

Supplementary Information

Highly confined magnetophononic polaritons and electron-phonon interactions in atomically thin MnBi_2Te_4

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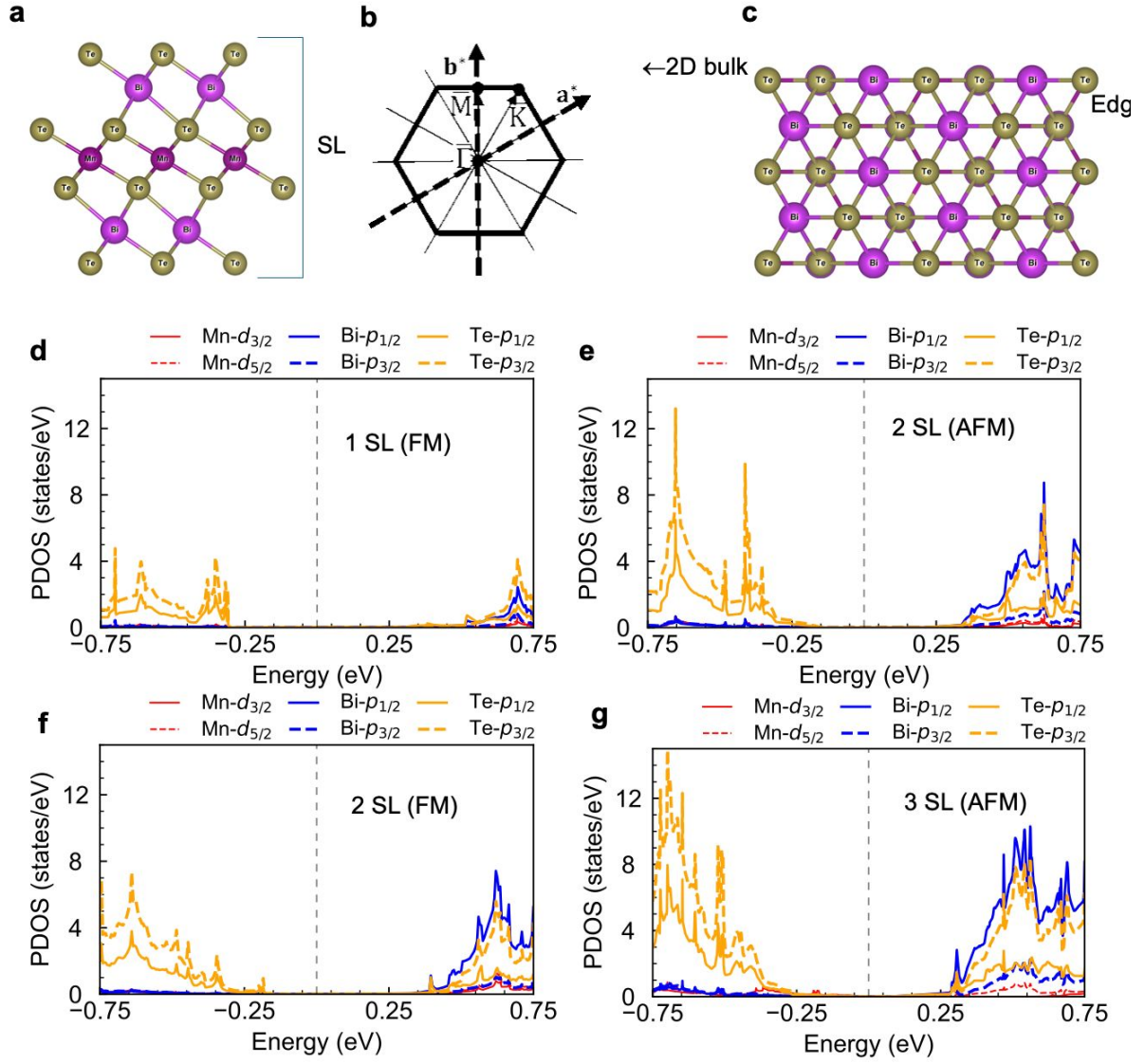


Figure S1. Septuple layer (SL) structure of MnBi_2Te_4 , its 2D Brillouin zone, 1D zigzag nanoribbon structure, and orbital decomposed density of states.

(a) Single septuple layer structure of MnBi_2Te_4 with ferromagnetic configuration. (b) Two dimensional (2D) Brillouin zone showing high symmetry points. (c) Zigzag nanoribbon (1D) of single septuple layer MnBi_2Te_4 . Calculated orbital decomposed density of states using fully relativistic effect of (d) 1 SL (FM), (e) 2 SL (AFM), (f) 2 SL (FM), and (g) 3 SL (AFM). The dashed lines (grey coloured) at zero energy indicate the Fermi level.

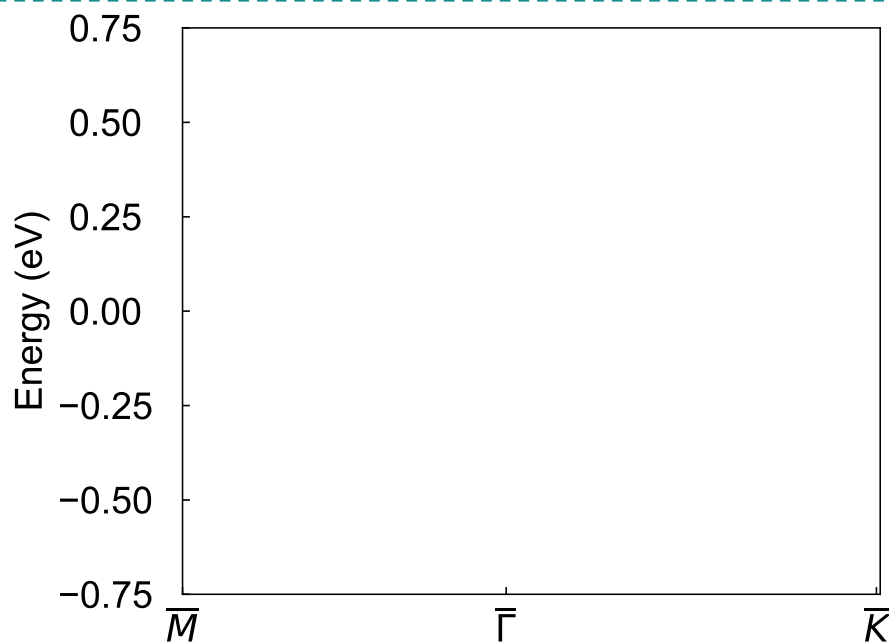


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Calculated electronic band structure of 1 SL by using standard DFT (blue), and 2D electron-phonon Wannier interpolation (red). We included Hubbard corrections in these calculations.

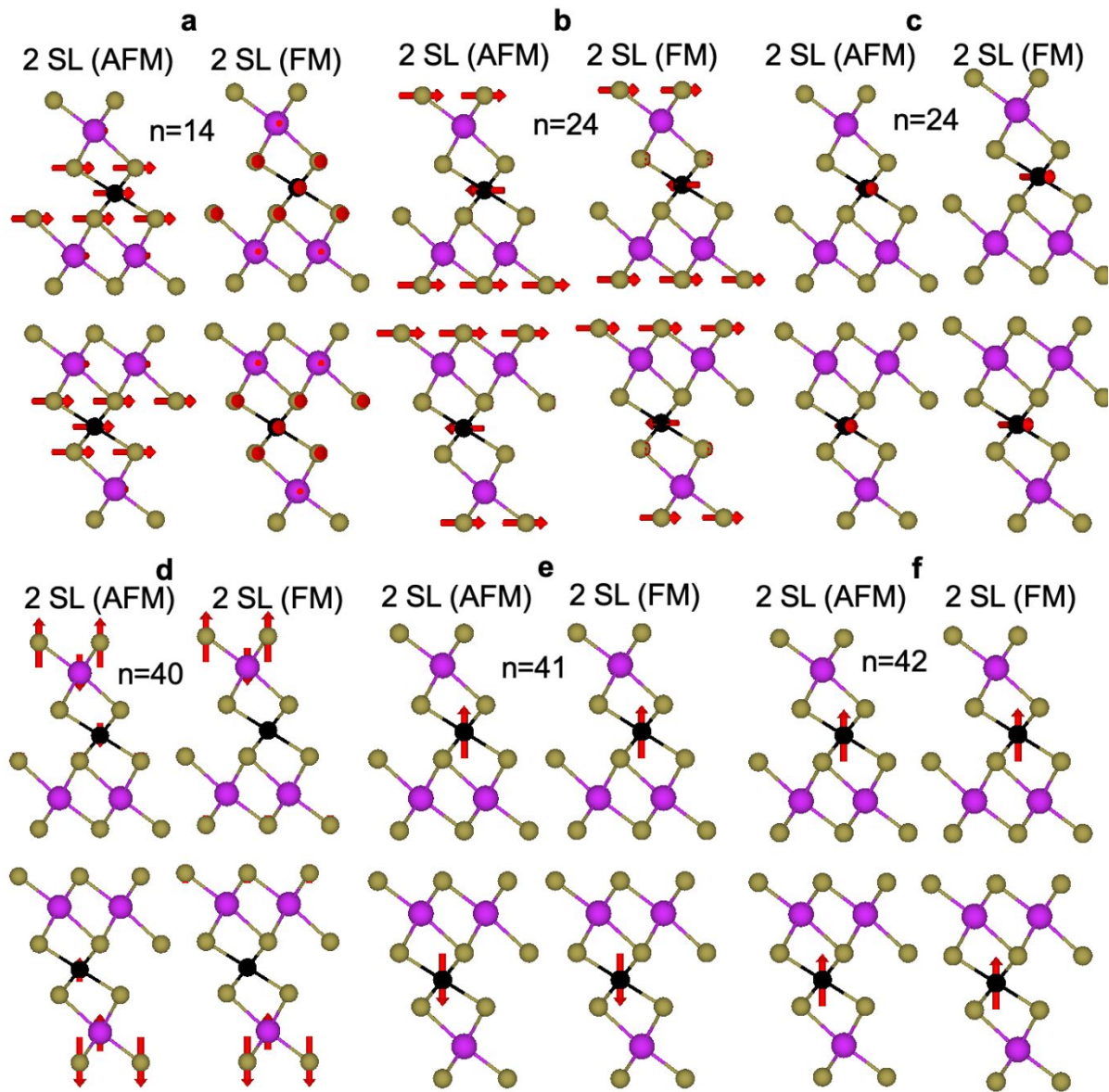


Figure S3. Vibrational visualization of six modes of 2 SL MnBi_2Te_4 in the AFM and FM configurations.

(a-c) Visualization of in-plane magnetophononic polaritonic vibrations at Γ -point in 2 SL MnBi_2Te_4 in AFM and artificial FM configurations. Two nearby columns correspond to the same phonon mode, shown for the AFM (left column) and AFM (right column) phases. Arrow lengths are scaled identically (based on the magnitude of the eigenvectors) in all panels. (d-e) Last three out-of-plane (first two are Raman modes, shown in (d-e), and third one is the IR mode ($A_{g,n}$)) mode vibrations at Γ -point in 2 SL MnBi_2Te_4 in AFM artificial FM configurations.

