

Highly Confined Magnetophononic Polaritons and Electron–Phonon Interactions in Atomically Thin MnBi_2Te_4

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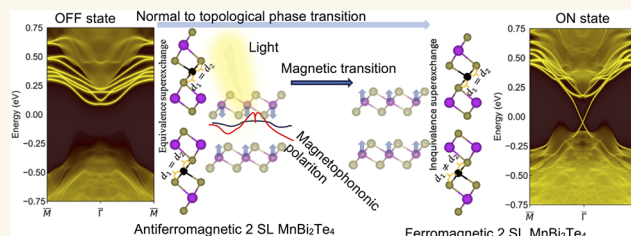
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ABSTRACT: Atomically thin materials can strongly confine light into quanta of lattice vibrations, leading to strong light–matter interactions. Some of these materials can exhibit topology, which protects the edge states embedded inside the bulk from backscattering, while some host magnetism, providing another degree of freedom to control topological properties. The simultaneous observation of all of these phenomena in a single material can lead to various applications by enabling light-controlled topological electronic and nanophotonic devices. Here we demonstrate via first-principles density functional perturbation theory calculations that atomically thin MnBi_2Te_4 is a promising platform for hosting three highly confined, tunable, and long-propagating magnetophononic polaritons. These polaritons strongly modulate exchange interactions, especially the second polariton in 2 SL MnBi_2Te_4 , inducing strong light–matter coupling in the presence of light. Intriguingly, this coupling not only induces an antiferromagnetic-to-ferromagnetic phase transition but also drives an electronic phase transition from trivial to topologically nontrivial states in 2 SL MnBi_2Te_4 . We also show that these polaritonic modes have relatively weak electron–phonon scattering rates and that these scattering rates change with layer thickness and magnetic ordering due to spin–phonon coupling. Overall, our results suggest that MnBi_2Te_4 is a promising material for topological electronics, nanophotonics, and nanophonics capable of providing greater control of data signals via light.

KEYWORDS: topological insulators, phonon polaritons, light–matter interactions, spin–phonon coupling, exchange-coupling modulation, topological switching, electron–phonon interactions



The interaction of light with phonons—quanta of lattice vibrations—can give birth to a hybrid state of matter by confining the wavelength of light into the matter. The formation of such a quasiparticle results in an increased electric field strength and an intense light–matter interaction. This hybrid quasiparticle of matter is called a phonon polariton, which compresses the wavelength of light by orders of magnitude smaller than the wavelength of light in free space.^{1–4} As a result, the phonon polariton leads to strong photonic forces, nonlinearity, large absorption probabilities,⁵ and enhanced dipolar-type emission⁶ in either three-dimensional (3D) or two-dimensional (2D) matter. In particular, in 2D polaritonic matter, extra confinement of the light's energy can be achieved due to the reduced dimensionality.⁷ For example, in 2D hexagonal boron nitride (hBN), the phonon polaritons largely trap light in the far-infrared region resulting in compression of the light wavelength by a factor of several hundred.⁸ Over the past decade, this feature of phonon polaritons has played a critical role in developing polaritonic and next-generation photonic devices, for example, photonic transistors,⁹ and in quantum computing.¹⁰ However, limited tunability, due to the intrinsic nature of phonons, and a narrow phonon energy spectrum¹¹ are two challenging barriers for these applications, which motivate the search for polaritonic materials with desired features along with enhanced tunability.

Recently, an experimental study discovered an intriguing feature in polaritonic materials—a topologically protected polaritonic edge state.¹² This polaritonic topology is similar to electronic topology, where electronic edge states are embedded inside the bulk bandgap and are protected from backscattering at low temperatures (typically up to 100 K).^{13–17} However, this protection can no longer be sustained against strong electron–phonon (e-ph) interactions at relatively high temperatures. For example, e-ph scattering can lead to a phase transition from the topological phase to a trivial phase via electronic band renormalization.^{18,19} Besides e-ph scattering, the topological phase transition can also be induced by strong light–matter interactions due to coupling between phonons and magnetism, modifying the interlayer exchange interaction. Recently, interlayer magnetophononic coupling has been experimentally observed in the magnetic topological insulator MnBi_2Te_4 ,²⁰ suggesting a pathway for achieving a topological

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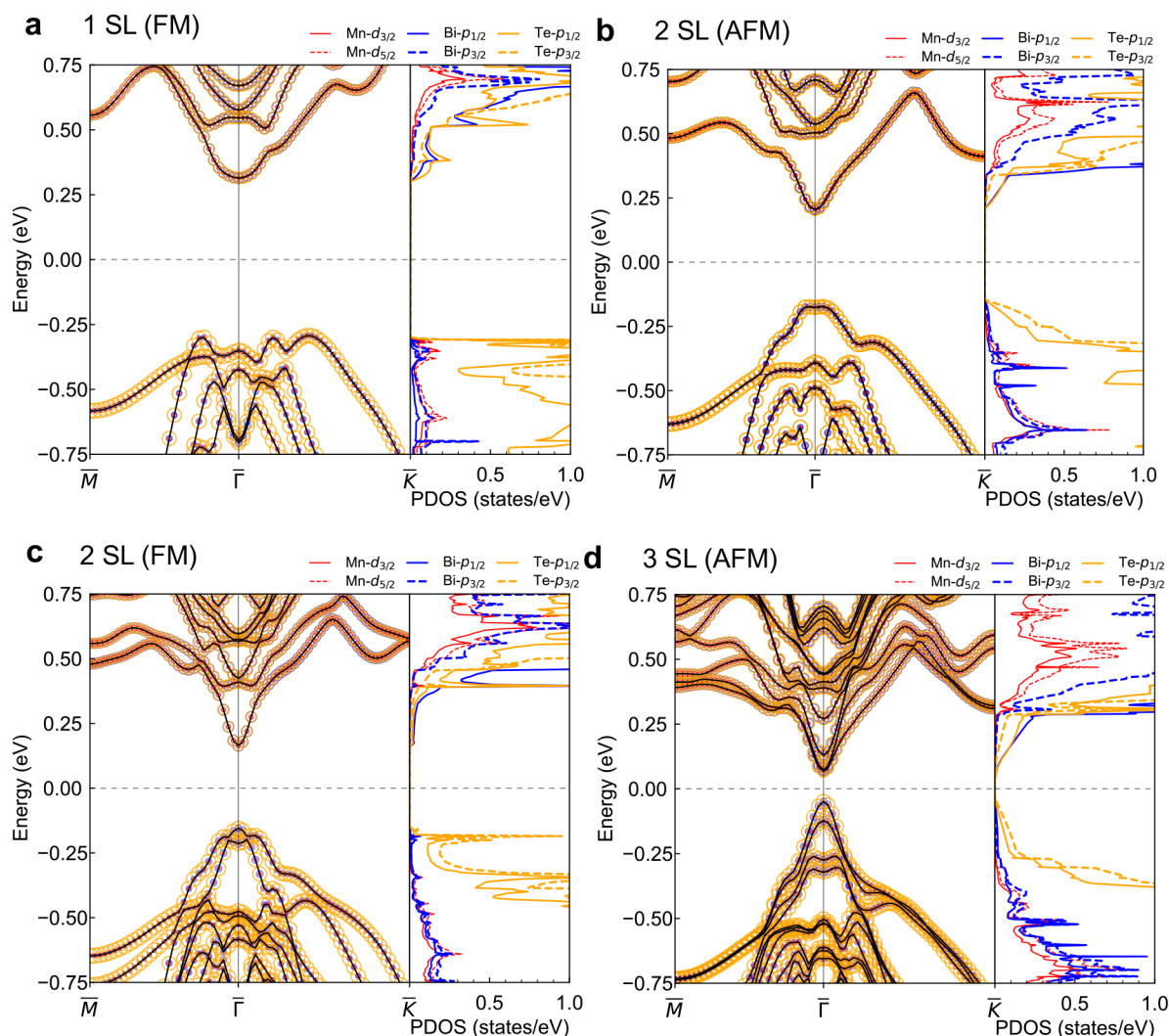


