Imaging the Néel vector switching in the monolayer antiferromagnet MnPSe$_3$ with strain-controlled Ising order

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Antiferromagnets are interesting materials for spintronics because of their faster dynamics and robustness against perturbations from magnetic fields. Control of the antiferromagnetic order constitutes an important step towards applications, but has been limited to bulk materials so far. Here, using spatially resolved second-harmonic generation, we show direct evidence of long-range antiferromagnetic order and Ising-type Néel vector switching in monolayer MnPSe$_3$ with large XY anisotropy. In addition to thermally induced switching, uniaxial strain can rotate the Néel vector, aligning it to a general in-plane direction irrespective of the crystal axes. A change of the universality class of the phase transition in the XY model under uniaxial strain causes this emergence of strain-controlled Ising order in the XY magnet MnPSe$_3$. Our discovery is a further ingredient for compact antiferromagnetic spintronic devices in the two-dimensional limit.

Detection and control of the spin order in ferromagnetic materials is the main principle in current information technology. Despite the great advantages of antiferromagnetic (AFM) spintronics, controlling and detecting Néel vectors have been limited in bulk materials$^{1,2}$. The discovery of two-dimensional (2D) ferromagnetic materials using the polar Kerr effect$^3$ has triggered tremendous interest in studying magnetism in the true 2D limit$^{4-11}$ and spintronic device applications in van der Waals heterostructures$^{12-18}$. Optical techniques are powerful tools to detect magnetism$^{19,20}$, but clear evidence for the AFM order in atomically thin 2D crystals has not been identified due to the lack of sensitive direct detection. For example, the polar Kerr effect is absent in AFM materials when the total magnetization is zero$^{19,15,20}$. And, although Raman spectroscopy is a powerful tool to study spin–photon coupling and collective magnons$^{21-24}$, their identification often does not provide unambiguous identification of the AFM order$^{19,20}$. Second-harmonic generation (SHG) has shown to be a sensitive tool to detect AFM orders due to inversion symmetry breaking from the spin order in magneto-electric materials including bulk Cr$_2$O$_3$ (ref. 25), few-layer MnPSe$_3$ (ref. 26) and a synthetic bilayer CrI$_3$ (ref. 27). Nevertheless, the detection of intrinsic AFM in the monolayer has not been demonstrated yet. In this work, we develop a sensitive SHG microscope and detect long-range Néel AFM order and Néel vector switching down to the monolayer in MnPSe$_3$ (ref. 28). Temperature-dependent SHG measurement in repetitive thermal cooling surprisingly collapses into two curves, which correspond to the switching of an Ising-type Néel vector reversed by the time-reversal operation, instead of the six-state clock ground state expected from the three-fold rotation symmetry in the structure$^{29-31}$. We image the spatial distribution of the Néel vectors across samples and rotate the Néel vectors by an arbitrary angle irrespective of the lattice in the sample plane by applying uniaxial strain. By studying both a Landau theory and a microscopic model that couples strain to nearest-neighbour exchange, we conclude that the phase transition of the XY model in the presence of strain falls into the Ising universality class instead of the XY class, which could explain the strain tunability. Finally, we find that the 180° AFM domain walls are highly mobile down to the monolayer after thermal cycles, paving the way for future control of the antiferromagnetic domains by strain or external fields on demand for ultra-compact 2D AFM terahertz spintronics.

Detecting AFM order and imaging AFM domains in thick flakes

MnPSe$_3$ belongs to the family of AFM transition metal phosphorous trichalcogenide MPX$_3$ (M=Mn, Ni, Fe, Co; X=S, Se), among which Mn compounds form inversion-breaking Néel order while others are in centrosymmetric zigzag ordered phases$^{32-36}$. In contrast to MnPSe$_3$, which has dominant out-of-plane moments$^{35,37}$, MnPSe$_3$ has in-plane spins with very large XY anisotropy according to the neutron scattering measurement$^{38}$ (Fig. 1a), which offers richer magnetic domain structures such as vortices and tunability$^{39}$. Above the Néel temperature, $T_N$, MnPSe$_3$ belongs to the point group 3 (S$_3$) and space group 148 (ref. 28), and has an inversion centre between two neighbouring Mn atoms but no mirror symmetry. The Mn atoms form a honeycomb lattice in one layer (Fig. 1a), and the honeycomb layers form the rhombohedral (ABC) stacking along the c axis with a three-fold rotational symmetry. Different from FeP$_3$ and NiP$_3$, which have a change from a two-fold rotational symmetry in the bulk to a three-fold rotational symmetry in the monolayer$^{21-23,38}$, MnPSe$_3$ is always three-fold symmetric.