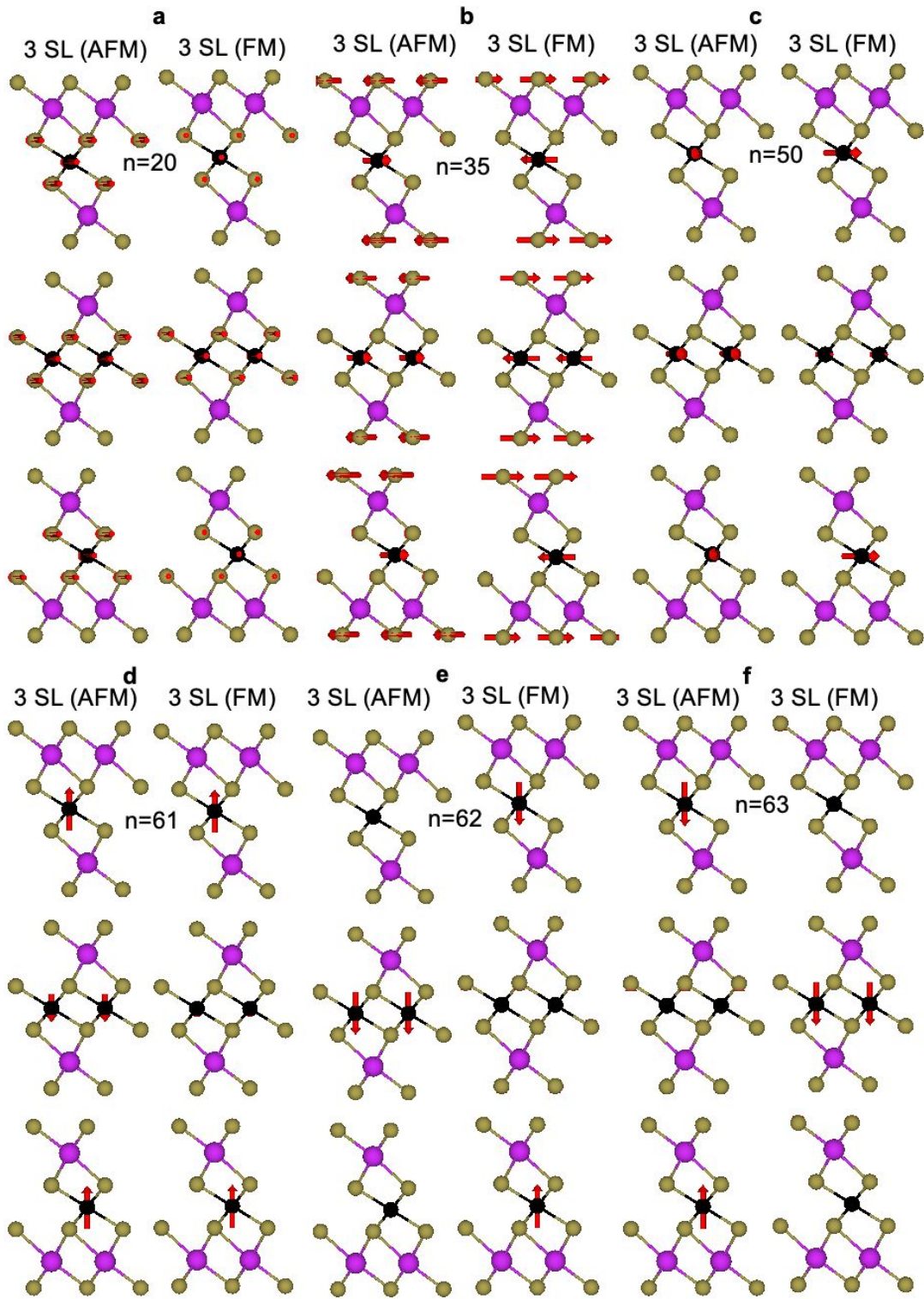


Figure S4. Vibrational visualization of six modes of 3 SL MnBi_2Te_4 in the AFM and FM configurations.

(a-c) Visualization of in-plane magnetophononic polaritonic vibrations at Γ -point in 3 SL MnBi_2Te_4 in AFM and artificial FM configurations. Two nearby columns correspond to the same phonon mode, shown for the AFM (left column) and FM (right column) phases. Arrow lengths are scaled identically (based on the magnitude of the eigenvectors) in all panels. (d-e) Last three out-of-plane (the third one is the Raman active mode and the first two of them are IR (A_{2u}) modes in AFM, while the first and third modes are IR (A_{2u}) modes and the second one is the Raman active mode in FM, shown in (d-e) at Γ -point in 3 SL MnBi_2Te_4 in AFM and artificial FM configurations.

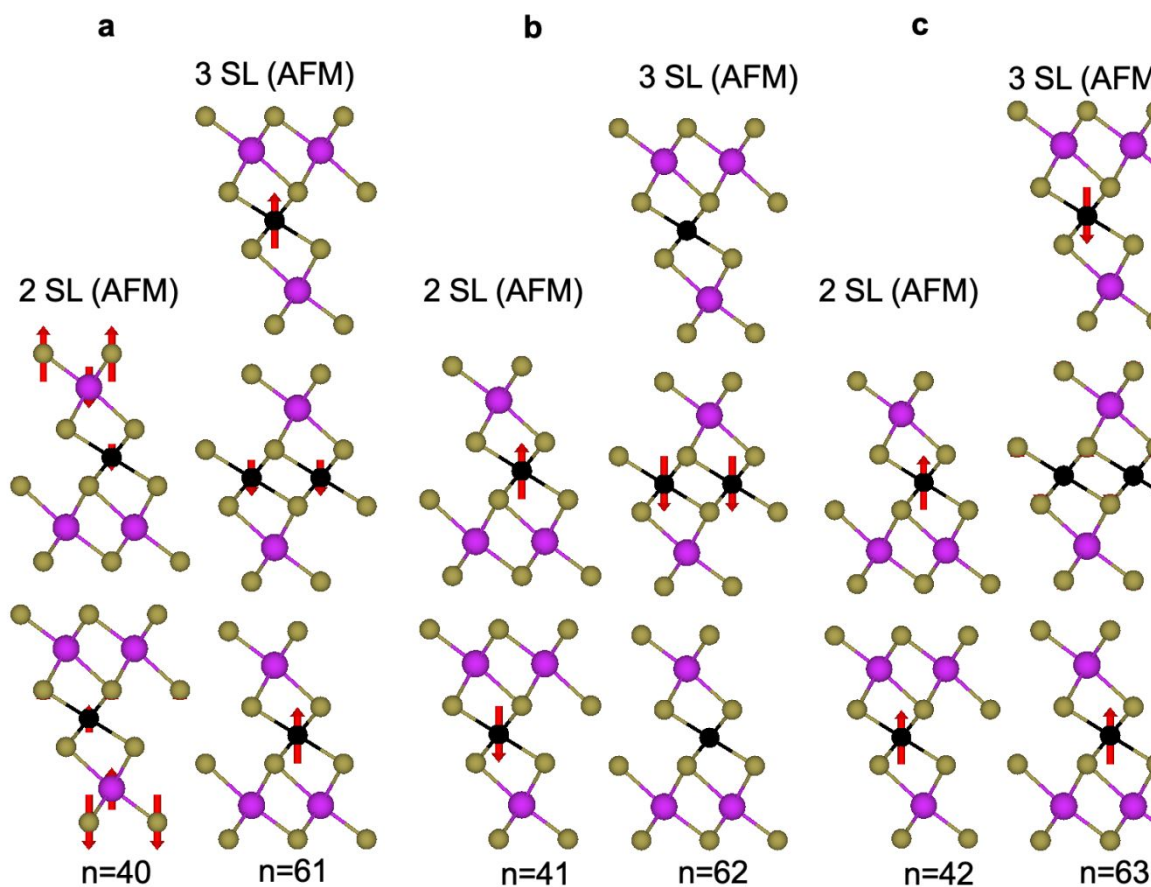


Figure S5. Comparative visualization of last three modes of 2SL and 3 SL MnBi_2Te_4 in the AFM phase.

(a-c) Visualization of last three out-of-plane vibrations at Γ -point in 2 SL and 3 SL MnBi_2Te_4 in AFM configuration. Two nearby columns correspond to the different phonon modes, shown for the 2 SL AFM (left column) and 3 SL AFM (right column) phases. Arrow lengths are scaled identically (based on the magnitude of the eigenvectors) in all panels. Phonon mode index has been labelled at the bottom of each subfigure. Here, first two modes presented above ($n=40, 41$) are Raman active modes, while the third one ($n=42$) is the IR active (A_{2u}) mode. On the other hand, the third one ($n=63$) is the Raman active mode and the first two ($n=61, 62$) of them are IR (A_{2u}) active modes.

Supplementary Note S1

In 3 SL AFM, 61, 62, and 63rd phonon modes originate from magnetic sublattices, where the 63rd mode is the Raman active but the 61 and 62nd are IR (A_{2u}) active modes (**Table S2**). These phonon branches form completely different vibrational patterns (**Figure S6**) compared to that for 40, 42, and 43rd phonon modes in 2 SL AFM, in which 40 and 41st are Raman active modes and 42nd is the IR active (A_{2u}) mode. The relaxed structure of 3 SL AFM retains spatial inversion symmetry. As a result, the two SLs Mn atoms are symmetry equivalent. The spatial

inversion symmetry enforces vibrational eigenmodes to form symmetric (even) and antisymmetric (odd) combinations of the two SLs atoms, while this symmetry does not require its projections on phonon modes to coincide each other. Thus, each mode can form different pattern of vibration compared to that of 2 SL AFM.

For example, in 61st mode (interlayer breathing mode), the bottom and middle Mn sublattices vibrate toward each other, while the top and middle sublattices vibrate in opposite directions. Compared to this mode, the 40th (breathing) mode in 2 SL AFM, top and bottom Mn sublattices vibrate toward each other, where significant contributions come from interlayer Bi and Te atoms. Unlike this mode, the 41st (breathing) mode originates from two Mn sublattices only that vibrate in opposite directions. On the other hand, the breathing 62nd mode in 3 SL AFM originates only from the middle of Mn sublattice. As the 3 SL AFM is very thin, the two SL still couple strongly due to their spin configuration, which produces a clear even–odd splitting of the Mn-derived phonon modes.

When we project phonon modes onto individual Mn atoms, these collective even and odd modes appear as two distinct sets associated with the top, bottom or middle SLs, even though **three SLs themselves are identical by symmetry**. Such behaviour can also be connected to the change in magnetic point group D_3 (32) (2 SL AFM) to $S_6(\bar{3})$ (3 SL AFM) with magnetic ordering, where 3 SL AFM is an uncompensated AFM with finite magnetic moment. However, this behaviour is absent in the 1 SL FM because only one Mn sublattice exists. This behaviour is also absent in the 2 SL AFM because strong interlayer constraints suppress the even–odd frequency separation. Thus, the 3 SL geometry is the minimal thickness where SL–SL vibrational coupling is still strong enough to produce a clearly resolvable and symmetry-allowed even–odd splitting in the Mn-projected phonon modes. However, between 2 SL AFM and FM phases, for example, the 40th breathing mode vibrational pattern changes, where the contributions from Mn sublattices become almost zero in FM phase (**Figure S4**), suggesting strong spin-phonon coupling. This is also valid for 3 SL AFM and FM phases (**Figure S5**). It is worth mentioning that spin-phonon coupling in MnBi₂Te₄ is both theoretically and experimentally well-established phenomenon¹.

Table S1. Computed phonon modes and their activity of 1-2 SL MnBi₂Te₄.