Figure 1. Layer-dependent electronic structures of 2D MnBi_2Te_4 . Atom-projected electronic band structure and orbital-decomposed density of states for 1 SL (a), 2 SL (AFM) (b), “high-field” 2 SL (FM) (c), and 3 SL (AFM) (d) of MnBi_2Te_4 . In the band structures, the red, blue, and orange circles indicate the contributions of Mn, Bi, and Te, respectively, to the energy bands. The left panels in each subfigure show the calculated orbital-decomposed density of states using fully relativistic effects, where the orbitals split based on the total angular momentum $j = l + s$. Here, l and s are the azimuthal and spin quantum numbers. In all cases, we set the Fermi level to zero energy. Figure S1 shows the density of states with the full range of values.

phase transition by strong light–matter interactions, in which one of the key determining factors is the phonon polariton. However, this polaritonic feature is yet to be explored in 2D topological insulators in general or even specifically in MnBi_2Te_4 .

Approaching the 2D limit, MnBi_2Te_4 exhibits various quantum electronic states, for instance, the quantum anomalous Hall (QAH) state as well as axion insulating states depending on its layer thickness.^{21–23} For example, 2D MnBi_2Te_4 with an odd number of layers (≥ 3) hosts the QAH phase, for which the QAH effect can be observed below 1.4 K at zero field, or up to the temperature of 6.5 K under an externally applied magnetic field.^{24,25} Fortunately, 2D MnBi_2Te_4 fulfills one of the important criteria for forming phonon polaritons by hosting Raman- and infrared-active optical phonon modes.^{26,27} However, if 2D MnBi_2Te_4 can host phonon polaritons, we could study not only the phonon polariton but also its relationship with topology, layer thickness, and magnetic ordering. 2D MnBi_2Te_4 is, therefore, an interesting playground for searching phonon polaritons and

their interaction with magnetism and topology. Besides, the phonons in 2D MnBi_2Te_4 can be influenced significantly by e-ph interactions and layer thickness. For instance, a recent study on e-ph and spin–phonon coupling, which was limited to studying only the A_{1g} phonon mode, predicted increased e-ph scattering in thinner samples below the magnetic transition temperature (24 K).^{28,29} Consequently, a detailed study of e-ph interactions for all phonon modes remains essential for achieving light-controlled topological phases as it can reveal their impact not only on topology and magnetism but also on phonon polaritons.

In this work, we search for phonon polaritons and investigate their interactions with electrons for all phonon modes of 2D MnBi_2Te_4 using density functional perturbation theory (DFPT). Our results reveal that MnBi_2Te_4 can host three long-propagating, highly confined, and tunable magneto-phononic polaritons in the far-infrared region. We demonstrate that these polaritons can parabolically modulate exchange interactions and weaken the intrinsic magnetic coupling in the presence of light. Consequently, strong light–matter coupling

Table 1. Computed In-Plane Lattice Parameters and Electronic Bandgap of MnBi₂Te₄ along with the Available Experimental Bandgap^a

Physical quantity	1 SL	2 SL (AFM)	2 SL (FM)	3 SL
<i>a</i> (Å)	4.212	4.217	4.218	4.232
<i>E_g</i> (This work) (meV)	612	380	254	109
<i>E_g</i> (Exp.) (meV)	780 (±100), ³³ 350, ³⁵ >300 ³⁶	300 (±100) ³³	-	109 (±15) ³³
<i>E_g</i> (Calc. by HSE ³³) (meV)	550	200	-	125
<i>E_g</i> (Calc. by PBE + U ²¹) (meV)	321	107	-	66

^aThe fully relaxed lattice parameters (*a*) were calculated by using the PBE functional, while the bandgap (*E_g*) was calculated by using the Engel–Vosko (EV93)³⁴ functional with Hubbard corrections.

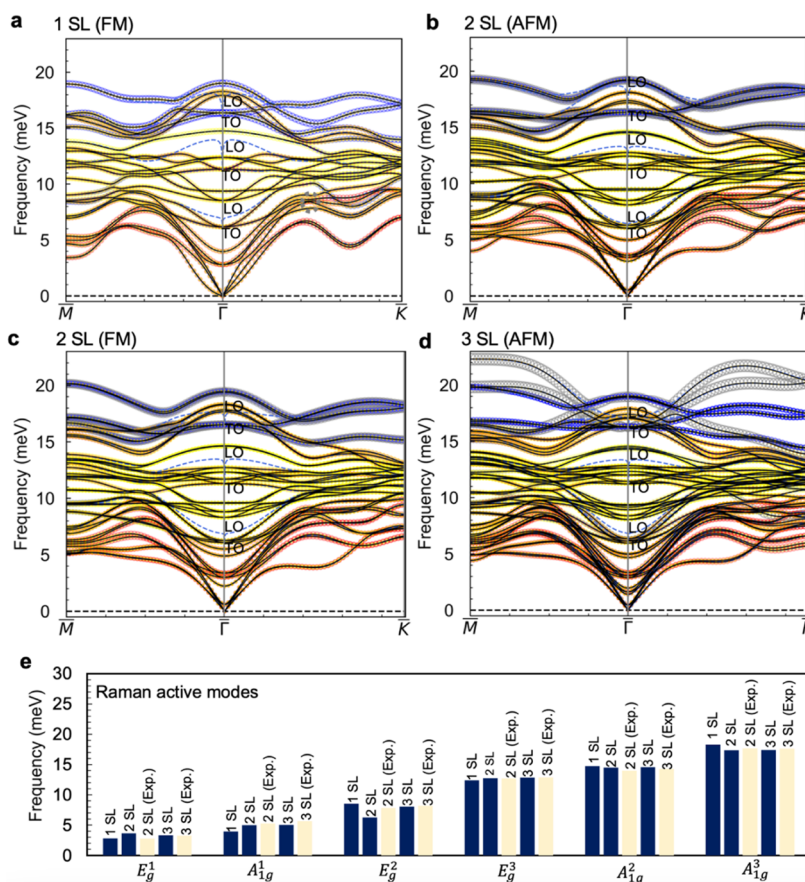


Figure 2. Layer-dependent phonon dispersions and Raman-active phonon modes for 2D MnBi₂Te₄ compared with available experimental data. Atom-projected phonon dispersions of (a) 1 SL (FM), (b) 2 SL (AFM), (c) 2 SL (FM), and (d) 3 SL (AFM) MnBi₂Te₄. In these subfigures, blue, red, and yellow circles indicate the mode contributions (eigenvectors) from Mn, Bi, and Te, respectively. In subfigures (b–d), the contributions of two magnetic Mn sublattices are indicated by blue (parallel spins) and gray (antiparallel spins) circles. The blue dashed lines are the calculated phonon dispersions including 2D nonanalytical corrections, which lead to transverse optical (TO) and longitudinal optical (LO) phonons splitting due to long-range Coulomb interactions. The dotted open circle in (a) near 7.5 meV frequency shows a linear band crossing. For clarity, only one linear band crossing is shown in the figure. (e) Computed phonon frequencies of five Raman-active modes of 2D MnBi₂Te₄ with different layer thicknesses, compared with the experimental data taken from ref 27. No data are available for 1 SL and 2 SL (FM).

