

# Magnetic order in 2D antiferromagnets revealed by spontaneous anisotropic magnetostriction

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The temperature dependent order parameter provides important information on the nature of magnetism. Using traditional methods to study this parameter in two-dimensional (2D) magnets remains difficult, however, particularly for insulating antiferromagnetic (AF) compounds. Here, we show that its temperature dependence in AF MPS<sub>3</sub> (M(II) = Fe, Co, Ni) can be probed via the anisotropy in the resonance frequency of rectangular membranes, mediated by a combination of anisotropic magnetostriction and spontaneous staggered magnetization. Density functional calculations followed by a derived orbital-resolved magnetic exchange analysis confirm and unravel the microscopic origin of this magnetization inducing anisotropic strain. We further show that the temperature and thickness dependent order parameter allows to deduce the material's critical exponents characterising magnetic order. Nanomechanical sensing of magnetic order thus provides a future platform to investigate 2D magnetism down to the single-layer limit.

Layered two-dimensional (2D) magnetic materials offer an emerging platform for fundamental studies of magnetism in the 2D limit. Their stackability into van der Waals heterostructures opens pathways to non-trivial magnetic phases and technological applications, including sensors, memories and spintronic logic devices [1]. In addition to ferromagnetism, first observed in CrI<sub>3</sub> [2] and Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub> [3], antiferromagnetism in 2D materials has also been studied in FePS<sub>3</sub> [4] and CrSBr [5]. Antiferromagnetic (AF) materials are of particular technological interest due to their high spin-wave propagation speed and lack of macroscopic stray fields, making them strong candidates for spintronic and magnonic applications [6–10].

For insulating, thin AF materials, such as MPS<sub>3</sub> (M(II) = Fe, Co, Ni), few methods are available to study their intrinsic magnetism. Conventional techniques, such as neutron scattering, magnetization measurement by a superconducting quantum interference device (SQUID) or vibrating sample magnetometry are challenging, due to the small volumes of exfoliated 2D materials. Other methods, suited to 2D materials, require electrical conductance, the presence of specific optical modes or ferromagnetic order; they are therefore difficult to apply [1]. In contrast, strain applied to 2D magnetic materials was shown to be extremely powerful, inducing magnetization reversal [11], reorientating the easy-axis [12], or reversing the exchange interaction [13]. In addition, the direct coupling between strain, resonance frequency and magnetization in membranes of 2D magnets, makes nanomechanical resonance a sensitive method for studying their

phase transitions [14–16]. Here, we show, guided by density functional theory (DFT), that the magnetic order parameter of MPS<sub>3</sub> AF membranes can be quantified through the anisotropy in their magneto-elastic response; from its temperature dependence the critical exponents are determined, and their thickness dependence is investigated.

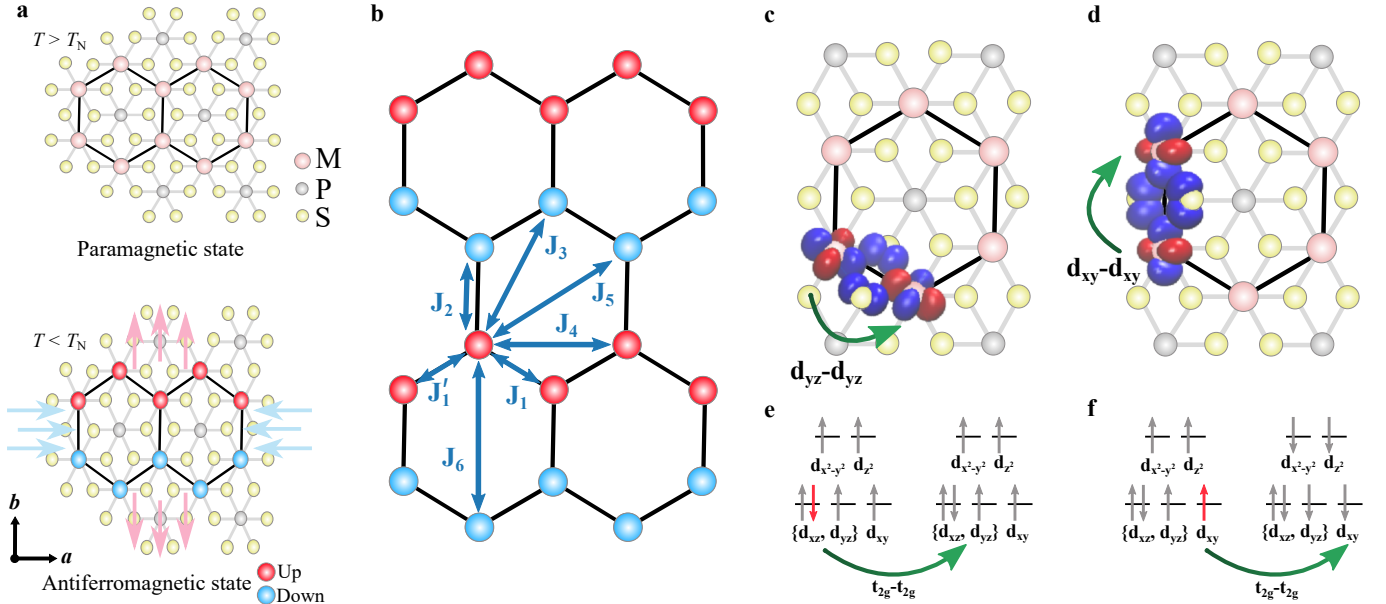
## RESULTS AND DISCUSSION

### First principles analysis of spontaneous magnetostriction in MPS<sub>3</sub>

Transition-metal phosphorus trisulphides, with general formula MPS<sub>3</sub>, are layered materials stacked in a monoclinic lattice with symmetry group C2/m [17], as shown in the top view of a single-layer in the paramagnetic phase, Fig. 1a, top panel. The spins of FePS<sub>3</sub> point out-of-plane, whereas both CoPS<sub>3</sub> and NiPS<sub>3</sub> are in-plane systems with their spins preferentially aligned along the *a* axis. The intralayer AF order forms a zigzag configuration, as shown in bottom panel of Fig. 1a, leading to two opposite aligned magnetic sub-lattices. The difference of the magnetisation between these sub-lattices is the Néel vector. In bulk CoPS<sub>3</sub> and NiPS<sub>3</sub>, these layers with this staggered magnetism are stacked in a ferromagnetic (FM) fashion with Néel transition temperatures, *T<sub>N</sub>*, around 119 and 155 K, respectively [18, 19]. The interlayer magnetic interactions in FePS<sub>3</sub> are AF with a transition around 118 K [20].

To analyse the effect of magnetic ordering on the lattice, we performed first principles structural optimiza-

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SUPPLEMENTARY FIG. 1. **Magnetostriction in  $\text{MPS}_3$  membranes.** **a**, **top panel**, Crystalline structure of  $\text{MPS}_3$  in the paramagnetic phase ( $T > T_N$ ). Black hexagons indicate the organisation of magnetic atoms in the lattice. **a**, **bottom panel**, Crystalline structure of  $\text{MPS}_3$  at the AF phase ( $T < T_N$ ) as it elongates in the  $b$  and contracts in the  $a$  direction. Light blue and red arrows indicate the axial lattice distortion. **b**, Illustration of the exchange interaction parameters included into the Heisenberg spin Hamiltonian. **c-d**, Calculated maximally localized Wannier orbitals. Green arrows illustrate the most relevant FM superexchange channels for  $J_1$  ( $J'_1$ ) (**c**) and  $J_2$  (**d**), corresponding with the  $d_{yz}$ - $d_{yz}$  ( $d_{xz}$ - $d_{xz}$ ) and  $d_{xy}$ - $d_{xy}$  orbitals, respectively. **e-f**, Electron configuration of the  $\text{Fe}^{2+}$  magnetic ions connected by  $J_1$  (**e**) and  $J_2$  (**f**), showing parallel and antiparallel spin orientations, respectively.

tions of  $\text{FePS}_3$ ,  $\text{CoPS}_3$  and  $\text{NiPS}_3$  based on density functional theory (DFT). For the ground state zigzag magnetic configuration, the calculations predict a compression of the  $a$  lattice parameter with respect to the crystallographic, non-magnetic structure of 2.545% and 1.328% for the Co and Fe derivatives respectively (see Table 1). In addition, the  $b$  axis expands by 0.402% (Co) and 0.359% (Fe). In contrast, in  $\text{NiPS}_3$  the lattice parameters remain almost unchanged. The crystal and magnetic structures are strongly connected in these compounds, which is further corroborated by simulations of different spin configurations (see Supplementary Note 1).

The microscopic mechanism governing the spontaneous magnetostriction in these materials is studied using orbital-resolved magnetic exchange analyses based on maximally localized Wannier functions, (see Supplementary Note 1). The analysis shows that the spontaneous magnetostriction calculated in  $\text{FePS}_3$  and  $\text{CoPS}_3$  arises from isotropic magnetic exchange interactions between  $t_{2g}$ - $t_{2g}$  orbitals. Specifically, for  $\text{FePS}_3$  the main magnetic exchange channels, substantially affected by the compression of the  $a$  and expansion of the  $b$  lattice parameters, are the ones involving  $t_{2g}$ - $t_{2g}$  interactions of FM nature. The changes in the lattice parameters result in an increase in  $J_1$  and  $J'_1$  due to a decrease in distance between the  $d_{yz}$ - $d_{yz}$  and  $d_{xz}$ - $d_{xz}$  orbitals, respectively (Fig. 1c). Simultaneously, these changes cause a decrease of  $J_2$  due to a larger separation of the  $d_{xy}$ - $d_{xy}$  orbitals

(Fig. 1d). This is compatible with the electron configuration of  $\text{Fe}^{2+}$  ( $d^6$ ), which has these orbitals partially filled and allows FM hopping between them (Fig. 1e,f).

This hopping effect also occurs for  $\text{Co}^{2+}$  ( $d^7$ ) although the additional electron present for Co blocks the  $d_{xy}$ - $d_{xy}$  pathway (Supplementary Note 1, Fig. S2). This results in a stronger effect along  $J_1$  and  $J'_1$  for the optimized structure, maximizing FM interactions in the zigzag chain, which involve the  $d_{yz}$ - $d_{yz}$  and  $d_{xz}$ - $d_{xz}$  orbitals, respectively. For the  $\text{Ni}^{2+}$  derivative ( $d^8$ ), the  $t_{2g}$  energy levels are fully occupied (Supplementary Note 1, Fig. S3), which results in a blocking of the  $t_{2g}$ - $t_{2g}$  magnetic super-exchange channels. This leads to an almost negligible modification in the lattice parameters of the optimized structure with respect to the crystallographic non-magnetic one.

### Resonance frequency changes due to spontaneous magnetostrictive strain

The predicted anisotropic change of lattice parameters when going from the paramagnetic to the AF phase, causes compressive stress,  $\sigma_a$ , and tensile stress,  $\sigma_b$ , along the  $a$  axis and  $b$  axis respectively, as illustrated in Fig. 1a, bottom panel. To quantify this anisotropy appearing at the phase transition, we use rectangular membranes, shown in Fig. 2b, to nanomechanically probe stress varia-

SUPPLEMENTARY TABLE 1. CoPS<sub>3</sub>, FePS<sub>3</sub> and NiPS<sub>3</sub> lattice parameters of the crystallographic non-magnetic (NM) and fully optimized zigzag antiferromagnetic (AF-zigzag) configurations, as calculated by DFT (see Supplementary Note 1).

	CoPS <sub>3</sub>		FePS <sub>3</sub>		NiPS <sub>3</sub>	
Lattice parameter (Å)	<i>a</i>	<i>b</i>	<i>a</i>	<i>b</i>	<i>a</i>	<i>b</i>
NM	5.895	10.19	5.947	10.301	5.812	10.07
AF-zigzag	5.745	10.231	5.868	10.338	5.817	10.061
Change (%)	-2.545	+0.402	-1.328	+0.359	+0.086	-0.089

tions, along a specific crystallographic axis [21] (see Supplementary Note 2). In the following analysis, we neglect the stress contribution from the thermal expansion of the substrate, as this is small compared to that of the MPS<sub>3</sub> compounds [14].

The resonance frequency of the fundamental mode of a rectangular membrane,  $f_{\text{res}}$ , is approximately given by [22]:

$$f_{\text{res}} \approx \frac{1}{2} \sqrt{\frac{1}{\rho} \left[ \frac{1}{w^2} \sigma_w + \frac{1}{l^2} \sigma_l \right]}, \quad (1)$$

where  $\rho$  is the mass density,  $w$  and  $l$  are respectively the width and length of the membrane, as indicated in Fig. 2b, and  $\sigma_w, l$  are the stresses parallel to these directions. For high-aspect-ratio membranes ( $w \ll l$ ), the mechanical resonance frequency is mostly determined by the stress along the shortest direction,  $\sigma_w$ .

We study the resonance frequency of thin MPS<sub>3</sub> flakes suspended over star-shaped cavities with 30° angular resolution, as shown in an example device in Fig. 2b. When the longest side of the cavity is aligned along a crystallographic axis ( $a$  or  $b$ ) and  $w \ll l$ , its fundamental resonance frequency ( $f_a$  or  $f_b$ ) is determined by the stress along the perpendicular axis ( $\sigma_b$  or  $\sigma_a$ ):

$$f_a \approx \frac{1}{2} \sqrt{\frac{1}{\rho w^2} \sigma_b} \text{ and } f_b \approx \frac{1}{2} \sqrt{\frac{1}{\rho w^2} \sigma_a}. \quad (2)$$

On cavities oriented at an intermediate angle,  $\theta$ , (defined with respect to the  $b$  axis), the resonance frequency is:

$$f_\theta(T) \approx \frac{1}{2} \sqrt{\frac{1}{\rho w^2} [\sigma_{a,\theta} + \sigma_{b,\theta}]}, \quad (3)$$

$$\sigma_{a,\theta} = \frac{E}{(1 - \nu^2)} (\cos^2 \theta + \nu \sin^2 \theta) (\bar{\epsilon} - \epsilon_{\text{ms},a}),$$

$$\sigma_{b,\theta} = \frac{E}{(1 - \nu^2)} (\sin^2 \theta + \nu \cos^2 \theta) (\bar{\epsilon} - \epsilon_{\text{ms},b}),$$