As shown in the inset of Fig. 1b, a small temperature-independent SHG from the electric quadruple (EQ) contribution, $P_i^{(2)} (2\omega) \propto |\sum_{ijkl}x_{ij}^{EQ}E_i (\omega) \nabla_k E_j (\omega)|^2$ follows the lattice three-fold

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rotational symmetry above $T_N$, where $E$ is the electric field, $\omega$ is the fundamental frequency, $\chi$ is the susceptibility and $i,j,k,l$ are Cartesian indices (Methods). Below the Néel temperature, the formation of the Néel AFM order with in-plane spins breaks the inversion symmetry ($P$), which allows an electric dipole (ED) contribution to the SHG, $I_{ij}^{\text{ED}}(2\omega) \propto \sum_{k} \langle k|L\rangle E_{k}(\omega) E_{k}(\omega)^{\ast}$. $\chi_{ijkl}^{\text{ED}}(L)$ is proportional to the order parameter, the Néel vector $L$ (ref. 28), and changes sign when $L$ flips by $180^\circ$ (L = $M_{i} - M_{i}$, where $M_{i}$ and $M_{i}$ are the magnetization of two neighbouring Mn atoms; Fig. 1a). In the AFM state, the product of the inversion symmetry ($P$) and the time-reversal symmetry ($T$), the so-called $P'T'$ symmetry, is still preserved40, even though both $P$ and $T$ are broken. This kind of ED term is often called non-reciprocal or c-type SHG allowed by the $P'T'$ symmetry, while the EQ term is an i-type SHG, where ‘c’ and ‘i’ mean changing and invariant under time-reversal symmetry, respectively28. Figure 1b shows a typical SHG response as a function of temperature on an ~100-nm-thick film exfoliated on SiO2/Si. A sharp turn-on of the ED SHG signal clearly indicates a phase transition at 67.9±0.2 K, agreeing well with the $T_N$ (68±0.5 K) determined from in-plane magnetization measurement. Below $T_N$, as shown in Fig. 1c, a giant two-fold signal emerges in the crossed configuration, which clearly breaks the three-fold rotation symmetry. Another surprising observation is that the peak of the parallel polar pattern is only 1/20 of that in the crossed pattern, which was not observed in previous $P'T'$ invariant van der Waals AFM materials26,27. The nodal direction of the two-fold crossed pattern is also shown to be close to the Néel vector direction (Supplementary Notes 1 and 2).

After confirming the detection of AFM order by SHG, we use scanning SHG microscopy with 2 μm spatial resolution to study the AFM domains. In MnPSe3, with in-plane spins and large XY anisotropy28,39, six energetically equal magnetic domains are expected due to the three-fold rotational crystalline anisotropy and the time-reversal operation29-31. Nevertheless, Fig. 2a shows that temperature-dependent SHG intensity under ten consecutive cooling runs across $T_N$ on the same spot of a 15-μm-thick sample collapses into two curves instead of six. The crossed and parallel polar patterns that respond to these two AFM domains are shown in Fig. 2b,c. To figure out the relation between these two domains, we performed spatial scanning SHG microscopy at 5 K with the angles of the two polarizers chosen near the maximum signal in the crossed pattern. An optical image of a region with uniform thickness (~100 nm, a second exfoliated thick sample) and SHG maps at 5 K after two cooling processes across $T_N$ are shown in Fig. 2d–f. Sharp dark lines with very low SHG intensity are observed, with bright domains of high and nearly equal SHG intensity on both sides. In one of the regions, we pick out a few points such as the blue dot in Fig. 2e and observe the same polar patterns shown as blue in the top right of Fig. 2e. Crossing the dark line to a different region, we pick out a few points such as the orange dot and observe the crossed polar patterns rotated by a small angle shown as orange on the top right of Fig. 2e. By keeping the laser spot at the orange point and performing a few thermal cycles, the polar pattern switches only between the blue and orange dots shown in Fig. 2e.