induced by the second polariton in 2 SL MnBi₂Te₄ drives an antiferromagnetic-to-ferromagnetic phase transition. We show that this magnetic transition brings topologically robust in-gap edge states in 2 SL MnBi₂Te₄, i.e., a normal-to-topological phase transition. We also show that e-ph scattering rates of these polaritonic modes with electrons change with magnetic ordering and topology due to spin–phonon coupling. We develop an in-depth understanding of the relationship among magnetophononic polaritons, e-ph interactions, and topology, underpinning a step forward toward realizing a light-controlled topological phase. A thorough understanding of such phase transitions can potentially enable read–write functionality in

topological insulators, leading to applications in ultralow-energy quantum electronics, nanophononics, and nanophotonics.

RESULTS AND DISCUSSION

Two-dimensional (2D) MnBi₂Te₄ forms a hexagonal layered crystal structure (Figure S1) with seven atomic Te–Bi–Te–Mn–Te–Bi–Te layers³⁰ (termed as “septuple layers”) stacked along the out-of-plane direction. The structure belongs to the *P3m1* (#166) space group and has *D*_{3d} (–3*m*) point-group symmetry.³¹ The adjacent septuple layers (SLs) are coupled together by weak van der Waals forces. In the case of the single

Table 2. Computed Dynamical and Polaritonic Parameters from DFPT^a

Physical quantity		1 SL		2 SL (AFM)		2 SL (FM)		3 SL	
ϵ_D		15.5		17.2		21.9		24.7	
ϵ		15.5		17.2		21.9		24.7	
ϵ_z		37.1		35.8		45.9		51.8	
P_d (Å ²)		6.08		12.77		13.51		19.90	
Z^*	Mn	4.02		5.65		4.12		3.78	
	Bi	6.26		5.03		6.36		6.57	
	Te	−4.13		−3.93		−4.21		−4.23	
ω_{LO} (meV)	$n = 8$	6.17	$n = 14$	6.04	$n = 14$	6.08	$n = 20$	5.96	
	$n = 13$	11.33	$n = 24$	11.33	$n = 24$	11.31	$n = 35$	11.22	
	$n = 18$	16.37	$n = 34$	16.33	$n = 34$	16.50	$n = 50$	16.25	
γ (meV)	0.09		0.11		0.12		0.15		
$D(c)$	$n = 8$	4.7×10^{-5}	$n = 14$	4.8×10^{-5}	$n = 14$	1.0×10^{-4}	$n = 20$	1.8×10^{-4}	
	$n = 13$	8.7×10^{-5}	$n = 24$	1.6×10^{-4}	$n = 24$	1.7×10^{-4}	$n = 35$	2.6×10^{-4}	
	$n = 18$	2.1×10^{-5}	$n = 34$	8.2×10^{-5}	$n = 34$	4.5×10^{-5}	$n = 50$	5.4×10^{-5}	

^aCalculated 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}), phonon damping factor (γ , which is the inverse of the phonon lifetime), and the polaritonic deceleration factor $D = v_g/c$, where v_g is the polariton group velocity as defined in the main text and c is the light speed in vacuum.

SL ($N_{SL} = 1$)), the spins of the Mn^{2+} ions in the magnetic sublattice align in parallel, resulting in a large magnetic moment of $\sim 5 \mu_B$. For the 2 SL structure ($N_{SL} = 2$), the spins of Mn^{2+} ions in two neighboring magnetic sublattices align in an antiparallel manner, resulting in a zero net magnetic moment with antiferromagnetic (AFM) order. However, in the 2 SL system, it is possible to rotate the spins of Mn^{2+} ions to lie parallel by applying a large external magnetic field to drive a so-called metamagnetic transition.³²

To demonstrate spin–phonon coupling and its impact on electron–phonon (e-ph) interactions in $MnBi_2Te_4$, we also consider the “high field” 2 SL (and 3 SL for a few properties only) structures with ferromagnetic (FM) order. While different spin configurations of Mn^{2+} introduce magnetism, they do not directly alter the electronic topology but can indirectly change topology due to hybridization of the Mn-*d* orbitals with the Bi-*p* and Te-*p* orbitals, which can be seen from the atom-projected band structures and the density of states shown in Figure 1. In the electronic structures, Mn-*d* states (red circles) are far from the Fermi level, while the Bi–Te (blue and yellow circles, respectively) dominate the band minima/maxima near the highest occupied and lowest unoccupied states which determine the topology. However, the topology depends on the SL thickness, and the Mn-*d* states indirectly influence the topology, determining the correct shape of bands in the vicinity of the Fermi level. In the case of 1 SL, the valence band maxima (VBM) and conduction band minima (CBM) have no Dirac-like dispersion (Figure 1a), suggesting the topologically trivial nature of 1 SL $MnBi_2Te_4$, with an indirect bandgap >600 meV.

Compared to 1 SL, the bandgap in 2 SL is substantially reduced (Table 1) due to strong hybridization of wave functions between the top and bottom SLs. Though the CBM has Dirac-like dispersion, the VBM has no Dirac-like dispersion (Figure 1b) in 2 SL AFM, suggesting its trivial nature. To induce a topological feature in this system, the hybridization of Mn-*d* orbitals with Bi-*p* and Te-*p* orbitals must be sufficiently weak.²² Turning the magnetic order from AFM to FM in 2 SL makes the hybridization weak enough (Figure 1c and Figure S1c) and the Dirac like dispersion appears at both the VBM and the CBM (Figure 1c), suggesting

a nontrivial nature, i.e., a zero-plateau QAH phase. In contrast to the 2 SL FM, the VBM and CBM in 3 SL exhibit Dirac-like dispersion (Figure 1d) with a small bandgap of about 100 meV (Table 1), indicating a nontrivial QAH phase (see Figure 5 for further confirmation). It is worth mentioning here that these thickness-dependent results including the bandgaps (Table 1) agree fairly well with experiments³³ and earlier theoretical predictions.²¹

The origin of thickness-dependent electronic structures can be explained via orbital-projected density of states (PDOS) (the right panel of each subfigure in Figure 1). For 1 SL $MnBi_2Te_4$, Bi- $p_{1/2}$ and Te- $p_{1/2}$ states predominantly contribute to the CBM, while Te- $p_{1/2}$ and Te- $p_{3/2}$ have dominant contributions to the VBM (right panel in Figure 1a). With increasing thickness to 2 SL, the contribution of Bi- $p_{1/2}$ increases so that both Bi- $p_{1/2}$ and Te- $p_{1/2}$ states equally contribute to the CBM. On the other hand, the contributions of Te- $p_{1/2}$ and Te- $p_{3/2}$ states to the VBM in 2 SL (AFM) are reduced compared to 1 SL, moving the VBM to the zone center (at Γ). Such alternations in orbital ordering induce a direct bandgap at the zone center. However, the VBM remains slightly flat around the zone center for 2 SL AFM order (Figure 1b). By turning 2 SL AFM order to FM order or by introducing another layer, these orbital alternations (or hybridization) become sufficiently stronger (or weaker) to make the dispersion Dirac-like at the zone center.