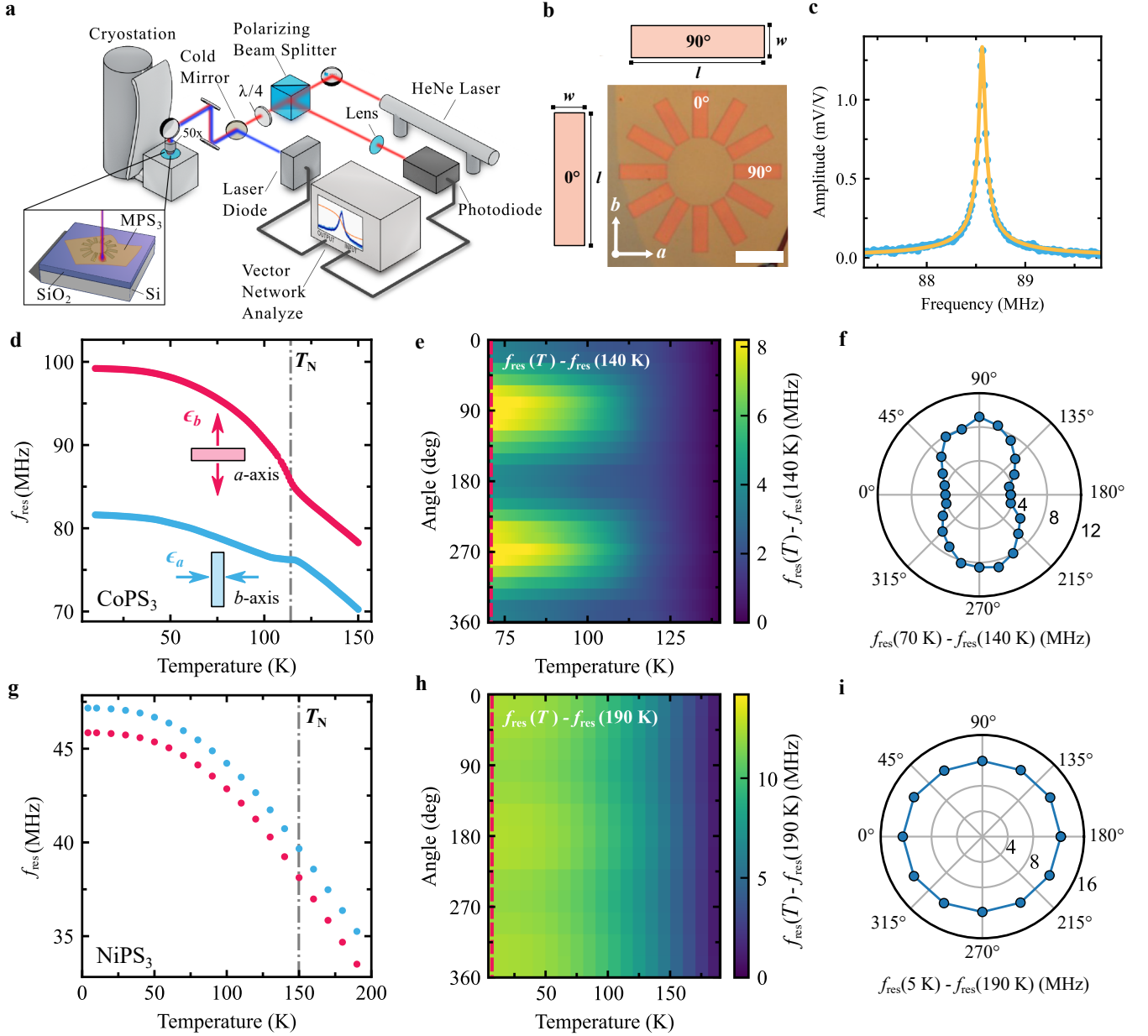
where we have used the constitutive equations for a magnetostrictive membrane with plane stress [23], while only keeping the anisotropy in the magnetostriction coefficient, Supplementary Note 2. Here,  $E$  is the Young's modulus and  $\nu$  is Poisson's ratio of the material. Moreover, we have  $\bar{\epsilon} = \epsilon_{\text{fab}} - \epsilon_{\text{th}}$ , with  $\epsilon_{\text{fab}}$  the residual fabrication strain and  $\epsilon_{\text{th}}$  the phononic thermal expansion induced strain variation. The magnetostrictive strain along

the  $a$  and  $b$ -axes is given by  $\epsilon_{\text{ms},a,b} = \lambda_{a,b} L^2$ , respectively (see Supplementary Note 3 for a detailed derivation of Eq. (3)), where  $\lambda_{a,b}$  are magnetostriction coefficients and  $L^2$  is the AF order parameter squared.

The temperature dependence of the resonance frequency comprises two contributions: one due to the phononic thermal expansion coefficient  $\alpha$ , given by  $\epsilon_{\text{th}}(T) = \int_{T_0}^T \alpha(\tilde{T}) d\tilde{T}$ , where  $T_0$  is a reference temperature and  $\tilde{T}$  the integration variable, and the magnetostrictive contribution  $\epsilon_{\text{ms},a,b}(T) = \lambda_{a,b} L^2(T)$ . The former contribution is a slowly varying function of  $T$ , while the latter term contains the staggered magnetization, which increases abruptly near the phase transition; it thus can be used to determine  $L(T)$ , as we will show below. We assume  $\lambda_{a,b}$  to be  $T$  independent, as its temperature dependence will be negligible when compared to that of  $L(T)$ .

#### Nanomechanical determination of the order parameter

To quantify the anisotropy in the magnetic membranes, a laser interferometry technique is used to measure their resonance frequency as a function of temperature [24]. A MPS<sub>3</sub> flake, suspended over holes in a patterned Si/SiO<sub>2</sub> chip, Fig. 2b, is placed inside a cryostat with optical access as shown in Fig. 2a. Both actuation and detection are done optically, by means of a power-modulated blue laser which opto-thermally excites the fundamental resonance, and a constant red laser which measures the change in the reflected signal resulting from the membrane's motion [14]. A typical resonance is shown in Fig. 2c, along with the damped harmonic oscillator model fit defining the resonance frequency. Figure 2d shows that in CoPS<sub>3</sub>  $f_a$  and  $f_b$  exhibit a similar temperature dependence for  $T > T_N$ , while opposite behaviour below the phase transition is visible, namely an increase of  $f_a$  and a relative decrease of  $f_b$ . This sudden change in  $f(T)$  for the perpendicular cavities, occurring near  $T_N$ , constitutes, in accordance with the DFT calculations, the central result of this work as it shows that the magnetic ordering in MPS<sub>3</sub> leads to anisotropic strain and thus spontaneous magnetostriction. We further note that strictly speaking,  $T_N$  should be replaced by  $T_N^*$  which includes the effects of strain (see Supplementary Note 2).



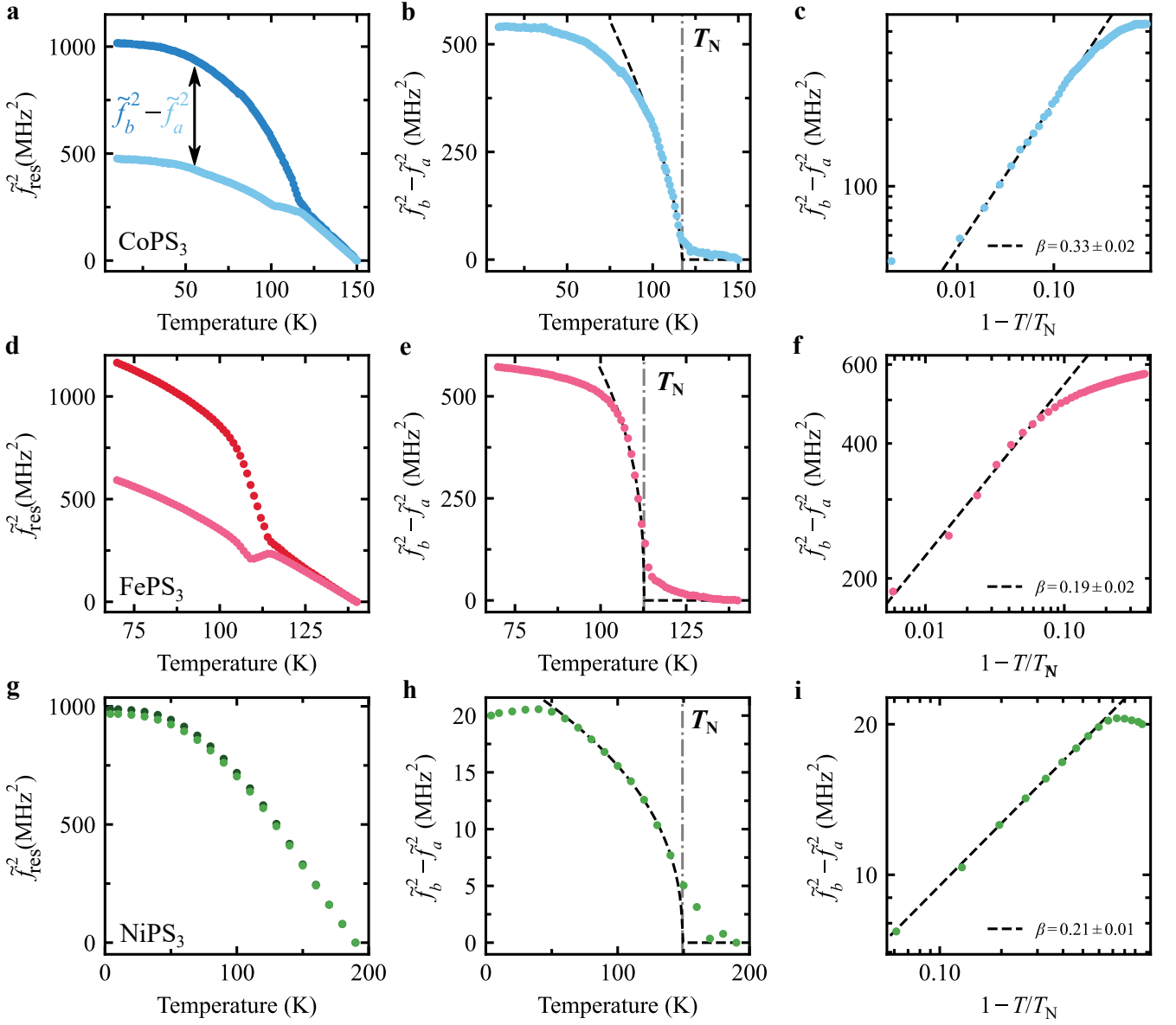
**SUPPLEMENTARY FIG. 2. Angle-resolved mechanical characterization via laser interferometry.** **a**, Schematic illustration of the laser interferometry setup and sample with rectangular cavity array. **b**, Optical image of the rectangular membranes array for a CoPS<sub>3</sub> sample. The  $a$  and  $b$  axis are determined from the resonance frequency behaviour. Scale bar: 12  $\mu\text{m}$ . Schematic of 0° and 90° membranes from the array where  $w$  is the width of the membrane and  $l$  its length. **c**, Measured amplitude of the fundamental resonance peak in a CoPS<sub>3</sub> drum at  $T = 10$  K and Lorentzian fit used to extract the fundamental resonance frequency,  $f_{\text{res}}$ , and quality factor,  $Q$ . **d**, Temperature dependence of  $f_{\text{res}}$  of a CoPS<sub>3</sub> rectangular membrane orientated along the  $a$  and  $b$  axes. The dashed line indicates the transition temperature  $T_N$  extracted from the data. **e**, Resonance frequency difference,  $f_{\text{res}}(T) - f_{\text{res}}(140 \text{ K})$ , as a function of angle and temperature. The dashed line indicates the transition as in **d**. **f**, Polar plot of  $f_{\text{res}}(T) - f_{\text{res}}(140 \text{ K})$  taken along the red dashed line in (e). Panels **g-i**, follow the same structure as (c-e) for NiPS<sub>3</sub> resonators with negligible anisotropy, measured between 5 K and 190 K.

For simplicity, we here use the notation  $T_N$  for the measured transition temperatures.

The anisotropic behavior of CoPS<sub>3</sub> in the AF state is even more evident in Fig. 2e, where  $f_{\text{res}}(T) - f_{\text{res}}(140 \text{ K})$  for the different cavities of the star-shaped sample are plotted as a function of  $\theta$  and temperature. The po-

lar plot in Fig. 2f shows the data along the red dashed line at  $T = 70 \text{ K}$  in Fig. 2e and results in a characteristic dumbbell-shape. Similar anisotropic behaviour is observed in FePS<sub>3</sub> as shown in Supplementary Note 4. On the contrary, for NiPS<sub>3</sub> negligible anisotropy is observed in the angle-resolved magnetostriction data in Fig. 2g-i.





**SUPPLEMENTARY FIG. 3. Anisotropy and critical behaviour in resonance frequency of MPS<sub>3</sub> (M(II) = Co, Fe, Ni) membranes.** **a**, Pretension corrected resonance frequency ( $\tilde{f}_a^2(T) = f_a^2(T) - f_a^2(150\text{K})$  and  $\tilde{f}_b^2(T) = f_b^2(T) - f_b^2(150\text{K})$ ) of rectangular membranes of CoPS<sub>3</sub> oriented along the  $b$  axis (blue) and  $a$  axis (red) **b**, Difference of the corrected frequency squared  $\tilde{f}_b^2 - \tilde{f}_a^2$  proportional to the order parameter  $L^2$  from Eq. 4. The dashed-dotted line indicates the measured transition temperature  $T_N$ . The dashed black line is a powerlaw fit through the data close to  $T_N$  (see Supplementary Note 6). **c**, Difference of the corrected frequency squared  $\tilde{f}_b^2 - \tilde{f}_a^2$  as a function of the reduced temperature  $1 - T/T_N$ . The dashed black line is the fit from **b** where the slope defines the critical exponent  $2\beta$ . **d-f**, and **g-i**, follow the same structure as (a-d) for FePS<sub>3</sub> and NiPS<sub>3</sub> resonators, respectively.

To obtain  $L(T)$  from the data, we first subtract the pretension contribution from the resonance frequency  $f_\theta(T_0)$  by calculating  $\tilde{f}_\theta^2(T) = f_\theta^2(T) - f_\theta^2(T_0)$ , for each angle, where  $T_0 = 150\text{ K}$  is the highest temperature in our measurements. The resulting values of  $\tilde{f}_\theta^2(T)$  along the crystalline axes  $a$  and  $b$  are shown in Fig. 3a,d,g for the three MPS<sub>3</sub> compounds. With Eq. (3), we then calculate the

difference  $\tilde{f}_b^2(T) - \tilde{f}_a^2(T)$  which yields

$$\tilde{f}_b^2 - \tilde{f}_a^2 = \frac{E}{4\rho w^2(1+\nu)} [\lambda_a - \lambda_b] L^2. \quad (4)$$

We can now use Eq. (4) to access the critical behaviour of  $L$  below  $T_N$  by plotting  $\tilde{f}_b^2 - \tilde{f}_a^2$  as a function of temperature. As shown in Fig. 3b,e,h, the trend presents the typical critical behaviour with a non-zero order pa-

parameter appearing in the ordered state for  $T < T_N$ . Figures 3c,f,i show the same critical curve as Fig. 3b,e,h respectively, plotted on a logarithmic scale against the reduced temperature  $(1 - T/T_N)$ . Note that the difference  $\tilde{f}_b^2 - \tilde{f}_a^2$  for NiPS<sub>3</sub>, is substantially smaller than that of the Fe/CoPS<sub>3</sub> membranes indicative of a weaker anisotropic magnetostrictive behaviour.

The angle dependence of the resonance frequencies allows us to estimate the ratio  $r_{ab} = \lambda_a/\lambda_b$  between the magnetostriction parameters,  $\lambda_{a,b}$ , (see Supplementary Note 3). This ratio we directly compare to DFT calculations: Experimentally, we find for FePS<sub>3</sub>,  $r_{ab} = -2.3 \pm 0.3$  while from the DFT calculations we estimate  $r_{ab} = -3.70$ . For CoPS<sub>3</sub> (taking [25]  $\nu_{\text{CoPS}_3} = 0.293$ ), the experimental value is  $-1.42 \pm 0.07$  and the DFT one  $-6.33$ . We conclude that although both the sign and order of magnitude of the magnetostrictive anisotropy in these compounds are well reproduced in the current work, more detailed studies will be needed to obtain full quantitative correspondence with theory.