We interpret the two regions with high SHG intensity as different AFM domains where the spins are reversely by 180° under the time-reversal operation, and the dark lines are domain walls due to destructive interference34,35). The arrows in Fig. 2c–f indicate the opposite directions of the Néel vectors in different regions determined by SHG polar pattern measurements at 5 K (Supplementary Note 1). A second SHG map after a thermal cycling across $T_N$ in Fig. 2f shows that the domain wall is not pinned and different regions still have only the two kinds of polar patterns shown in Fig. 2e. (The mapping on a 30-μm-thick naturally grown sample is shown in Supplementary Figure 6.) The reason why we could observe the domain wall between two AFM regions with a π phase shift by the destructive SHG interference is that the SHG has both ED and EQ contributions and only the ED term is sensitive to the π phase shift. One could write the signal we observe as $I_{ij}(2\omega) \propto \sum_{k} \langle k|L\rangle E_{k}(\omega) E_{k}(\omega)^{\ast} + \langle k|\chi_{ijkl}^{\text{EQ}}(L)\rangle E_{k}(\omega) E_{k}(\omega)^{\ast} \nabla_{k} E_{k}(\omega)$. where the ± signs indicate the sign change of the ED term under time-reversal operation. As shown in Figs. 1b and 2a, the ED contribution at 5 K is ~200 times larger than the EQ part, and therefore the two AFM domains have high and nearly equal SHG while the domain wall has very low SHG with the EQ contribution only.

**Detecting AFM order and imaging Néel vector switching down to the monolayer**

Before we discuss the origin of the two-state Ising order instead of the six-state clock order, we investigate whether the AFM order exists and whether direct imaging of Néel vector switching could be detected.
detected in the monolayer first. The ultra-thin flakes down to the monolayer are exfoliated on SiO2/Si wafers. The number of layers is determined by a combination of atomic force microscopy and optical contrast measurements4 (Extended Data Fig. 1). To probe the intrinsic properties of the materials, we exfoliate samples down to the monolayer in a glove box. Figure 3a shows the layer-dependent square root of the SHG intensity measured as a function of the temperature. All of the thin flakes show a clear phase transition down to the monolayer with the layer-dependent transition temperature shown in Fig. 3b. The transition temperature decreases from 66 K in the nine-layer sample to 56 K in the bilayer sample, and it is 40 K measured in three different monolayer samples (Extended Data Figs. 2 and 3 for the other two monolayer samples). The decrease of the transition temperature in atomically thin samples occurs because of the increasing fluctuation when approaching the 2D limit, which is expected for breaking the inversion symmetry in all of these samples4,5,44. This is different from synthetic layered AFM CrI3, which supports SHG signals only with even numbers of layers27.

Figure 3d shows an optical image of the monolayer MnPSe3 sample 1 (S1). We performed twelve thermal cycles at the green dot shown in Fig. 3d with the temperature-dependent SHG collapse on the thick sample. Light and dark green arrows show the direction nodes in the crossed polar pattern, which is close to the Néel vector direction. Scale bars, 50 μm. Inset: crossed patterns on two sides of a domain wall with positions marked by the orange and blue dots. The colour bar indicates the SHG intensity.

Demonstration of strain-controlled Ising order
We observed the same two-state Ising order on all of the samples with a thickness from a monolayer to ~30 μm on different substrates. Also, all of the samples show two-fold crossed polar patterns, regardless of whether they are prepared in air or in a glove box (Extended Data Figs. 2–6 and Supplementary Figs. 2, 4, 7 and 9). Since the flakes are exfoliated on SiO2, and the bulk micrometre-thick crystals are glued on the metal platform directly, a certain amount of strain is inevitable. Therefore, we hypothesized that the Ising anisotropy is induced by the strain. In order to verify it, we deliberately applied an ~2% uniaxial strain by exfoliating a 15 nm flake on the polymer polydimethylsiloxane (PDMS)45,46 and then stretched the PDMS as shown in Fig. 4a. The strain strength is determined by measuring the length of the optical image of the sample along the elongation direction shown in Fig. 4b before and after stretching (Supplementary Fig. 12 and Supplementary Note 4). The SHG mapping with ~2% strain along the x direction, shown in Fig. 4c, is quite homogeneous, and the crossed polar patterns at different positions all point along the same direction (Fig. 4d), which indicates that the strain aligns the Néel vector.

