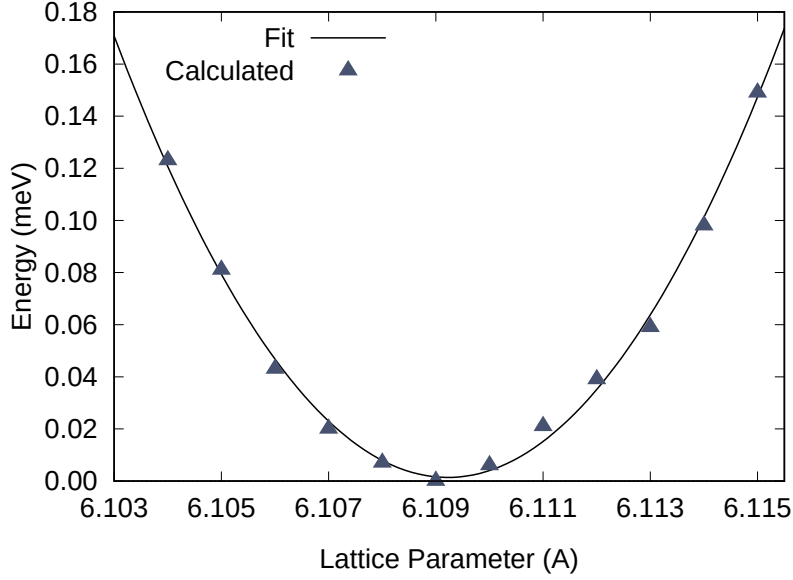


Figure S5: Optimization of the lattice constant of monolayer MnPS₃.

77 4 Method for extraction of parameters for micromagnetic simulations

The method to extract magnetic parameters was based on the XXZ anisotropic Heisenberg Hamiltonian:

$$\mathcal{H}_{bilinear} = - \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_{\langle i,j \rangle} \lambda_{ij} S_i^z S_j^z - D \sum_i (S_i^z)^2 \quad (1)$$

where $J_{i,j}$ is the bilinear (BL) exchange interactions between spins $\mathbf{S}_i$ and $\mathbf{S}_j$ at sites i and j , λ_{ij} is the anisotropic exchange and D is the on-site magnetic anisotropy. The model in Eq. 30 only includes bilinear terms since biquadratic exchange interaction is negligible⁶ in MnPS₃, and the $\langle i, j \rangle$ pairs are extended up to the third nearest neighbors (3NN). This spin Hamiltonian is then applied to each of the eight magnetic configurations considered in this work, resulting in a different equation given the relative orientation of neighbouring spins on each magnetic configuration (Supplementary Figure S6 and Supplementary Table S1). Moreover, each spin configuration corresponds to

one energy calculation obtained with the DFT+ U method including spin-orbit coupling. We can systematically determine all parameters in Eq.30 through a linear system of eight equations, one for each magnetic configuration.

$$E_{DFT}(eV) = g_{linear}(D, J_{1NN}, J_{2NN}, J_{3NN}, \lambda_{1NN}, \lambda_{2NN}, \lambda_{3NN}) \quad (2)$$

It is worth mentioning that the meaningful quantities that one can obtain from the ab initio simulations are the energy differences rather than the absolute values. By subtracting these equations in pairs, one can arrive to a linear system of six unknowns and eight equations of the form:

$$\Delta E(eV) = f_{linear}(D, J_{1NN}, J_{2NN}, J_{3NN}, \lambda_{1NN}, \lambda_{2NN}, \lambda_{3NN}) \quad (3)$$

78 where the various ΔE are defined as follows

$$\begin{aligned} \Delta E_1^x &= E_{AFM}^x - E_{FM}^x \\ \Delta E_2^x &= E_{zzAFM}^x - E_{strAFM}^x \\ \Delta E_3^x &= E_{strAFM}^x - E_{FM}^x \\ \Delta E_4^x &= E_{AFM}^x - E_{zzAFM}^x \\ \Delta E_1^z &= E_{AFM}^z - E_{FM}^z \\ \Delta E_2^z &= E_{zzAFM}^z - E_{strAFM}^z \\ \Delta E_3^z &= E_{strAFM}^z - E_{FM}^z \\ \Delta E_4^z &= E_{AFM}^z - E_{zzAFM}^z \end{aligned} \quad (4)$$

79 In the simplest case, where every spin moment ($\mathbf{S}_i$) is either perfectly out of plane or in plane, these
80 equations take simple forms and the system can be solved for the magnetic parameters analytically.

81 However, our calculations reveal that the in-plane configurations are never perfectly in plane, they
 82 are rather *canted* configurations due to the high out-of-plane anisotropy of the system. That is, the
 83 spins deviated from the xy -plane by a finite angle ($\approx 0.2^\circ - 5.7^\circ$). In a more general case, when the
 84 canted configurations are taken into account, the system of equations must be solved in its matrix
 85 form. To this end, a Fortran code was written as part of this work. The right-hand side of these
 86 equations is then obtained by reading the spin orientation from the OUTCAR output from VASP and
 87 calculating each sum in Eq.30 in its vectorial form, evaluating the dot product between neighboring
 88 (canted) spin moments ($\mathbf{S}_i$). Alternatively, the *Fortran* code allows to select *perfect* spins, where
 89 every spin moment is considered to be either completely in-plane or out-of-plane. The results
 90 obtained are presented in Table S1. The results in the canted and perfect spins configurations differ
 91 at most in the order of tens of μeV . Furthermore, the results with perfect spins agree with those
 92 obtained via the analytical solution of these equations, confirming that the matrix resolution of the
 93 system implemented in the code is correct.

	Perfect spins		Canted configuration	
	$S^2 meV$	meV	$S^2 meV$	meV
J_{1NN}	-4.946	-0.791	-4.950	-0.792
J_{2NN}	-0.211	-0.034	-0.210	-0.034
J_{3NN}	-1.959	-0.313	-1.958	-0.313
λ_{1NN}	0.071	0.011	0.075	0.012
λ_{2NN}	-0.104	-0.017	-0.105	-0.017
λ_{3NN}	-0.109	-0.017	-0.109	-0.017
D	0.684	0.109	0.677	0.108

Table S1: Calculated magnetic parameters for MnPS₃ using DFT+ U methods (Supplementary Note 1). Bilinear exchange J_{ij} and the anisotropic exchange λ_{ij} are calculated up to the 3rd-nearest neighbors (NN): J_{1NN} , J_{2NN} , J_{3NN} , λ_{1NN} , λ_{2NN} and λ_{3NN} . D represent the on-site magnetic anisotropy. $S = 5/2$ for Mn. We include both values of each parameters with and without the S^2 factor in meV's. *Perfect spins* parameters assumed that the spins are fully aligned in-plane or out-of-plane, while *Canted configuration* include a deviation by a small finite angle ($\lesssim 4^\circ - 6^\circ$).