The aforementioned changes in the electronic structure with layer thickness and magnetism also hint at potential changes in the phonons in 2D $MnBi_2Te_4$. We now pivot to phonons to explore the changes with thickness and magnetic order. For the 1 SL structure, $MnBi_2Te_4$ has a total of 7 atoms in the unit cell, giving rise to 3 acoustic and 18 optical phonon modes (Figure 2a). Introducing another SL into the structure (i.e., 2 SL) causes some phonon bands to become doubly degenerate, though the dispersions remain quite similar (Figure 2b). Also, the phonon energies are either blue- or red-shifted in the 2 SL (AFM) structure depending on the specific branch of phonons.

Turning the 2 SL AFM to FM also induces either blue or red shifts in the energies of specific phonon modes (Figure 2c), suggesting a spin–phonon coupling.²⁹ A similar effect of layer thickness can be observed in 3 SL (Figure 2d). In particular,

high-energy optical phonons at the zone boundary are blue-shifted significantly. In all cases, Bi atoms predominantly contribute to the acoustic phonons (in which Te and Mn have small contributions) and induce low-energy optical phonon modes (around ~ 0 – 10 meV indicated by red circles in Figure 2), while medium-energy optical phonons of ~ 10 – 15 meV are induced by Te atoms (yellow circles in Figure 2). The magnetic sublattice (Mn^{2+}) gives rise to the highest-energy optical phonons (as indicated by either blue or gray circles for parallel- or antiparallel-spin Mn^{2+}). Intriguingly, the contributions of the two magnetic sublattices become well distinguishable in 3 SL (Figure 2d) compared to all other layer thicknesses and other forms of magnetic ordering (see Note S1 and Figures S3–5 in the SI). It is worth mentioning that the phonon dispersions in Figure 2(a–d) have no negative frequencies, i.e., no imaginary phonon modes, which ensures the dynamical stability of 1–3 SL MnBi_2Te_4 considered here.

Phonon dispersions presented in Figure 2 were obtained with (blue-colored dashed lines) and without (black-colored solid lines) considering long-range interactions. The high-frequency dielectric constant (ϵ_∞) and Born effective charges (Z^*) used in the calculations are listed in Table 2. With increasing thickness of MnBi_2Te_4 , we can tune its dielectric properties, as can be seen from the thickness-dependent dielectric constants. Also, the zone-center polar modes exhibit thickness- (or magnetic-order-) dependent contributions to the dielectric permittivity. Like the dielectric constant, the calculated Born effective charge changes with layer thickness, while the dynamic polarizability increases with layer thickness and depends on the magnetic order. Large dynamic polarizabilities ($P_d > 10 \text{ \AA}^2$) suggest strong long-range interactions in MnBi_2Te_4 . Such long-range interactions can generally split longitudinal optical (LO) and transverse optical (TO) phonons by a few meV, termed as “LO–TO splitting”. The effect of Coulomb interactions on LO–TO splitting can be understood by using the following equation,^{7,37}

$$\omega_{n,\text{LO}}^2 - \omega_{n,\text{TO}}^2 = V(q) \frac{q^2}{\Omega} |\hat{q} \cdot Z_n|^2 \quad (1)$$

where $\omega_{n,\text{LO}}$ and $\omega_{n,\text{TO}}$ are the LO and TO phonon frequencies of n th mode; $V(q)$ is the screened Coulomb interaction, which depends on the dimensionality of the material; Ω is the unit cell area (for 2D); $\hat{q}$ is the phonon unit vector; and $Z_n = \sum_j Z_j^* \hat{\epsilon}_{nj} / \sqrt{M_j}$ is the mode effective charge and is only nonzero for infrared-active LO modes. For a 2D material, $V(q)$ is given by $1/(2q\epsilon_0\epsilon_{2D})$, where ϵ_{2D} is the in-plane dielectric function of the 2D system and ϵ_0 is the free-space permittivity. Consequently, the LO and TO phonons are merged at the zone center for $q \rightarrow 0$ (eq 1), i.e., LO–TO splitting vanishes at Γ for 2D materials.^{7,37} Indeed, the phonon dispersions of MnBi_2Te_4 calculated based on 2D nonanalytic corrections confirm the vanishing LO–TO splitting at Γ , as shown by the blue dashed lines in Figure 2, while the splitting exists outside the zone center. For further clarity, we also plot the LO phonons of MnBi_2Te_4 in Figure S6 (see SI) and compare them with those of a well-studied polar semiconductor-hexagonal boron nitride (hBN). The existence of well-distinguishable LO–TO phonons confirms the presence of long-range Coulomb interactions in 2D MnBi_2Te_4 regardless of its layer thickness.

To perform further analysis of LO phonons in 2D MnBi_2Te_4 , we calculate phonon modes and their IR or

Raman activity, as listed in Table S1 and Table S2. The zone-center phonon modes in irreducible representations can be written as $\Gamma_{\text{vib}} = \Gamma_{\text{equiv}} \oplus \Gamma_{\text{vect}}$ ³⁸ where Γ_{equiv} stands for the equivalent representation and represents the modes for the invariant atoms under group symmetry operations. For the point group symmetry D_{3d} , the vector representation becomes $\Gamma_{\text{vect}} = E_u \oplus A_{2u}$. Then, we find the mode representation by expanding it into irreducible representations for 1 SL MnBi_2Te_4 as

$$\Gamma_{\text{vib}} = 3A_{1g} \oplus 4A_{2u} \oplus (2 \times 3)E_g \oplus (2 \times 4)E_u$$

where E_g and E_u are the doubly degenerate bands and the representation gives a total of $3N = 21$ modes. Here, g and u stand for even and odd transformations under the inversion symmetry of MnBi_2Te_4 . For layer thickness $N_{\text{SL}} > 1$, we can generalize the above representations as follows,

$$\begin{aligned} \Gamma_{\text{vib}} = & (3N_{\text{SL}} + 1)A_{1g} \oplus (4N_{\text{SL}} - 1)A_{2u} \\ & \oplus ((2 \times 3N_{\text{SL}}) + 2)E_g \oplus ((2 \times 4N_{\text{SL}}) - 2)E_u \end{aligned} \quad (2)$$