We further applied the strain along 0, 45, 90 and 135 degrees with respect to the x axis defined in Fig. 4b and found that the crossed polar pattern follows the rotation of the strain as shown in Fig. 4d–g within the experimental accuracy of ±10°, which indicates that the Néel vector is locked to the strain. We also demonstrated the Néel vector rotation by strain in a three-layer (3L) sample (Extended Data Fig. 7). Because PDMS is transparent and reduces colour contrast, the 3L sample is almost invisible on PDMS. Instead of direct straining of a monolayer on PDMS, we exfoliated a monolayer sample S3 with a long wavy shape on a SiO2/Si substrate to induce different strain directions in different regions, and find that Néel vector direction is also locked to the local strain and points to different directions in different regions (Extended Data Fig. 3).
We also find that the parallel polar patterns are different between monolayers S1 and S2. Fittings of the patterns indicate that the relative angles between the Néel vector and the crystal axis are different between samples S1 and S2, indicating that the strain directions in the two samples are different (Extended Data Fig. 2). The Néel vector could be rotated to any direction by the strain in atomically thin MnPSe₃ due to the strain-locked Ising order, which is drastically different from a non-XY system, where Néel vectors are switched between principal crystal axes only. We also noticed that the strain-induced anisotropy is much smaller than the large XY anisotropy in this system (Extended Data Fig. 8, Supplementary Fig. 13).

**Origin of the Ising order in an XY magnet under strain**

In order to understand why strain leads to an Ising order instead of a six-state clock model, we first employ a Landau expansion for free energy as a function of the Néel vector L, which applies close to the critical temperature when L is small. The lowest critical order that accounts for the three-fold crystalline anisotropy, along with \( P^T \) symmetry, is \( L^0 \cos 6\theta \), where \( \theta \) is the polar angle of the Néel vector measured from the a axis in Fig. 3a, and \( L \) is the magnitude of the Néel vector. The strain is described by a second-rank tensor whose principal axes designate the directions of tensile and compressive strain. It can be diagonalized with a rotation about the c axis by an angle \( \theta_0 \), where \( \theta_0 \) is along the principal strain axis, and it provides the term \( u_e L_0^2 \cos 2(\theta - \theta_0) \). Therefore, the \( \theta \) dependent terms in the Landau free energy then take the form,

\[
F(\theta) = bu_e L_0^2 \cos 2(\theta - \theta_0) + cL_0^2 \cos 6\theta \tag{1}
\]

where \( b \) and \( c \) are coefficients, \( u_e \) is the amplitude of the strain and \( L_0 \) is the magnitude of the Néel vector at the minimum free energy (Supplementary Note 5). As shown in Fig. 5, for \( u_e = 0 \), in the absence of strain, this describes a six-state clock model, whose critical behaviour is expected to be in the XY universality class. For \( u_e \neq 0 \), the Ising anisotropy dominates, and the critical behaviour is in the Ising universality class²⁹,³⁰. In general, the direction of the Néel vector will be determined by the competition between the strain and the crystalline anisotropy (Supplementary Note 5–6). This model explains why an Ising order is also induced in the ~15-μm-thick bulk sample mounted by glue, as the strain is quite small.

These conclusions also follow from a more microscopic model of spins on a honeycomb lattice with interactions that reflect the symmetries of the crystal. We consider an XY model with nearest-neighbour couplings that are modified by strain and have the form,

\[
H = J_1 |s_i| \sum_j s_{i,j} + J_{\perp} \sum_i s_{i,\perp} + J_2 \sum_i (s_i| s_{i,\perp} + s_{i,\perp} s_{i,\perp}) \tag{2}
\]
Fig. 4 | Strain-tunable Néel vector in MnPSe₃. a, A schematic of the method to control the in-plane strain. A MnPSe₃ flake is first exfoliated on a PDMS substrate. A tunable stretching force is then applied by a micro-manipulator on the PDMS to transfer strain to the sample, which is maintained by attaching the PDMS to a gold-coated sample holder. b, An optical image of a stretched 15-nm-thick MnPSe₃ flake with a uniaxial strain ~2% along the x axis. Scale bar, 10 µm. c, SHG spatial mapping with a 2% strain applied along the x axis at nominally 5 K. Scale bar, 10 µm. d–g, Crossed polar patterns with strain direction along 0° (d), 45° (e), 90° (f) and 135° (g) with respect to the x axis. The red arrow represents the strain direction. The measured data are shown in blue open circles, with fits shown in blue lines.