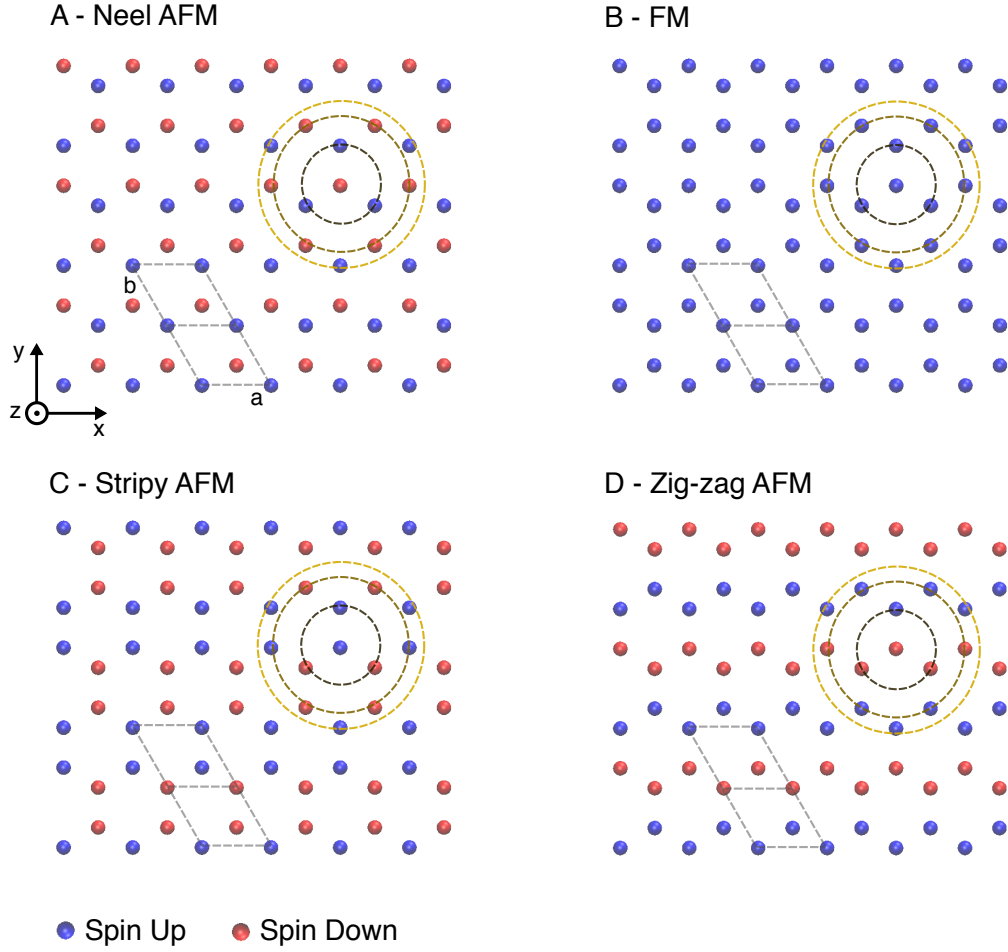


Figure S6: Spin configurations utilized in the DFT+ U simulations to calculate the exchange parameters (J_{ij} , λ_{ij}) and on-site magnetic anisotropy (D) in Eq.30 for monolayer MnPS₃. All four magnetic configurations were calculated considering non-collinear magnetism with the quantization axis in either z or x , which resulted in Eqs.4. The different circle sizes correspond to first-, second- and third-nearest neighbors accordingly to the diameter length (e.g. from smallest to largest) respectively. The honeycomb unit cell is drawn in each configuration. The total energy for each configurations is the following: Néel AFM _{x} = −111.90757 eV, Néel AFM _{z} = −111.90825 eV, FM _{x} = −111.74187 eV, FM _{z} = −111.74165 eV, Stripy AFM _{x} = −111.82776 eV, Stripy AFM _{z} = −111.82974 eV, Zigzag AFM _{x} = −111.83520 eV, Zigzag AFM _{z} = −111.84036 eV.

5 Monte Carlo simulations

Macromagnetic quantities such as magnetization, susceptibility or Néel temperature were calculated by using Monte Carlo method implemented in a home-brewed version of the VAMPIRE atomistic spin dynamics package⁷. A 50 nm×50 nm square flake (15580 sites in total) with periodic boundary conditions was utilized for these simulations. The temperature was sampled by 1 K in the range 0-200 K, with a finer mesh around the critical temperature. At each temperature, 1×10^6 Monte Carlo steps were done to obtain the magnetic configuration. This is due to a slow convergence of the susceptibility with respect to the number of steps and in order to avoid scattering of the data. The normalized Néel vector ranging from 0 to 1 was plotted as a function of temperature (Supplementary Fig. S7). According to our simulations, the transition temperature is 44 K in the monolayer limit. As regards the magnetic susceptibility, plots were produced for the whole AFM material including both sublattices (Supplementary Fig. S8) as well as for one sub-lattice (Supplementary Fig. S9). The latter shows a ferromagnetic (FM) response, i.e. a peak in the susceptibility at the critical temperature.

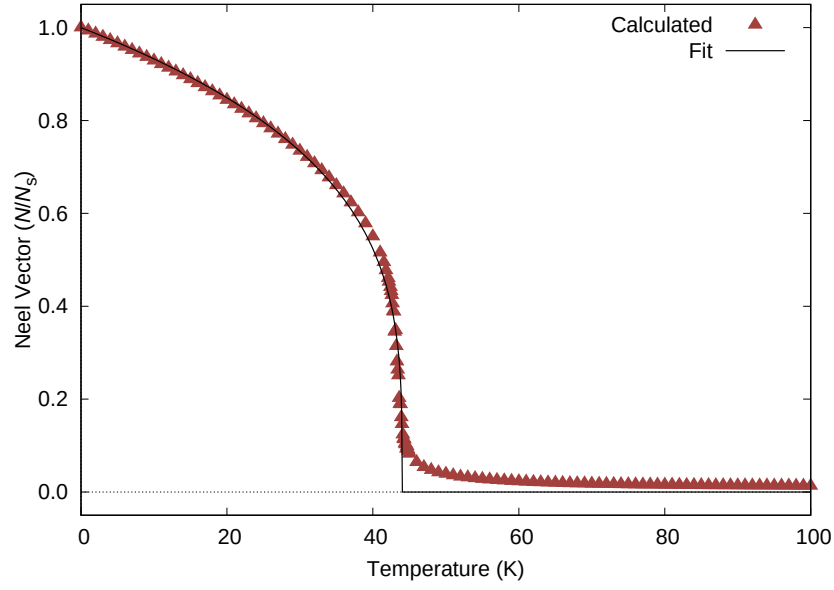


Figure S7: Néel vector (a.u.) versus temperature (in K) for monolayer MnPS₃ including up to 3rd nearest-neighbors as described in Supplementary Note 4.

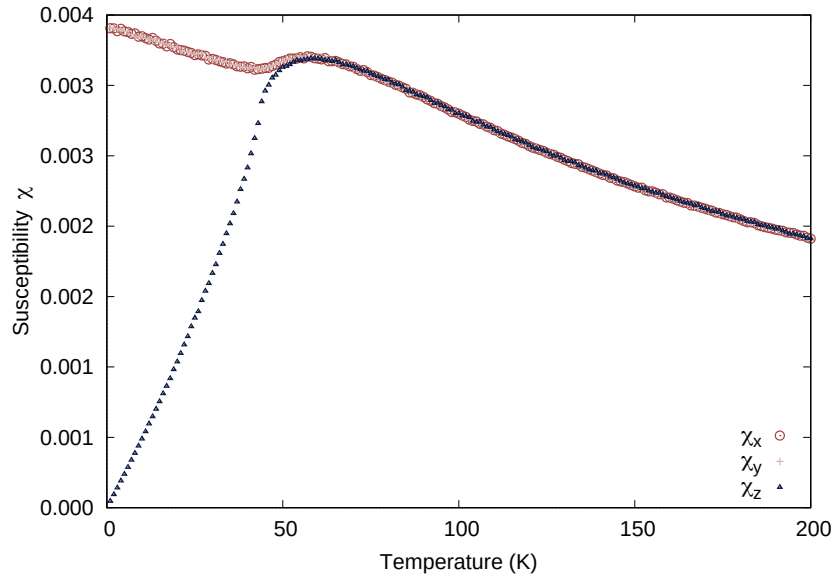


Figure S8: Magnetic susceptibility χ of monolayer MnPS₃ as a function of temperature (in K) projected on in-plane (χ_x , χ_y) and out-of-plane (χ_z) coordinates. Both sub-lattices are included in the simulation. This calculation included the formalism described in Supplementary Note 4.

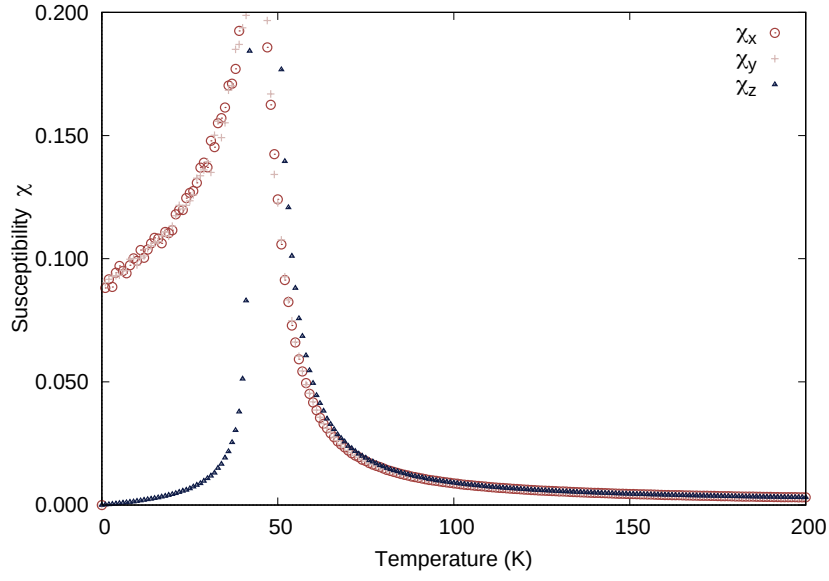


Figure S9: Magnetic susceptibility χ of monolayer MnPS_3 as a function of temperature (in K) projected on in-plane (χ_x , χ_y) and out-of-plane (χ_z) coordinates. Just one sub-lattice is included in the simulation which represents a ferromagnetic coupling between the spins. This calculation included the formalism described in Supplementary Note 4.