which leads to a total of $3N = 42$ and $3N = 63$ modes for $N_{\text{SL}} = 2$ and $N_{\text{SL}} = 3$, respectively. From the above representations, we can easily identify IR- and Raman-active modes, i.e., the modes with even transformation under the inversion operation are Raman-active and modes with odd transformation are IR-active. In MnBi_2Te_4 , the A_{1g} and doubly degenerate E_g modes (eq 2) are Raman-active, while the nondegenerate A_{2u} and doubly degenerate E_u modes are IR-active. However, the degenerate (IR) LO and TO phonons (E_u) can only be split by the (nonanalytic) long-range Coulomb interactions. The E_g modes in 2–3 SL MnBi_2Te_4 also exhibit a clear splitting for finite in-plane momentum (Figure S6) while they remain strictly doubly degenerate at the Γ point. This behavior cannot be regarded as LO–TO splitting, as E_g is a Raman-active gerade mode with zero effective charge. Therefore, this mode can neither couple to long-range dipole–dipole interactions (as these systems are centrosymmetric) nor long-range Coulomb interactions, rather it originates from interlayer hybridization and spin–phonon coupling (see Note S2 in the SI for further detailed information). In particular, the 1 SL has 3 infrared (IR)-active LO phonons, while the 2 SL (and others) has more than 3 IR-active LO phonons (where only 3 are prominent) and 3 Raman-active LO phonons (red lines in Figure S3). We also calculate the frequency of 5 Raman modes (A_{1g}, E_g) and compare with the experimental frequencies at 5 K²⁷ (Figure 2e) which shows excellent agreement between the computed and experimental results. Currently, there are no available experimental data for IR-active modes; the calculated IR spectra for these modes including IR-active LO phonons are presented in Supplemental Figure S7d.

Both theory⁷ and experiments^{11,38} have shown that IR-active LO phonons in 2D polaritonic materials exactly satisfy all the necessary conditions for a strongly confined and evanescent electromagnetic mode, i.e., a quasiparticle–phonon polariton (see Note S4 in the SI). This quasiparticle is similar to that of the IR-active surface phonon polariton in a 3D material, in which phonon dispersions exhibit LO–TO splitting. In the case of 2D MnBi_2Te_4 , only IR-active degenerate E_u modes can therefore fulfill all the necessary conditions for phonon polaritons (see Note S4 in SI for further explanation). We identify three such magnetophononic polaritonic modes (Figure S6 in the SI) in 2D MnBi_2Te_4 , where we refer to

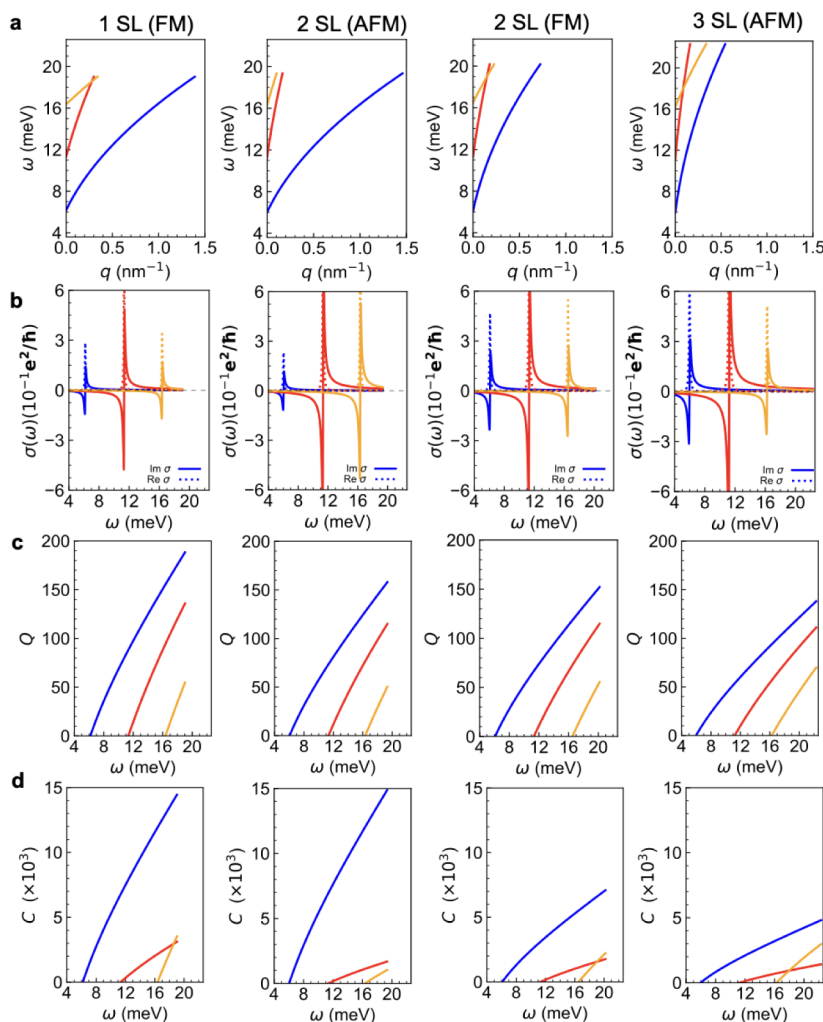


Figure 3. Layer-dependent properties of phonon polaritons in 2D MnBi_2Te_4 . (a) Polariton dispersions of three LO phonons in 2D MnBi_2Te_4 for different layer thicknesses and magnetic configurations. First ($n = 8, 14, 20$ for 1 SL, 2 SL, and 3 SL), second ($n = 13, 24, 35$ for 1 SL, 2 SL, and 3 SL), and third ($n = 18, 34, 50$ for 1 SL, 2 SL, and 3 SL) polaritons are shown by blue, red, and orange color lines, respectively. The three polariton branches plotted in Figure 3a are intrinsic and not an artifact of our representation. (b) Real (dashed lines) and imaginary (solid lines) parts of the lattice optical conductivity. (c) Propagation quality factor $Q = \text{Im } \sigma / \text{Re } \sigma$, which determines the distance a 2D phonon polariton can travel before it decays in wavenumber. (d) Confinement factor (C), which measures the amount of compression of the light wavelength λ confined inside a phonon polariton (2D) compared to the wavelength of light in free space (λ_0).

magnetically coupled phonon polaritons as magnetophononic polaritons. We now turn to explore the figures of merit for these polaritons in 2D MnBi_2Te_4 . In the microscopic theory, three quantities mainly determine the figures of merit of polaritons, namely,

$$Q = \frac{\text{Im } \sigma}{\text{Re } \sigma}, \quad C = \frac{q}{q_0}, \quad D = \frac{v_g}{c} \quad (3)$$

where Q is the propagation-quality factor, which is defined as the ratio of the imaginary and real parts of the phonon optical conductivity (σ) and measures the propagation distance traveled by the polariton within its lifetime. In eq 3, C is the confinement factor, defined as the ratio of the polariton momentum (q) and the free light momentum (q_0), and measures the amount of compression of the wavelength of light confined within the phonon polariton compared to its wavelength in vacuum. D in eq 3 is the deceleration factor, which is defined as the ratio of the IR-active 2D LO phonon group velocity (v_g) and the light velocity in vacuum (c), and

measures the deceleration of light confined within the phonon polariton. To evaluate the figures of merit in eq 3, we therefore need to calculate the phonon optical conductivity (σ) and the dispersion of phonon polaritons within the long-wavelength limit.