Fig. 5 | Renormalization group flow of the Landau theory. When μₓ = 0, the system flows into the three-dimensional (3D) XY critical point and develops the six-state clock order at low temperature. When μₓ ≠ 0, the system flows into the 3D Ising critical point and develops the two-state Ising order at low temperature.

where we distinguish the coupling for spin polarizations along (J₁₂) and perpendicular to (J₁₃) the ij-th Mn–Mn bond; Jᵥ is a symmetry-allowed cross coupling between orthogonal spin components sₓ₁₂ and sₓ₁₃ in each bond. The total energy is proportional to \( E = J_x \cos(2(\theta - \theta_0 - \alpha/2)) \), where \( \alpha/2 \) defines a misalignment angle of the spin orientation from the principal strain axis. As long as the response is linear, decreasing the amount of strain while keeping the same direction will not change the tipping angle (Supplementary Note 6 for more details).

Conclusions
The direct imaging of Néel vector switching in the monolayer goes beyond previous works of domain imaging in bulk AFM samples²⁻²³ and opens the possibility of ultra-compact AFM spintronics. Our work also creates a method to control the 2D antiferromagnetism other than electric gating¹⁵,¹⁶ and magnetic fields¹⁷,⁴⁸. The monolayer MnPSe₃ also provides a platform to study the Kosterlitz–Thouless transition in a truly 2D XY magnet with six-state clock order if the strain could be tuned to zero by a voltage-controlled piezo-stage⁴⁹. Additionally, our imaging and strain-tuning methods are generally applicable to other van der Waals AFM materials including the intrinsic AFM topological insulator⁵⁰. Looking forward, we hope that the discovery of strain-tunable AFM order in atomically thin MnPSe₃ will stimulate further investigations in designing spatial strain distribution to create spin patterns on demand for magnon propagation in low-dissipation terahertz spintronic devices.

Online content
Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of authorship and code availability are available at https://doi.org/10.1038/s41565-021-00885-5.

Received: 6 December 2020; Accepted: 2 March 2021; Published online: 19 April 2021

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Methods

Sample preparation. Single crystals of MnPSe₃ were grown by the chemical vapour transport method. Elemental powders of high purity Mn, P and Se were pressed into a pellet and sealed inside a quartz tube under vacuum. The tube was then annealed for a week at 730°C to form polycrystalline MnPSe₃ powder, the composition of which was verified with powder X-ray diffraction. Crystals were then grown using the chemical vapour transport method with iodine as the transport agent: 2 g of the powder and 0.4 g of iodine crystals were placed at the end of a quartz tube, which was sealed off at 13 cm length under vacuum. The sealed tube was then set in a temperature gradient of 650/525°C for four days to transport the starting materials placed at the hot end to the cold end. The Mn/P/Se ratio was measured to be 1.00(1):0.96(1):3.07(1) with energy-dispersive X-ray spectroscopy. The ultra-thin samples were prepared by a standard mechanical exfoliation process on Si substrates with 90-nm-thick SiO₂ from a few MnPSe₃ bulk crystals. Due to the small interlayer van der Waals coupling, one could easily exfoliate MnPSe₃ crystal down to atomically thin layers. The total exposure time in air for samples exfoliated in a glove box was less than one minute before the samples were loaded into the cryostat in vacuum. The samples studied in this work were as follows: (1) 15-μm-thick and 30-μm-thick bulk crystals mounted on the metal platform of the cryostats by glue; (2) one ~50-nm-thick flake and four ~100-nm-thick flakes exfoliated on SiO₂/Si in air; (3) an 8L sample and at least three samples of each thickness from monolayer to five-layer exfoliated on SiO₂/Si in air; (4) a 9L sample and at least three samples of each thickness from monolayer to four-layer exfoliated on SiO₂/Si in a glove box; and (5) a 3L sample, an 8L sample and more than five thick (10–20 nm) samples exfoliated on PDMS for the strain tuning experiment. The data on a 10-nm-thick sample and a 15-nm-thick sample exfoliated on PDMS are shown in the paper.