6 Spin dynamics based on Landau-Lifshitz-Gilbert equation methods

Spin dynamics simulations were conducted to address the formation of magnetic domains and domain walls. The formalism is based on Landau-Lifshitz-Gilbert (LLG) equations methods⁸ incorporating temperature effects through Langevin dynamics. A modified Heun integration method⁹ was utilized in the calculations to allow longer simulation time (several nanoseconds). Time step used of 0.01 fs was used in all simulations. We used a Gilbert damping α parameter of $\alpha = 0.01$ to simulate the dynamics of domains. α is typically in the range of 0.005-0.1 in most materials¹⁰⁻¹³.

7 Winded and unwinded domain walls in MnPS₃

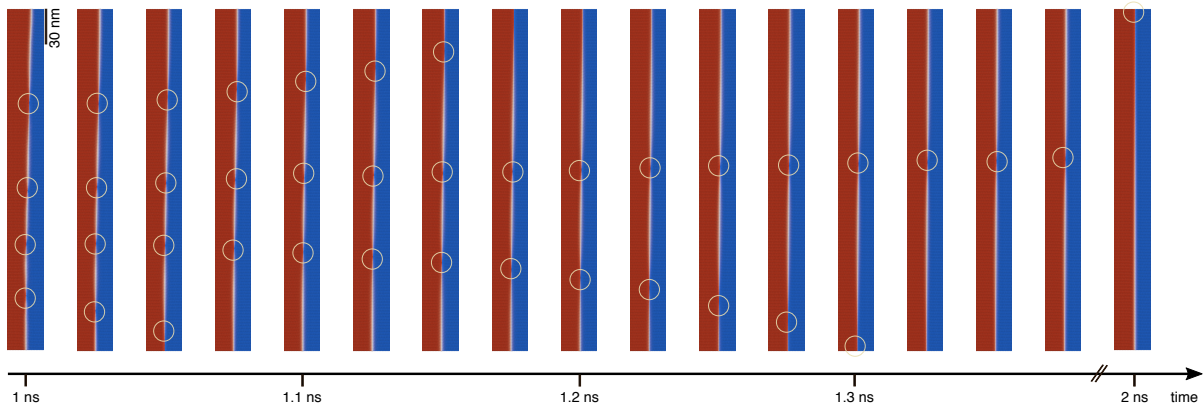


Figure S10: Snapshots at different simulation times (ns) of the evolution of the domain walls in MnPS₃. The circles highlighted the formation of nodes (regions where spins alternate dynamically along the wall profile rather than between magnetic domains) in a winded state which progressively moved out of the domain wall boundary as time progresses. At longer times, the domain wall becomes unwinded with no presence of nodes.

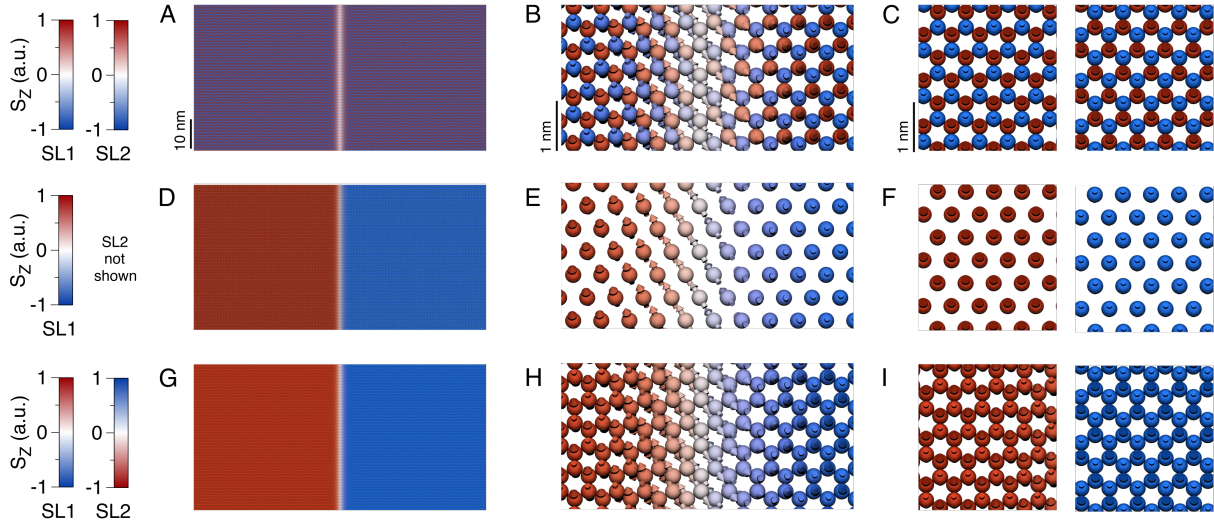


Figure S11: Schematic of the color convention utilized to plot the magnetic domains in the anti-ferromagnetic MnPS_3 .

A-C, Show how both magnetic domains (faint violet) apart from the domain wall (faint white) will look the same (**A**) if red (blue) is assigned to up (down) irrespective of the sub-lattice a given site belongs to. Detailed renders are shown at (**B**) the domain wall and (**C**) away from it. In this case, the domains are hard to distinguish away from the wall. **D-F**, Show the result if only one sub-lattice is rendered. The desired visual effect is achieved at the expense of losing half of the atoms. If the other sub-lattice is to be represented including all atoms in the system then the colour scheme for the second sub-lattice should be inverted. **G-I**, The final result used throughout the figures in this study. Colour scales are shown on the left for each convention.

We model the motion of electrons through the system using a circuit theory approach, considering a finite difference discretization of the material into $(1 \text{ nm})^3$ cells. The total resistance of a device is given by the summation of pairwise cell resistances $R = \sum_{ij} R_{ij}$ along the current direction. Considering two neighboring cells i, j with reduced magnetizations $\mathbf{m}_i, \mathbf{m}_j$, where $\mathbf{m} = \mathbf{M}(T)/M_S(0)$, the resistance can be expressed as:

$$R_{ij} = R_{\text{ep}} + \frac{R_{\text{es}}}{2} (1 - \mathbf{m}_i \cdot \mathbf{m}_j); \quad (5)$$

where R_{ep} is the resistance arising from electron-phonon scattering, and R_{es} is the resistance arising from electron-spin ($s-d$) scattering. For ferromagnetic materials, any misalignment of magnetization leads to an increase in the resistance due to electron-spin scattering. The current flow is computed from the sum of all resistances in all cells parallel to the current path, given a defined applied voltage V using Ohm's law $I = V/R$. At the same time, the misalignment of magnetization also leads to spin-transfer torque on the magnetization in that cell due to the conservation of angular momentum. We express this in terms of adiabatic and non-adiabatic spin transport coefficients using the Slonczewski approach¹⁴. For computational efficiency the additional torques are included within the effective field calculation for the LLG equation¹⁵, with the total effective field given by:

$$\mathbf{B}_{\text{eff}}^i = -\frac{1}{\mu_S} \frac{\partial \mathcal{H}}{\partial \mathbf{S}} + \mathbf{B}_{\text{th}} + a_J (\mathbf{S} \times \mathbf{p}) + b_J \mathbf{p} \quad (6)$$

where $\mathcal{H}$ is the spin Hamiltonian, $\mathbf{B}_{\text{th}}$ is the stochastic Langevin thermal field^{7,16}, a_J and b_J are the adiabatic and non-adiabatic spin transfer torque coefficients respectively, and $\mathbf{p}$ is the electron spin

120 polarization vector. The polarization vector is defined on a cell-wise basis as the magnetization
121 from the previous cell along the current flow direction¹⁷. In the case of non-magnetic or param-
122 agnetic materials, the spin-dependent scattering is zero and so the total resistance depends only
123 on spin independent (electron-phonon) scattering. A ferromagnetic sample including a domain
124 wall will show a higher resistance due to electron-spin scattering at the wall, while the conserva-
125 tion of angular momentum will lead to a motion of the domain wall along the current direction.
126 Our model gives a natural configurational dependence of the total resistance and thus the dynamic
127 spatial variations of current flow.