In the long-wavelength limit ($q \rightarrow 0$), the phonon optical conductivity can be calculated by using the random-phase approximation based on the LO phonon frequency with its corresponding ionic displacements, as follows:^{7,26}

$$\sigma(\omega) = \frac{-i\omega |\hat{q} \cdot Z_n|^2}{\Omega(\omega_{LO}^2 - \omega^2 - 2i\gamma\omega)} \quad (4)$$

where ω is the phonon-momentum-dependent frequency and γ is the phonon linewidth (also termed the phonon damping factor) due to phonon–phonon interactions. Based on this, we can easily calculate the dispersion of 2D phonon polaritons by using the following relation:^{7,26}

$$q = \frac{2i\omega\epsilon_0}{\sigma} = \frac{-2\Omega\epsilon_0(\omega_{LO}^2 - \omega^2 - 2i\gamma\omega)}{l\hat{q}\cdot Z_n l^2} \quad (5)$$

Then, we can define the group velocity of a 2D LO phonon by using microscopic parameters as follows,

$$v_g = \frac{l\hat{q}\cdot Z_n l^2}{4\epsilon_0\omega_{LO}\Omega} \quad (6)$$

This microscopic group velocity is directly proportional to the deceleration factor ($D = v_g/c$). We study the deceleration of light in three magnetophononic-polariton modes in 2D MnBi₂Te₄ by calculating the group velocity (v_g) defined in eq 6, as listed in Table 2. The deceleration factors of all polaritons in 2D MnBi₂Te₄ are ultralow ($10^{-5} - 10^{-4}$), with group velocities ~ 4 – 5 orders of magnitude lower than the speed of light in free space (10^8). The values are comparable to those of ordinary phonon polaritons in monolayer hBN (1.8×10^{-5}).²⁶ These phonon polaritons can therefore drastically slow down light by a factor of 10^{-5} – 10^{-4} .

Due to the ultralow deceleration factors—that are directly proportional to the phonon optical conductivity—the monotonic increase of the magnetophononic-polariton momentum with frequency directly follows from the dispersion relation (eq 5). In 2D MnBi₂Te₄, the polariton momentum satisfies $q(\omega) \propto \omega/|\sigma(\omega)|$; the sharp fall of the conductivity ($|\sigma(\omega)|$) near the resonance leads to the very rapid growth of $q(\omega)$ in the long-wavelength limit ($q \rightarrow 0$). Away from the resonance frequency, the conductivity varies smoothly with phonon energy. Consequently, such variation leads to produce a gradual increase of the polariton momentum with energy (Figure 3a). In contrast, the magnetophononic polaritons with the largest momentum (i.e., the first blue line) and the third (yellow line) in Figure 3a have the lowest phonon optical conductivity (Figure 3b).

According to eq 4, the phonon damping factor, mode effective charges, LO phonon frequency, and the unit-cell area mainly determine the shape of the phonon optical conductivity curves (Figure 3b). In all cases, the second polariton has higher phonon optical conductivity (both real and imaginary parts) (Figure 3b) due to its large mode effective charge (Figure S7c) than all other polaritons. Although the damping factor (0.09–0.15 meV) is comparable to that of hBN (0.14 meV),²⁶ MnBi₂Te₄ has an approximately smaller value of the mode effective charges for the first and third polaritons, i.e., $Z_n \approx 0.46$ – 0.89 e/(amu)^{1/2}, as shown in Figure S7c, than that of 2D hBN (~ 1.1 e/(amu)^{1/2}),²⁶ while the second polariton has a comparable mode effective charge, i.e., $Z_n \approx 0.84$ – 1.17 e/(amu)^{1/2}. Also, MnBi₂Te₄ has a 10–25 times lower LO frequency (Figure S6) and a 2 times larger unit-cell area (Table 1) than that of hBN.²⁶ For the lower LO frequency and larger unit cell of 2D MnBi₂Te₄, the peak value of the phonon optical conductivity is ~ 1.5 times lower than that of 2D hBN (Figure S8c).²⁶ Also, the phonon optical conductivity becomes higher for larger Z_n (Figure S7c), while the wavelength of the phonon polariton becomes shorter (Figure 3a) for larger (eq 5) Z_n .

In particular, the second magnetophononic polariton with a frequency of 19 meV (shown in red in Figure 3a) has a significantly shorter wavelength than the measured wavelength of any phonon polariton so far and even much shorter than the wavelength of the plasmon excitation in graphene (>200 nm).³⁹ We specifically highlight this frequency because it lies in a suitable range for approximately describing the magneto-

phononic polariton (frequency and wavelength), which has the highest oscillator strength and highest optical conductivity. This frequency also lies in the terahertz (THz) and far-infrared (FIR) spectral ranges that are suitable for practical experimental measurements and potential applications, making it particularly relevant for our study. Moreover, the second polariton has a momentum of 0.32 nm^{-1} , indicating that the polariton's effective wavelength ($\lambda_{\text{effective}} = 2/\pi k \approx 18 \text{ nm}$) is extremely small. This is significantly smaller than the free-space wavelength of a photon at 19 meV, which is approximately $65 \mu\text{m}$. Such a large momentum-to-energy ratio indicates that the electromagnetic field of this magnetophononic polariton is highly confined to the nanoscale, making it an “ultraslow” light-matter quasiparticle. It is worth to mention that polariton wavelengths depend on the in-plane wavevector, specific measurement geometry, and experimental conditions. Our comparison above with the plasmon in graphene therefore aims only to indicate the relative confinement scale and expected experimental detectability. We do not intend to equate the absolute plasmon wavevector or conditions between our theory and real experiments in the referenced paper.³⁹ This is also valid in the rest of the other comparisons here.

Despite having a short wavelength, the widely used experimental technique, scattering scanning near-field optical microscopy (s-SNOM), can measure these 2D magnetophononic polaritons, which are closer to the LO frequency, where dissipation near the LO phonon is low and confinement is small.⁷ Fortunately, the dissipation near the LO phonon in 2D MnBi₂Te₄ is lower compared to hBN, i.e., a higher propagation quality factor (e.g., for the first polariton, $Q \approx 100$ – 150 at $q = 1 \text{ nm}^{-1}$) in 2D MnBi₂Te₄ (Figure 3c) than in 2D hBN ($Q < 25$,⁷ $Q < 75$,²⁶ $Q \approx 45$ (Figure S7 in the SI)). The large values of the imaginary part of the phonon optical conductivity (Figure 3c) lead to higher propagation quality factors. Also, the propagation quality factor is reduced with layer thickness and (AFM to FM in 2 SL) magnetic ordering. Increasing number of layers leads to multiple phonon modes and strong phonon–phonon scattering that increases the damping factor (Table 2). As the quality factor Q depends on the phonon modes (eq 4), the values of Q change with the layer thickness and magnetic ordering. However, this effect gets weaker for the high-energy magnetophononic polaritons, i.e., for the second and third polaritons.