### Thickness dependence of critical behaviour

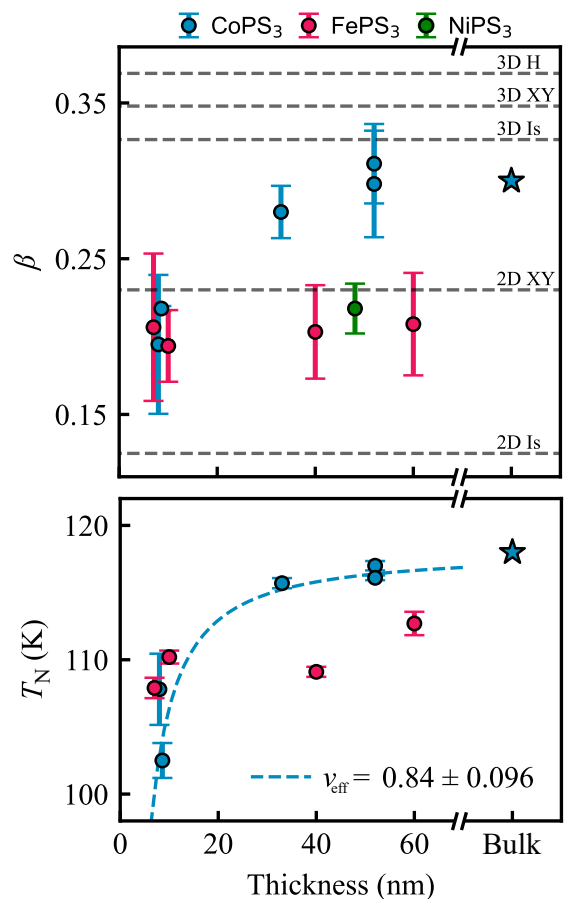
As follows from Landau's theory of phase transitions (see Supplementary Note 2),  $L(T)$  near  $T_N$  is given by

$$L^2(T) = \begin{cases} 0 & \text{if } T > T_N \\ \frac{A}{2B}(T_N - T)^{2\beta} & \text{if } T < T_N, \end{cases} \quad (5)$$

where  $A$  and  $B$  are constants and  $\beta$  is a critical exponent representative of the magnetic order. We fit Eq. (5) to the data in Fig. 3b,e,h in the region close to  $T_N$  (indicated by the black dashed line in Fig. 3b,e,h) to extract the critical exponent  $\beta$  and  $T_N$  for the three materials (see Supplementary Note 6 for more details on the fitting procedure). In the logarithmic plot of the critical curve the fitting of a straight line shows good agreement to the data points, consistent with the result of Eq. (5). The values for  $\beta$  and  $T_N$  are plotted in Fig. 4 as a function of thickness,  $t$ , and listed in Supplementary Note 6, Table 49.

For the weakly anisotropic NiPS<sub>3</sub>,  $\beta = 0.218 \pm 0.016$ , comparable to the value ( $\beta = 0.22 \pm 0.02$ ) found in Ref. [27], and consistent with the expected 2D XY magnetic dimensionality ( $\beta_{2\text{DXY}} = 0.233$ ) of NiPS<sub>3</sub> [28]. For FePS<sub>3</sub> we find  $\beta = 0.208 \pm 0.033$ , comparable with literature values [29]. For both  $\beta$  and  $T_N$  no appreciable thickness dependence is observed, similar to what has previously been reported in Ref. [30], where changes in the critical behaviour mostly become visible in the monolayer limit.

For thicker CoPS<sub>3</sub> samples ( $t = 40 - 60$  nm) we find  $\beta = 0.289 \pm 0.034$  close to what is reported in literature for the bulk ( $\beta_{\text{bulk}} = 0.3 \pm 0.01$  [18]) and consistent with the 3D Ising model. For samples with  $t < 10$  nm the measured  $\beta$ , on the other hand, is  $0.195 \pm 0.045$ , closer



**SUPPLEMENTARY FIG. 4. Thickness dependence of critical behaviour.** Average critical exponent,  $\beta$ , and critical temperature,  $T_N$ , of MPS<sub>3</sub> resonators plotted as a function of thickness. The blue stars indicate CoPS<sub>3</sub> bulk values from [26]. Critical parameters have been determined from power law fits to  $\tilde{f}_b^2 - \tilde{f}_a^2$ , as shown in Fig. 3b,e,h, and then taking the average value over the fit parameter for all angles  $\theta \neq 0$ . Error bars are calculated from standard deviation of fit results for all  $\theta$ . The horizontal gray dashed lines in the upper plot indicate the expected values of  $\beta$  for the 3D or 2D versions of the Heisenberg (H), XY or Ising (Is) models. The blue dashed line in the lower panel indicates a fit to Eq. (6) through the CoPS<sub>3</sub> data with  $\nu_{\text{eff}} = 0.84 \pm 0.13$ .

to  $\beta_{2\text{DXY}}$  as shown in the top panel of Fig. 4. This constitutes a noticeable change in  $\beta$  while going from bulk to thinner samples. Similarly, we observe for CoPS<sub>3</sub> a decrease in  $T_N$  from the bulk value of 118 K down to  $\sim 100$  K, similar to what was previously reported in Ref. [26]. We fit a power law to the dependence of  $T_N$  on thickness,

$$T_N(t)/T_N^{3\text{D}} \propto 1 - (C/t)^{1/\nu_{\text{eff}}}, \quad (6)$$

where  $C$  is a non-universal constant related to the interlayer coupling, and  $\nu_{\text{eff}}$  is an effective critical exponent related to the correlation length [31]. Fitting the CoPS<sub>3</sub> data points with  $T_N^{3\text{D}} = 118$  K [26] yields  $C = 1.43 \pm 0.457$  nm and  $\nu_{\text{eff}} = 0.84 \pm 0.096$ . This

value of  $\nu_{\text{eff}}$  is intermediate between the expected values of  $\nu_{\text{eff}} = 0.630$  for the 3D Ising and  $\nu_{\text{eff}} = 1$  for the 2D Ising models, and indicative of a transition regime [32].

## CONCLUSIONS

In conclusion, we provide a comprehensive analysis of the anisotropic magnetostriction effect in  $\text{MPS}_3$  compounds and its implications to the dynamics of membrane made from them. DFT calculations provide a microscopic explanation for the anisotropic lattice deformation in  $\text{CoPS}_3$ ,  $\text{FePS}_3$  and  $\text{NiPS}_3$  which are consistent with our measurements. We further demonstrate the relation between magnetic ordering and anisotropy in the mechanical resonance frequency of suspended  $\text{MPS}_3$  resonators, providing a direct measure of the AF order parameter in absence of an external magnetic field. We observe a thickness dependence in the critical behaviour of  $\text{CoPS}_3$  resonators [18, 26], which is absent in the case of  $\text{FePS}_3$ . The presented technique is of particular interest for the study of 2D magnetism given the scarcity of methods available to investigate critical phenomena of van der Waals materials in the atomically thin limit.

## METHODS

**Sample fabrication.** Substrates consist of thermal  $\text{SiO}_2$  of 285 nm thickness, grown on highly doped ( $\text{Si}^{++}$ ) silicon. The rectangular cavities are defined via e-beam lithography using AR-P 6200 resist. After development, the exposed  $\text{SiO}_2$  areas are fully etched via reactive ion etching. The AR-P 6200 resist is stripped in PRS-3000 and the sample is cleaned in an  $\text{O}_2$  plasma before stamping. The exfoliation and transfer of multi-layer  $\text{MPS}_3$  flakes is done using a polydimethylsiloxane (PDMS) transfer method. First,  $\text{MPS}_3$  crystals are exfoliated onto the PDMS through scotch tape. Selected flakes are then transferred on the star-shaped cavities in the  $\text{SiO}_2/\text{Si}$  substrate.

**Laser interferometry.** Samples are mounted on a heater stage which is cooled down to 4 K using a Montana Instruments Cryostation s50 cryostat with optical access. A blue diode laser ( $\lambda = 405$  nm) is used to excite the membrane optothermally via AC power-modulation from a vector network analyzer (VNA) [33]. Displacements are detected by focusing a red He-Ne laser beam ( $\lambda = 632$  nm) on the cavity formed by the membrane and Si substrate. The reflected light, which is modulated by the position-dependent membrane motion, is recorded by a photodiode and processed by a phase-sensitive VNA. Laser spot size is  $\sim 1 \mu\text{m}$ .

**DFT calculations.** First principles spin-polarized DFT calculations in the plane wave formalism are performed as implemented in the Quantum ESPRESSO

package [34]. The exchange-correlation energy is calculated using the generalized gradient approximation using the Perdew–Burke–Ernzerhof functional [35] and standard Ultra-soft (USPP) solid-state pseudopotentials. The electronic wave functions are expanded with well-converged kinetic energy cut-offs for the wave functions (charge density) of 75 (800), 85 (800) and 85 (800) Ry for Fe, Co and Ni, respectively. The crystal structures are fully optimized using the Broyden–Fletcher–Goldfarb–Shanno (BFGS) algorithm [36] until the forces on each atom are smaller than  $1 \times 10^{-3}$  Ry/au and the energy difference between two consecutive relaxation steps is less than  $1 \times 10^{-4}$  Ry. In order to avoid unphysical interactions between images along the non-periodic direction, we add a vacuum of 18 Å in the  $z$  direction for the monolayer calculations. The Brillouin zone is sampled by a fine  $\Gamma$ -centered  $5 \times 5 \times 1$   $k$ -point Monkhorst–Pack [37]. A tight-binding Hamiltonian derived from first-principles is constructed in the base of Maximally-localized Wannier functions, as implemented in the Wannier90 code [38]. For that, we select the  $d$  orbitals of the metal centre (Fe, Co, Ni) and the  $s$  and  $p$  orbitals of P and S to construct the connected subspace. Magnetic interactions are determined using the Green’s function method in the TB2J software [39]. The orbital resolved analysis is performed after rotating the coordinate system of the crystal to align the metal-sulfur bonds direction of the octahedra with the cartesian axes.

**Crystal growth** Crystal growth of  $\text{MPS}_3$  ( $\text{M(II)} = \text{Ni, Fe, Co}$ ) is performed following a solid-state reaction inside a sealed evacuated quartz tube (pressure  $\sim 5 \times 10^{-5}$  mbar).  $\text{I}_2$  was used as a transport agent to obtain large crystals. A three-zone furnace is used, where a tube with the material was placed in the leftmost zone. This side is then heated up to 700 °C in 3 hours so that a temperature gradient of 700/650/675 °C is established. The other two zones are heated up in 24 hours from room temperature to 650 °C and kept at that temperature for one day. The temperature is kept constant for 28 days and cooled down naturally. With this process crystals with a length up to several centimeters are obtained. Detailed description of the crystal growth and characterization can be found in earlier work [14].

## Data availability

All data supporting the findings of this article and its Supplementary Information will be made available upon request to the authors.

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## Author contributions

D.L.E., A.M.R. performed the DFT and orbital resolved tight-binding calculations, supervised by J.J.B. M.Š., M.J.A.H. and G.B. performed the laser interferometry measurements and fabricated and inspected the samples. M.L. and M.J.A.H. fabricated the substrates. S.M.-V., C.B.-C. and E.C. synthesized and characterized the FePS<sub>3</sub>, CoPS<sub>3</sub> and NiPS<sub>3</sub> crystals, supervised by E.C. M.Š., M.J.A.H. and G.B. analysed the experimental data. M.Š., M.J.A.H., Y.M.B., and P.G.S. modeled the experimental data. H.S.J.v.d.Z. and P.G.S. supervised the project. The paper was jointly written by all authors with a main contribution from M.J.A.H. All authors discussed the results and commented on the paper.

## Competing interests

The authors declare no competing interests.

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# **SUPPLEMENTARY INFORMATION: Magnetic order in 2D antiferromagnets revealed by spontaneous anisotropic magnetostriction**

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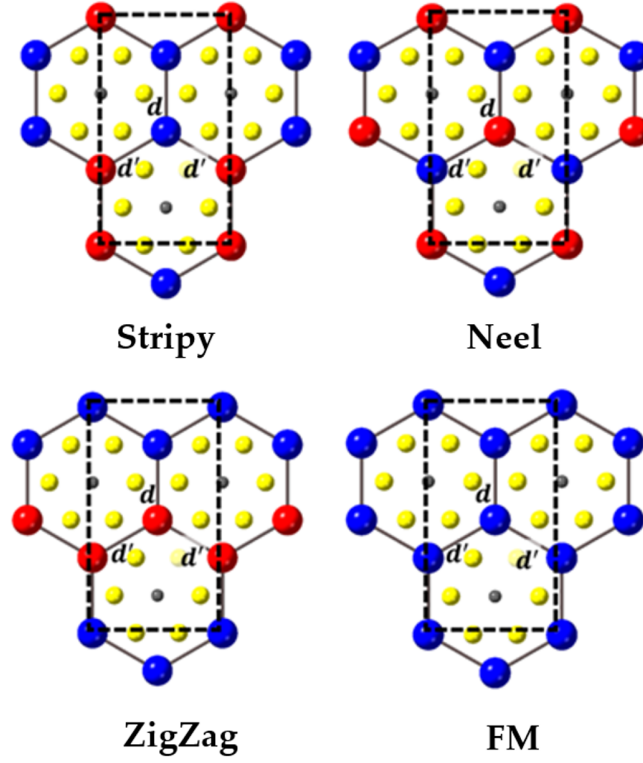
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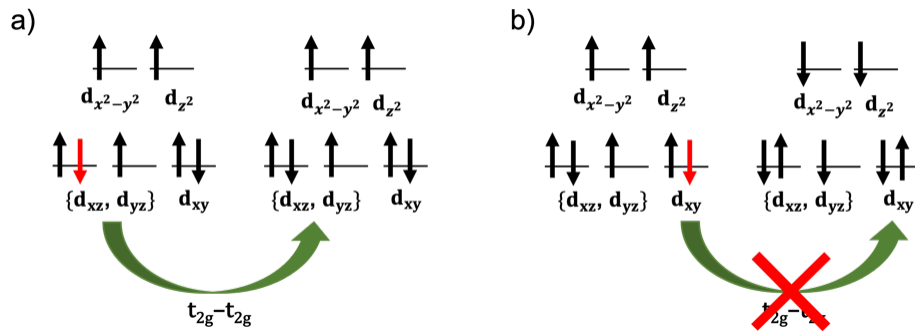


# Supplementary Note 1. DENSITY FUNCTIONAL THEORY CALCULATIONS

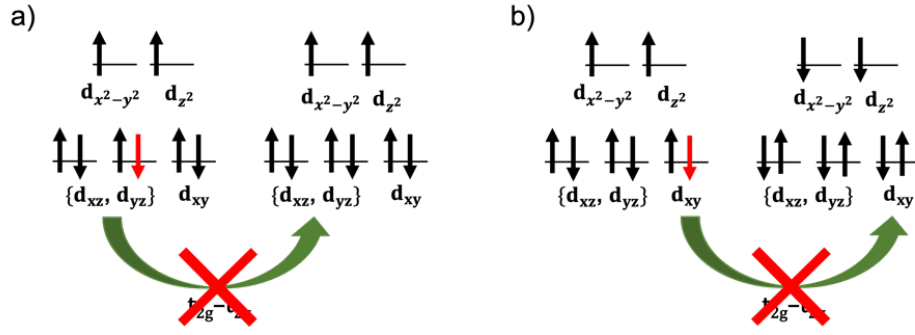
This section summarizes the results of orbital-resolved magnetic exchange analyses based on maximally localized Wannier functions, for both the crystallographic and optimized structures. These results are included in Supplementary Tables 7 – 42. Supplementary Tables 43 – 48 display the hopping integrals with the S atoms (Supplementary Fig. 8) that mediate the super-exchange interactions between magnetic centers for each channel.



SUPPLEMENTARY FIG. 5. Top view of a single-layer of  $\text{MPS}_3$  for different magnetic configurations, namely stripy, Néel, zigzag and ferromagnetic (FM). Blue and red balls represent transition metal atoms with spin up and down components, respectively.



SUPPLEMENTARY FIG. 6. Electron configuration of the  $\text{Co}^{2+}$  magnetic ions connected by  $J_1$  (a) and  $J_2$  (b), showing parallel and antiparallel spin orientations, respectively. Green arrows illustrate the most relevant ferromagnetic superexchange channels, namely  $d_{yz}-d_{yz}$  and  $d_{xy}-d_{xy}$  (cancelled), for  $J_1$  and  $J_2$  respectively



SUPPLEMENTARY FIG. 7. Electron configuration of the  $\text{Ni}^{2+}$  magnetic ions connected by  $J_1$  (a) and  $J_2$  (b), showing parallel and antiparallel spin orientations, respectively. Green arrows illustrate the most relevant ferromagnetic superexchange channels, namely  $d_{yz}$ - $d_{yz}$  (cancelled) and  $d_{xy}$ - $d_{xy}$  (cancelled), for  $J_1$  and  $J_2$  respectively.