SHG microscopy. The sample was loaded on a metal platform in a closed-cycle cryostat, and the temperature of the metal platform was controlled by a local heater, which induces only a submicrometre shift of the sample position between 5 K and 100 K. An ultrafast 800 nm Ti–sapphire laser pulse with a duration of ~50 fs at a repetition rate of 80 MHz was focused onto a 2 μm beam spot on the sample by a x50 objective under normal incidence. A typical laser power of 200 μW was used except for the following cases: 500 μW was used for thick flakes and bulk crystals. In the bilayer sample in Extended Data Fig. 6, 400 μW was used. No sample damage was observed during the measurement. The reflected SHG light was collected by a dichroic mirror into a photomultiplier tube connected with a lock-in amplifier or a photon counter. The photon counter is locked to 80 MHz in order to reduce the dark count below 0.2 c.p.s. without cooling the photomultiplier by cryogen. The detection sensitivity in the experiments is 0.2 c.p.s. Because the parallel signal is one order of magnitude smaller than the crossed signal, the polarization extinction ratio is important when measuring parallel patterns. The polarization of the fundamental light is controlled by a half-wave plate as well as a linear polarizer. The polarization of second-harmonic light is analysed by a linear polarizer. The SHG imaging microscopy is achieved by moving the sample with three Attocube nano-positioners.

Strain tuning. We exfoliated MnPSe₃ on PDMS with a square shape and applied tensile strain on two sides of the PDMS by a micro-manipulator. The stretched PDMS was then attached to a gold-coated sample platform. The strain was estimated by measuring the length change along the stretching direction in the optical image. A low transfer ratio (~13%) from PDMS to the sample was observed. To change the sample’s strain direction, we peeled off the PDMS from the sample platform and then stretched it in another direction. To apply strain along the 45° and 135° directions, we cut the four corners of the PDMS to form a smaller square shape in order to reduce twisting while applying strain. The error bar of the estimation of the sample’s strain strength was ±10°. The error bar of the strain direction, we cut the four corners of the PDMS to form a smaller square shape in order to reduce twisting while applying strain. The error bar of the estimation of the sample’s strain strength was ±10°.

Symmetry analysis for SHG polar patterns. For the SHG patterns above Tₜₙ, the angle dependencies are described by,

\[ I_{\text{parallel}}(2\omega, \phi) \propto \left| J_{\text{even}}^{\text{E}\times\text{E}} \cos 3\phi + J_{\text{odd}}^{\text{E}\times\text{E}} \sin 3\phi \right|^2, \quad (\text{a}) \]

\[ I_{\text{crossed}}(2\omega, \phi) \propto \left| J_{\text{even}}^{\text{Q}\times\text{Q}} \sin 3\phi - J_{\text{odd}}^{\text{Q}\times\text{Q}} \cos 3\phi \right|^2. \quad (\text{b}) \]

Here, \( \phi \) is the angle of the incident linear polarization with respect to the \( a \) axis of the crystal. Note that in the fit, there is also a constant angle shift in \( \phi \), which is the angle between the horizontal axis in the lab and the crystal \( a \) axis. For the SHG patterns below \( T_{\text{tn}} \), we fit the crossed polar pattern by,

\[ I_{\text{crossed}}(2\omega, \phi, \theta) \propto L^2 \sin^2(\phi - \theta)^2. \quad (\text{c}) \]

and denote the node direction in the polar pattern as the Néel vector direction. Here, \( \theta \) is the angle of the direction of the Néel vector with respect to the crystal \( a \) axis. Supplementary Note 1 contains more details.

Data availability

All data needed to evaluate the conclusions in the paper are present in the paper and the Supplementary Information. Additional data related to this paper could be requested from the authors.

Acknowledgements

We thank S. W. Cheong and O. Tchernyshyov for helpful discussions. The project design, data collection and analysis, and preparation of the manuscript are supported by L.W’s startup package at the University of Pennsylvania. The development of the SHG photon counter is supported by the ARO YIP award under grant W911NF1910342 to L.W. The measurement by atomic force microscopy is supported by the ARO MURI under grant W911NF2020166 to L.W. The acquisition of the oscillator laser for the SHG experiment is supported by the National Science Foundation through Penn MRSEC (DMR-1720530). E.I.M. acknowledges support from National Science Foundation EAGER 1838456. C.L.K is supported by a Simons Investigator grant from the Simons Foundation. D.G.M acknowledges support from the Gordon and Betty Moore Foundation’s EPiQS Initiative, grant GBMF9069. H.W. and X.Q. acknowledge support from National Science Foundation DMR-1753054 and the Texas A&M University President’s Excellence Fund X-Grants Program. B.X. and C.B. are supported by the Schweizerische Nationalfonds by grant no. 200020-172611. The density functional theory calculations were conducted with the advanced computing resources provided by Texas A&M High Performance Research Computing.