In contrast to the propagation-quality factor, the confinement factor (C) in 2D MnBi₂Te₄ increases with layer thickness and magnetic ordering in a different manner due to their corresponding dispersions (Figure 3a). In particular, the confinement factor reduces from single to three septuple layers (1 SL $\rightarrow$ 2 SL FM $\rightarrow$ 3 SL AFM); however, this trend applies only to the second and third polaritons in the 2 SL AFM. This reduction trend arises not only from differences in magnetic ordering due to spin–phonon coupling but also from the strongest increase in interlayer hybridization and dielectric screening when the first (the second in the case of 3 SL) additional SL is introduced. The hybridization leads to a delocalization of the electronic states across the SLs (Figure 1). When it is 2 SL AFM, however, the electronic structure reorganizes due to the different magnetic symmetry of even-layer systems, reduced incremental screening changes for the first polariton, and the different magnetic interlayer exchange interactions between the top and bottom SLs. As a result, the electronic wave function of the corresponding mode becomes

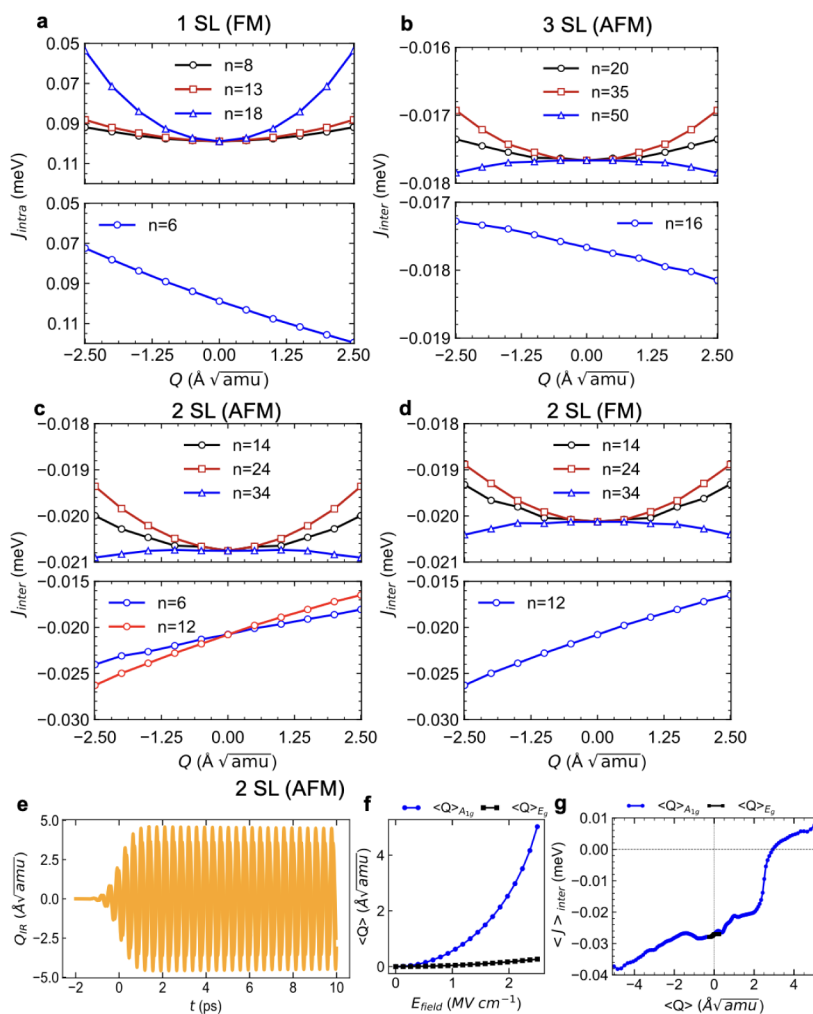


Figure 4. Exchange coupling modulations in MnBi_2Te_4 and magnetic phase transition under light. (a) Computed intralayer exchange coupling (J_{intra}) of 1 SL MnBi_2Te_4 as a function of the mode displacement amplitude (Q) for polaritonic modes (labeled by the mode index (n)) (top subplot) and one Raman mode (A_{1g}) (bottom panel). (b–d) Calculated interlayer magnetic exchange coupling (J_{inter}) of 3–2 SL MnBi_2Te_4 as a function of the corresponding mode displacement amplitude (Q) for polaritonic modes (top subplot) and one Raman mode (modes) (A_{1g}) except for two Raman modes ($A_{1g}^{(1)}$ (6th mode) and $A_{1g}^{(2)}$ (12th mode)) of 2 SL AFM (bottom of panel c). (e) Infrared (IR) mode displacement amplitude under an incident laser light pulse in 2 SL AFM as a function of time (at a 1.0 MV/cm pump field and a 0.7 ps pulse). (f) Time-averaged displacement amplitude of the nonlinearly light-coupled breathing ($\langle Q \rangle_{A_{1g}}$) and shear $\langle Q \rangle_{E_g}$ mode with driving frequency $\Omega = \Omega_{\text{IR}}$ in 2 SL AFM as a function of the pump electric field. (g) Time-averaged interlayer exchange coupling as a function of the averaged breathing and shear mode displacements in 2 SL AFM. As the intrinsic phonon-polariton in 2D systems occurs at long wavelengths, the corresponding Γ -point IR phonon eigenvector represents its ionic displacement pattern here.^{7,26} This figure determines how the magnetic exchange interactions $J(Q_{\text{on}})$ depend on the actual ionic coordinates Q_{IR} and Q_{R} (we have omitted the subscript R in some cases within the manuscript for simplicity), but it does not specify the absolute ionic displacement for a given polariton occupation or for normalized magnetophononic polariton excitation amplitudes. Converting from a normalized polariton amplitude or occupation to Q_{IR} would require the phononic Hopfield component of the polariton. Here, the IR-active mode is the polariton-forming driver, while the Raman-active mode is an anharmonically activated assistant. Thus, the AFM-to-FM transition in 2 SL MnBi_2Te_4 occurs at a specific ionic displacement of the phononic component, with assistance from the Raman mode. However, this result does not imply stabilization of an equilibrium FM ground state; the process is reversible upon removal of light illumination.

more localized again, leading to enhanced confinement (C) of the first polariton in the 2 SL AFM.

Typically, the C factor for the second polariton (red line in Figure 3d) in 2D MnBi_2Te_4 is of the same order of magnitude ($\sim 10^3$ at $q = 1 \text{ nm}^{-1}$) compared to 2D hBN at the same q (Figure S8e in the SI).²⁶ Recent benchmark studies for the confinement factor in hBN found $C = 60$ in an SNOM experiment⁸ and $C = 500$ in an electron energy loss spectroscopy (EELS) experiment,⁴⁰ and suggested good agreement between theory and experiments. As the confinement becomes extreme, the polaritonic mode slows down and

stores energy more efficiently, leading to very large q . At the same time, the effective wavelength becomes very short, making the confinement very high. These trends of increase do not violate fundamental conservation laws, though there will always be in practice some limit (material losses, nonlocality, saturation, etc.) that will eventually prevent extremely large q or an unbounded confinement factor.