SUPPLEMENTARY TABLE 2.  $\text{CoPS}_3$ ,  $\text{FePS}_3$  and  $\text{NiPS}_3$  lattice parameters of the crystallographic non-magnetic (NM) and Stripy antiferromagnetic (AF) configurations

Lattice parameter ( $\text{\AA}$ )	$\text{CoPS}_3$		$\text{FePS}_3$		$\text{NiPS}_3$	
	a	b	a	b	a	b
Crystallographic NM	5.895	10.190	5.947	10.301	5.812	10.070
AF-stripy	5.772	10.003	5.978	10.172	5.826	10.084
Change (%)	-2.086	-1.835	+0.521	-1.252	+0.241	+0.139

SUPPLEMENTARY TABLE 3.  $\text{CoPS}_3$ ,  $\text{FePS}_3$  and  $\text{NiPS}_3$  lattice parameters of the crystallographic non-magnetic (NM) and Néel antiferromagnetic (AF) configurations.

Lattice parameter ( $\text{\AA}$ )	$\text{CoPS}_3$		$\text{FePS}_3$		$\text{NiPS}_3$	
	a	b	a	b	a	b
Crystallographic NM	5.895	10.190	5.947	10.301	5.812	10.070
AF-Néel	5.885	10.187	5.947	10.303	5.834	10.106
Change (%)	-0.169	-0.029	+0.000	+0.019	+0.378	+0.357

SUPPLEMENTARY TABLE 4.  $\text{CoPS}_3$ ,  $\text{FePS}_3$  and  $\text{NiPS}_3$  lattice parameters of the crystallographic non-magnetic (NM) and ferromagnetic (FM) configurations.

Lattice parameter ( $\text{\AA}$ )	$\text{CoPS}_3$		$\text{FePS}_3$		$\text{NiPS}_3$	
	a	b	a	b	a	b
Crystallographic NM	5.895	10.190	5.947	10.301	5.812	10.070
FM	5.836	10.103	5.932	10.278	5.824	10.089
Change (%)	-1.001	-0.854	-0.252	-0.223	+0.207	+0.189

SUPPLEMENTARY TABLE 5.  $\text{CoPS}_3$ ,  $\text{FePS}_3$  and  $\text{NiPS}_3$  distances (d and d') of the crystallographic non-magnetic (NM) and zigzag antiferromagnetic (AF) configurations.

Distance ( $\text{\AA}$ )	$\text{CoPS}_3$		$\text{FePS}_3$		$\text{NiPS}_3$	
	d	d'	d	d'	d	d'
Crystallographic NM	3.397	3.402	3.433	3.433	3.357	3.356
AF-zigzag	3.723	3.192	3.626	3.316	3.353	3.359
Change (%)	+9.596	-6.172	+5.621	-3.408	-0.119	+0.089

SUPPLEMENTARY TABLE 6. CoPS<sub>3</sub>, FePS<sub>3</sub> and NiPS<sub>3</sub> distances (d and d') of the crystallographic non-magnetic (NM) and Stripy antiferromagnetic (AF) configurations.

	CoPS <sub>3</sub>		FePS <sub>3</sub>		NiPS <sub>3</sub>	
Distance (Å)	d	d'	d	d'	d	d'
Crystallographic NM	3.397	3.402	3.433	3.433	3.357	3.356
AF-stripy	3.337	3.332	2.670	3.843	3.373	3.357
Change (%)	-1.766	-2.058	-22.225	+11.943	+0.477	+0.029

SUPPLEMENTARY TABLE 7. CoPS<sub>3</sub>, FePS<sub>3</sub> and NiPS<sub>3</sub> distances (d and d') of the crystallographic non-magnetic (NM) and Néel antiferromagnetic (AF) configurations.

	CoPS <sub>3</sub>		FePS <sub>3</sub>		NiPS <sub>3</sub>	
Distance (Å)	d	d'	d	d'	d	d'
Crystallographic NM	3.397	3.402	3.433	3.433	3.357	3.356
AF-Néel	3.393	3.398	3.436	3.433	3.369	3.369
Change (%)	-0.118	-0.118	+0.087	+0.000	+0.357	+0.387

SUPPLEMENTARY TABLE 8. CoPS<sub>3</sub>, FePS<sub>3</sub> and NiPS<sub>3</sub> distances (d and d') of the crystallographic non-magnetic (NM) and ferromagnetic (FM) configurations.

	CoPS <sub>3</sub>		FePS <sub>3</sub>		NiPS <sub>3</sub>	
Distance (Å)	d	d'	d	d'	d	d'
Crystallographic NM	3.397	3.402	3.433	3.433	3.357	3.356
FM	3.368	3.368	3.429	3.424	3.363	3.362
Change (%)	-0.854	-0.999	+0.116	+0.262	+0.179	+0.179

SUPPLEMENTARY TABLE 9. Orbital resolved exchange parameters for the experimental structure of FePS<sub>3</sub>.

J <sub>1</sub> (meV)	d <sub>z2</sub>	d <sub>xz</sub>	d <sub>yz</sub>	d <sub>x2y2</sub>	d <sub>xy</sub>
d <sub>z2</sub>	-0.467	-0.154	-0.157	-0.047	-0.027
d <sub>xz</sub>	-0.154	7.091	0.063	-0.307	0.288
d <sub>yz</sub>	-0.157	0.063	7.215	-0.103	0.109
d <sub>x2y2</sub>	-0.047	-0.307	-0.103	-0.194	-0.017
d <sub>xy</sub>	-0.027	0.288	0.109	-0.017	1.612

SUPPLEMENTARY TABLE 10. Orbital resolved exchange parameters for the experimental structure of FePS<sub>3</sub>.

J <sub>2</sub> (meV)	d <sub>z2</sub>	d <sub>xz</sub>	d <sub>yz</sub>	d <sub>x2y2</sub>	d <sub>xy</sub>
d <sub>z2</sub>	-0.045	-0.013	-0.011	-0.008	-1.18
d <sub>xz</sub>	-0.013	0.334	0.14	0.0	0.791
d <sub>yz</sub>	-0.011	0.14	0.418	0.036	0.634
d <sub>x2y2</sub>	-0.008	0.0	0.036	-0.783	0.086
d <sub>xy</sub>	-1.18	0.791	0.634	0.086	11.016

SUPPLEMENTARY TABLE 11. Orbital resolved exchange parameters for the experimental structure of FePS<sub>3</sub>.

J <sub>3</sub> (meV)	d <sub>z2</sub>	d <sub>xz</sub>	d <sub>yz</sub>	d <sub>x2y2</sub>	d <sub>xy</sub>
d <sub>z2</sub>	0.01	-0.058	-0.065	-0.028	-0.011
d <sub>xz</sub>	-0.058	-0.02	-0.037	-0.101	-0.076
d <sub>yz</sub>	-0.065	-0.037	-0.022	-0.122	0.089
d <sub>x2y2</sub>	-0.028	-0.101	-0.122	-0.004	-0.041
d <sub>xy</sub>	-0.011	-0.076	-0.089	-0.041	0.405

SUPPLEMENTARY TABLE 12. Orbital resolved exchange parameters for the experimental structure of FePS<sub>3</sub>.

<b>J<sub>4</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.121	0.002	0.003	0.013	-0.234
<b>d<sub>xz</sub></b>	0.002	-0.093	-0.022	-0.005	-0.146
<b>d<sub>yz</sub></b>	0.003	-0.022	-0.081	0.002	0.122
<b>d<sub>x2y2</sub></b>	0.012	-0.005	0.002	-0.01	-0.18
<b>d<sub>xy</sub></b>	-0.234	-0.146	-0.122	-0.18	-2.509

SUPPLEMENTARY TABLE 13. Orbital resolved exchange parameters for the experimental structure of FePS<sub>3</sub>.

<b>J<sub>5</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-2.323	0.019	0.015	-1.175	0.148
<b>d<sub>xz</sub></b>	0.019	0.035	0.024	-0.01	-0.239
<b>d<sub>yz</sub></b>	0.015	0.024	0.022	-0.032	0.154
<b>d<sub>x2y2</sub></b>	-1.175	-0.01	-0.032	-0.154	-0.138
<b>d<sub>xy</sub></b>	0.148	-0.239	-0.154	-0.138	-2.491

SUPPLEMENTARY TABLE 14. Orbital resolved exchange parameters for the experimental structure of FePS<sub>3</sub>.

<b>J<sub>6</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.156	0.03	0.025	-0.0	-0.05
<b>d<sub>xz</sub></b>	0.03	-0.012	0.004	0.053	-0.073
<b>d<sub>yz</sub></b>	0.025	0.004	-0.01	0.04	0.042
<b>d<sub>x2y2</sub></b>	-0.0	0.053	0.04	-5.914	-0.044
<b>d<sub>xy</sub></b>	-0.05	-0.073	-0.042	-0.044	0.325

SUPPLEMENTARY TABLE 15. Orbital resolved exchange parameters for the spin polarized optimized structure of FePS<sub>3</sub>.

<b>J<sub>1</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.899	-0.192	-0.188	-0.127	-0.017
<b>d<sub>xz</sub></b>	-0.192	8.323	0.035	-0.29	0.306
<b>d<sub>yz</sub></b>	-0.188	0.035	8.383	-0.088	0.12
<b>d<sub>x2y2</sub></b>	-0.127	-0.29	-0.088	-0.28	-0.044
<b>d<sub>xy</sub></b>	-0.017	0.306	0.12	-0.044	1.823

SUPPLEMENTARY TABLE 16. Orbital resolved exchange parameters for the spin polarized optimized structure of FePS<sub>3</sub>.

<b>J<sub>2</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.104	-0.021	-0.019	-0.009	-2.114
<b>d<sub>xz</sub></b>	-0.021	0.161	0.019	-0.005	0.5
<b>d<sub>yz</sub></b>	-0.019	0.019	0.182	0.002	0.432
<b>d<sub>x2y2</sub></b>	-0.009	-0.005	0.002	0.128	0.015
<b>d<sub>xy</sub></b>	-2.114	0.5	0.432	0.015	5.022

SUPPLEMENTARY TABLE 17. Orbital resolved exchange parameters for the spin polarized optimized structure of FePS<sub>3</sub>.

<b>J<sub>3</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	0.012	-0.083	-0.093	-0.059	-0.013
<b>d<sub>xz</sub></b>	-0.083	-0.025	-0.044	-0.088	-0.059
<b>d<sub>yz</sub></b>	-0.093	-0.044	-0.027	-0.105	-0.064
<b>d<sub>x2y2</sub></b>	-0.059	-0.088	-0.105	0.007	-0.038
<b>d<sub>xy</sub></b>	-0.013	-0.059	-0.064	-0.038	0.278

SUPPLEMENTARY TABLE 18. Orbital resolved exchange parameters for the spin polarized optimized structure of FePS<sub>3</sub>.

$\mathbf{J}_4$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-0.11	0.002	0.003	0.036	-0.212
$\mathbf{d}_{xz}$	0.002	-0.112	-0.039	-0.014	-0.061
$\mathbf{d}_{yz}$	0.003	-0.039	-0.105	0.0	-0.066
$\mathbf{d}_{x2y2}$	0.036	-0.014	0.0	-0.004	-0.164
$\mathbf{d}_{xy}$	-0.212	-0.061	-0.066	-0.164	-1.186

SUPPLEMENTARY TABLE 19. Orbital resolved exchange parameters for the spin polarized optimized structure of FePS<sub>3</sub>.

$\mathbf{J}_5$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-2.613	-0.016	-0.014	-1.223	0.154
$\mathbf{d}_{xz}$	-0.016	-0.008	0.032	-0.059	-0.105
$\mathbf{d}_{yz}$	-0.014	0.032	-0.016	-0.075	-0.06
$\mathbf{d}_{x2y2}$	-1.223	-0.059	-0.075	-0.181	-0.122
$\mathbf{d}_{xy}$	0.154	-0.105	-0.06	-0.122	-1.114

SUPPLEMENTARY TABLE 20. Orbital resolved exchange parameters for the spin polarized optimized structure of FePS<sub>3</sub>.

$\mathbf{J}_6$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-0.196	0.064	0.056	-0.001	-0.236
$\mathbf{d}_{xz}$	0.064	-0.026	0.005	0.083	-0.092
$\mathbf{d}_{yz}$	0.056	0.005	-0.022	0.083	-0.071
$\mathbf{d}_{x2y2}$	-0.001	0.083	0.083	-5.386	-0.025
$\mathbf{d}_{xy}$	-0.236	-0.092	-0.071	-0.025	1.036

SUPPLEMENTARY TABLE 21. Orbital resolved exchange parameters for the experimental structure of CoPS<sub>3</sub>.

$\mathbf{J}_1$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	0.168	-0.907	-0.922	0.108	-0.221
$\mathbf{d}_{xz}$	-0.907	7.514	0.105	-0.977	-0.417
$\mathbf{d}_{yz}$	-0.922	0.105	7.465	-1.266	-0.066
$\mathbf{d}_{x2y2}$	0.108	-0.977	-1.266	-0.094	0.073
$\mathbf{d}_{xy}$	-0.221	-0.417	-0.065	0.073	-0.36

SUPPLEMENTARY TABLE 22. Orbital resolved exchange parameters for the experimental structure of CoPS<sub>3</sub>.

$\mathbf{J}_2$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-0.371	0.078	0.066	-0.002	-0.242
$\mathbf{d}_{xz}$	0.078	0.524	0.022	0.115	-0.093
$\mathbf{d}_{yz}$	0.066	0.022	0.54	-0.106	0.126
$\mathbf{d}_{x2y2}$	-0.002	0.115	-0.106	-0.687	-0.034
$\mathbf{d}_{xy}$	-0.242	-0.093	0.126	-0.034	-0.068

SUPPLEMENTARY TABLE 23. Orbital resolved exchange parameters for the experimental structure of CoPS<sub>3</sub>.

$\mathbf{J}_3$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	0.069	-0.117	-0.114	-0.047	-0.004
$\mathbf{d}_{xz}$	-0.117	-0.06	-0.024	-0.326	-0.003
$\mathbf{d}_{yz}$	-0.114	-0.024	-0.059	-0.367	0.01
$\mathbf{d}_{x2y2}$	-0.047	-0.326	-0.367	-0.17	0.003
$\mathbf{d}_{xy}$	-0.004	-0.003	0.01	0.004	-0.022

SUPPLEMENTARY TABLE 24. Orbital resolved exchange parameters for the experimental structure of CoPS<sub>3</sub>.

<b>J<sub>4</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.025	-0.011	-0.001	0.015	0.046
<b>d<sub>xz</sub></b>	-0.011	-0.062	-0.004	0.002	0.047
<b>d<sub>yz</sub></b>	-0.001	-0.004	-0.05	-0.006	0.046
<b>d<sub>x2y2</sub></b>	0.015	0.002	-0.006	-0.082	-0.013
<b>d<sub>xy</sub></b>	0.046	0.047	0.046	-0.013	0.055

SUPPLEMENTARY TABLE 25. Orbital resolved exchange parameters for the experimental structure of CoPS<sub>3</sub>.

<b>J<sub>5</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-3.482	-0.181	-0.163	-0.862	0.13
<b>d<sub>xz</sub></b>	-0.181	-0.018	0.001	-0.263	0.034
<b>d<sub>yz</sub></b>	-0.163	0.001	-0.044	-0.18	0.004
<b>d<sub>x2y2</sub></b>	-0.862	-0.263	-0.18	-0.016	0.097
<b>d<sub>xy</sub></b>	0.13	0.034	0.004	0.097	0.051

SUPPLEMENTARY TABLE 26. Orbital resolved exchange parameters for the experimental structure of CoPS<sub>3</sub>.