Calculated phonon mode index (n), frequency (ω), mode symbol in the group representation (GR), and Raman (R) or Infrared (IR) activity.

n	1 SL		2 SL (AFM)			2 SL (FM)			
	ω (meV)	IR\ R	GR	ω (meV)	IR\ R	GR	ω (meV)	IR\ R	GR
1	0.00	IR	A _{2u}	0.00	IR	A _{2u}	0.00	R	A _{1g}
2	0.00	IR	E _u	0.00	R	E _g	0.00	IR	A _{2u}
3	0.00	IR	E _u	0.00	R	E _g	0.00	R	E _g
4	2.79	R	E _g	0.47	IR	E _u	0.59	R	E _g
5	2.79	R	E _g	0.47	IR	E _u	0.59	IR	E _u
6	3.96	R	A _{1g}	3.08	R	A _{1g}	2.22	IR	E _u
7	6.17	IR	E _u	3.42	IR	E _u	3.00	IR	E _u
8	6.17	IR	E _u	3.42	IR	E _u	3.00	IR	E _u
9	8.53	R	E _g	3.62	R	E _g	3.38	R	E _g
10	8.53	R	E _g	3.62	R	E _g	3.38	R	E _g
11	9.37	IR	A _{2u}	4.96	IR	A _{2u}	4.89	IR	A _{2u}
12	11.33	IR	E _u	4.97	R	A _{1g}	5.51	R	A _{1g}
13	11.32	IR	E _u	6.04	IR	E _u	6.08	IR	E _u
14	12.38	R	E _g	6.04	IR	E _u	6.08	IR	E _u
15	12.38	R	E _g	6.27	R	E _g	6.26	R	E _g
16	14.75	R	A _{1g}	6.27	R	E _g	6.26	R	E _g
17	16.36	IR	E _u	8.10	IR	E _u	8.31	IR	E _u
18	16.78	IR	E _u	8.10	IR	E _u	8.31	IR	E _u
19	17.93	IR	A _{2u}	8.49	R	E _g	8.81	R	E _g
20	18.28	R	A _{1g}	8.49	R	E _g	8.81	R	E _g
21	19.02	IR	A _{2u}	9.40	IR	A _{2u}	9.56	R	A _{1g}
22				9.53	R	A _{1g}	9.61	IR	A _{2u}
23				11.33	IR	E _u	11.31	IR	E _u
24				11.33	IR	E _u	11.31	IR	E _u
25				11.63	R	E _g	11.70	R	E _g
26				11.63	R	E _g	11.70	R	E _g
27				12.44	IR	E _u	12.32	IR	E _u
28				12.44	IR	E _u	12.32	IR	E _u
29				12.75	R	E _g	12.72	R	E _g
30				12.75	R	E _g	12.72	R	E _g
31				14.51	R	A _{1g}	14.61	R	A _{1g}
32				14.62	IR	A _{2u}	14.66	IR	A _{2u}
33				16.32	IR	E _u	16.50	IR	E _u
34				16.32	IR	E _u	16.50	IR	E _u
35				16.33	R	E _g	16.52	R	R
36				16.33	R	E _g	16.52	R	E _g
37				17.19	IR	A _{2u}	17.67	IR	A _{2u}
38				17.37	R	A _{1g}	17.83	R	A _{1g}
39				18.09	R	A _{1g}	18.27	IR	A _{2u}
40				18.12	IR	A _{2u}	18.34	R	A _{1g}
41				19.17	R	A _{1g}	19.37	R	A _{1g}
42				19.24	IR	A _{2u}	19.48	IR	A _{2u}

Supplementary Note S2

The crystallographic point-group symmetry (D_{3d}) in 2D MnBi₂Te₄ does not necessarily prevent the observed splitting of E_g modes in 2-3 SL MnBi₂Te₄. Group theory enforces that an E_g mode must remain exactly doubly degenerate at the Γ point. Phonon dispersions of E_g fully respect this requirement. We mainly find that the E_g modes are doubly degenerate at Γ

but split for finite in-plane momentum. This is entirely consistent with symmetry, where irreducible representations apply strictly at Γ but the symmetry cannot protect degeneracy away from Γ . In nonpolar E_g case, the analogous splitting at finite q originates from two physical mechanisms that are well established in the literature. The first mechanism is interlayer hybridization. When we add one more SL into 1 SL system, the interlayer interactions lead to induce strong coupling between E_g vibrations of each SL, producing symmetric and antisymmetric vibrational combinations. These hybridized E_g modes exhibit different dispersions at finite q , even though group theory enforces their degeneracy at Γ . However, 2D non-analytic corrections to the dynamical matrix enforce appearance of such splitting in the dispersions. This suggests that the 2D non-analytic corrections are essential to account for the interlayer hybridization or coupling in 2-3 SL MnBi_2Te_4 . Similar E_g mode splitting behaviour is well-studied fact in layered materials²⁻⁴.

The second contribution comes from the magnetic ordering due to spin-phonon coupling. Different spin configurations of different magnetic orderings change the effective force constants via spin-phonon coupling, where interlayer exchange coupling depends on atomic displacements induced by the corresponding mode (**Figure 4** in the main text). Then the correction to the dynamical matrix will depend on the multiple of first derivative of exchange coupling relative to the displacement and spin operator $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$. This correction does not necessarily require any preservation of symmetries of the dynamical matrix, unlike that must hold in nonmagnetic systems. Such correction can occur in 2D MnBi_2Te_4 , even though its crystallographic space group remains untouched. The well-known effect of spin-phonon coupling are that it can remove mode degeneracies, change frequencies, or lead to anisotropic dispersions, while the crystallographic point group remains at Γ ⁵⁻⁷. In 2-3 SL MnBi_2Te_4 , the two components of the E_g doublet modulate interlayer magnetic superexchange paths differently (as a result, total modulation is very weak (**Figure 4g**)). This should lead to E_g modes splitting at finite q in 2-3 SL systems. However, this effect cannot induce any E_g modes splitting in 1 SL because the interlayer exchange path is absent in 1 SL FM system.

Table S2. Computed magnetic order dependent phonon modes and their activity of 3 SL MnBi₂Te₄.

Calculated phonon mode index (n), frequency (ω), mode symbol in the group representation (GR), and Raman (R) or Infrared (IR) activity.

n	3 SL AFM			3 SL FM		
	ω (meV)	IR\ R	GR	ω (meV)	IR\ R	GR
1	0.00	IR	E _u	0.00	IR	A _{2u}
2	0.00	IR	E _u	0.00	R	A _{1g}
3	0.00	IR	A _{2u}	0.00	IR	E _u
4	0.43	R	E _g	1.05	IR	E _u
5	0.43	R	E _g	1.05	R	E _g
6	1.39	R	A _{1g}	1.09	R	E _g
7	1.61	IR	E _u	1.76	IR	A _{2u}
8	1.61	IR	E _u	1.76	IR	E _u
9	1.95	IR	A _{2u}	2.05	IR	E _u
10	2.89	R	E _g	3.12	R	E _g
11	2.89	R	E _g	3.12	R	E _g
12	3.07	IR	E _u	3.29	IR	E _u
13	3.07	IR	E _u	3.29	IR	E _u
14	3.34	R	E _g	3.42	R	E _g
15	3.34	R	E _g	3.42	R	E _g
16	4.68	R	A _{1g}	4.52	R	A _{1g}
17	4.97	IR	A _{2u}	4.77	IR	A _{2u}
18	5.00	R	A _{1g}	5.01	R	A _{1g}
19	5.96	IR	E _u	6.11	IR	E _u
20	5.96	IR	E _u	6.11	IR	E _u
21	6.13	IR	E _u	6.34	R	E _g
22	6.13	IR	E _u	6.34	R	E _g
23	6.35	R	E _g	6.52	IR	E _u
24	6.35	R	E _g	6.52	IR	E _u
25	8.05	R	E _g	7.92	R	E _g
26	8.05	R	E _g	7.92	R	E _g
27	8.39	IR	E _u	8.25	IR	E _u
28	8.39	IR	E _u	8.25	IR	E _u
29	8.92	R	E _g	8.99	IR	A _{2u}
30	8.92	R	E _g	8.99	R	E _g
31	9.14	IR	A _{2u}	9.21	R	E _g
32	9.30	IR	A _{2u}	9.36	R	A _{1g}
33	9.38	R	A _{1g}	9.37	IR	A _{2u}
34	11.21	IR	E _u	11.09	IR	E _u
35	11.21	IR	E _u	11.09	IR	E _u
36	11.45	R	E _g	11.45	R	E _g
37	11.45	R	E _g	11.45	R	E _g
38	12.00	IR	E _u	11.96	IR	E _u
39	12.00	IR	E _u	11.96	IR	E _u
40	12.33	R	E _g	12.31	R	E _g
41	12.33	R	E _g	12.31	R	E _g
42	12.64	IR	E _u	12.67	IR	E _u
43	12.64	IR	E _u	12.67	IR	E _u
44	12.84	R	E _g	12.87	R	E _g
45	12.84	R	E _g	12.87	R	E _g
46	14.30	R	A _{1g}	14.51	R	A _{1g}
47	14.42	IR	A _{2u}	14.58	IR	A _{2u}
48	14.57	R	A _{1g}	14.66	R	A _{1g}
49	16.24	IR	E _u	16.05	IR	E _u
50	16.24	IR	E _u	16.05	IR	E _u
51	16.28	IR	A _{2u}	16.08	IR	E _u
52	16.28	IR	E _u	16.08	IR	E _u
53	16.32	IR	E _u	16.19	R	E _g
54	16.33	R	E _g	16.19	R	E _g
55	16.33	R	E _g	16.84	R	A _{2u}
56	17.11	IR	A _{2u}	17.23	IR	A _{2u}
57	17.11	R	A _{1g}	17.27	R	A _{1g}
58	17.40	R	A _{1g}	17.56	R	A _{1g}
59	17.90	R	A _{1g}	17.88	R	A _{1g}
60	17.90	IR	A _{2u}	17.88	IR	A _{2u}
61	18.91	IR	A _{2u}	18.81	IR	A _{2u}
62	18.95	IR	A _{2u}	18.86	R	A _{1g}
63	19.03	R	A _{1g}	18.90	IR	A _{2u}

Table S3. Computed magnetic order dependent dynamical parameters of 3 SL MnBi₂Te₄ from DFPT. Calculated high frequency dielectric constant (ϵ_D), electronic dielectric permittivity (ϵ), zone center polar mode contributions to the permittivity (ϵ_z), dynamic polarizability (P_d) with Clausius-Mossotti corrections, in-plane Born effective charges (Z^*), frequency of (infrared active) longitudinal optical (LO) phonons (ω_{LO}).

Physical quantity	3 SL AFM		3 SL FM	
ϵ_D	24.7		28.5	
ϵ	24.7		28.5	
ϵ_z	51.8		81.7	
$P_d(\text{\AA}^2)$	19.90		20.40	
Z^*	Mn	3.78	4.18	
	Bi	6.57	6.25	
	Te	-4.23	-4.17	
$\omega_{LO}(\text{meV})$	n=14	6.08	n=20	6.11
	n=24	11.31	n=35	11.09
	n=34	16.50	n=50	16.05

Supplementary Note S3

In nonmagnetic crystal structures, the crystallographic point group determines vibrational mode representations. On the other hand, the magnetic point group (also known as the Shubnikov group) combines the crystallographic symmetry operations with time-reversal symmetry.^{8, 9} These combined symmetry operations govern phonon selection rules in crystal structures with magnetic order. The onset of magnetic order can reduce the overall symmetry even when the atomic positions and point group symmetry remain unchanged. As a result, two different magnetic configurations (e.g., AFM vs FM stacking) may have different magnetic point groups and therefore different allowed IR-active phonon modes. This happens in magnetic crystal structure because the phonon-induced polarization must follow the magnetic symmetry in its transformation.⁹ Consequently, lowering magnetic symmetry can activate or deactivate IR modes by altering their irreducible representations. Such magnetic-symmetry-induced changes in IR activity are well documented, e.g., in Co₂Mo₃O₈, where the AFM transition modifies IR and Raman selection rules without changing the underlying crystallographic point group.¹⁰ Thus, changes in IR activity in 2 SL MnBi₂Te₄ are fully consistent with magnetic group theory.

In particular, the crystallographic point group is D_{3d} for all SLs and magnetic orderings in 2D MnBi₂Te₄. However, during the AFM to FM transition in 2 SL MnBi₂Te₄, the magnetic point group changes from $D_3(32)$ to $S_6(\bar{3})$. Such symmetry lowering modifies the IR activity of certain phonon modes. We also notice that the symmetry operations with time reversal

symmetry are altered due to AFM to FM transition. For instance, 2 SL AFM phase is symmetric under 180-degree rotation symmetry operation (#2), while 2 SL FM phase is invariant under combined 180-degree rotation and time reversal symmetries. More importantly, the 2 SL AFM phase is invariant under the combined inversion and time reversal symmetries, while the inversion symmetry is a good symmetry alone in 2 SL FM. However, these alternations of symmetry operations do not affect the crystallographic point group, but they can change the phonon dynamical matrix and its related properties (**Table S2** and **S3**).