Domain wall velocity vs. current density: low-velocity regime The equation for the domain wall (DW) velocity v as a function of current density j can be written as¹⁸:

$$v = \frac{\gamma\sigma B_D}{2\alpha} \quad (7)$$

where γ is the gyromagnetic ratio, σ is the DW width, α is the Gilbert damping factor, and B_D is the field-like component of the spin-orbit torque. B_D is defined as:

$$B_D = \frac{\mu_B \theta_{SH}}{\gamma e m_s t_z} j \quad (8)$$

where μ_B is the Bohr magneton, θ_{SH} is the effective spin Hall angle, e is the electron charge, $m_s = 5\mu_B/\Omega_{cell}$ is the modulus of the magnetization per sub-lattice, t_z is the layer thickness and j is the current density. At the low-velocity regime, σ is constant ($\sigma \approx \sigma_o = 3.4188$ nm), which results in:

$$v = C_{model} j \quad (9)$$

where $C_{model} = \frac{\sigma\mu_B\theta_{SH}}{|e|m_s t_z 2\alpha}$. Even though we have the majority of the parameters in C_{model} , the value of θ_{SH} is unknown for monolayer MnPS₃ which ramps up the determination of C_{model} . However, we can assume a similar linear dependence between v and j , and determine the fitting coefficient from our dataset via:

$$v = C_{fitting} j \quad (10)$$

129 with $C_{fitting} = 67.11 \times 10^{-13} \frac{m^3}{C}$.

Domain wall velocity vs. current density: relativistic regime Now, we consider relativistic corrections to the DW width σ ^{18,19}:

$$\sigma = \sigma_o \sqrt{1 - (v/v_{g2})^2} \quad (11)$$

where v_{g2} is the maximum group velocity of the spin-waves in MnPS₃. Including Eq.11 in Eq. 7 we obtain:

$$v = \frac{\sigma_o \mu_B \theta_{SH}}{|e| m_s t_z 2 \alpha} \sqrt{1 - (v/v_{g2})^2} j \quad (12)$$

130 Notably, v is on both sides of Eq.12 which means that it must be obtained self-consistently.
 131 Doing so, we can satisfactorily reproduce the non-linear trend observed for the wall velocities at
 132 large current densities as shown in Figure 3d.

Domain wall mass vs domain wall velocity The effective inertial mass M_{DW} of an antiferromagnetic wall is given by¹⁹:

$$M_{DW} = \rho \int \frac{\partial \mathbf{n}}{\partial r} \cdot \frac{\partial \mathbf{n}}{\partial r} dV \quad (13)$$

where $\mathbf{n} = \mathbf{m}_1(\mathbf{r}) - \mathbf{m}_2(\mathbf{r})$, with $\mathbf{m}_{1,2}(\mathbf{r})$ is the magnetization in each sublattice, $\rho = \frac{1}{J_{1NN} \gamma^2}$, and J_{1NN} is the exchange parameter for the first nearest-neighbours. Supplementary Eq. 13 can be solved using the Walker ansatz²⁰:

$$M_{DW} = \frac{2 \rho w t_z \pi}{\sigma} \quad (14)$$

where w is the DW length (or the width of the stripe of the material), and an additional factor π is included because of the definition of σ in the main text. Relativistic corrections appeared via

Lorentz contraction factor in σ (Supplementary Eq.11) resulting in:

$$M_{DW} = \frac{2\rho wt_z \pi}{\sigma_o \sqrt{1 - (v/v_{max})^2}} \quad (15)$$

Estimation of θ_{SH} From Eqs. 7-10, and assuming $C_{fitting} \approx C_{model}$ we can estimate the value of θ_{sh} using $C_{model} = \frac{\sigma \mu_B \theta_{SH}}{|e| m_s t_z 2 \alpha}$ and $C_{fitting} = 67.11 \times 10^{-13} \frac{m^3}{C}$. The parameters in C_{model} are $\sigma = \sigma_o = 3.4188 \text{ nm}$, $|e| = 1.602 \times 10^{-19} \text{ C}$, $t_z = 0.64 \text{ nm}$ and the Gilbert damping $\alpha = 0.01$. As regards the modulus of the magnetization per lattice m_s , it is $m_s = 5\mu_B/\Omega_{cell}$. Since the volume of the cell is $\Omega_{cell} = \cos(\pi/6)a_o^2 t_z$, it cancels out the thickness, arriving to:

$$C_{model} = \frac{\lambda \theta_{SH} \cos(\pi/6) a_o^2}{10|e|\alpha} \quad (16)$$

133 where the lattice parameter is $a_o = 6.109 \text{ \AA}$. From Eq.16 and the value of $C_{fitting}$, we can find

134 $\theta_{SH} = 1.0 \times 10^{-5}$.

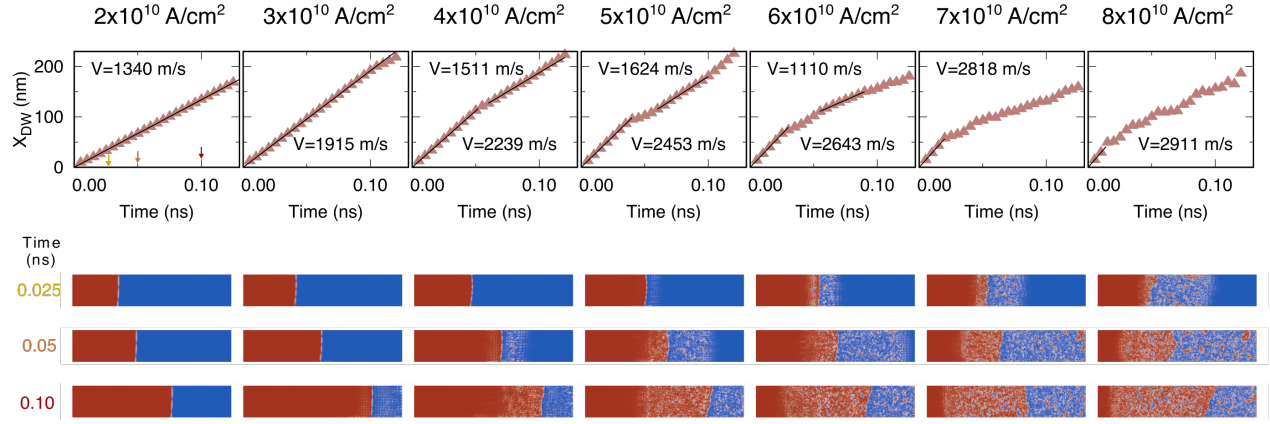


Figure S12: Top row: Variation of the position of the domain-wall X_{DW} (nm) with time(ns) at different current densities ($2 - 8 \times 10^{10} \text{ A cm}^{-2}$). For densities between $4 - 6 \times 10^{10} \text{ A cm}^{-2}$, two velocities could be extracted corresponding to two states of the domain-wall before and after it starting loosing its coherence over the dynamics. For densities above $6 \times 10^{10} \text{ A cm}^{-2}$, the domain wall remains for few fraction of ns with a more complex pattern of the spins over the wall. Bottom row: Snapshots of the domain wall motion at the different current densities stated at the top panels, and times. The different colours correspond to convention utilized to display the magnetic domains in an antiferromagnetic material (see Supplementary Note 8).

12 Magnon dispersion model for Néel AFM bipartite lattice

Spin Hamiltonian The model used in this work corresponds to an anisotropic Heisenberg Hamiltonian, where anisotropic exchange is included within an XXZ model. This implies neglecting off-diagonal terms of the exchange tensor $\mathbb{J}_{ij}$ such that two type of exchange parameters are retained, i.e., an isotropic exchange ($J^{XX} = J^{YY} = J$) and an out-of-plane anisotropic contribution ($J^{ZZ} = J + \lambda$). Only bilinear terms were included in the Hamiltonian, since biquadratic anisotropy as well as biquadratic exchange are negligible for MnPS₃⁶. These assumptions result in the spin Hamiltonian shown in Eq. 17.

$$\hat{\mathcal{H}} = -D \sum_i (S_i^z)^2 - \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_{\langle i,j \rangle} \lambda_{ij} S_i^z S_j^z \quad (17)$$

where D is the on-site anisotropy, λ_{ij} the inter-site anisotropy and J_{ij} represents the isotropic exchange. The hat is omitted in the spin angular momentum operators for simplicity. From this equation, we can include the x , y and z components of the spin angular momentum operators explicitly and arrive to Eq. 18.