<b>J<sub>3</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.321	0.054	0.045	0.001	0.114
<b>d<sub>xz</sub></b>	0.054	-0.018	-0.023	0.138	0.072
<b>d<sub>yz</sub></b>	0.045	-0.023	-0.01	0.224	0.01
<b>d<sub>x2y2</sub></b>	0.001	0.138	0.224	-3.745	-0.026
<b>d<sub>xy</sub></b>	0.114	0.072	0.01	-0.026	0.029

SUPPLEMENTARY TABLE 27. Orbital resolved exchange parameters for the spin polarized optimized structure of CoPS<sub>3</sub>.

<b>J<sub>1</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.029	-0.889	-0.868	-0.449	-0.182
<b>d<sub>xz</sub></b>	-0.889	10.043	0.01	-0.658	0.055
<b>d<sub>yz</sub></b>	-0.868	0.01	10.011	-0.76	0.152
<b>d<sub>x2y2</sub></b>	-0.449	-0.658	-0.76	0.024	-0.03
<b>d<sub>xy</sub></b>	-0.182	0.055	0.152	-0.03	-0.173

SUPPLEMENTARY TABLE 28. Orbital resolved exchange parameters for the spin polarized optimized structure of CoPS<sub>3</sub>.

<b>J<sub>2</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.213	0.02	0.018	0.0	-0.113
<b>d<sub>xz</sub></b>	0.02	0.106	-0.008	-0.014	0.122
<b>d<sub>yz</sub></b>	0.018	-0.008	0.109	-0.038	0.128
<b>d<sub>x2y2</sub></b>	0.0	-0.014	-0.038	-0.035	0.0
<b>d<sub>xy</sub></b>	-0.113	0.122	0.128	0.0	-0.199

SUPPLEMENTARY TABLE 29. Orbital resolved exchange parameters for the spin polarized optimized structure of CoPS<sub>3</sub>.

<b>J<sub>3</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	0.029	-0.135	-0.136	-0.162	-0.027
<b>d<sub>xz</sub></b>	-0.135	-0.049	-0.018	-0.2	0.051
<b>d<sub>yz</sub></b>	-0.136	-0.018	-0.049	-0.215	0.051
<b>d<sub>x2y2</sub></b>	-0.162	-0.2	-0.215	-0.089	-0.007
<b>d<sub>xy</sub></b>	-0.027	0.051	0.051	-0.007	0.011



SUPPLEMENTARY TABLE 30. Orbital resolved exchange parameters for the spin polarized optimized structure of CoPS<sub>3</sub>.

$\mathbf{J}_4$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-0.014	0.002	0.005	0.068	-0.048
$\mathbf{d}_{xz}$	0.002	-0.019	-0.027	-0.002	0.009
$\mathbf{d}_{yz}$	0.005	-0.027	-0.014	-0.013	0.016
$\mathbf{d}_{x2y2}$	0.068	-0.002	-0.013	-0.071	-0.021
$\mathbf{d}_{xy}$	-0.048	0.009	0.016	-0.021	0.002

SUPPLEMENTARY TABLE 31. Orbital resolved exchange parameters for the spin polarized optimized structure of CoPS<sub>3</sub>.

$\mathbf{J}_5$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-3.726	-0.232	-0.228	-1.283	0.014
$\mathbf{d}_{xz}$	-0.232	-0.055	0.0	-0.211	0.002
$\mathbf{d}_{yz}$	-0.229	0.0	-0.058	-0.183	-0.004
$\mathbf{d}_{x2y2}$	-1.283	-0.211	-0.183	-0.06	0.002
$\mathbf{d}_{xy}$	0.014	0.002	-0.004	0.002	0.002

SUPPLEMENTARY TABLE 32. Orbital resolved exchange parameters for the spin polarized optimized structure of CoPS<sub>3</sub>.

$\mathbf{J}_6$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-0.483	0.099	0.087	0.001	0.316
$\mathbf{d}_{xz}$	0.099	-0.068	0.0	0.08	0.064
$\mathbf{d}_{yz}$	0.087	0.0	-0.066	0.108	0.041
$\mathbf{d}_{x2y2}$	0.001	0.08	0.108	-3.258	-0.01
$\mathbf{d}_{xy}$	0.316	0.064	0.041	-0.01	0.196

SUPPLEMENTARY TABLE 33. Orbital resolved exchange parameters for the experimental structure of NiPS<sub>3</sub>.

$\mathbf{J}_1$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-0.055	-0.008	-0.012	-0.038	0.002
$\mathbf{d}_{xz}$	-0.008	0.014	-0.002	0.011	0.001
$\mathbf{d}_{yz}$	-0.012	-0.002	0.012	0.007	0.0
$\mathbf{d}_{x2y2}$	-0.038	0.011	0.007	-0.312	-0.004
$\mathbf{d}_{xy}$	0.003	0.001	0.0	-0.004	-0.002

SUPPLEMENTARY TABLE 34. Orbital resolved exchange parameters for the experimental structure of NiPS<sub>3</sub>.

$\mathbf{J}_2$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-0.161	-0.012	-0.01	0.0	-0.011
$\mathbf{d}_{xz}$	-0.012	-0.006	0.001	0.001	0.005
$\mathbf{d}_{yz}$	-0.01	0.001	-0.006	0.001	0.005
$\mathbf{d}_{x2y2}$	0.0	0.001	0.001	-0.1	-0.008
$\mathbf{d}_{xy}$	-0.011	0.005	0.005	-0.008	0.017

SUPPLEMENTARY TABLE 35. Orbital resolved exchange parameters for the experimental structure of NiPS<sub>3</sub>.

$\mathbf{J}_3$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	0.015	-0.0	0.0	0.004	0.001
$\mathbf{d}_{xz}$	-0.0	-0.0	-0.001	0.011	-0.0
$\mathbf{d}_{yz}$	0.0	-0.001	-0.0	0.012	-0.0
$\mathbf{d}_{x2y2}$	0.004	0.011	0.012	0.053	0.007
$\mathbf{d}_{xy}$	0.001	-0.0	-0.0	0.007	-0.0

SUPPLEMENTARY TABLE 36. Orbital resolved exchange parameters for the experimental structure of NiPS<sub>3</sub>.

<b>J<sub>4</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	0.204	-0.001	-0.0	0.024	0.003
<b>d<sub>xz</sub></b>	-0.001	-0.001	0.0	-0.0	0.0
<b>d<sub>yz</sub></b>	-0.0	0.0	-0.001	0.0	0.0
<b>d<sub>x2y2</sub></b>	0.024	-0.0	0.0	-0.028	-0.009
<b>d<sub>xy</sub></b>	0.003	0.0	0.0	-0.009	-0.001

SUPPLEMENTARY TABLE 37. Orbital resolved exchange parameters for the experimental structure of NiPS<sub>3</sub>.

<b>J<sub>5</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-5.191	-0.074	-0.061	-2.334	-0.042
<b>d<sub>xz</sub></b>	-0.074	-0.006	-0.0	-0.031	0.0
<b>d<sub>yz</sub></b>	-0.061	-0.0	-0.006	-0.024	-0.0
<b>d<sub>x2y2</sub></b>	-2.334	-0.031	-0.024	-0.38	-0.003
<b>d<sub>xy</sub></b>	-0.042	0.0	-0.0	-0.003	-0.001

SUPPLEMENTARY TABLE 38. Orbital resolved exchange parameters for the experimental structure of NiPS<sub>3</sub>.

<b>J<sub>6</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.058	-0.002	-0.002	0.0	0.028
<b>d<sub>xz</sub></b>	-0.002	0.0	0.0	-0.003	0.0
<b>d<sub>yz</sub></b>	-0.002	0.0	0.0	-0.004	0.0
<b>d<sub>x2y2</sub></b>	0.0	-0.003	-0.004	-10.997	-0.092
<b>d<sub>xy</sub></b>	0.028	0.0	0.0	-0.092	-0.008

SUPPLEMENTARY TABLE 39. Orbital resolved exchange parameters for the spin polarized optimized structure of NiPS<sub>3</sub>.

<b>J<sub>1</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	0.099	0.009	0.007	-0.05	0.007
<b>d<sub>xz</sub></b>	0.009	0.004	-0.002	0.044	-0.0
<b>d<sub>yz</sub></b>	0.007	-0.002	0.002	0.038	-0.0
<b>d<sub>x2y2</sub></b>	-0.05	0.044	0.038	-0.219	-0.003
<b>d<sub>xy</sub></b>	0.007	-0.0	-0.0	-0.003	-0.004

SUPPLEMENTARY TABLE 40. Orbital resolved exchange parameters for the spin polarized optimized structure of NiPS<sub>3</sub>.

<b>J<sub>2</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	-0.17	-0.009	-0.007	0.001	0.08
<b>d<sub>xz</sub></b>	-0.009	-0.008	-0.002	0.007	0.005
<b>d<sub>yz</sub></b>	-0.007	-0.002	-0.009	0.007	0.004
<b>d<sub>x2y2</sub></b>	0.001	0.007	0.007	0.054	-0.004
<b>d<sub>xy</sub></b>	0.08	0.005	0.004	-0.004	0.002

SUPPLEMENTARY TABLE 41. Orbital resolved exchange parameters for the spin polarized optimized structure of NiPS<sub>3</sub>.

<b>J<sub>3</sub> (meV)</b>	<b>d<sub>z2</sub></b>	<b>d<sub>xz</sub></b>	<b>d<sub>yz</sub></b>	<b>d<sub>x2y2</sub></b>	<b>d<sub>xy</sub></b>
<b>d<sub>z2</sub></b>	0.012	-0.002	-0.001	-0.013	0.001
<b>d<sub>xz</sub></b>	-0.002	-0.001	-0.001	0.008	-0.0
<b>d<sub>yz</sub></b>	-0.001	-0.001	-0.0	0.008	-0.001
<b>d<sub>x2y2</sub></b>	-0.013	0.008	0.008	0.028	0.01
<b>d<sub>xy</sub></b>	0.001	-0.0	-0.001	0.01	-0.001

SUPPLEMENTARY TABLE 42. Orbital resolved exchange parameters for the spin polarized optimized structure of NiPS<sub>3</sub>.

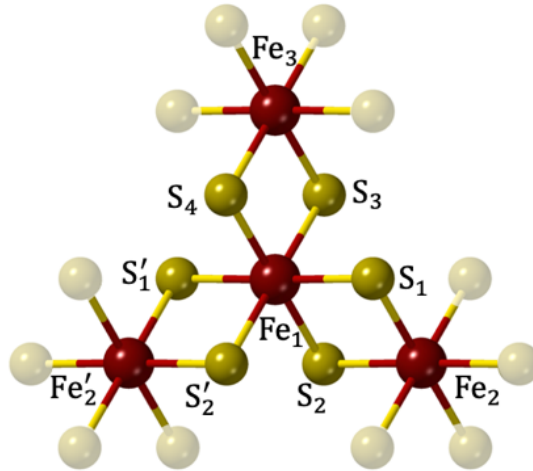
$\mathbf{J}_4$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	0.116	0.002	0.002	0.028	-0.006
$\mathbf{d}_{xz}$	0.002	-0.002	0.0	0.0	0.0
$\mathbf{d}_{yz}$	0.002	0.0	-0.002	0.0	0.0
$\mathbf{d}_{x2y2}$	0.028	0.0	0.0	-0.028	-0.01
$\mathbf{d}_{xy}$	-0.006	0.0	0.0	-0.01	0.0

SUPPLEMENTARY TABLE 43. Orbital resolved exchange parameters for the spin polarized optimized structure of NiPS<sub>3</sub>.

$\mathbf{J}_5$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-4.91	-0.056	-0.046	-2.12	-0.032
$\mathbf{d}_{xz}$	-0.056	-0.009	0.0	-0.013	0.0
$\mathbf{d}_{yz}$	-0.046	0.0	-0.008	-0.007	-0.0
$\mathbf{d}_{x2y2}$	-2.12	-0.013	-0.007	-0.353	-0.003
$\mathbf{d}_{xy}$	-0.032	0.0	-0.0	-0.003	-0.001

SUPPLEMENTARY TABLE 44. Orbital resolved exchange parameters for the spin polarized optimized structure of NiPS<sub>3</sub>.

$\mathbf{J}_6$ (meV)	$\mathbf{d}_{z2}$	$\mathbf{d}_{xz}$	$\mathbf{d}_{yz}$	$\mathbf{d}_{x2y2}$	$\mathbf{d}_{xy}$
$\mathbf{d}_{z2}$	-0.04	-0.002	-0.002	0.0	0.048
$\mathbf{d}_{xz}$	-0.002	0.0	0.0	-0.003	0.0
$\mathbf{d}_{yz}$	-0.002	0.0	0.0	-0.004	0.0
$\mathbf{d}_{x2y2}$	0.0	-0.003	-0.004	-10.334	-0.07
$\mathbf{d}_{xy}$	0.048	0.0	0.0	-0.07	-0.012

SUPPLEMENTARY FIG. 8. Top view of a cluster of single-layer FePS<sub>3</sub> centered in Fe<sub>1</sub> labelling the first Fe atoms neighbours and the S atoms that mediate super-exchange magnetic interactions.

SUPPLEMENTARY TABLE 45. Hopping parameters for the crystallographic structure of FePS<sub>3</sub>.

		$\mathbf{S}_1(\mathbf{p}_z)$	$\mathbf{S}_1(\mathbf{p}_x)$	$\mathbf{S}_1(\mathbf{p}_y)$	$\mathbf{S}_2(\mathbf{p}_z)$	$\mathbf{S}_2(\mathbf{p}_x)$	$\mathbf{S}_2(\mathbf{p}_y)$
<b>Fe<sub>1</sub></b>	$\mathbf{d}_{z2}$	1.089	-0.079	-0.243	0.054	-0.012	-0.608
	$\mathbf{d}_{xz}$	-0.124	-0.331	-0.029	-0.041	-0.021	0.028
	$\mathbf{d}_{yz}$	-0.254	-0.100	-0.357	-0.427	0.127	-0.002
	$\mathbf{d}_{x2y2}$	0.002	-0.005	-0.037	0.127	-0.086	-0.987
	$\mathbf{d}_{xy}$	0.003	0.021	0.038	0.037	-0.341	0.181
<b>Fe<sub>2</sub></b>	$\mathbf{d}_{z2}$	-0.054	0.012	0.608	-1.089	0.079	0.243
	$\mathbf{d}_{xz}$	0.041	0.021	-0.028	0.124	0.331	0.029
	$\mathbf{d}_{yz}$	0.427	-0.127	0.002	0.254	0.100	0.357
	$\mathbf{d}_{x2y2}$	-0.127	0.086	0.987	-0.002	0.005	0.037
	$\mathbf{d}_{xy}$	-0.037	0.341	-0.181	-0.003	-0.021	-0.038

SUPPLEMENTARY TABLE 46. Hopping parameters for the crystallographic structure of FePS<sub>3</sub>.

		$\mathbf{S}_3(\mathbf{p}_z)$	$\mathbf{S}_3(\mathbf{p}_x)$	$\mathbf{S}_3(\mathbf{p}_y)$	$\mathbf{S}_4(\mathbf{p}_z)$	$\mathbf{S}_4(\mathbf{p}_x)$	$\mathbf{S}_4(\mathbf{p}_y)$
<b>Fe<sub>1</sub></b>	$\mathbf{d}_{z2}$	0.015	-0.525	-0.037	-0.015	0.040	0.525
	$\mathbf{d}_{xz}$	-0.274	-0.023	-0.031	-0.020	-0.008	-0.029
	$\mathbf{d}_{yz}$	0.037	0.030	0.010	0.276	0.030	0.026
	$\mathbf{d}_{x2y2}$	-0.036	0.942	0.032	-0.028	0.078	0.939
	$\mathbf{d}_{xy}$	-0.070	-0.043	-0.325	0.075	0.319	-0.078
<b>Fe<sub>3</sub></b>	$\mathbf{d}_{z2}$	0.023	-0.056	-0.562	-0.022	0.562	0.053
	$\mathbf{d}_{xz}$	0.028	0.018	0.062	0.334	-0.016	0.012
	$\mathbf{d}_{yz}$	-0.336	-0.011	0.011	-0.049	-0.062	-0.018
	$\mathbf{d}_{x2y2}$	0.041	-0.117	-1.048	0.052	-1.050	-0.058
	$\mathbf{d}_{xy}$	-0.096	-0.414	0.080	0.090	0.056	0.423

SUPPLEMENTARY TABLE 47. Hopping parameters for the crystallographic structure of CoPS<sub>3</sub>.