Compared to 2 SL AFM, both symmetry operations and the magnetic point group remain the same after AFM to FM transition in 3 SL AFM ($S_6(\bar{3})$), because the 3 SL AFM is an uncompensated AFM ($\uparrow\downarrow\uparrow$) configuration with a finite magnetic moment. Despite no change in crystallographic and magnetic point group, we still observe the change in phonon irreducible representations and IR/Raman activities when going from the AFM ($\uparrow\downarrow\uparrow$) to the FM configuration (**Table S2**). In the 3 SL FM spin ($\uparrow\uparrow\uparrow$) configuration, some of the symmetry operations act as purely unitary, as these operations project each SL onto an equivalent SL with the same spin configuration. On the other hand, In the 3 SL (uncompensated) AFM spin ($\uparrow\downarrow\uparrow$) configuration, several of these symmetry operations exchange spin-up and spin-down configured SLs. Consequently, these spatial symmetry operations become symmetries only if they are combined with time reversal symmetry operation. These combined symmetry operations act as antiunitary parts of the magnetic space group. In standard treatments of magnetic point groups, the classification of phonon mode representations and activity selection rules of Raman and IR modes are controlled by the unitary part, while the full magnetic point group including antiunitary part does not necessarily control the mode representation and mode activity.⁹ As a result, the AFM and FM orders have different unitary symmetry groups for the selection rules and mode representation.

Therefore, the change of magnetic order from AFM to FM differently decomposes phonon modes at Γ into irreducible representations. Such different mode decomposition leads to change activities of IR and Raman modes in 3 SL MnBi_2Te_4 . Additionally, magnetic ordering induced change in spin-phonon coupling can modify the dynamical matrix and long-range interactions related parameters, e.g., macroscopic dielectric constant and mode effective charge. Consequently, it is expected that the change in magnetic order shifts the phonon frequencies and activities of some IR and Raman modes. Similar behaviour was reported in CrI_3 ¹¹⁻¹³ and related systems^{6, 14-16}. Also, strong spin-phonon coupling is well-established phenomenon (both theoretically and experimentally) in MnBi_2Te_4 .^{1, 17}

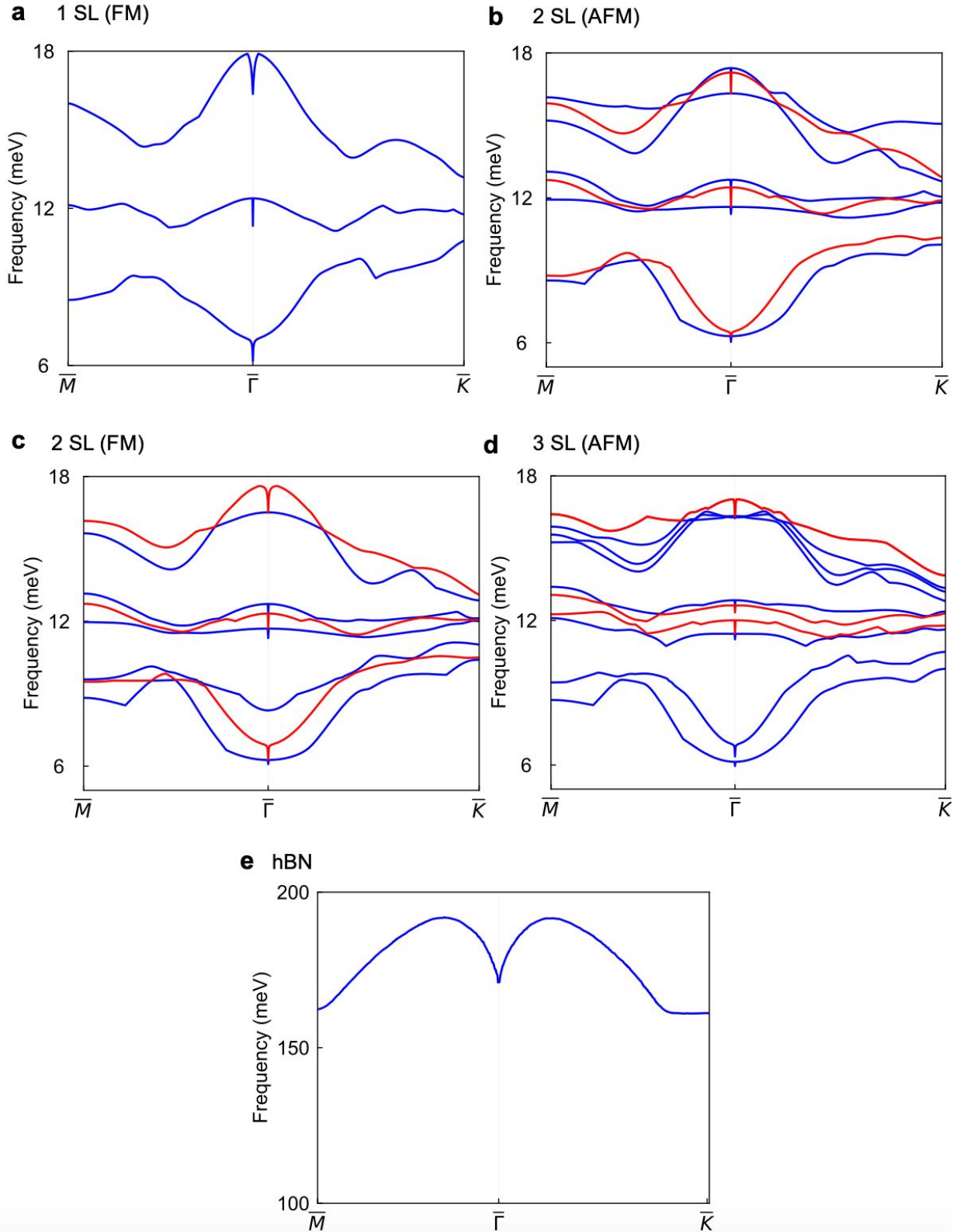


Figure S6. Longitudinal optical (LO) phonon modes in MnBi_2Te_4 and hBN obtained using 2D non-analytical Coulomb corrections.

Longitudinal optical (LO) phonon modes in 1 SL (a), 2 SL AFM (b), artificial 2 SL FM (c), and 3 SL MnBi_2Te_4 (d), and hexagonal boron nitride (hBN) (e), respectively. The blue lines are the infrared active LO phonon modes, while red lines are the Raman active LO phonon modes.

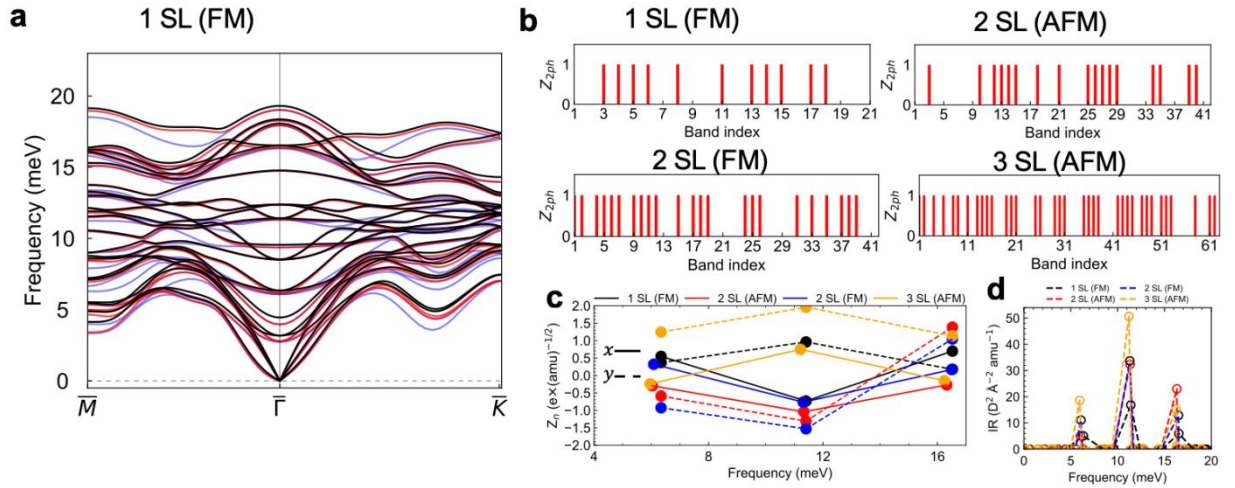


Figure S7. Comparison of phonon dispersions calculated with different methods, topological Z_{2ph} invariant, and mode effective charges for 2D $MnBi_2Te_4$.

(a) Computed phonon dispersions of 1 SL $MnBi_2Te_4$, by using DFPT (blue), 2D EPW interpolated (red), and tight binding (black) based green function method. (b) Calculated topological Z_{2ph} invariant based on Wannier charge center calculations. (c) Calculated anisotropic mode effective charges of polaritonic modes in $MnBi_2Te_4$ by using equation defined in the main text and DFPT. It's worth mentioning here that we did not consider Hubbard corrections in these calculations, as phonons and electron-phonon interactions for magnetic systems with Hubbard and SOC are not yet implemented in DFPT calculations. (d) Calculated IR spectra of 2D $MnBi_2Te_4$ with different thickness.

Supplementary Note S4

In 2D $MnBi_2Te_4$, the IR-active optical phonon branch that carries macroscopic charge (the LO mode) becomes essentially the phonon–polariton mode in the long-wavelength limit ($q \rightarrow 0$). This is consistent with the recent theory and experiments that for 2D systems compared to the conventional three-dimensional systems, the LO–TO splitting vanishes at the Γ -point ($q = 0$) and the LO and TO modes become degenerate at the zone center.¹⁸ However, the LO phonon branch rises in frequency with increasing q due to a 2D Coulomb interaction. Thus, the LO mode acquires its character as a purely mechanical mode (i.e., a normal phonon) once $q \gg q_c$, where q_c is the extremely small photon-phonon crossing momentum).¹⁹ Therefore, magnetophononic polariton behaviour in 2D $MnBi_2Te_4$ is confined to an extremely narrow momentum region near Γ . Although a finite LO-TO splitting via 2D Coulomb interaction persists for $q > 0$, for larger q , the same branch behaves as a normal phonon. Reader should not get confused that phonon polariton does not induce a different phonon frequency. The polariton only modifies the behaviour of an existing IR-active phonon mode at very small q . Although the IR-active phonon mode can span the entire Brillouin zone in frequency as a normal phonon, the polaritonic character can persist only in a very-small region near zone center (Γ).

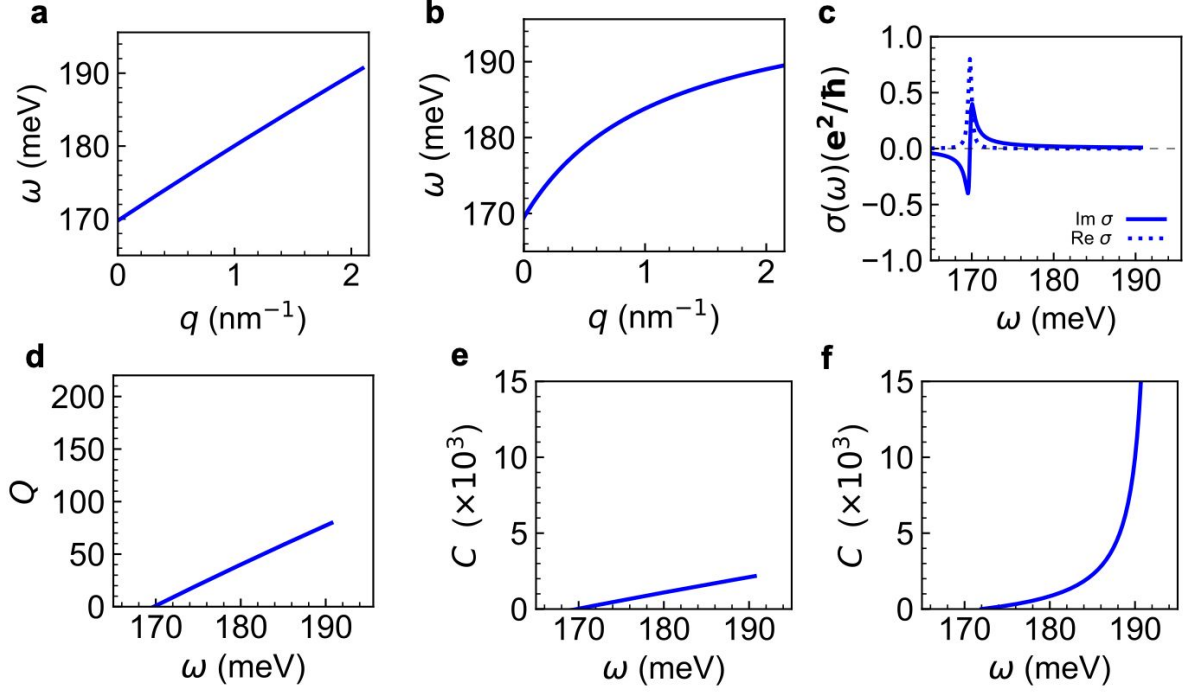


Figure S8. Calculated properties of phonon polaritons in 2D hBN.