Author contributions

L.W. conceived the project and coordinated the experiments and theoretical work. L.W. designed the SHG imaging set-up and built it with Z.N.; Z.N. performed the experiments and analysed the data under the supervision of L.W.; L.W., Z.N., E.I.M. and C.L.K. discussed and interpreted the data. E.I.M. performed the spin model calculation. C.L.K. performed the Landau theory calculation. A.V.H. and D.G.M. grew the crystals and performed the magnetization measurements. H.W. and X.Q. performed the first-principles calculation. B.X. and C.B. performed the optical conductivity measurement. L.W. and Z.N. wrote the manuscript with the input of all authors. All authors edited the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

Extended data is available for this paper at https://doi.org/10.1038/s41565-021-00885-5.

Supplementary information

The online version contains supplementary material available at https://doi.org/10.1038/s41565-021-00885-5.

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Peer review information

Nature Nanotechnology thanks the anonymous reviewers for their contribution to the peer review of this work.

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Extended Data Fig. 1  | Thickness characterization of atomically thin MnPSe$_3$ samples. a, Optical image of a multilayer MnPSe$_3$ exfoliated on the SiO$_2$/Si substrate. Scale bar: 10 $\mu$m. b, Atomic force microscopy image of the same sample in a. The step between monolayer and bilayer is around 0.78 nm. Scale bar: 10 $\mu$m. c, Optical contrast of samples with different layer numbers. Green circles are data extracted from sample shown in a and orange circles are data from other samples. The black line is a linear fit.
Extended Data Fig. 2 | SHG data for monolayer S2 exfoliated on SiO/$\text{Si}$ in a glove box. **a**, Optical image of the monolayer S2. There are some small bilayer/trilayer islands inside the monolayer sample. Scale bar: 10 μm. The dashed region is the area for SHG mapping. **b**, SHG intensity mapping of the monolayer S2 at 5 K. **c**, Temperature dependence of SHG intensity measured at the green point marked in **a** for one thermal cycle. **d–e**, Polarization-dependent SHG patterns measured at the green point in (d) crossed and (e) parallel configuration after two different thermal cycles and at 5 K. Data from both domains are shown. The dots are experimental data and the solid lines are the best fit. The patterns are different from the monolayer S1, indicating the angles between the Néel vector and the crystalline axis are different in the two samples.
Extended Data Fig. 3 | SHG data for monolayer S3 exfoliated on SiO$_2$/Si in a glove box.  
a, Optical image of the monolayer S3. Scale bar: 10 μm. The dashed box denotes the region of SHG mapping. b, SHG intensity mapping of the monolayer S3. Scale bar: 10 μm.  
c–d, Polar patterns at the two points denoted by purple and green dots are measured after thermal cycles. c, Crossed patterns of two domains of the purple point measured at 5 K after two different thermal cycles. d, Crossed patterns of two domains of the green point measured at 5 K after two different thermal cycles. Different crossed polar patterns also with different orientations at the purple and the green points indicate that their Néel vectors have different directions. e, Temperature dependence of SHG intensity at the purple point for one thermal cycle.  
f, Temperature dependence of SHG intensity at the green point for one thermal cycle.
Extended Data Fig. 4 | More data of the monolayer S1 shown in the main text. a, Optical image of the sample S1. Scale bar: 10 μm. The dashed region is the area for SHG mapping. b, SHG intensity mapping at $\phi = 120^\circ$ (the peak of domain 1) in the crossed pattern. c, SHG intensity mapping at $\phi = 160^\circ$ (the peak of domain 2) in the crossed pattern. Maps in b, c are after the same thermal cycle. The domain walls are highlighted by the green dashed lines. d–i, Crossed patterns of six different points marked by different colors in b after the same thermal cycle. P1–P5 are the same points in Fig. 3f in the main text. P6 is very close to sample corner and displays smaller SHG intensity with a slightly different orientation, which indicates the Néel vector at P6 has a slightly different direction. All the SHG measurements are performed at 5 K.
Extended Data Fig. 5 | SHG data of atomically thin samples exfoliated on SiO₂/Si in air. a, Temperature dependence of square root of the SHG intensity in samples with different thickness. All of the samples in this figure are exfoliated in air. b, SHG intensity at 5 K as a function of layer numbers in a log-log plot. Data are shown in yellow dots. The solid line is a fit for $I \propto N^2$. c, Néel temperature as a function of layer numbers. Note that there is a large reduction of both SHG signal and Néel temperature of monolayer compared to those exfoliated in the glove box due to the aging effect. See more data on the aging effect on a bilayer sample in Supplementary Figure 10. d, SHG intensity mapping of a monolayer MnPSe₃ exfoliated in air at 5 K. Scale bar: 3 μm. Inset: Optical image of the monolayer sample. f, Crossed polar pattern measured at the center of the monolayer sample shown in d at 5 K.
Extended Data Fig. 6 | Néel vector switching of a bilayer sample exfoliated on SiO₂/Si in air. a, Optical image of the bilayer sample. Scale bar: 10 μm. b, SHG intensity mapping of the bilayer sample at 5 K. Note that the SHG intensity of this sample is quite uniform, which indicates that the Néel vector direction is nearly aligned. c, SHG intensity of 6 consecutive cooling runs across Tₘ. The curves collapse into two, indicating the existence of two domains. d, Crossed and e, parallel polar patterns measured after each cooling at 5 K. The measured data are shown in dots. The blue and red shaded regions are guides for the eye, corresponding to the two different AFM domains.
Extended Data Fig. 7 | Strain tunability of Néel vector of a trilayer sample on a PDMS substrate. The strained sample is prepared using the same method as in the main text. The measured thickness is around 3.2 nm, which is a typical thickness for a trilayer sample. The measured Néel temperature is also consistent with a typical trilayer sample. 