$$\hat{\mathcal{H}} = -D \sum_i (S_i^z)^2 - \sum_{\langle i,j \rangle} J_{ij} S_i^x S_j^x - \sum_{\langle i,j \rangle} J_{ij} S_i^y S_j^y - \sum_{\langle i,j \rangle} (\lambda_{ij} + J_{ij}) S_i^z S_j^z \quad (18)$$

We now introduce the rising and lowering ladder operators of the spin angular momentum as in Eq. 3. Using these definitions to express the Hamiltonian, Eq. 20 is obtained.

$$S^\pm = S^x \pm iS^y \quad S^x = \frac{S^+ + S^-}{2} \quad S^y = \frac{S^+ - S^-}{2i} \quad (19)$$

$$\mathcal{H} = -D \sum_i (S_i^z)^2 - \sum_{\langle i,j \rangle} J_{ij} \frac{1}{2} (S_i^+ S_j^- + S_i^- S_j^+) - \sum_{\langle i,j \rangle} (\lambda_{ij} + J_{ij}) S_i^z S_j^z \quad (20)$$

150 We now extend the exchange interactions up to the third nearest neighbours (3NNs), obtain-
 151 ing Eq. 21.

$$\begin{aligned} \mathcal{H} = & \underbrace{-D \sum_i (S_i^z)^2}_{\mathcal{H}_D} - \underbrace{J_1 \sum_{\langle i,j \rangle_1} \frac{1}{2} (S_i^+ S_j^- + S_i^- S_j^+)}_{\mathcal{H}_{1NN}} - \underbrace{(\lambda_1 + J_1) \sum_{\langle i,j \rangle_1} S_i^z S_j^z}_{\mathcal{H}_{1NN}^\lambda} \\ & - \underbrace{J_2 \sum_{\langle i,j \rangle_2} \frac{1}{2} (S_i^+ S_j^- + S_i^- S_j^+)}_{\mathcal{H}_{2NN}} - \underbrace{(\lambda_2 + J_2) \sum_{\langle i,j \rangle_2} S_i^z S_j^z}_{\mathcal{H}_{2NN}^\lambda} \\ & - \underbrace{J_3 \sum_{\langle i,j \rangle_3} \frac{1}{2} (S_i^+ S_j^- + S_i^- S_j^+)}_{\mathcal{H}_{3NN}} - \underbrace{(\lambda_3 + J_3) \sum_{\langle i,j \rangle_3} S_i^z S_j^z}_{\mathcal{H}_{3NN}^\lambda} \end{aligned} \quad (21)$$

152 **Holstein-Primakoff transformation** We now introduce the Holstein-Primakoff (HP) transformation²¹,
 153 by which we express the spin angular momentum operators in terms the bosonic creation and anni-
 154 hilation operators. We do so considering a bipartite lattice, where the 1NNs of a site in sub-lattice
 155 A(B) always belong to sub-lattice B(A). The letter assigned to each creation and annihilation op-
 156 erator denotes the sub-lattice to which a particular site belongs. This transformation allows us to

157 parametrise small fluctuations about the Néel state.

For $i \in A$ sub-lattice:

$$S_i^z = (s - a_i^\dagger a_i) \quad (22a)$$

$$S_i^+ \sim \sqrt{2s} a_i \quad (22b)$$

$$S_i^- \sim a_i^\dagger \sqrt{2s} \quad (22c)$$

For $i \in B$ sub-lattice:

$$S_i^z = (b_i^\dagger b_i - s) \quad (22d)$$

$$S_i^+ \sim \sqrt{2s} b_i^\dagger \quad (22e)$$

$$S_i^- \sim b_i \sqrt{2s} \quad (22f)$$

159 When applying the HP transformation to the Hamiltonian operator in Eq. 21, careful con-
 160 sideration should be given to whether a site and its neighbours belong to the same or opposite
 161 sub-lattices in a Néel AFM configuration. For instance, given a site in sub-lattice A, the 2NNs will
 162 be in the same sublattice A, while the 1NN and 3NNs will belong to the opposite sublattice B. Via
 163 this transformation, Eqs. 23 are obtained for each constituent part of the Hamiltonian in Eq. 21.
 164 We note that terms of order 4 or higher in the bosonic operators ($\mathcal{O}(a^4)$) were neglected.

$$\frac{H_D}{D} = - \sum_{i \in A} s^2 - 2s(a_i^\dagger a_i) - \sum_{j \in B} s^2 - 2s(b_j^\dagger b_j) \quad (23a)$$

$$\frac{H_{1NN}}{J_1} = - \sum_{i \in A} \sum_{j \in 1NN_i} \frac{1}{2} 2s(a_i b_j + a_i^\dagger b_j^\dagger) \quad (23b)$$

$$\frac{H_{1NN}^\lambda}{\lambda_1 + J_1} = - \sum_{i \in A} \sum_{j \in 1NN_i} -s^2 + s(a_i^\dagger a_i + b_j^\dagger b_j) \quad (23c)$$

$$\frac{H_{2NN}}{J_2} = - \sum_{i \in A} \sum_{j \in 2NN_i} \frac{1}{2} 2s(a_i a_j^\dagger + a_i^\dagger a_j) - \sum_{j \in B} \sum_{i \in 2NN_j} \frac{1}{2} 2s(b_i b_j^\dagger + b_i^\dagger b_j) \quad (23d)$$

$$\frac{H_{2NN}^\lambda}{\lambda_2 + J_2} = - \sum_{i \in A} \sum_{j \in 2NN_i} s^2 - s(a_i^\dagger a_i + a_j^\dagger a_j) - \sum_{j \in B} \sum_{i \in 2NN_j} s^2 - s(b_i^\dagger b_i + b_j^\dagger b_j) \quad (23e)$$

$$\frac{H_{3NN}}{J_3} = - \sum_{i \in A} \sum_{j \in 3NN_i} \frac{1}{2} 2s(a_i b_j + a_i^\dagger b_j^\dagger) \quad (23f)$$

$$\frac{H_{3NN}^\lambda}{\lambda_3 + J_3} = - \sum_{i \in A} \sum_{j \in 3NN_i} -s^2 + s(a_i^\dagger a_i + b_j^\dagger b_j) \quad (23g)$$

165 **Bosonic operators in k-space** We now express the Hamiltonian in momentum space to simplify
 166 its diagonalization, for which we must first Fourier-transform the creation and annihilation oper-
 167 ators (see Eqs. 24). The normalisation factor results from the fact that each set of operators is
 168 defined only on $N/2$ sites, where N is the total number of spins in the lattice.

For $i \in A$ sublattice:

$$a_i = \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} A_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{r}_i} \quad (24a)$$

$$a_i^\dagger = \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger e^{+i\mathbf{k} \cdot \mathbf{r}_i} \quad (24b)$$

For $i \in B$ sublattice:

$$b_i = \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} B_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{r}_i} \quad (24c)$$

$$b_i^\dagger = \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} B_{\mathbf{k}}^\dagger e^{+i\mathbf{k} \cdot \mathbf{r}_i} \quad (24d)$$

170

The products of bosonic operators that appear in Eqs. 23 can now be re-written in k-space

171

applying the definitions in Eqs. 24, thus obtaining Eqs. 25.

$$\sum_{\langle i,j \rangle} a_i^\dagger a_i = z \sum_i \frac{2}{N} \sum_{\mathbf{k}, \mathbf{k}'} A_{\mathbf{k}}^\dagger A_{\mathbf{k}'} e^{i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{r}_i} = \sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger A_{\mathbf{k}} \quad (25a)$$

$$\sum_{\langle i,j \rangle} b_j^\dagger b_j = z \sum_j \frac{2}{N} \sum_{\mathbf{k}, \mathbf{k}'} B_{\mathbf{k}}^\dagger B_{\mathbf{k}'} e^{i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{r}_j} = \sum_{\mathbf{k}} B_{\mathbf{k}}^\dagger B_{\mathbf{k}} \quad (25b)$$

$$\sum_{\langle i,j \rangle} a_i b_j = \sum_i \frac{2}{N} \sum_{\mathbf{k}, \mathbf{k}'} A_{\mathbf{k}} B_{\mathbf{k}'} e^{-i(\mathbf{k}+\mathbf{k}') \cdot \mathbf{r}_i} \sum_j e^{-i\mathbf{k}' \cdot (\mathbf{r}_j - \mathbf{r}_i)} = \sum_{\mathbf{k}} A_{\mathbf{k}} B_{-\mathbf{k}} \gamma(\mathbf{k}) \quad (25c)$$