		$\mathbf{S}_1(\mathbf{p}_z)$	$\mathbf{S}_1(\mathbf{p}_x)$	$\mathbf{S}_1(\mathbf{p}_y)$	$\mathbf{S}_2(\mathbf{p}_z)$	$\mathbf{S}_2(\mathbf{p}_x)$	$\mathbf{S}_2(\mathbf{p}_y)$
<b>Co<sub>1</sub></b>	$\mathbf{d}_{z2}$	1.019	-0.281	-0.178	0.032	-0.174	-0.581
	$\mathbf{d}_{xz}$	-0.229	-0.339	0.098	-0.069	-0.032	0.039
	$\mathbf{d}_{yz}$	-0.284	0.040	-0.341	-0.431	0.024	-0.056
	$\mathbf{d}_{x2y2}$	-0.003	0.003	-0.043	0.059	-0.405	-0.902
	$\mathbf{d}_{xy}$	-0.001	0.017	0.036	-0.089	-0.35	0.381
<b>Co<sub>2</sub></b>	$\mathbf{d}_{z2}$	-0.032	0.174	0.581	-1.019	0.279	0.178
	$\mathbf{d}_{xz}$	0.069	0.032	-0.039	0.229	0.339	-0.097
	$\mathbf{d}_{yz}$	0.431	-0.025	0.056	0.284	-0.041	0.341
	$\mathbf{d}_{x2y2}$	-0.059	0.406	0.901	0.003	-0.003	0.043
	$\mathbf{d}_{xy}$	0.089	0.350	-0.382	0.001	-0.017	-0.036

SUPPLEMENTARY TABLE 48. Hopping parameters for the crystallographic structure of CoPS<sub>3</sub>.

		$\mathbf{S}_3(\mathbf{p}_z)$	$\mathbf{S}_3(\mathbf{p}_x)$	$\mathbf{S}_3(\mathbf{p}_y)$	$\mathbf{S}_4(\mathbf{p}_z)$	$\mathbf{S}_4(\mathbf{p}_x)$	$\mathbf{S}_4(\mathbf{p}_y)$
<b>Co<sub>1</sub></b>	$\mathbf{d}_{z2}$	0.017	-0.489	-0.033	-0.018	0.035	0.488
	$\mathbf{d}_{xz}$	-0.241	-0.043	-0.014	-0.029	-0.013	-0.034
	$\mathbf{d}_{yz}$	0.044	0.037	0.014	0.243	0.014	0.046
	$\mathbf{d}_{x2y2}$	-0.042	0.904	0.011	-0.034	0.051	0.908
	$\mathbf{d}_{xy}$	-0.072	-0.094	-0.299	0.077	0.296	-0.019
<b>Co<sub>3</sub></b>	$\mathbf{d}_{z2}$	0.015	-0.031	-0.534	-0.015	0.534	0.029
	$\mathbf{d}_{xz}$	0.037	0.020	0.056	0.282	0.011	0.015
	$\mathbf{d}_{yz}$	-0.285	-0.015	-0.015	-0.054	-0.057	-0.020
	$\mathbf{d}_{x2y2}$	0.037	-0.036	-1.011	0.046	-1.005	0.009
	$\mathbf{d}_{xy}$	-0.089	-0.341	0.012	0.083	0.113	0.341

SUPPLEMENTARY TABLE 49. Hopping parameters for the crystallographic structure of NiPS<sub>3</sub>.

		$\mathbf{S}_1(\mathbf{p}_z)$	$\mathbf{S}_1(\mathbf{p}_x)$	$\mathbf{S}_1(\mathbf{p}_y)$	$\mathbf{S}_2(\mathbf{p}_z)$	$\mathbf{S}_2(\mathbf{p}_x)$	$\mathbf{S}_2(\mathbf{p}_y)$
<b>Ni<sub>1</sub></b>	$\mathbf{d}_{z2}$	1.023	-0.279	-0.112	0.009	-0.116	-0.553
	$\mathbf{d}_{xz}$	-0.288	-0.316	0.066	-0.055	-0.038	0.047
	$\mathbf{d}_{yz}$	-0.166	0.053	-0.352	-0.367	-0.011	0.015
	$\mathbf{d}_{x2y2}$	0.032	0.025	-0.022	0.044	-0.237	-0.890
	$\mathbf{d}_{xy}$	-0.017	-0.008	0.053	-0.057	-0.320	0.262
<b>Ni<sub>2</sub></b>	$\mathbf{d}_{z2}$	-0.008	0.118	0.553	-1.026	0.275	0.113
	$\mathbf{d}_{xz}$	0.055	0.038	-0.047	0.288	0.316	-0.065
	$\mathbf{d}_{yz}$	0.366	0.009	-0.015	0.166	-0.055	0.353
	$\mathbf{d}_{x2y2}$	-0.043	0.241	0.889	-0.032	-0.025	0.022
	$\mathbf{d}_{xy}$	0.058	0.320	-0.263	0.017	0.008	-0.053

SUPPLEMENTARY TABLE 50. Hopping parameters for the crystallographic structure of NiPS<sub>3</sub>.

		$\mathbf{S}_3(\mathbf{p}_z)$	$\mathbf{S}_3(\mathbf{p}_x)$	$\mathbf{S}_3(\mathbf{p}_y)$	$\mathbf{S}_4(\mathbf{p}_z)$	$\mathbf{S}_4(\mathbf{p}_x)$	$\mathbf{S}_4(\mathbf{p}_y)$
<b>Ni<sub>1</sub></b>	$\mathbf{d}_{z2}$	0.034	-0.526	-0.061	-0.035	0.064	0.525
	$\mathbf{d}_{xz}$	-0.331	-0.053	-0.030	-0.011	-0.016	-0.031
	$\mathbf{d}_{yz}$	0.039	0.036	0.019	0.333	0.030	0.057
	$\mathbf{d}_{x2y2}$	-0.069	0.971	0.090	-0.057	0.160	0.955
	$\mathbf{d}_{xy}$	-0.068	0.003	-0.389	0.080	0.369	-0.170
<b>Ni<sub>3</sub></b>	$\mathbf{d}_{z2}$	0.032	-0.05	-0.556	-0.031	0.557	0.047
	$\mathbf{d}_{xz}$	0.015	0.017	0.042	0.352	0.032	0.043
	$\mathbf{d}_{yz}$	-0.355	-0.042	-0.036	-0.045	-0.045	-0.020
	$\mathbf{d}_{x2y2}$	0.051	-0.140	-1.025	0.062	-1.036	-0.067
	$\mathbf{d}_{xy}$	-0.082	-0.393	0.159	0.071	0.019	0.408

## Supplementary Note 2. LANDAU THEORY OF SECOND-ORDER PHASE TRANSITIONS AND SPONTANEOUS MAGNETOSTRICTION

Magnetostriction is a coupling between magnetic and mechanical parts of our system. This coupling can be described by an energy term in the total free energy of our system [1]. We can then write the total free energy as

$$F - F_0 = U_{\text{el}}(z) + a(T - T_N)^{2\beta} L_i L_i + B L_i L_i L_i L_i - \sigma_{ij}(z) \lambda_{ijkl} L_k L_l. \quad (7)$$

Here  $F$  is the total free energy of in the AF phase at zero magnetic field,  $F_0$  is the free energy of paramagnetic phase,  $U_{\text{el}}(z)$  is the elastic energy of a membrane with deflection  $z$  at its centre,  $T$  is the temperature of our system and  $T_N$  is the Néel temperature,  $L_i$  are the components of the Néel vector,  $\beta$  is a critical exponent,  $a$  and  $B$  are positive constants,  $\sigma_{ij}(z)$  is the stress tensor and  $\lambda_{ijkl}$  is the magnetostriction tensor. The last term couples the stress to the Néel vector thereby describing the magnetostriction. If we assume the Néel vector to be fully aligned with the easy axis, Eq. (7) simplifies to:

$$F - F_0 = U_{\text{el}}(z) + a(T - T_N)^{2\beta} L^2 + B L^4 - \sigma_{ij}(z) \lambda_{ij} L^2, \quad (8)$$

where  $L$  is the magnetic order parameter (i.e. the magnitude of the Néel vector). For notational convenience we write  $\lambda_{ij}$  in dropping the third and fourth index of  $\lambda_{ijkl}$  as only the component where  $kl$  corresponds to the easy axis contributes. The elastic energy in a homogeneous membrane is given by [2]

$$U_{\text{el}} = \int \int \frac{S_{ijkl}}{2} \sigma_{ij}(x, y, z) \sigma_{kl}(x, y, z) dx dy, \quad (9)$$

where the integration runs over the in plane dimensions of the membrane,  $z$  is the membrane deflection at its centre and should not be confused with the out of plane coordinate. For ease of notation we will not explicitly write the integration and coordinate dependence from here on. Assuming the membrane thickness does not vary significantly we can take the out of plane component stress component to vanish,  $\sigma_{zx} = \sigma_{zy} = \sigma_{zz} = 0$ . Eq. (9) then simplifies to

$$U_{\text{el}} = \frac{S_{xxxx}}{2} \sigma_{xx} \sigma_{xx} + \frac{S_{yyyy}}{2} \sigma_{yy} \sigma_{yy} + S_{xxyy} \sigma_{xx} \sigma_{yy} + S_{xxxy} \sigma_{xx} \sigma_{xy} + S_{yyxy} \sigma_{yy} \sigma_{xy} + \frac{S_{xyxy}}{2} \sigma_{xy} \sigma_{xy}. \quad (10)$$

Taking our coordinates such that  $x, y$  correspond with the principle stress directions all terms containing  $\sigma_{xy}$  vanish. This simplifies the elastic energy further to

$$U_{\text{el}} = \frac{S_{xxxx}}{2} \sigma_{xx} \sigma_{xx} + \frac{S_{yyyy}}{2} \sigma_{yy} \sigma_{yy} + S_{xxyy} \sigma_{xx} \sigma_{yy}. \quad (11)$$

Assuming the material has isotropic elastic properties the relevant compliance tensor components are

$$S_{xxxx} = S_{yyyy} = \frac{1}{E} \text{ and } S_{xxyy} = \frac{-\nu}{E}. \quad (12)$$

Substituting this in to Eq. (11) we find

$$U_{\text{el}} = \frac{1}{2E} \sigma_{xx} \sigma_{xx} + \frac{1}{2E} \sigma_{yy} \sigma_{yy} - \frac{\nu}{E} \sigma_{xx} \sigma_{yy}. \quad (13)$$

By taking the derivative of the free energy with respect to either  $z$  or  $L$  we find the forces acting on these degrees of freedom,  $\phi_L$  and  $\phi_z$  respectively to be given by

$$-\phi_L = \frac{d(F - F_0)}{dL} = 2a(T - T_N)^{2\beta} L + 4BL^3 - 2\sigma_{ij}(z) \lambda_{ij} L, \quad (14)$$

$$-\phi_z = \frac{d(F - F_0)}{dz} = (S_{ijkl} \sigma_{ij}(z) + \lambda_{kl} L^2) \frac{d\sigma_{kl}(z)}{dz}. \quad (15)$$

### Order parameter and critical exponent

In order to find an equation that describes the order parameter as a function of temperature, we find a solution for equation (14) for the case where  $\phi_L = 0$ . Aside from the trivial solution  $L = 0$ , we find for below the transition the additional solution:

$$L^2 = -\frac{a}{2B} (T - T_N)^{2\beta} + \frac{\sigma_{ij} \lambda_{ij}}{2B}, \quad (16)$$



which could be rewritten for  $T_N^* = T_N - (\frac{\sigma_{ij}\lambda_{ij}}{a})^{\frac{1}{2\beta}}$  [3] as:

$$L^2 = \frac{a}{2B}(T_N^* - T)^{2\beta}. \quad (17)$$

This equation now describes the temperature dependence of the order parameter in a critical region near  $T_N$  with a corresponding critical exponent  $\beta$ .

### Magnetostrictive strain and resonance frequency

To assess the magnetostriction contribution to strain and thus the frequency of a rectangular membrane resonator, we need to find stiffness of the membrane from its force-deflection equation. In doing that we analyse equation (15). First, we describe strain equation for the rectangular membrane at its centre as:

$$\epsilon_{xx}(z) = \epsilon_{0,x} + \frac{c_1}{2} \frac{z^2}{l^2} \quad (18a)$$

$$\epsilon_{yy}(z) = \epsilon_{0,y} + \frac{c_1}{2} \frac{z^2}{w^2} \quad (18b)$$

where  $c_1$  is a geometrical pre-factor that describes the deflection shape of the fundamental mode of vibration [4, 5]. For  $w \ll l$  we can neglect the  $z$  dependence of  $\epsilon_{xx}(z)$ . Now, we substitute (18) to (15), using the relation  $\sigma_{ij} = C_{ijkl}\epsilon_{kl}$ , we find

$$-\phi_z = \left( \frac{E}{1-\nu^2} \epsilon_{0,y} + \frac{\nu E}{1-\nu^2} \epsilon_{0,x} - \lambda_{yy} L^2 \right) \frac{c_1}{w^2} z + \frac{E}{1-\nu^2} \frac{c_1^2}{2} \frac{z^3}{w^4}, \quad (19)$$

where we used that  $C_{xxxx} = C_{yyyy} = \frac{E}{1-\nu^2}$  and  $C_{yyxx} = C_{xxyy} = \frac{\nu E}{1-\nu^2}$ . Eq. (19) becomes

$$-\phi_z = k_1 z - \frac{\lambda_{ij} c_1}{w^2} L^2 z + k_3 z^3 \quad (20)$$

where  $k_1$  is the elastic linear stiffness and  $k_3$  is the cubic elastic stiffness, given by

$$k_1 = \frac{E}{1-\nu^2} (\epsilon_{0,y} + \nu \epsilon_{0,x}) \frac{c_1}{w^2} \quad (21)$$

$$k_3 = \frac{E}{1-\nu^2} \frac{c_1^2}{2w^4}. \quad (22)$$

Assuming small deflections we can neglect the  $z^3$  contribution in eq. (19) and find that the linear stiffness is changed with respect to the purely elastic case. If we consider a rectangular cavity with its long axis is parallel to the crystalline axis  $b$  or  $a$  respectively we find:

$$-\phi_{z \text{ b,a}} = \left( k_1 - \frac{c_1}{w^2} \lambda_{a,b} L^2 \right) z, \quad (23)$$

where  $\lambda_{a,b}$  are the phenomenological magnetostriction coefficients, chosen such that to couple  $a$  and  $b$  crystalline directions and  $L$ . That leads to a change in the effective linear stiffness  $k_{b,a}$ :

$$k_{b,a} = k_1 - \frac{c_1}{w^2} \lambda_{a,b} L^2, \quad (24)$$

which can be used to write down the frequency equations using  $f_{a,b} = \frac{1}{2\pi} \sqrt{\frac{k_{a,b}}{m}}$ ,

$$f_b \approx \frac{1}{2\pi} \sqrt{\frac{1}{m} \frac{c_1}{w^2} \left[ \frac{E}{1-\nu^2} (\epsilon_{0,a} + \nu \epsilon_{0,b}) - \lambda_a L^2 \right]}, \quad (25a)$$

$$f_a \approx \frac{1}{2\pi} \sqrt{\frac{1}{m} \frac{c_1}{w^2} \left[ \frac{E}{1-\nu^2} (\epsilon_{0,b} + \nu \epsilon_{0,a}) - \lambda_b L^2 \right]}, \quad (25b)$$

where  $m$  is the mass of the membrane. And define magnetostrictive strain by:

$$\epsilon_{\text{ms},a} = \frac{c_1}{mw^2} \lambda_a L^2, \quad (26)$$

$$\epsilon_{\text{ms},b} = \frac{c_1}{mw^2} \lambda_b L^2. \quad (27)$$

Taking a difference of the squares of equations (25) and assuming  $\epsilon_{0,a} = \epsilon_{0,b}$ , we arrive at the final equation:

$$f_b^2 - f_a^2 = -\frac{1}{4\pi^2} \frac{c_1}{mw^2} [\lambda_b - \lambda_a] L^2, \quad (28)$$