(a), Optical image of the sample exfoliated on the PDMS before adding strains. The trilayer sample is marked by the black box. Scale bar: 10 μm. 

(b–c), Optical images of the sample when vertical and horizontal strain are added (marked by the red arrows), respectively. The sample and PDMS are mounted on the metal platform and the contrast of the sample is low. A 5% strain is added on the PDMS by a micro-manipulator. The directions of the cracks of the ~60-nm thick flake on the top also indicate the strain directions. 

(d), SHG intensity mapping of the dashed area in (a) under different strain directions (marked by the red arrows) and φ in the crossed pattern. The dark intensity maps in (d) indicate that the Néel vectors are mainly along the strain direction, and rotated by around 90° when the strain direction is switched from vertical to horizontal. 

(e, f), Crossed pattern at the center of the trilayer sample with vertical strain direction. All of the above SHG measurements are operated at 5 K.
Extended Data Fig. 8 | Strain dependence on the Néel temperature of an 8-L sample on a PDMS substrate. The MnPSe$_3$ samples in the strain-tuning experiment (Fig. 4 in the main text, extended Fig. 7 and supplementary Fig. 13) are exfoliated on a ~30 μm PDMS, which is too thick to have the best thermal conductance. One needs to calibrate the sample temperature by measuring a thick (> 50 nm) flake on the same PDMS each time. To measure the sample temperature without calibration, we use thinner home-made (~ 5 μm) PDMS to increase the thermal conductance and encapsulate the sample after exfoliation and attach the ~ 5 μm thick PDMS to the metal platform after applying strain (shown in a). b, Optical image of an as-exfoliated sample (~ 8 L) on the PDMS substrate before being put onto the metal platform. Scale bar: 10 μm. c, Optical image of the same sample under a horizontal strain. A 5% strain on PDMS is added by a micromanipulator and then the PDMS is attached to the metal platform. Scale bar: 10 μm. d, SHG intensity mappings of the as-exfoliated and strained sample measured at $\phi = 0^\circ$ and $\phi = 90^\circ$ in the crossed polar pattern. The as-exfoliated sample shows that the Néel vectors are not oriented horizontally while the strained sample favors a horizontal Néel vector orientation. The data is collected at 5 K. e, f, Temperature dependence of SHG intensity of the (e) as-exfoliated and the (f) strained sample. The difference of the Néel temperature is within 1 K.