$$\sum_{\langle i,j \rangle} a_i^\dagger b_j^\dagger = \sum_i \frac{2}{N} \sum_{\mathbf{k}, \mathbf{k}'} A_{\mathbf{k}}^\dagger B_{\mathbf{k}'}^\dagger e^{+i(\mathbf{k}+\mathbf{k}') \cdot \mathbf{r}_i} \sum_j e^{+i\mathbf{k}' \cdot (\mathbf{r}_j - \mathbf{r}_i)} = \sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger B_{-\mathbf{k}}^\dagger \gamma^*(\mathbf{k}) \quad (25d)$$

$$\sum_{\langle i,j \rangle} a_i a_j^\dagger = \sum_i \frac{2}{N} \sum_{\mathbf{k}, \mathbf{k}'} A_{\mathbf{k}} A_{\mathbf{k}'}^\dagger e^{-i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{r}_i} \sum_j e^{+i\mathbf{k}' \cdot (\mathbf{r}_j - \mathbf{r}_i)} = \sum_{\mathbf{k}} A_{\mathbf{k}} A_{\mathbf{k}}^\dagger \gamma(\mathbf{k}) \quad (25e)$$

$$\sum_{\langle i,j \rangle} a_i^\dagger a_j = \sum_i \frac{2}{N} \sum_{\mathbf{k}, \mathbf{k}'} A_{\mathbf{k}}^\dagger A_{\mathbf{k}'} e^{+i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{r}_i} \sum_j e^{-i\mathbf{k}' \cdot (\mathbf{r}_j - \mathbf{r}_i)} = \sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger A_{\mathbf{k}} \gamma^*(\mathbf{k}) \quad (25f)$$

$$\sum_{\langle i,j \rangle} b_i b_j^\dagger = \sum_i \frac{2}{N} \sum_{\mathbf{k}, \mathbf{k}'} B_{\mathbf{k}} B_{\mathbf{k}'}^\dagger e^{-i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{r}_i} \sum_j e^{+i\mathbf{k}' \cdot (\mathbf{r}_j - \mathbf{r}_i)} = \sum_{\mathbf{k}} B_{\mathbf{k}} B_{\mathbf{k}}^\dagger \gamma(\mathbf{k}) \quad (25g)$$

$$\sum_{\langle i,j \rangle} b_i^\dagger b_j = \sum_i \frac{2}{N} \sum_{\mathbf{k}, \mathbf{k}'} B_{\mathbf{k}}^\dagger B_{\mathbf{k}'} e^{+i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{r}_i} \sum_j e^{-i\mathbf{k}' \cdot (\mathbf{r}_j - \mathbf{r}_i)} = \sum_{\mathbf{k}} B_{\mathbf{k}}^\dagger B_{\mathbf{k}} \gamma^*(\mathbf{k}) \quad (25h)$$

172

These expressions initially include double sums over $\mathbf{k}$ and $\mathbf{k}'$. However, they have only

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non-zero values when either $\mathbf{k} = \mathbf{k}'$ (when only one operator is dagger) or $\mathbf{k} = -\mathbf{k}'$ (if both are

174

either dagger or non-dagger). We note that the sums over i turned into an $N/2$ factor that cancelled

175

out the pre-factors in Eqs. 25. The sums over j , i.e., the NNs of i , turn into a pre-factor denoting

176

the number of NNs to the site i (z_1 , z_2 or z_3). The exception to this is when an exponential term

177

with $(\mathbf{r}_j - \mathbf{r}_i)$ appears in the sum in Eqs. 25. In such cases, we express them in terms of the $\gamma(\mathbf{k})$

178 factors, as defined in Eqs. 26.

$$\gamma_1(\mathbf{k}) = \sum_{j \in 1\text{NN}_i} e^{+i\mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_i)} \quad (26a)$$

$$\gamma_2(\mathbf{k}) = \sum_{j \in 2\text{NN}_i} e^{+i\mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_i)} \quad (26b)$$

$$\gamma_3(\mathbf{k}) = \sum_{j \in 3\text{NN}_i} e^{+i\mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_i)} \quad (26c)$$

179 **Spin Hamiltonian in k-space** We now replace the products of bosonic operators in Eqs. 23 by
 180 the corresponding expressions in terms of their Fourier transforms (see Eqs. 25), arriving to a
 181 Hamiltonian in k-space as shown in Eqs. 27.

$$\frac{H_D}{D} = -Ns^2 + 2s \left(\sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger A_{\mathbf{k}} + \sum_{\mathbf{k}} B_{\mathbf{k}}^\dagger B_{\mathbf{k}} \right) \quad (27a)$$

$$\frac{H_{1\text{NN}}}{J_1} = +Nz_1s^2 - s \left(\sum_{\mathbf{k}} A_{\mathbf{k}} B_{-\mathbf{k}} \gamma_1(\mathbf{k}) + \sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger B_{-\mathbf{k}}^\dagger \gamma_1^*(\mathbf{k}) \right) \quad (27b)$$

$$\frac{H_{1\text{NN}}^\lambda}{\lambda_1 + J_1} = +Nz_1s^2 - z_1s \left(\sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger A_{\mathbf{k}} + \sum_{\mathbf{k}} B_{\mathbf{k}}^\dagger B_{\mathbf{k}} \right) \quad (27c)$$

$$\frac{H_{2NN}}{J_2} = -Nz_2s^2 - 2s \sum_{\mathbf{k}} \left(A_{\mathbf{k}}^\dagger A_{\mathbf{k}} + B_{\mathbf{k}} B_{\mathbf{k}}^\dagger \right) \gamma_2(\mathbf{k}) \quad (27d)$$

$$\frac{H_{2NN}^\lambda}{\lambda_2 + J_2} = -Nz_2s^2 + 2z_2s \left(\sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger A_{\mathbf{k}} + \sum_{\mathbf{k}} B_{\mathbf{k}}^\dagger B_{\mathbf{k}} \right) \quad (27e)$$

$$\frac{H_{3NN}}{J_3} = +Nz_3s^2 - s \left(\sum_{\mathbf{k}} A_{\mathbf{k}} B_{-\mathbf{k}} \gamma_3(\mathbf{k}) + \sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger B_{-\mathbf{k}}^\dagger \gamma_3^*(\mathbf{k}) \right) \quad (27f)$$

$$\frac{H_{3NN}^\lambda}{\lambda_3 + J_3} = +Nz_3s^2 - z_3s \left(\sum_{\mathbf{k}} A_{\mathbf{k}}^\dagger A_{\mathbf{k}} + \sum_{\mathbf{k}} B_{\mathbf{k}}^\dagger B_{\mathbf{k}} \right) \quad (27g)$$

182

183 It can be proven that $\gamma_2(\mathbf{k})$ is a real function due to the symmetric array of the 2NNs around a
 184 given site i . This results in the vectors $(\mathbf{r}_j - \mathbf{r}_i)$ being multiplicative inverses of one another. Then,
 185 as the imaginary part of $\gamma_2(\mathbf{k})$ is an odd function of $(\mathbf{r}_j - \mathbf{r}_i)$, it vanishes for the 2NNs. This is not
 186 the case for 1NN and 3NNs, so the complex conjugates $\gamma_1^*(\mathbf{k})$ and $\gamma_3^*(\mathbf{k})$ are also present in Eqs.
 187 27.

188 Finally, the Hamiltonian in Eq. 27 can be expressed as the vector-matrix multiplication
 189 shown in Eq. 28, where all the terms that do not depend on $\mathbf{k}$ are confined to H_0 . In addition,

190 the various magnetic constants have been grouped in $\mathcal{C}_{\text{mag}} = -2D + z_1 J_1 + z_1 \lambda_1 - 2z_2 J_2 - 2z_2 \lambda_2 +$
 191 $z_3 J_3 + z_3 \lambda_3$. In this step, we also relabelled $\sum_{\mathbf{k}} B_{\mathbf{k}} B_{\mathbf{k}}^{\dagger} \rightarrow \sum_{\mathbf{k}} B_{-\mathbf{k}} B_{-\mathbf{k}}^{\dagger}$ and made use of the commu-
 192 tation relation $B_{\mathbf{k}}^{\dagger} B_{\mathbf{k}} = B_{\mathbf{k}} B_{\mathbf{k}}^{\dagger} - 1$.