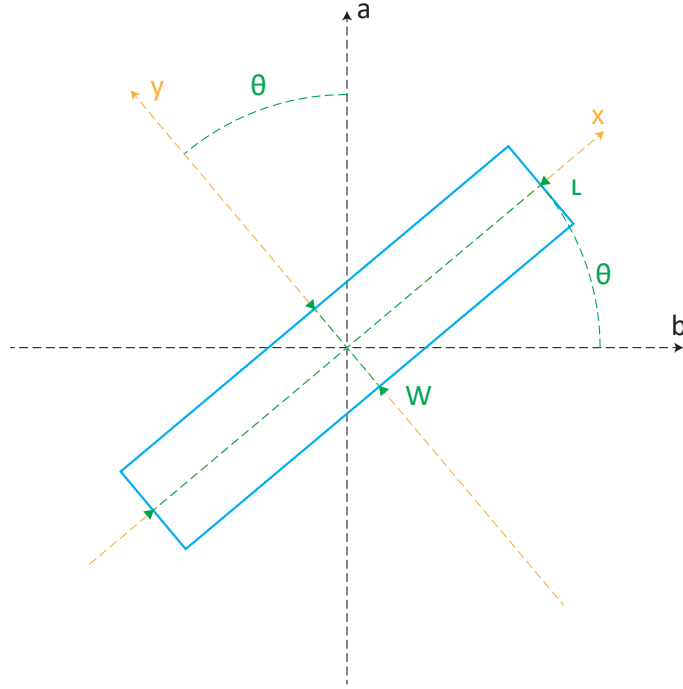
which relates antiferromagnetic order parameter  $L$  and measured resonance frequencies of orthogonal resonators aligned to crystalline axes  $f_{a,b}$  in ordered phase. Finally, one can show that by plugging equation (17) to (28):

$$f_b^2 - f_a^2 \propto (T_N^* - T)^{2\beta}, \quad (29)$$

which could be used to fit experimental data to extract critical exponent  $\beta$  near the phase transition temperature  $T_N^*$ .

### Supplementary Note 3. DERIVATION OF ANISOTROPIC RESONANCE FREQUENCY

Here, we derive the general equation of the resonance frequency of a rectangular cavity oriented at an angle  $\theta$  with respect to the crystalline axes, as schematically shown in Fig. 9. The global coordinate system is defined by the crystallographic axes  $a$  and  $b$ , along which the material deforms resulting in stresses  $\sigma_{aa}$  and  $\sigma_{bb}$ . The longest side of the cavity, with length  $l$ , can be oriented at an arbitrary angle  $\theta$  with respect to  $b$ .



**SUPPLEMENTARY FIG. 9. Schematic illustration of rectangular membrane** Rectangular membrane of width  $w$  and length  $l$  oriented with its long side at an angle  $\theta$  with respect to the crystalline direction  $b$ . The  $x$ - $y$  direction refer to the main directions of the rectangular membrane.

Let us first consider a cavity oriented parallel to a crystallographic axis. Since the membranes are very thin, we can assume that the stress in the direction perpendicular to the plane is zero,  $\sigma_{cc} = 0$ , the membrane's stress tensor can be expressed as

$$\boldsymbol{\sigma} = \begin{pmatrix} \sigma_{aa} & \sigma_{ab} \\ \sigma_{ba} & \sigma_{bb} \end{pmatrix}_{ab}, \quad (30)$$

where the subscript  $(\ )_{ab}$  indicates we expressed the stress tensor in the basis of the crystallographic coordinate system. If we assume that there are no shear forces acting on the crystal lattice,  $\sigma_{ab} = \sigma_{ba} = 0$ , there will be no shear on cavities oriented along the main crystallographic axes. Now, if we consider a rectangular cavity rotated by  $\theta$  with respect to the crystallographic axes, we can define a rotated  $xy$ -coordinate system oriented along the main axis of the rectangle. To express  $\sigma$  in this coordinate system we use the tensor transformation rule,  $\sigma'_{ij} = q_{ki}q_{lj}\sigma_{kl}$  where  $q_{ij}$  are components of the rotation tensor transforming the  $ab$ -coordinate system,  $\mathbf{e}$ , into the  $xy$ -coordinate system,  $\mathbf{e}'$  as  $\mathbf{e}'_i = q_{ij}\mathbf{e}_j$ . We then get:

$$\sigma = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{yx} & \sigma_{yy} \end{pmatrix}_{xy} = \begin{pmatrix} \cos^2(\theta)\sigma_{aa} + \sin^2(\theta)\sigma_{bb} & -\cos(\theta)\sin(\theta)\sigma_{aa} + \sin(\theta)\cos(\theta)\sigma_{bb} \\ -\sin(\theta)\cos(\theta)\sigma_{aa} + \sin(\theta)\cos(\theta)\sigma_{bb} & \sin^2(\theta)\sigma_{aa} + \cos^2(\theta)\sigma_{bb} \end{pmatrix}_{xy}, \quad (31)$$

Then, the resonance frequency of a rectangular membrane oriented at an angle  $\theta$  with respect to the crystallographic axis can be expressed as

$$f_\theta \approx \frac{1}{2} \sqrt{\frac{1}{\rho} \left( \frac{\sigma_{xx}}{l^2} + \frac{\sigma_{yy}}{w^2} \right)}. \quad (32)$$

In the case of high-aspect ratio membranes ( $w \ll l$ ), Eq. 32 can be approximated to

$$f_\theta \approx \frac{1}{2} \sqrt{\frac{1}{\rho} \frac{\sigma_{yy}}{w^2}} = \frac{1}{2} \sqrt{\frac{1}{\rho w^2} (\sin^2(\theta)\sigma_{aa} + \cos^2(\theta)\sigma_{bb})}, \quad (33)$$

which is Eq. 3 of the main text.

Now, let us consider the constitutive equations of the material:

$$c_1 = E(\epsilon_{\text{fab},aa} - \epsilon_{\text{th},aa} - \epsilon_{\text{ms},aa}) = E \left( \epsilon_{\text{fab},aa} - \int_{T_0}^{T_1} \alpha_a(T) dT - \lambda_a L^2(T_1) \right) = \sigma_{aa}(T_1) - \nu \sigma_{bb}(T_1) \quad (34)$$

$$c_2 = E(\epsilon_{\text{fab},bb} - \epsilon_{\text{th},bb} - \epsilon_{\text{ms},bb}) = E \left( \epsilon_{\text{fab},bb} - \int_{T_0}^{T_1} \alpha_b(T) dT - \lambda_b L^2(T_1) \right) = \sigma_{bb}(T_1) - \nu \sigma_{aa}(T_1), \quad (35)$$

where  $\epsilon_{\text{fab}}$  is residual fabrication strain at  $T = T_0$ ,  $\epsilon_{\text{th}}$  and  $\epsilon_{\text{ms}}$  are respectively the thermal expansion and magnetostriction contributions to strain,  $\alpha$  is the thermal expansion coefficient,  $\lambda$  the magnetostriction coefficient and  $E$  is the Young's modulus, which is assumed to be isotropic. We can thus write

$$\sigma_{aa} = c_1 + \nu \sigma_{bb} \quad (36)$$

$$\sigma_{bb} = c_2 + \nu \sigma_{aa}, \quad (37)$$

which can be combined in the following expressions for  $\sigma_{aa}$  and  $\sigma_{bb}$ :

$$\sigma_{aa} = \frac{c_1 + \nu c_2}{1 - \nu^2} \quad (38)$$

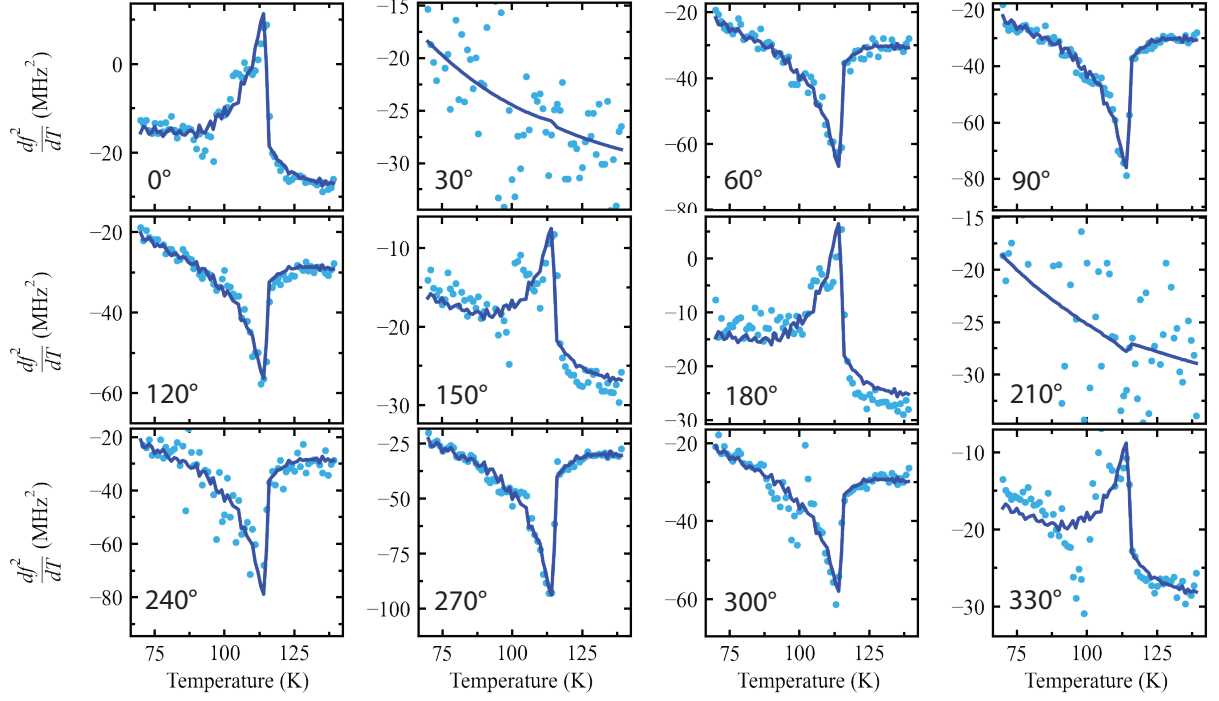
$$\sigma_{bb} = \frac{c_2 + \nu c_1}{1 - \nu^2}. \quad (39)$$

We can now rewrite Eq. 33 in terms of the different contributions to strain, i.e. residual strain from fabrication ( $\epsilon_{\text{fab}}$ ), thermal expansion ( $\propto \alpha$ ) and magnetostriction ( $\propto \lambda$ ):

$$f_\theta(T) = \frac{1}{2} \sqrt{\frac{E}{\rho w^2(1 - \nu^2)}} [\sin^2 \theta (c_1 + \nu c_2) + \cos^2 \theta (c_2 + \nu c_1)] \quad (40)$$

$$= \frac{1}{2} \sqrt{\frac{E}{\rho w^2(1 - \nu^2)}} [(\sin^2 \theta + \nu \cos^2 \theta)(\epsilon_{\text{fab},aa} - \epsilon_{\text{th},aa} - \epsilon_{\text{ms},aa}) + (\cos^2 \theta + \nu \sin^2 \theta)(\epsilon_{\text{fab},bb} - \epsilon_{\text{th},bb} - \epsilon_{\text{ms},bb})]. \quad (41)$$

Which is consistent with Eq. (25). We can eliminate the pretension  $\epsilon_{\text{fab}}$  terms by considering  $\tilde{f}_\theta(T) = f_\theta(T) - f_\theta(T_0)$ . In the following, we assume that the only anisotropic temperature-dependent contribution to the total strain comes



SUPPLEMENTARY FIG. 10. **Angle-resolved**  $\frac{df^2}{dT}$ : Plot of measured (light blue dots)  $\frac{df^2}{dT}$  and fit to  $b_1 C_{\text{Debye}} + b_2 \frac{dL^2}{dT}$  (blue full line) for all angles of a star-shaped array of CoPS<sub>3</sub>.

from magnetostriction, thus we take  $\epsilon_{\text{th},aa} = \epsilon_{\text{th},bb} = \epsilon_{\text{th}}$ . By definition of  $\theta$  we have  $b \rightarrow \theta = 0^\circ$  and  $a \rightarrow \theta = 90^\circ$ . From Eq. (41), we then find that  $\tilde{f}_a^2 - \tilde{f}_b^2$  becomes

$$\tilde{f}_a^2 - \tilde{f}_b^2 = \frac{E}{4\rho w^2(1+\nu)} (-\epsilon_{\text{ms},aa} + \epsilon_{\text{ms},bb}) \quad (42)$$

$$= -\frac{E}{4\rho w^2(1+\nu)} (\lambda_a - \lambda_b) L^2 \quad (43)$$

from which we can directly extract the order parameter. The thermal expansion contribution to strain  $\epsilon_\alpha$  is proportional to the integral over the temperature of the thermal expansion coefficient  $\alpha$ , which is proportional to the Debye specific heat,  $C_{\text{Debye}}$ , via the Grünesen parameter. Thus, the derivative with respect to temperature of  $f_\theta^2$

$$\frac{df_\theta^2}{dT} = \frac{E}{4\rho w^2(1-\nu^2)} \left[ (\sin^2 \theta + \nu \cos^2 \theta) \left( -\alpha - \lambda_a \frac{dL^2}{dT} \right) + (\cos^2 \theta + \nu \sin^2 \theta) \left( -\alpha - \lambda_b \frac{dL^2}{dT} \right) \right] \quad (44)$$

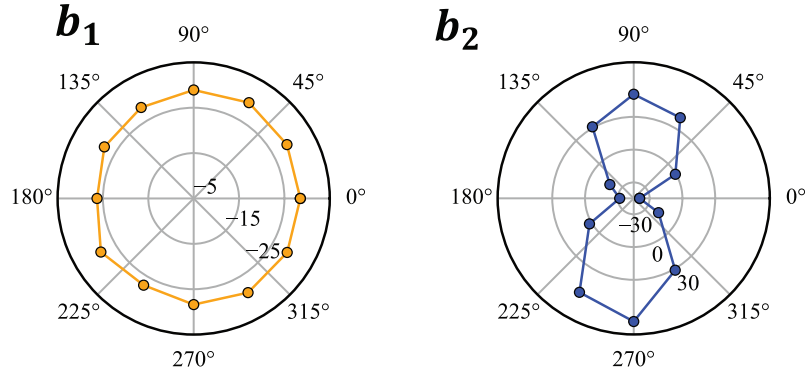
$$= \frac{-E}{4\rho w^2(1-\nu^2)} \left[ \alpha(1+\nu) + \left( \sin^2 \theta (\lambda_a + \nu \lambda_b) \frac{dL^2}{dT} + \cos^2 \theta (\lambda_b + \nu \lambda_a) \frac{dL^2}{dT} \right) \right]. \quad (45)$$

can be fitted to  $b_1 C_{\text{Debye}} + b_2 \frac{dL^2}{dT}$  where  $b_1$  and  $b_2$  are fit parameters, and  $\frac{dL^2}{dT}$  is estimated from Eq. 43. The results of these fits along to measured data of  $\frac{df_\theta^2}{dT}$  are shown in Fig. 10. Polar plot of the resulting  $b_1(\theta)$  and  $b_2(\theta)$  are shown in Fig. 11, which confirm that the thermal contribution to strain does not exhibit significant anisotropic behavior. From Eq. 42 and 40, the expected angle dependance of the parameter  $b_2$  is

$$b_2(\theta) = \left( \frac{E}{4\rho w^2(1-\nu^2)} \right)^2 (1-\nu)(\lambda_a - \lambda_b) [(\sin^2 \theta (\lambda_a + \nu \lambda_b) + \cos^2 \theta (\lambda_b + \nu \lambda_a))], \quad (46)$$

which we use to fit  $b_2(\theta)$  in Fig.11 to  $A \sin^2 \theta + B \cos^2 \theta$  where

$$\frac{A}{B} = \frac{\lambda_a + \nu \lambda_b}{\lambda_b + \nu \lambda_a}. \quad (47)$$