(a) Computed polariton dispersions of LO phonons by using microscopic model (eq. 5 in the main text) (b) Computed polariton dispersions of LO phonons by using Lorentz like 3D model with asymptotic behaviour (eq. 13 in the Ref.¹⁸). However, due to much lower frequency and purely long-range 2D Coulomb interaction in 2D MnBi₂Te₄ compared to 2D hBN, this model failed to produce correct shape of the magnetophononic polaritons. Although both theoretical models produce similar momentum values, the microscopic model cannot produce asymptotic behaviour (shape of the curve). On the other hand, this model can produce such shape of the dispersion in the case of 2D MnBi₂Te₄, especially for the first magnetophononic polariton. (c) Real (dashed lines) and imaginary (solid lines) part of the lattice optical conductivity based on microscopic model. (d) Propagation quality factor $Q = \text{Im } \sigma / \text{Re } \sigma$. (e) Confinement factor (C) calculated by using microscopic model and Lorentz like 3D model (f). Again, the asymptotic behaviour is absent in the C curve that calculated by using microscopic model. Here the C seems to increase infinitely after asymptotic frequency. To compute all these parameters plotted in this figure, we use phonon damping factor value of $\gamma = 0.49 \text{ meV}$ for 2D hBN.

Near the optical-phonon resonance, hybridization with the lattice creates highly confined, dispersive magnetophononic polaritons in 2D MnBi₂Te₄. Consequently, their group velocity and spatial evolution differ substantially from those of an ordinary photon in the same medium (Table 1). The group velocity (equivalently, the deceleration factor) of these magnetophononic polaritons ranges from $\sim 10^{-5}c$ to $10^{-4}c$, indicating that they move at an extremely slow speed of $\sim 0.003\text{-}0.030 \text{ m/ps}$, i.e., ~ 3 to 30 nm/ps . The confinement factor illustrated in Fig. 3 reinforces that these magnetophononic polaritons are highly confined within the crystal space. Nonetheless, they are not fixed at any specific point. To induce an AFM-to-FM and topological phase transition in 2 SL MnBi₂Te₄ through anharmonic coupling between these polaritons and a Raman mode with light, we can use a pulse duration approximately equal to the polariton lifetime, i.e., $\tau_{\text{pulse}} \approx \tau_{\text{polariton}}$.

A recent Raman experiment measured the lifetime of the breathing A_{1g} mode to be approximately 13.3 ps.¹ Although the average phonon lifetime in 2D $MnBi_2Te_4$ ranges from ~26 to 45 ps, depending on layer thickness (Table 1), the polariton should travel 39.6 to 408 nm during its lifetime (13.3 ps). This distance is much smaller than the wavelength of mid-infrared light used for excitation, which is typically 10 to 50 μm due to the diffraction limit.²⁰ Therefore, the light confines the interaction within the laser spot, making these magnetophononic polaritons localized enough to respond to light, induce an AFM-to-FM transition, and trigger a topological phase change.

Notably, the relevant nonlinear displacement is not determined solely by the rapidly varying phase of a propagating polariton. If we represent the IR-active polariton coordinate in $MnBi_2Te_4$ locally as $Q_{IR}(\mathbf{r}, t) = A(\mathbf{r}, t) \cos(\mathbf{q} \cdot \mathbf{r} - \omega t)$. Through the anharmonic interaction $V_{anh} = \gamma Q_{IR}^2 Q_R$, the Raman assistant mode experiences a force proportional to Q_{IR}^2 . This quantity contains a rectified component proportional to the local polariton intensity envelope, $\langle Q_{IR}^2(\mathbf{r}, t) \rangle = \frac{1}{2} |A(\mathbf{r}, t)|^2$, because if the polaritonic E_u mode vibrates with a frequency ω , then $\cos^2(\mathbf{q} \cdot \mathbf{r} - \omega t) = \frac{1}{2} + \frac{1}{2} \cos(2(\mathbf{q} \cdot \mathbf{r} - \omega t))$. The $\frac{1}{2} \cos(2(\mathbf{q} \cdot \mathbf{r} - \omega t))$ part is a fast oscillation, but the $1/2$ term is a pure DC directional shift (rectification). This indicates that the A_{1g} Raman mode doesn't oscillate rapidly; instead, it is abruptly shifted in one direction and remains there, similar to a latched switch. Additionally, this shift doesn't immediately disappear after the 0.6 ps (which was used in Fig. 4f) light pulse ends. The atoms stay confined in this position and can only return slowly, at a rate determined by the polariton's natural decay. With a magnetophononic polariton lifetime of about 13.3 ps, the structural change persists in that region for a significantly extended time—around twenty-two times longer than the laser pulse duration. Therefore, the nonlinear displacement stays relatively stable, indicating a prolonged change in atomic positions and bond lengths. This stability provides enough time for the material to undergo its magnetic or electronic transition in 2 SL $MnBi_2Te_4$.

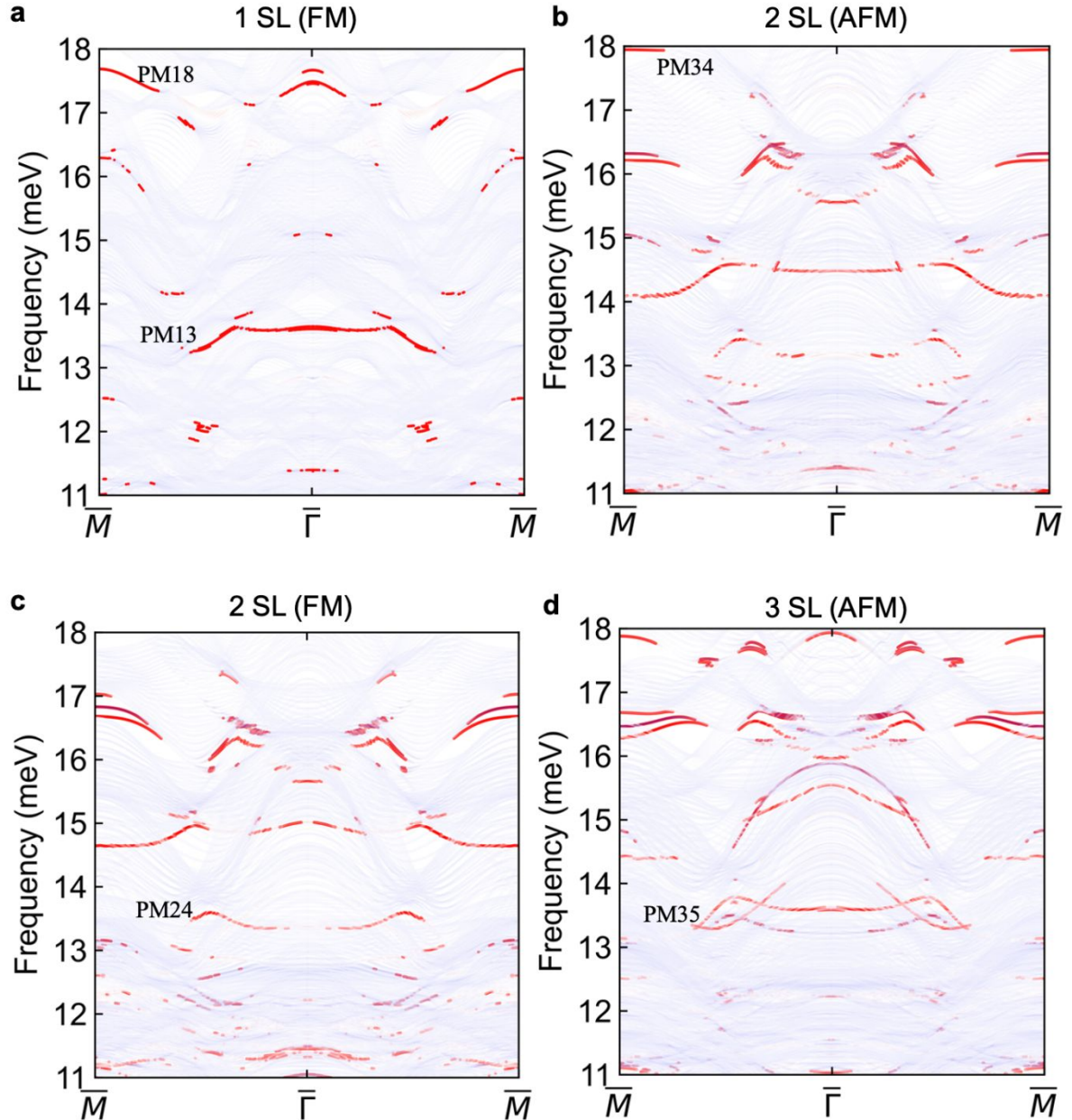


Figure S9. Phonon dispersion of zigzag (1D nanoribbon) edge of 2D MnBi_2Te_4 .

Edge phonon dispersion of 1 SL (a), 3 SL (b), 2 SL AFM (c) and artificial 2 SL FM (d) of MnBi_2Te_4 . Full range phonon dispersions are not shown here for clarity. No imaginary modes observed in the phonon dispersions. Red phonon bands are induced from the edge states and are topologically protected phononic edge states. The pink coloured bands are the trivial surface phonon bands. The label PM indicates the edge mode corresponding the bulk surface mode with nonzero Z_{ph} and the number indicates the bulk surface polaritonic mode index, i.e., $n = 13, 18$ for 1 SL, $n = 34$ for 2 SL AFM, $n = 24$ for 2SL FM, and $n = 35$ for 3 SL AFM. Only the edge phonons corresponding to the polaritonic modes that fall within this frequency range (11-18 meV) are shown here for clarity.

Supplementary Note S5

Recently, topological feature of phonon polariton has been experimentally (based on mid-IR spectroscopy) observed in the metasurface of 2D hBN²¹. In this note, we focus on the phonon

dispersions of 2D MnBi₂Te₄ (**Figure 2**) to explore this feature in 2D MnBi₂Te₄. As a careful analysis of phonon dispersions points out that several phonon modes linearly cross or touch other modes, as indicated by the red coloured-dashed circle (**Figure 2a**). This suggests the possibility of topologically protected phononic states, similar to the topological electronic states. To confirm whether MnBi₂Te₄ can host such nontrivial states or not, we calculate topological invariant (Z_{2ph}) for phonons²². The calculated nonzero values (**Figure S8**) of Z_{2ph} indicates that for several phonon modes, 2D MnBi₂Te₄ may host nontrivial topologically protected edge states. Surprisingly, some of them with nonzero Z_{2ph} in 2D MnBi₂Te₄ are also polaritonic modes: 3 modes (n=8,13,18) for 1 SL, 2 modes (n=14, 34) for 2 SL AFM, 1 mode (n=24) for 2 SL FM, and 2 mode (n=20, 35) for 3 SL AFM. As increasing number of layers leads more phonon modes and modifies its symmetries, the Z_{2ph} changes with the (FM to AFM) magnetic ordering and SL thickness of 2D MnBi₂Te₄.

Supplementary Note S6

For further confirmation topological polaritons described in **Note 5**, we calculate nanoribbon's (1D structure with both zigzag and armchair orientations) phonon dispersions (**Figure S9**), which shows few phonon edge states (in red). These red coloured phonon modes are induced by the edge atoms. These phononic states are topologically protected edge states, i.e., have a nonzero Z_{2ph} invariant. Aforementioned polaritonic modes also have the corresponding topologically protected edge states, as labelled in the **Figure S9**. To our best knowledge, this is the first ever reported topological magnetophononic polariton in magnetic topological insulators. These polaritonic modes are expected to be free from polariton induced backscattering within the finite q and corresponding frequency window. Additionally, the phononic edge states are shifted either by the increase of layer thickness or changing the magnetic order (in 2 SL from AFM to FM) (**Figure S9**). Such changes can provide a better control of the phononic states in the practical applications, for example, in the phononic diodes, waveguide modules and devices based on nanophononics²³⁻²⁵. However, several questions still remain unanswered, for example, “are these phonons chiral?”, “do they have angular momentum or magnetic moment?”, which would be future study directions.