$$\mathcal{H} = H_0 - s \sum_{\mathbf{k}} \begin{pmatrix} A_{\mathbf{k}}^{\dagger} & B_{-\mathbf{k}} \end{pmatrix} \begin{pmatrix} \mathcal{C}_{\text{mag}} + 2J_2 \gamma_2(\mathbf{k}) & J_1 \gamma_1^*(\mathbf{k}) + J_3 \gamma_3^*(\mathbf{k}) \\ J_1 \gamma_1(\mathbf{k}) + J_3 \gamma_3(\mathbf{k}) & \mathcal{C}_{\text{mag}} + 2J_2 \gamma_2(\mathbf{k}) \end{pmatrix} \begin{pmatrix} A_{\mathbf{k}} \\ B_{-\mathbf{k}}^{\dagger} \end{pmatrix} - \mathcal{C}_{\text{mag}} \quad (28)$$

193

194 From this matrix form, the eigenvalues can be straightforwardly obtained as

$$\varepsilon_{\pm}(\mathbf{k}) = -s 2J_2 \gamma_2(\mathbf{k}) \pm s |J_1 \gamma_1(\mathbf{k}) + J_3 \gamma_3(\mathbf{k})| \quad (29)$$

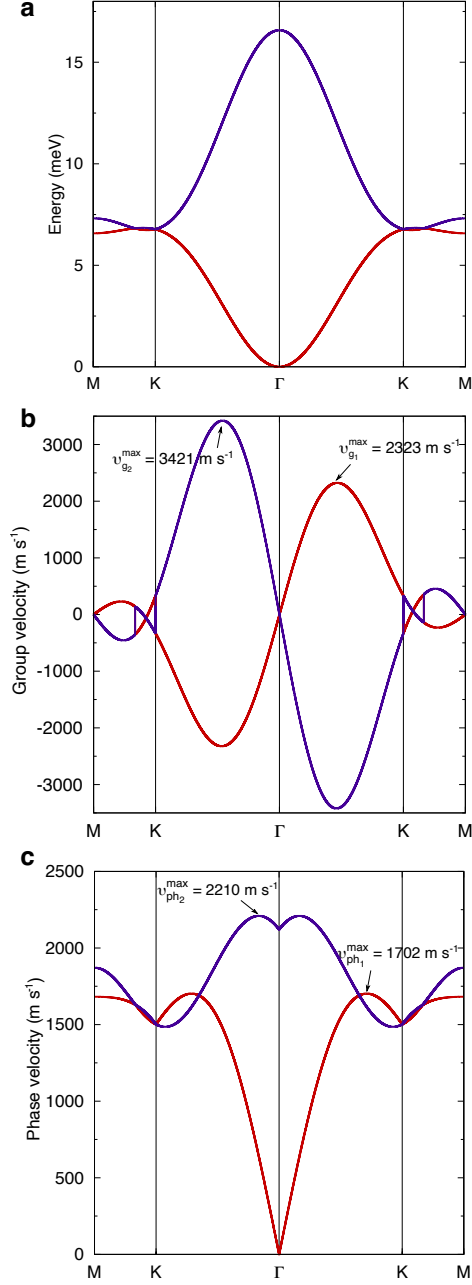


Figure S13: **a**, Magnon spectra from Supplementary Eq.29 using parameters from Supplementary Table S1 calculated over $M-K-\Gamma-K-M$. Branches are coloured differently for highlighting features on the velocities. **b-c**, Group and phase velocities, respectively extracted from the magnon dispersion in **a**. Group velocities are extracted as the first-derivative of Supplementary Eq. 29 ($v_g = \frac{\partial \epsilon_{\pm}(\mathbf{k})}{\partial \mathbf{k}}$) and the phase velocity as the ratio of magnon dispersion over the k -points ($v_{ph} = \frac{\epsilon_{\pm}(\mathbf{k})}{|\mathbf{k}|}$). The maximum values of v_g and v_{ph} at different branches are shown.

195 13 Spin-flop analytic model for monolayer MnPS₃

The domain wall dynamics induced by magnetic fields in MnPS₃ is edge-mediated as mentioned in the main text. The variation of the wall velocity v with the field B follows a linear behaviour up to 20 T (Fig. 3i) which can be fitted by Eq. 7 (linear regression coefficient $R^2 = 0.9996$). We observed however that for $B > 20$ T there is a gradual decrease of v with the spins showing some initial features of a spin-flop (SF) transition for fields above $B_{\text{SF}} = 22$ T. In this case, B_{SF} determines the highest field that can be applied to MnPS₃ without the domain wall being completely disrupted but with more curvature appreciated during the motion (Supplementary Movie 12). For $B > B_{\text{SF}}$, the domain wall still propagates throughout the layer but it systematically changes its width, curvature and orientation of the spins as the layers enters in a SF state. Supplementary Movie 13 shows the domain-wall dynamics at $B=25$ T where several of these features can be appreciated. To further understand the effect of high-fields on the antiferromagnetic (AFM) coupling of MnPS₃ we have developed an analytical model taking into account the main interactions present into the system. We start with the following XXZ spin Hamiltonian:

$$\hat{\mathcal{H}} = -D \sum_i (S_i^z)^2 - \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_{\langle i,j \rangle} \lambda_{ij} S_i^z S_j^z - \sum_i \mu_s \mathbf{S}_i \cdot \mathbf{B}_{\text{app}} \quad (30)$$

196 For this analysis, we will consider SF states as nearly in-plane AFM orientation, where spins
 197 in both sub-lattices are slightly canted towards the same out-of-plane direction ($+z$ or $-z$). We
 198 define this SF canting angle α as the angle formed between the spins and the monolayer plane in
 199 a spin-flop state (see Supplementary Figure S14). In this model, a state with $\alpha = 0$ would be an
 200 in-plane AFM state, while deviations from non-zero values of α represent SF states. In the limit,

201 $\alpha = \pm\pi$ leads to an out-of-plane FM state. Hence, this representation would be valid in the range
 202 $-\pi < \alpha < \pi$, although we will be mainly concerned with states around $\alpha = 0$.

We can now express the spin Hamiltonian (Supplementary Eq. 30) as a function of α , where the external magnetic field $\mathbf{B}_{\text{app}}$ is applied along $+z$. Assuming that α is the same for every spin in the lattice we obtain:

$$\begin{aligned} \hat{\mathcal{H}} = & -D \sum_i \sin^2 \alpha - (z_1 J_1 + z_3 J_3) \sum_i \cos(\pi - 2\alpha) - z_2 J_2 \sum_i \cos(0) \\ & - (z_1 \lambda_1 + z_2 \lambda_2 + z_3 \lambda_3) \sum_i \sin^2 \alpha - \sum_i \mu_s B \cos(\pi - \alpha) \end{aligned} \quad (31)$$

203 Supplementary Eq. 31 takes into account the fact that, given a site i , its first (1NNs) and third
 204 (3NNs) nearest neighbours will belong to the opposite sublattice while the second nearest neigh-
 205 bours (2NNs) will be in the same sub-lattice as i . We can now evaluate the energy of different SP
 206 configurations by varying α around zero. Furthermore, we can do so parametrically on the external
 207 magnetic field. Ultimately, what we want to address is whether any SP configuration is stable, i.e.,
 208 has a lower energy than the out-of-plane AFM configuration (AFM_z), which is the ground state of
 209 the system. Hence, we choose AFM_z as the reference energy and plot the $E(\alpha, \mathbf{B}_{\text{app}})$ curves rel-
 210 ative to it as displayed in Supplementary Figure S14. The curve at $\mathbf{B}_{\text{app}} = 0$ T shows a minimum
 211 at $\alpha = 0$, which is the in-plane AFM state (AFM_x). Indeed, AFM_x is higher in energy than the
 212 ground state configuration AFM_z. As $\mathbf{B}_{\text{app}} > 0$, there is a two-fold implication: First, the minimum
 213 of the curves shifts towards a positive value of α , revealing a certain degree of alignment with the
 214 external magnetic field. It is worth mentioning that the minimum would move towards negative
 215 values of α if the magnetic field was applied in $-z$. Second, the energy of that minimum lowers