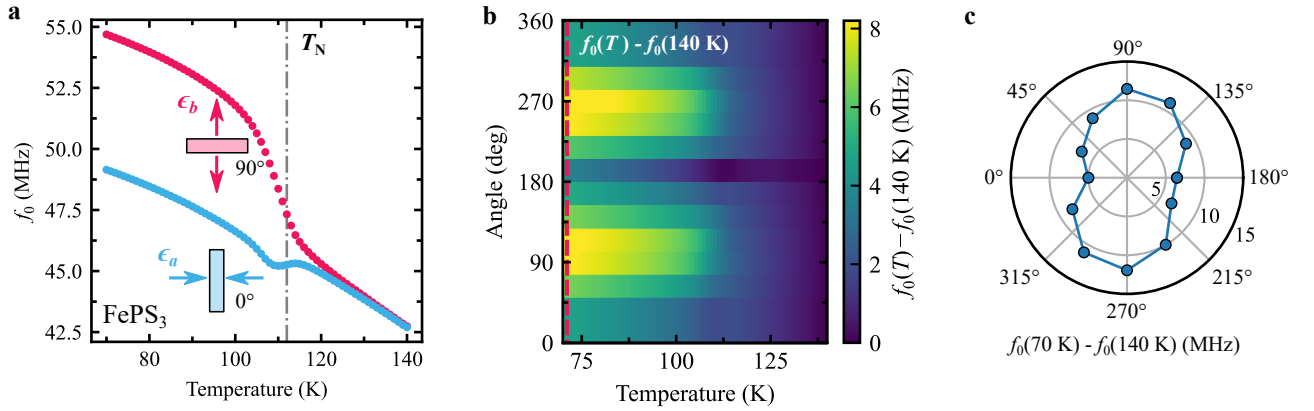


SUPPLEMENTARY FIG. 11. **Polar plot of fit parameters  $b_1$  and  $b_2$ :** Polar plot of fit parameters  $b_1$  and  $b_2$  from the fits to  $df^2/dT$  shown in Fig. 10.

The fit yields  $A/B = -2.062$  and  $-1.798$  for  $\text{CoPS}_3$  and  $A/B = -5.025$  and  $-8.695$  for  $\text{FePS}_3$ .

#### Supplementary Note 4. ANISOTROPIC RESONANCE FREQUENCY OF $\text{FePS}_3$ RESONATORS

Figure 12 shows resonance frequency data measured on  $\text{FePS}_3$  star-cavity resonators, as presented in Fig. 2 of the main text for  $\text{CoPS}_3$  and  $\text{NiPS}_3$  samples. Figure 12a shows the temperature dependence of the resonance frequency of membranes orientated along the  $a$ -axis (in red) and  $b$ -axis (in blue). Similarly to  $\text{CoPS}_3$ , the opposite strain along  $a$  and  $b$  arising from spontaneous magnetostriction results in opposite behaviour of the resonance frequency near the transition temperature  $T_N$ . The resulting anisotropic response is further illustrated in the map plot of resonance frequency as a function of temperature and angles in Fig. 12b and in the polar plot of  $f_{\text{res}}(T) - f_{\text{res}}(140 \text{ K})$  in Fig. 12c.

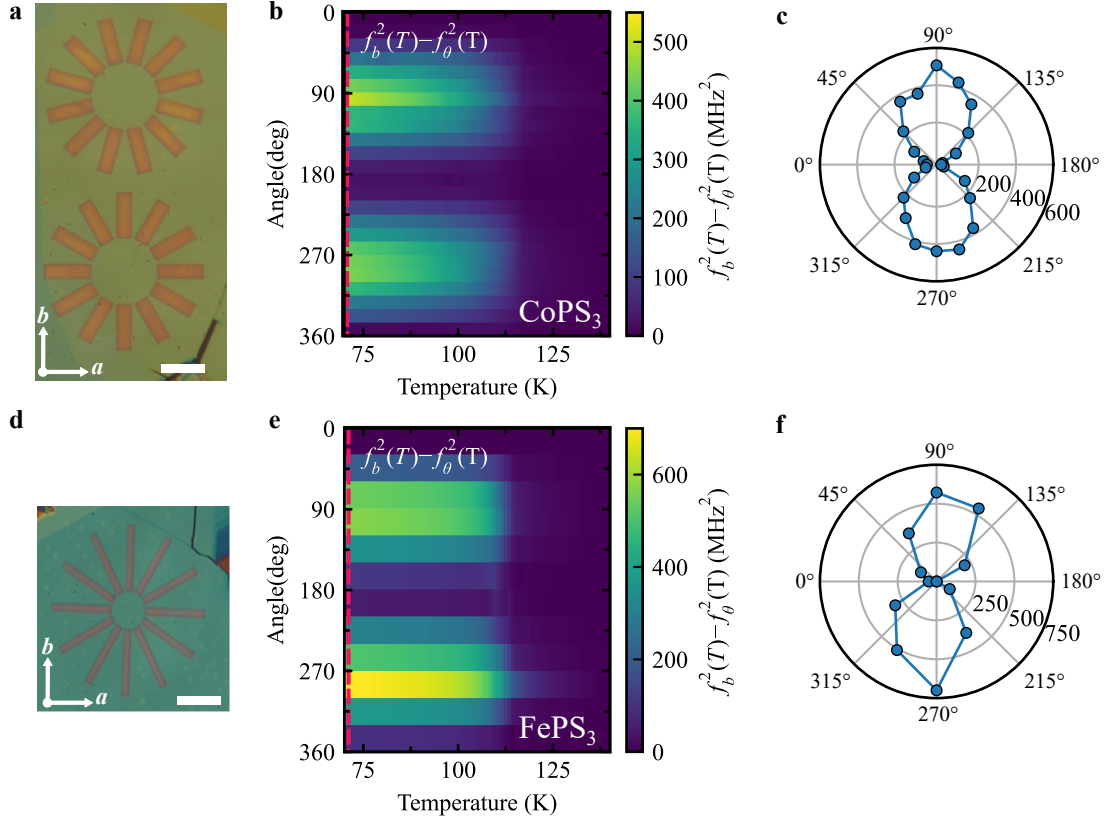


SUPPLEMENTARY FIG. 12. **Angle-resolved resonance frequency data of  $\text{FePS}_3$  membranes.** **a** Temperature dependence of  $f_{\text{res}}$  of a  $\text{FePS}_3$  rectangular membrane orientated at  $0^\circ$  (blue) and  $90^\circ$  (red) with respect to the  $b$  crystallographic axis. Note that  $f_{0,0^\circ}$  and  $f_{0,90^\circ}$  are proportional to the strain  $\epsilon_a$  and  $\epsilon_b$  respectively (see Eq. 2). The dashed-dotted grey line indicates the transition temperature  $T_N$ . **b** Resonance frequency difference,  $f_{\text{res}}(T) - f_{\text{res}}(140 \text{ K})$ , as a function of angle  $\theta$  with respect to  $b$ -axis and temperature. **c** Polar plot of  $f_{\text{res}}(T) - f_{\text{res}}(140 \text{ K})$  taken along the red dashed line in (b).

#### Supplementary Note 5. ORDER PARAMETER RELATED FREQUENCY DIFFERENCE $\tilde{f}_b^2 - \tilde{f}_\theta^2$

In the main text, we have shown how to relate the difference  $f_b^2 - f_a^2$  to the antiferromagnetic order parameter through the magnetostriction induced strain at the phase transition  $\epsilon_{\text{ms},aa} = \lambda_a L^2$  and  $\epsilon_{\text{ms},bb} = \lambda_b L^2$ . In general (see

derivation in Supplementary Note 2),  $\tilde{f}_b^2 - \tilde{f}_\theta^2$  is also proportional to  $L^2$ , where  $\tilde{f}_\theta^2$  is the pretension corrected resonance frequency of a rectangular cavity oriented at an angle  $\theta$  with respect to the  $b$ -axis. We show this quantity for the CoPS<sub>3</sub> and FePS<sub>3</sub> star-cavity resonators in Fig. 13a,d by plotting  $\tilde{f}_b^2 - \tilde{f}_\theta^2$  as a function of angle and temperature in Fig. 13b,e. Figure 13c,f shows the polar plot of  $\tilde{f}_b^2 - \tilde{f}_\theta^2$  taken along the red dashed line in 13b,e.



**SUPPLEMENTARY FIG. 13. Angle-resolved data of  $f_b^2 - f_\theta^2 \propto L^2$  of CoPS<sub>3</sub> and FePS<sub>3</sub> membranes.** **a,c** Optical image of the CoPS<sub>3</sub> (a) and FePS<sub>3</sub> (c) resonators. Scale bar 12  $\mu\text{m}$ . **b** Resonance frequency difference,  $f_b^2 - f_\theta^2 \propto L^2$ , as a function of angle  $\theta$  and temperature of the CoPS<sub>3</sub> sample in (a). **c** Polar plot of  $f_b^2 - f_\theta^2$  taken along the red dashed line in (b). **e,f** follows the same structure as (b,c) for the FePS<sub>3</sub> sample in (c).

This behaviour is observed for the thicker samples ( $t > 10$  nm) and it is exploited to have a better estimate of the critical parameters  $\beta$  and  $T_N$  as discussed in Supplementary Note 6. For thinner resonators every irregularity, like wrinkles or tears, can strongly affect their mode shapes. In some cases, these imperfections can drastically change the resonance frequency of the fundamental mode, as well as its temperature dependence. Therefore, when analysing the critical behaviour of thin flakes, we choose only the most pristine and unaffected membranes out of all fabricated out of a single flake to be sure we are not affected by these irregularities.

### Supplementary Note 6. CRITICAL CURVE FIT

To extract critical parameters  $\beta$  and  $T_N$  shown in Fig. 3 and 4 of the main text, we fit the order parameter related difference  $f_b^2 - f_a^2 \propto L^2$  to the power law  $A_\theta(1 - T/T_N)^{2\beta}$ . The experimental determination of critical parameters is often debated due to the difficulty of extracting from one set of data, three strongly correlated parameters,  $\beta$ ,  $T_N$  and  $A_\theta$ . In addition, finite size effects are known to smear the transition which usually results in a non-zero tail of the order parameter in the disordered state and makes it harder to unambiguously determine the critical temperature. Also, the choice of the temperature interval for the fit is not universal and it is often arbitrary.

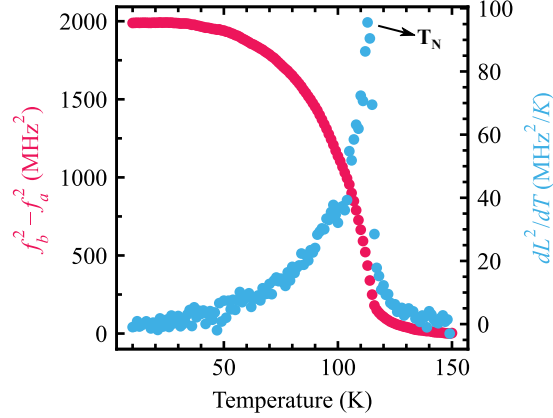
In order to have a better estimate of the critical exponents from our experiments, we compute  $f_b^2 - f_\theta^2$  for all  $\theta$  in a star and fit the data to  $A_\theta(1 - T/T_N)^{2\beta}$ . For each star, we then calculate the average value and standard deviation of the critical parameters  $T_N$  and  $\beta$  weighted by the error from the fit  $T_{N,\text{err}}$  and  $\beta_{\text{err}}$ .



We start with an initial guess,  $T_N^*$ , for  $T_N$  by extracting the maximum of the derivative of  $f_b^2 - f_a^2$  with respect to temperature as shown in Fig. 14. We then fit the  $A_\theta(1 - T/T_N)^{2\beta}$  to  $f_b^2 - f_a^2$  over the range  $[\alpha T_N^*, T_N^*]$ , for  $\alpha$  varying between  $[0.85, 0.95]$  allowing  $A, T_N, \beta$  to vary. We then define the total error for each  $\alpha$  to be  $T_{N,\text{err}} + \beta_{\text{err}}$ , where  $T_{N,\text{err}}, \beta_{\text{err}}$  are the standard deviation errors of the fit. We then take the extracted  $T_N, \beta$  with corresponding  $T_{N,\text{err}}, \beta_{\text{err}}$  to be ones given the fit corresponding to the  $\alpha$  minimizing the total error. We repeat this process for each  $\theta$  yielding a distribution of  $T_N, \beta$ . We then extract a weighted mean of this distribution as follows:

$$\bar{\beta} = \frac{1}{N} \sum_{\theta} \beta \frac{\beta_{\text{err},\text{min}}}{\beta_{\text{err}}} \quad (48)$$

Where  $N$  is the number of cavity pairs and  $\beta_{\text{err},\text{min}}$  the  $\beta_{\text{err}}$  of the pairing with smallest  $\beta_{\text{err}}$ . We then fit a normal distribution with  $\bar{\beta}$  as mean to the distribution of  $\beta$  where weigh each  $\beta$  by  $\frac{\beta_{\text{err},\text{min}}}{\beta_{\text{err}}}$ , from which we extract the standard deviation. Repeating this process for  $T_N$ . The resulting parameters for each sample are listed in Table 51.



SUPPLEMENTARY FIG. 14. **First estimate of  $T_N$  from the derivative of the order parameter.**

Material	Sample	$t$ (nm)	$\beta$	$T_N$ (K)
CoPS <sub>3</sub>	1	33	$0.28 \pm 0.017$	$115.7 \pm 0.38$
CoPS <sub>3</sub>	2	52	$0.311 \pm 0.025$	$117 \pm 0.35$
CoPS <sub>3</sub>	3	52	$0.298 \pm 0.034$	$116.1 \pm 0.18$
CoPS <sub>3</sub>	4	8	$0.195 \pm 0.0446$	$107.8 \pm 2.65$
CoPS <sub>3</sub>	5	8.6	$0.218 \pm 0.002$	$102.5 \pm 1.3$
FePS <sub>3</sub>	1	60	$0.208 \pm 0.0328$	$112.7 \pm 0.87$
FePS <sub>3</sub>	2	40	$0.203 \pm 0.03$	$109.1 \pm 0.37$
FePS <sub>3</sub>	3	10	$0.194 \pm 0.023$	$110.2 \pm 0.48$
FePS <sub>3</sub>	4	7	$0.206 \pm 0.047$	$107.9 \pm 0.76$
NiPS <sub>3</sub>	1	48	$0.218 \pm 0.016$	$150.7 \pm 0.7$

SUPPLEMENTARY TABLE 51. **Critical exponents of MPS<sub>3</sub> samples.** Critical exponents,  $\beta$  and  $T_N$ , for CoPS<sub>3</sub>, FePS<sub>3</sub> and NiPS<sub>3</sub> samples of different thicknesses, extracted following the procedure described in Supplementary Information Supplementary Note 6

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