Supplementary Note S7

To parameterize the potential (defined in **equation 9** in the main text), we extract the cubic coefficients from DFT based forces calculations considering SOC coupling and Hubbard corrections. Then these forces were projected onto phonon eigenvectors of the corresponding modes. For a Raman mode R with eigenvector $\zeta_{\kappa\alpha}^{(R)}$ (where α is the Cartesian direction, *i.e.*, x, y, z) and atomic masses m_a , we can define the normal-coordinate displacement as $u_{a\alpha}(Q_R) = Q_R e_{a\alpha}^{(R)} / \sqrt{m_a}$. Now we can define the generalized force along the Raman coordinate based on the DFT forces $F_{\kappa\alpha}$ as ²⁶

$$F_R(Q) \equiv \sum_{\kappa\alpha} \frac{\zeta_{\kappa\alpha}^{(R)}}{\sqrt{m_a}} F_{a\alpha}, \quad (S1)$$

For $A_{1g}^{(2)}$ mode, we obtain the cubic self-anharmonicity ($\beta_3^{(A_{1g})}$) by setting the displacement $u = 0$ for E_u by fitting $-F_{A_{1g}}(Q_a) = \Omega_{A_{1g}}^2 Q_a + \beta_3^{(A_{1g})} Q_a^2$, while we treat E_g as the doubly degenerate subspace and we explicitly project forces onto both part of eigenvectors, which yields (F_X, F_Y) . Thus, for a chosen displacement direction, we obtain the cubic self-anharmonic coefficient ($\beta_3^{(E_g)}$) by using the same scalar fit $-F_{\parallel} = \Omega_{E_g}^2 Q + \beta_3^{(E_g)} Q^2$, as the full 2D invariant $\propto (Q_X^3 - 3Q_X Q_Y^2)$ already validates the correct symmetry of the anharmonic force. We extract polaritonic (IR)–Raman couplings γ_3 from Raman forces induced by *pure* polaritonic mode displacements. For this, we set the displacement for the Raman mode to zero and create displacements via polaritonic E_u mode only (with amplitude $Q_{E_u} = \sqrt{Q_x^2 + Q_y^2}$). Then, we can define the couplings based on the fitting (from **equation 9** in the main text) as,

$$F_{A_{1g}}(Q_{E_u}) = -\gamma_3^{(A_{1g})} Q_{E_u}^2; (F_X, F_Y)(Q_{E_u}) = -\gamma_3^{(E_g)} (Q_x^2 - Q_y^2, 2Q_x Q_y). \quad (S2)$$

Now we can obtain $\gamma_3^{(A_{1g})}$ from a linear fit of $F_{A_{1g}}$ versus $Q_{E_u}^2$, and $(F_X, F_Y)(Q_{E_u})$ versus $\gamma_3^{(E_g)}$ from the corresponding fit for the E_g mode. The calculated values of these coefficients are listed in **Table S4**.

Table S4. Calculated nonlinear anharmonic coupling coefficients of 2 SL MnBi₂Te₄

Anharmonic coefficient	Calculated value in eV/(Å√amu) ³	Coupling modes
$\gamma_3^{(E_g)}$	-3.583×10^{-5}	$E_u - E_g$
$\gamma_3^{(A_{1g})}$	-1.966×10^{-4}	$E_u - A_{1g}$
$\beta_3^{(E_g)}$	-3.738×10^{-4}	E_g
$\beta_3^{(A_{1g})}$	-5.093×10^{-6}	A_{1g}

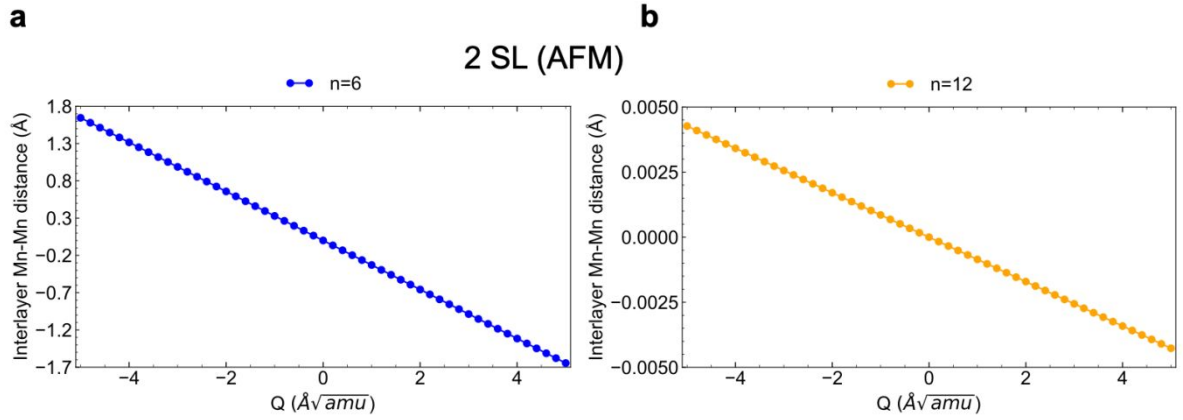


Figure S10: Comparative Raman modes induced interlayer Mn-Mn distance in 2 SL MnBi₂Te₄ (AFM).

(a) Changes in interlayer Mn-Mn distance induced by the Raman mode $A_{1g}^{(1)}$ (6th mode) amplitude Q . (b) Changes in interlayer Mn-Mn distance induced by the Raman mode $A_{1g}^{(2)}$ (12th mode) amplitude Q . In both cases, we used intrinsic directions of vibrations from DFPT data.

Based on the calculated nonlinear coefficients, we solve the following equations of motion:

$$\begin{aligned}
\ddot{Q}_x &= -\Omega_{E_u}^2 Q_x - 2\Gamma_{IR} \dot{Q}_x + Z_x^* E_x(t) - \gamma_3^{(E_g)} (Q_x Q_x + Q_y Q_y) - \gamma_3^{(A_{1g})} Q_a Q_x, \\
\ddot{Q}_y &= -\Omega_{E_u}^2 Q_y - 2\Gamma_{IR} \dot{Q}_y + Z_y^* E_y(t) - \gamma_3^{(E_g)} (-Q_y Q_x + Q_x Q_y) - \gamma_3^{(A_{1g})} Q_a Q_y, \\
\ddot{Q}_X &= -\Omega_{E_g}^2 Q_X - 2\Gamma_{E_g} \dot{Q}_X - \frac{1}{2} \gamma_3^{(E_g)} (Q_x^2 - Q_y^2) - \beta_3^{(E_g)} (Q_X^2 - Q_Y^2), \\
\ddot{Q}_Y &= -\Omega_{E_g}^2 Q_Y - 2\Gamma_{E_g} \dot{Q}_Y - \gamma_3^{(E_g)} Q_x Q_y + 2\beta_3^{(E_g)} Q_X Q_Y, \\
\ddot{Q}_a &= -\Omega_{A_{1g}}^2 Q_a - 2\Gamma_{A_{1g}} \dot{Q}_a - \frac{1}{2} \gamma_3^{(A_{1g})} (Q_x^2 + Q_y^2) - \beta_3^{(A_{1g})} Q_a^2.
\end{aligned} \tag{S3}$$

Readers should note that these displacements represent ionic displacements generated based on the zone-center phonon eigenvectors. While the fundamental crystal lattice geometry determines the base anisotropy, lattice vibrations (phonons) can dynamically alter the crystal's symmetry and properties in two ways. Phonons involve atomic displacements that can temporarily reduce the overall symmetry, affecting properties such as electrical conductivity, optical response, and magnetic behavior. When light hits a crystal, it can impulsively excite atoms, creating coherent phonons that stretch or compress lattice bonds. These displacements transiently alter the optical index matrix, modulating the crystal's optical anisotropy (birefringence) and sometimes even reversing its sign.

A crystal's anisotropy is limited by its space-group symmetry. When a strong light pulse excites a phonon mode that breaks the lattice's symmetry, it causes atomic displacements that alter the crystal's original symmetry. This can transform an isotropic cubic crystal into an anisotropic tetragonal shape, creating directional differences in properties. For example, in materials like Bismuth (Bi) or Antimony (Sb), a laser pulse can pump a specific A_{1g} optical phonon mode.²⁷ Femtosecond X-ray pulses capture images of atoms moving to lower-symmetry positions, oscillating while the crystal remains intact below its melting point.

In the case of 2 SL MnBi_2Te_4 , the A_{1g} mode is symmetric, reflecting the crystal's symmetry. Driving it shifts bond lengths in 2 SL AFM without changing the overall symmetry, only affecting anisotropy. To alter the symmetry, the polariton must interact with a broken-symmetry mode like E_u , requiring higher laser intensity to overcome lattice forces. Low-amplitude A_{1g} modes don't break symmetry, but intense infrared-active phonon driving can transfer energy through nonlinear coupling, shifting atoms and breaking symmetry. When low-intensity mid-IR or terahertz light excites a phonon-polariton, nonlinear phononics limit mode interaction energy, causing tiny displacements and weak oscillations. To break symmetry, laser

intensity must exceed a high threshold, inducing large displacements. Large amplitude Q_{IR} creates a strong, rectified force that pushes the A_{1g} mode out of its potential well, as Q_{IR} appears squared in the coupling potential equation (eq. 9 in the main text).

The internal electric field (~ 1 V/Å or 100 MV/cm) holds atoms in a solid. To alter the lattice or symmetry, an external laser must produce a comparable field. Fields of 10 MV/cm or more cause symmetry-breaking phase transitions, while nonlinear phononics occur between 1 MV/cm and 50 MV/cm. The 1-5 MV/cm range signals the onset of nonlinear phononics, with weak nonlinear shifts. Exciting a symmetric mode (e.g., A_{1g} in Bismuth) moves atoms along a path preserving the space group $R\bar{3}m$. At low amplitudes, anisotropy changes quantitatively (slight electronic shifts) but not qualitatively (no change in tensor elements). The symmetry-breaking regime (10-30 MV/cm) can push atoms across boundaries, as in Gallium Selenide (13 MV/cm) affecting light-electron-lattice interactions,²⁸ or Lithium Niobate (30 MV/cm)²⁹ inducing nonlinear phononics that reverse ferroelectric polarization. High laser intensities can deform the electronic potential surface, moving atoms to cubic positions, collapsing Peierls distortion, and causing an electronically driven structural phase transition.³⁰ Under a 10 MV/cm DC field, most insulators would break down, but ultrafast pulses (femtoseconds) can pass through, avoiding ionization and reversing the lattice without damage.

Since the AFM-to-FM transition in 2 SL MnBi_2Te_4 requires a $3 \text{ Å}\sqrt{\text{amu}}$ displacement—which is within the threshold—this displacement leads only to a quantitative change in anisotropy, bandgap, and topology by altering bond length (Fig. 5).

To assess whether the required displacement ($3 \text{ Å}\sqrt{\text{amu}}$) is realistic relative to the structural stability of the crystal, we have calculated the relevant bond lengths in the ground-state structure and compared them with those in the displaced structure under the A_{1g} mode at amplitude $3 \text{ Å}\sqrt{\text{amu}}$. We have kept the lattice parameters fixed and allowed only the atomic coordinates to be displaced. If we denote the ground-state AFM structure by GS and the displaced structure by DS, the bond difference between them is given by $\Delta d = d_{GS} - d_{DS}$, where d is the bond length in Å. In percentage, we calculated the change in bond length using the equation: $\% \Delta d = \frac{d_{GS} - d_{DS}}{d_{GS}} \times 100$. While the Mn-Mn interlayer distance changes negligibly, the displacement leads to 0.44% and 1.15% bond length changes for Mn-Te and Bi-Te bond pairs. Thus, the atomic motion remains very small compared with the equilibrium bond lengths

and does not produce significant bond stretching, compression, or local structural damage. This indicates that the required displacement is well within the structurally feasible regime, as indicated by a Lindemann-type comparison of atomic motion with the characteristic bond length.