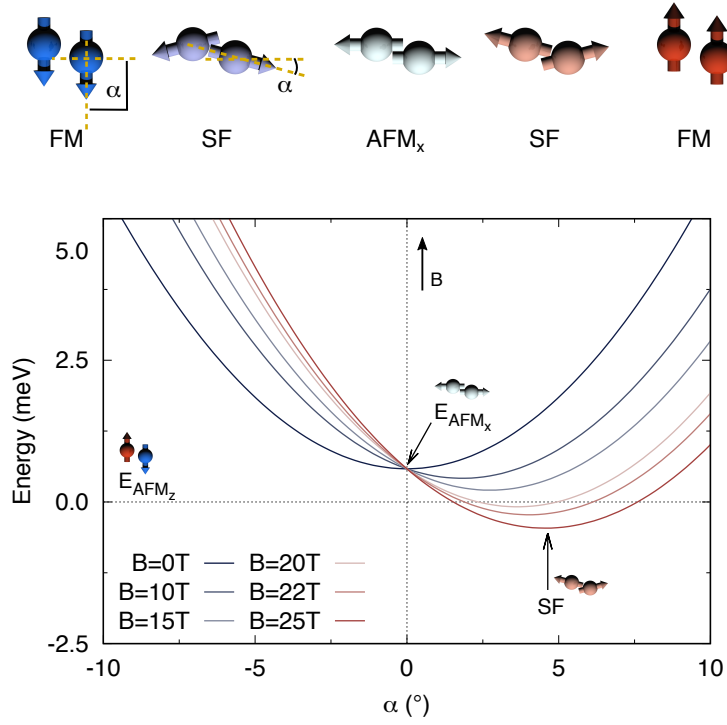


Figure S14: Energy relative to the out-of-plane AFM ground state (AFM_z) as a function of the spin-flop (SP) angle α , at different values of $B(\text{T})$.

as $\mathbf{B}_{\text{app}}$ increases. Admittedly, there will be an external magnetic field strong enough to lower the minimum energy of the curve below AFM_z . At that point, we expect a SF transition to occur. It is noteworthy that the energy of AFM_z does not depend on $\mathbf{B}_{\text{app}}$, since every term of the form $\mathbf{S}_i \cdot \mathbf{B}_{\text{app}}$ will be cancelled out by the opposite sub-lattice.

The curve at $\mathbf{B}_{\text{app}} = 20 \text{ T}$ is ever so slightly lower than the ground state, thus predicting that SF transitions should be observed above 20 T. Our Monte-Carlo simulations show an incipient SF state that competes with the ground state but is at last overtaken by the latter, probably due to border effects. Nevertheless, simulations above 22 T show stable SF states (Supplementary Movie

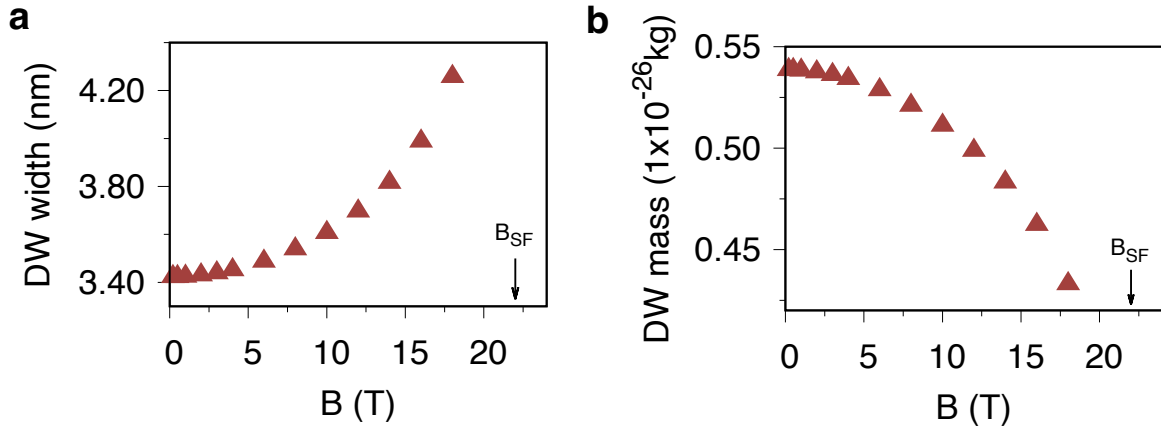


Figure S15: **a, b**, Variation of the domain wall (DW) width and DW wall mass, respectively, with the applied magnetic field. A spin-flop (SF) state is observed at fields above 22 T. This determines the critical field B_{SF} that can still be applied into the system with minor interferences on the DW motion. See Supplementary Movie S12.

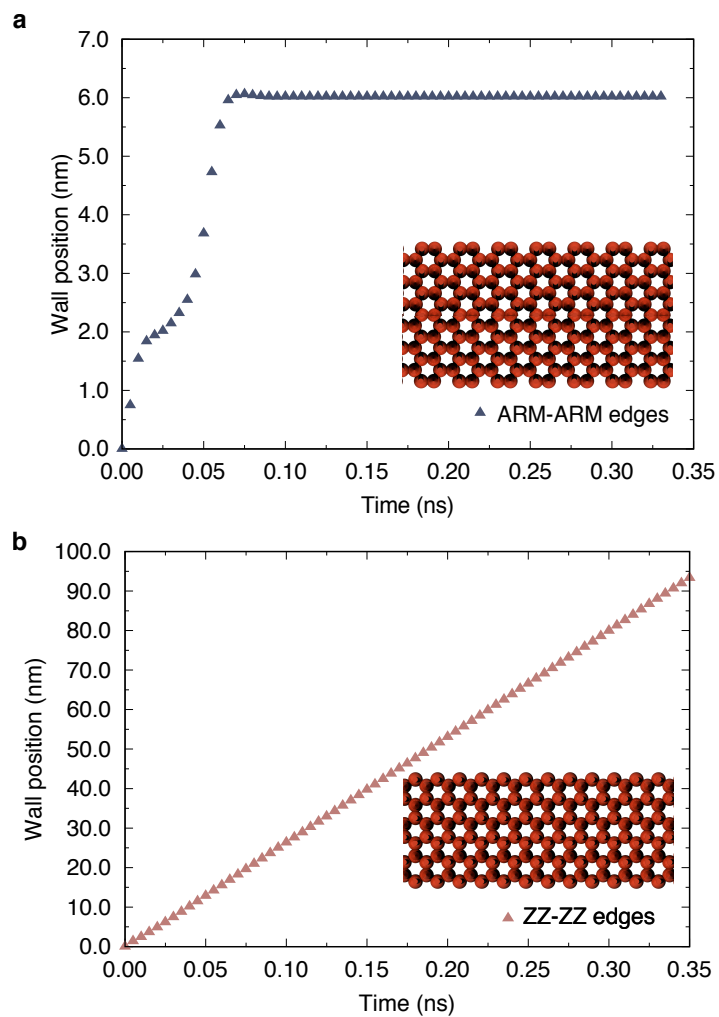


Figure S16: **a, b**, Variation of the domain wall position (nm) as a function of time (ns) induced by a current density of 4×10^9 A/cm² for a monolayer MnPS₃ with armchair (ARM) and zig-zag (ZZ) edges, respectively. The insets show the respective edges away from the domain wall at both sides of the flake, e.g. ARM-ARM and ZZ-ZZ.

15 Supplementary Movies

- Supplementary Movie 1: Spin dynamics showing the cooling down process at zero magnetic field from 80 K down to 0 K for monolayer MnPS₃.
- Supplementary Movie 2: Similar as Movie S1, but at a 0.20 T applied magnetic field.
- Supplementary Movie 3: Close look of the effect of nodes at domain walls.
- Supplementary Movie 4: Domain wall dynamics at a current density of $8 \times 10^9 \text{ A cm}^{-2}$.
- Supplementary Movie 5: Emission of spin-waves during domain wall motion at a current density of $5 \times 10^{10} \text{ A/cm}^2$. Turbulent states are observed after the propagation of the wall.
- Supplementary Movie 6: Close look of the formation of different spin-textures (e.g vortex, antivortex) at $5 \times 10^{10} \text{ A/cm}^2$.
- Supplementary Movie 7: Field-induced domain wall motion at 2.0 T for a flake with zig-zag and dangling-bond edges.
- Supplementary Movie 8: Similar as Supplementary Movie 7, but with a close look of the domain wall motion at 2.0 T showing the formation of a curvature on the domain wall profile induced by the edges.
- Supplementary Movie 9: Edge-induced restrictions of the domain wall under current density of $4 \times 10^9 \text{ A cm}^{-2}$.

- Supplementary Movie 10: Similar as Supplementary Movie 9, but using a side-view perspective.
- Supplementary Movie 11: Edges terminating the layer with atoms at different sublattices. No motion is observed despite of the intensity of the current utilised.
- Supplementary Movie 12: Field driven domain wall motion at B=22 T.
- Supplementary Movie 13: Field driven domain wall motion at B=25 T. The bright colours observed throughout the layer at longer times in the dynamics show the spins in a spin-flop state, where the spins tend to be slightly orientated in-plane with a relative angle α between them (Supplementary Figure S14).

All movies can be found at the following repository link.

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