For additional stability evaluation, we have conducted an analysis based on the static Lindemann stability criterion.³¹ We have calculated $u_i = |\mathbf{R}_i^{DS} - \mathbf{R}_i^{GS}|$ for each atom using periodic boundaries, where $\mathbf{R}_i^{GS}$ and $\mathbf{R}_i^{DS}$ are Cartesian position vectors of atom i in the ground-state and displaced structure, respectively, and u_i is the displacement magnitude of atom i . Then, we obtained the local Lindemann-type ratio as $L_i = \frac{u_i}{d_{GS}}$. The Lindemann concept compares RMS atomic motion to nearest-neighbor distance, typically using a characteristic scale of about 0.1, though this is empirical. We have calculated RMS atomic displacement by using the following equation,

$$L_{\text{RMS}} = \frac{\sqrt{\frac{1}{N} \sum_i u_i^2}}{d_{\text{NN},i}},$$

where d_{NN} is the nearest-neighbor distance, and N is the total number of atoms in the unit cell. While L_{RMS} is approximately zero for Mn, L_{RMS} is approximately 0.025 for Bi and Te, which is much smaller than the characteristic scale of about 0.1.³² This ensures the stability of the displaced structure and suggests its experimental feasibility. However, it should be noted that the experimental optical damage threshold is also influenced by factors such as pulse duration, fluence, absorption, and heat dissipation, which are beyond the scope of the current static first-principles calculations. Nonetheless, the minimal bond-length variation and the very small L_{RMS} indicate that the predicted AFM to FM transition in 2 SL MnBi₂Te₄ does not require a significant or destructive lattice distortion. Thanks to the wonderful geometric pattern of 2 SL MnBi₂Te₄ and its A_{1g} (#12) mode displacement pattern.

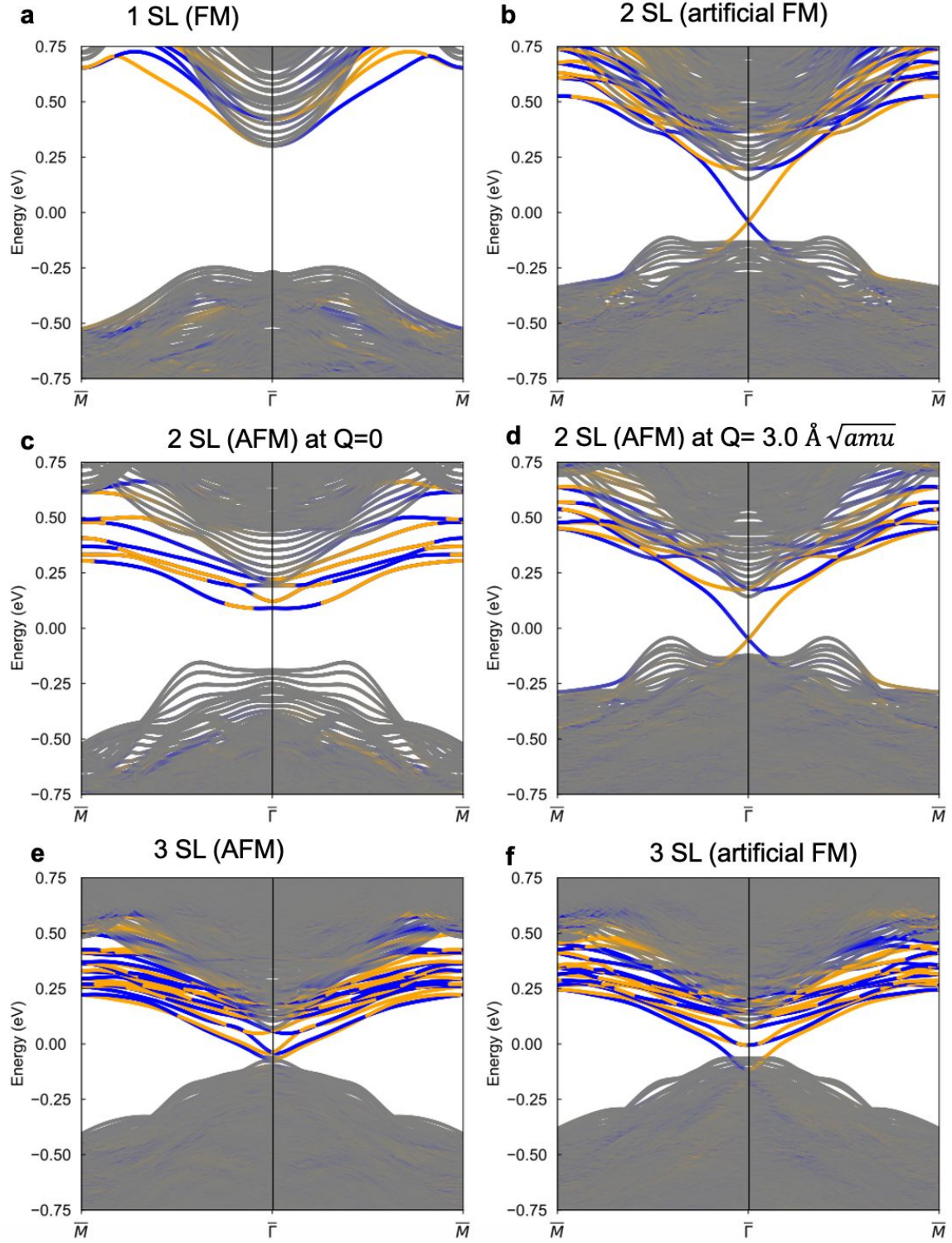


Figure S11. Evolution of topology in 2D MnBi_2Te_4 via magnetic order, thickness, and light.

(a-b) Nanoribbon bandstructure along a defined momentum path of 1 SL FM, 2 SL artificial FM. Nanoribbons used in these calculations are shown in Fig. 1 of the SI. (c-d) Nanoribbon electronic dispersions of 2 SL AFM in the absence of light (i.e., $Q=0$, no light-induced displacement) and in the presence of light (i.e., at $Q=3.0 \text{ \AA} \sqrt{amu}$), respectively. (e-f) Nanoribbon (one dimensional—1D) electronic dispersions of 3 SL AFM MnBi_2Te_4 and 3 SL (artificial) FM MnBi_2Te_4 . In all figures, the blue coloured lines represent left edge states, grey coloured lines represent bulk surface bands, and the orange-coloured lines represent right edge states. The zero energy represents the Fermi level.

Supplementary Note S8

We calculate intralayer coupling of 1 SL FM phase with 211 supercells. Then, we calculate total energy of each displaced structure in AFM and FM configurations by including SOC and Hubbard corrections. Finally, we calculate exchange coupling based on the energy mapping method. We use this same energy mapping method to calculate interlayer exchange coupling in 2-3 SL systems (with 111 supercell). These calculations can capture static displacements effect on the exchange interactions. To account dynamic effect accurately, we evaluate the interlayer exchange coupling using the magnetic force theorem (MFT). We formulate this based on a Wannier tight-binding representation of the Kohn–Sham Hamiltonian. For two between magnetic subspaces (SLs) 1 and 2, we obtain the interlayer exchange coupling from the second-order variation of the potential relative to infinitesimal rotations of the local magnetic moments. This leads to the interlayer exchange coupling as ^{33, 34}

$$J_{SL1-SL2} = \frac{1}{\pi} \text{Im} \int_{-\infty}^E d\varepsilon \frac{1}{N_k} \sum_{\mathbf{k}} \text{Tr} [\Delta_{SL1}(\mathbf{k}) G(\mathbf{k}, \varepsilon) \Delta_{SL2}(\mathbf{k}) G(\mathbf{k}, \varepsilon)],$$

where ε is the energy (E) integration variable, N_k : number of $\mathbf{k}$ -points used for Brillouin-zone integration, $\mathbf{k}$ is the crystal momentum, $\text{Tr}[\dots]$ represent trace over the Wannier spinor basis, $H^W(\mathbf{k})$ represent Wannier-interpolated fully relativistic tight-binding Hamiltonian, $G(\mathbf{k}, \varepsilon)$ is Green's function ($G(\mathbf{k}, \varepsilon) = [(\varepsilon + i0^+) \mathbb{I} - H^W(\mathbf{k})]^{-1}$, where $i0^+$ is the enforcing the retarded and $\mathbb{I}$ is the identity operator), and $\Delta_\ell(\mathbf{k})$ is the exchange-splitting operator (which is projected onto magnetic sublattice ℓ).

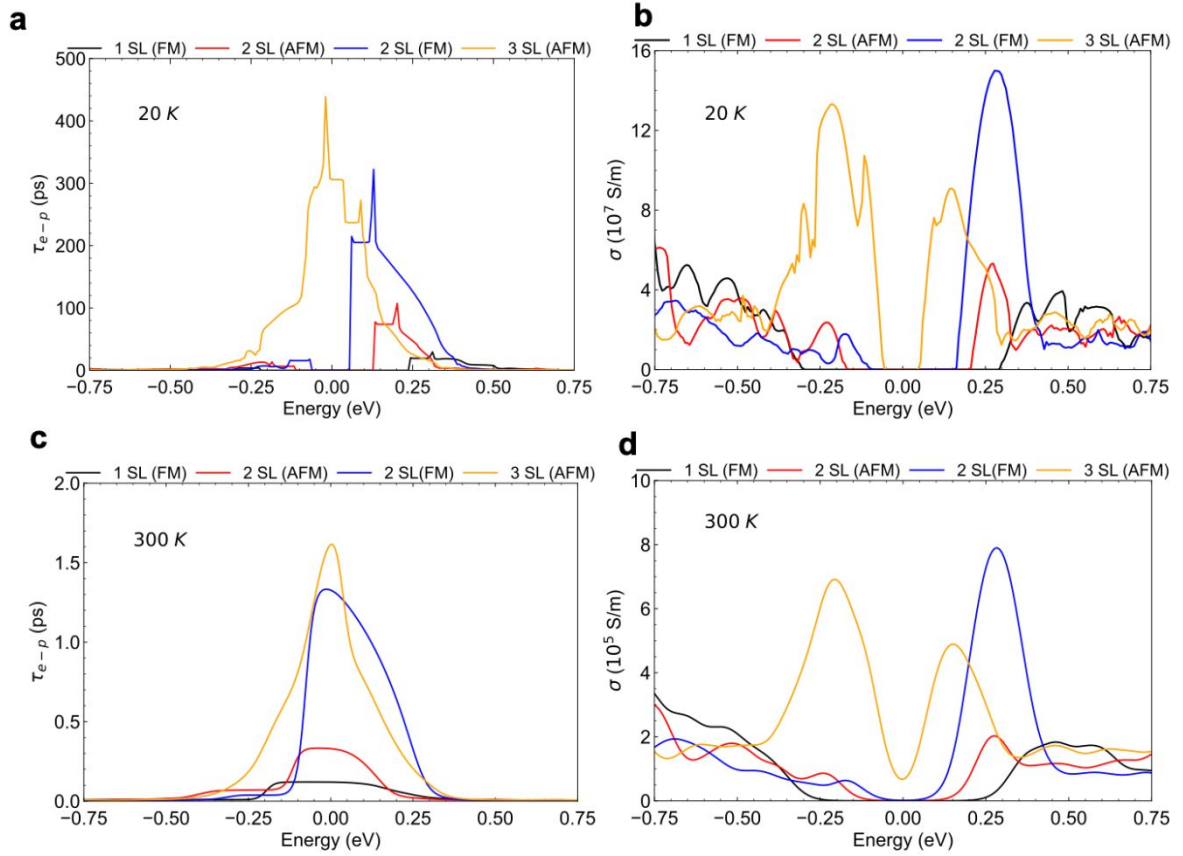


Figure S12. Carrier lifetime and electrical conductivity at 20 K and 300 K in 2D MnBi_2Te_4 .

Calculated carrier lifetime at 20 K (a) and 300 K (c) and electrical conductivity at 20 K (b) and 300 K (d) as a function of electronic chemical potential by considering electron-phonon scattering. In these calculations, we used electronic eigenvalues calculated with Hubbard corrections and EV93 functional.

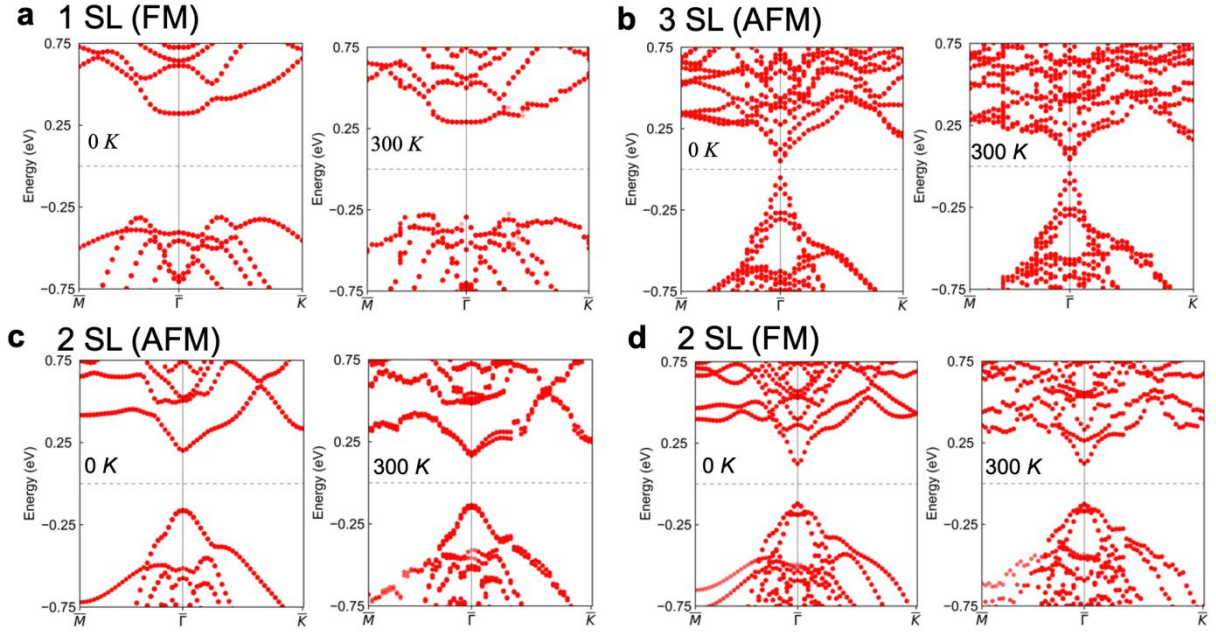


Figure S13. Temperature dependent band structures of 2D MnBi_2Te_4 from electron-phonon scattering. Electron dispersions at 0 K and 300 K for 1 SL (a), 3 SL (b), 2 SL AFM (c) and artificial 2 SL FM (d) of MnBi_2Te_4 . The dashed lines at zero energy indicate the Fermi level. In these calculations, we considered Hubbard corrections explicitly.

Supplementary Note S9

Figure S13 shows the electronic band structures of 2D MnBi_2Te_4 obtained via explicitly considering e-ph interactions. The bandgap at 0 K (**Figure S13a**) is almost same compared to the standard DFT band structures in **Figure 1**, suggesting no bandgap renormalization by e-ph scattering at 0 K, as expected. Interestingly, the bandgap renormalization in 1 SL at 300 K by is about 100 meV, while the renormalization is about 60 meV in 2 SL (AFM). This indicates that the e-ph scattering is much stronger in 1 SL than in 2 SL. By changing the magnetic order from AFM to FM in 2 SL, the renormalized bandgap is reduced to 20 meV, which is significantly smaller than that in 2 SL AFM, indicating that the e-ph scattering gets much weaker due to changing the magnetic ordering (spin-phonon coupling). In 3 SL MnBi_2Te_4 , the renormalized bandgap renormalization is only 10 meV, which is the lowest among all other thicknesses or phases studied here, pointing out that QAH phase of 3 SL (AFM) has negligibly weak e-ph scattering.

Supplementary Note S10

Rodríguez-Vega *et al.*'s work presents a compelling theoretical proposal for **non-equilibrium light-driven switching** of magnetic and topological states in 2 SL MnBi₂Te₄ as a prime material candidate³⁵. However, the specific methodological framework of Ref. ³⁵ cannot be directly compared to our calculations, because several technical approximations limit the quantitative reliability of its conclusions for MnBi₂Te₄. For example, all Γ -point phonons were computed without spin-orbit coupling (main text page #4153, lines 10–13), even though SOC plays a central role in spin-lattice coupling in MnBi₂Te₄ [Ref.³⁵]. The work also identifies the highest-frequency A_{2u} mode as having the largest mode effective charge (page #4153, lines 33–35). However, in SI, **Figure 1** (page #6 in the SI) shows that this mode actually has one of the smallest Z^* , making its selection inconsistent with the authors' own data. Moreover, several phonon irreps and mode indices are not matched consistently between the main text and SI. For example, the paper reported one of the A_{1g} mode with index #14 and frequency 2.3 THz in **Figure 2a** but **Figure S1** shows that this 14th mode has $Z^* \sim -2 e/\sqrt{\text{amu}}$. This indicates that this 14th mode is an IR-active mode, and we find that it is indeed an IR-active E_u mode. Besides, the electronic band gap (~ 60 meV) used (**Figure 5a** in page #4156) in the topological analysis is much smaller than experimentally established values (>200 meV). These issues make it difficult to use Ref.³⁵ as a comparative or contextual purpose for the present study.

Compared to the reference paper, this work uses a fully SOC included highly sophisticated DFPT based phonon analysis. The dynamical analysis shows excellent agreement with the experimentally measured data. This work also uses correct 'irreducible representation' identification including magnetic and point group symmetry, identification of magnetophononic phonon polaritons including damping factor, an accurate electronic structures, and electron-phonon coupling analysis, making our predictions robust and distinct from few of those available in Ref.³⁵. Thus, while the Rodríguez-Vega *et al.*'s study may provide conceptual inspiration, these results in our work represent a intriguing study of magnetophononic polaritons, their connections with magnetism and topology, and influence of electron-phonon interactions on the topology and their connections with magnetism.

It is also worth mentioning that some previous studies have demonstrated magnetic exchange modulation via nonlinear phononics in 3D materials, using phonon eigenvectors to analyze lattice distortions and changes in exchange coupling. In Ref. ³⁶, magnetism is tuned by indirectly exciting Raman-active phonons through infrared-active modes in LaTiO₃ and

YTiO₃, thereby affecting TiO₆ rotations and Ti-O distortions. This reduces the rotational angle, triggering an FM-to-G-type AFM transition driven by a small displacement (~ 1.6 Å). However, achieving this requires a high laser pulse intensity (22 MV/cm), which the authors sought to reduce by applying external strain.

Refs. ^{37, 38} reported a nonlinear phonon-displacement-induced magnetic phase transition in bilayer CrI₃ and 3D GdTiO₃, requiring high pulse fields ($E_0=4$ MV/cm for $\tau = 0.2$ ps and $E_0=20$ MV/cm for $\tau = 0.8$ ps in CrI₃; and $E_0>7$ MV/cm in 3D GdTiO₃). Meanwhile, Ref. ³⁹ found that quasistatic structural distortion caused by nonlinear phonon-phonon interactions (at $E_0=0.6$ MV/cm) in 3D Cr₂O₃ can shift the magnetic state from antiferromagnetic to one with ferromagnetically coupled spins. Conversely, Ref. ⁴⁰ reports multiple ferroic orders in 3D BiFeO₃ from nonlinear phonon-driven structural modulations.

Compared with these studies, the behavior shown in our **Fig. 4** is highly distinct. Specifically, Figs. 4a-d illustrate three magnetophononic polariton modes and Raman modes that modulate magnetic exchange in 2D MnBi₂Te₄ across various SL thicknesses and magnetic orders. These data pertain to static displacements. Because light-matter interactions are inherently dynamic, we show dynamic effects in Figs. 4e-g for one polariton mode and two Raman modes, selected for their intensity and symmetry. **Fig. 3** illustrates how polaritonic functionality varies with magnetic ordering. To connect these phenomena, we present the modulation of exchange coupling as a function of phononic displacements in **Fig. 4**. Two of our SL systems are 2D magnetic topological insulators, requiring about $3 \text{ Å}\sqrt{\text{amu}}$ of displacement for a magnetic phase transition, achievable with an electric field of 2.09 MV/cm and a 0.6-ps pulse. We demonstrate that exchange modulation can induce a reversible topological phase transition with topologically protected edge states. Thus, our study is entirely distinct within these contexts.

Supplementary Note S11

Here we discover that MnBi₂Te₄ hosts three highly confined, ultraslow, long-propagating, and tunable magnetophononic polaritons, characteristic of strong light-matter interactions in MnBi₂Te₄. The second magnetophononic polariton can induce magnetic phase transition in 2 SL MnBi₂Te₄, leading to a normal to topological phase transition. Such feature can not only lead to subwavelength confinement of light but also be highly useful in topological switching

applications, which can be exploited in future study. We also find that these magnetophononic polaritons in MnBi_2Te_4 have strong dependency on SL thickness, topology, and magnetic order (antiferromagnetic to ferromagnetic). This feature indicates the tunability of these magnetophononic polaritons, which open avenues for studying thickness, topology, and magnetism-driven phonon polaritonic behaviour, topology and magnetism-modified selection rules, and the possibility of topological polaritonic transport or edge polaritonic states. These represent distinct physical phenomena beyond the basic magnetophononic polaritons properties characterized in this study. We believe that such tunability feature will motivate further investigation of light–matter coupling driven topological switching application in real devices. These magnetophononic polaritons have also strong connection with electron-phonon interactions due to spin-phonon coupling. The electron-phonon interactions relate strongly with SL thickness, topology, and magnetic order. This featured result will naturally motivate several avenues for future research. For example, magnetophononic polaritons induced or assisted recombination that can enhance photocurrent saturation and lead to phonon assisted radiative or non-radiative process, temperature-dependent polariton renormalization, and potential polariton-driven modifications of carrier dynamics. All these phenomena extend beyond the scope of our current study here.

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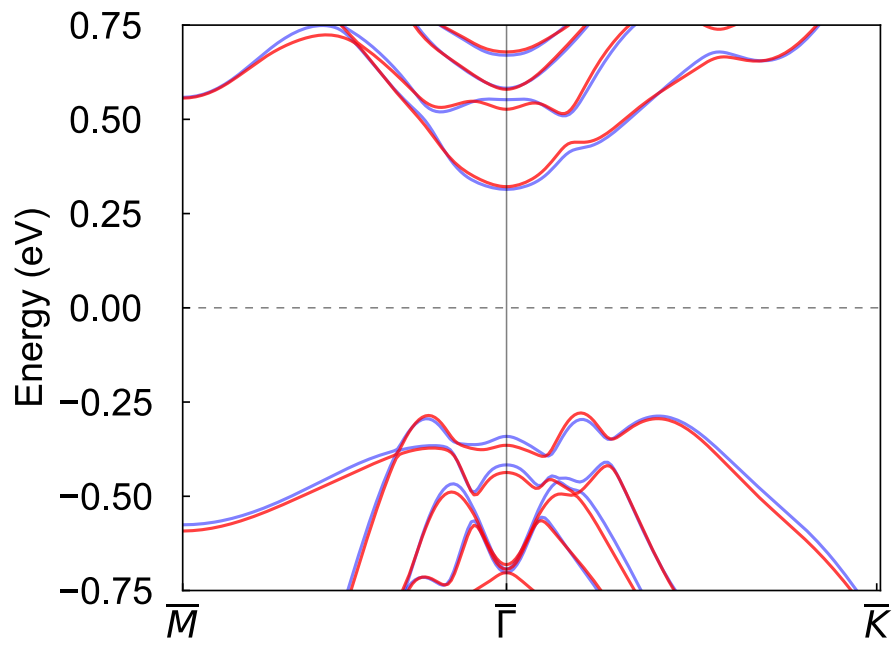


Figure S2. Electronic band structure of 1 SL MnBi₂Te₄, DFT (blue), and 2D electron-phonon Wannier interpolated (red).

Calculated electronic band structure of 1 SL by using standard DFT (blue), and 2D electron-phonon Wannier interpolation (red). We included Hubbard corrections in these calculations.