The first polariton (blue line) for $N_{\text{SL}} = 1$ and the first and third (yellow line) polaritons for $N_{\text{SL}} \geq 2$ have 1 order of magnitude higher ($\approx 10^4$ at $q = 1 \text{ nm}^{-1}$ for trivial insulating phases, i.e., 1 SL FM and 2 SL AFM) confinement than 2D

hBN at the same q (Figure S8e in the SI).²⁶ This points to the possibility that larger values of the confinement factor in 2D MnBi₂Te₄ than in 2D hBN can be experimentally measured. Besides, for different thicknesses and magnetic orders, the confinement factors in 2D MnBi₂Te₄ exhibit a different rate of increase with frequency (Figure 3d). Besides, the high confinement mentioned above, phonon polaritons can have topological protection.¹² A more careful analysis of phonon dispersions (Figure 2a) by calculating the topological invariant (Z_{2ph})²⁸ points out that some of the polaritonic modes have nonzero Z_{2ph} and the corresponding phononic edge states (Figure S8 in SI), suggesting their topological nature. These polaritonic modes (see Note S5 and S6 in the SI for detailed information) are expected to be free from any polariton-induced backscattering within a finite q and corresponding frequency window.¹² However, the topological protection can exist as long as there is no excitation, perturbation, or mode mixing, which can break the symmetry responsible for the degeneracy of the polaritonic mode. As these polaritonic features change with magnetic ordering through spin–phonon coupling, it can be quite intriguing to explore how these polaritonic modes modulate magnetic exchange interactions and topological properties. A useful way to quantify the static modulation of magnetic exchange induced by the polaritonic modes is to map DFT total energies, which should be calculated along the displacement amplitude q for the corresponding mode onto a Heisenberg Hamiltonian,

$$\hat{H}(Q) = E_{nm}(Q) - \sum_{\langle ij \rangle} J_{ij}(Q) \hat{S}_i \cdot \hat{S}_j \quad (7)$$

Here $E_{nm}(Q)$ represents the nonmagnetic ground state energy (that is canceled eventually) and $\hat{S}_i$ stands for localized spin operators. In this case, we calculate total energies $E^{(con)}(Q)$ for a set of distinct magnetic configurations (con = FM/AFM patterns) at static displacements Q , and then we evaluate the exchange coupling ($J_{ij}(Q)$) for each displacement by solving the equation (see Note S8 in the SI for further detailed information):

$$E^{(con)}(Q) = E_{nm}(Q) - \sum_{\langle ij \rangle} J_{ij}(Q) (\mathbf{S}_i \cdot \mathbf{S}_j)_{con} \quad (8)$$

This method can precisely capture (static) phonon-induced magnetic exchange modulation. At zero displacement, the intralayer exchange coupling (J_{intra}) is 0.1 meV (with excellent agreement with the experimental value $SJ_1 = 0.26$ meV, $S = 5/2$),⁴¹ suggesting intralayer ferromagnetic coupling and consistency with the earlier studies [ref 41]. A clear modulation in $J_{intra}(Q)$ directly encodes the strength of the light–matter coupling (Figure 4a). Intriguingly, all polaritonic modes induce a parabolic (quadratic) dependence $J_{intra}(Q_{IR}) \approx J_{intra}(0) + \alpha Q_{IR}^2$ (Figure 4), where α is the spin–phonon coupling coefficient. This quadratic behavior indicates that symmetry suppresses the linear part of Q_{IR} contribution or filters it out, remaining the modulation effect at second order. As 2D MnBi₂Te₄ is a centrosymmetric system, the two opposite displacements $+Q_{IR}$ and $-Q_{IR}$ induced by the polaritonic modes can be nearly equivalent symmetrically for the exchange-coupling geometry, despite their ability to reverse the dipole. Consequently, the exchange coupling becomes an even function of the polaritonic mode displacement to second order. On the other hand, linear variations may cancel in competing hopping paths, while the change of hybridization,

which contributes to the exchange nonlinearly, varies the coupling as Q_{IR}^2 . Such deviations from linear behavior also suggest the presence of higher order magnetoelastic effects beyond the harmonic approximation.

However, $J_{intra}(Q)$ changes linearly for the Raman active (A_{1g}) mode (bottom panel of Figure 4a), suggesting that A_{1g} mode can directly change the interlayer Te–Te separation (superexchange mediators) and the Mn–Te–Mn exchange geometry (e.g., bond length or bond angle). Analogous to the 1 SL FM phase, the breathing A_{1g} mode leads to linear modulation of the interlayer exchange coupling ($J_{inter}(Q)$ in all other phases (bottom panels in Figure 4b–d), while between $A_{1g}^{(1)}$ (6th mode) and $A_{1g}^{(2)}$ (12th mode) in 2 SL AFM, the latter one has much stronger interlayer exchange modulation than the former one, which is consistent with an earlier experimental report,²⁰ even though $A_{1g}^{(1)}$ mode leads to a significant change in the interlayer Mn–Mn distance (Figure S10). Although polaritonic modes lead to parabolic modulation, the shape of the parabola for the third polaritonic mode (blue-colored curves in Figure 4) is inverted in all other cases due to spin-layer symmetry compared to the 1 SL FM phase.

To explore this intriguing feature, we model the light-driven polaritonic dynamics within the 2D nonlinear-phononics approach,⁴² where we define the potential energy ($V(Q_x, Q_y, Q_a, Q_x, Q_y; t)$) in terms of the mass-weighted mode displacement amplitude Q (in $\text{\AA}\sqrt{\text{amu}}$). As 2D MnBi₂Te₄ crystallizes in a trigonal structure with the D_{3d} point group, the symmetry condition allows cubic coupling between Raman and infrared modes, where $(\Gamma_{IR} \otimes \Gamma_{IR}) \otimes \Gamma_{IR} \supset A_{1g}$. Thus, $E_u \otimes E_u$ IR modes coupling consists of both A_{1g} and E_g Raman modes, which enables simultaneous rectification of shear (in-plane E_g) and breathing (out-of-plane A_{1g}) Raman modes under E_u driving. As a result, we consider a resonantly driven IR-active E_u mode (we denote the displacement amplitude for this mode as $\mathbf{Q}_{E_u} = (Q_x, Q_y)$) coupled to two Raman-active phonon modes—one is the fully symmetric A_{1g} mode (with displacement amplitude Q_a), and the other is the doubly degenerate E_g mode (with displacement amplitude $\mathbf{Q}_{E_g} = (Q_x, Q_y)$).

The potential energy can therefore be written as⁴²

$$\begin{aligned} V(Q_x, Q_y, Q_a, Q_x, Q_y; t) = & \frac{1}{2} \Omega_{E_u}^2 (Q_x^2 + Q_y^2) + \frac{1}{2} \Omega_{A_{1g}}^2 Q_a^2 \\ & + \frac{1}{2} \Omega_{E_g}^2 (Q_x^2 + Q_y^2) \\ & + \gamma_3^{(A_{1g})} Q_a (Q_x^2 + Q_y^2) + \gamma_3^{(E_g)} \\ & [Q_x (Q_x^2 - Q_y^2) + 2Q_y Q_x Q_y] \\ & + \frac{1}{3} \beta_3^{(A_{1g})} Q_a^3 + \frac{1}{3} \beta_3^{(E_g)} (Q_x^3 - 3Q_x Q_y^2) \\ & - \mathbf{Z}^* \cdot \mathbf{E}(t) \cdot \mathbf{Q}_{E_u} \end{aligned} \quad (9)$$

Here $\mathbf{Z}^*$ stands for the mode-effective charge (obtained from DFPT and shown in Figure S2 in the SI), $\mathbf{E}(t)$ stands for a pulsed electric field, Ω stands for the corresponding phonon mode frequency, γ_3 denotes the infrared–Raman cubic coupling, and β_3 stands for intrinsic Raman anharmonicities. Then, we solve the damped, driven Euler–Lagrange equations ($\dot{Q}_i(t) + 2\Gamma_i \dot{Q}_i(t) + \frac{\partial V}{\partial Q_i} = 0$) for each mode displacement with a Gaussian-like electric-field profile

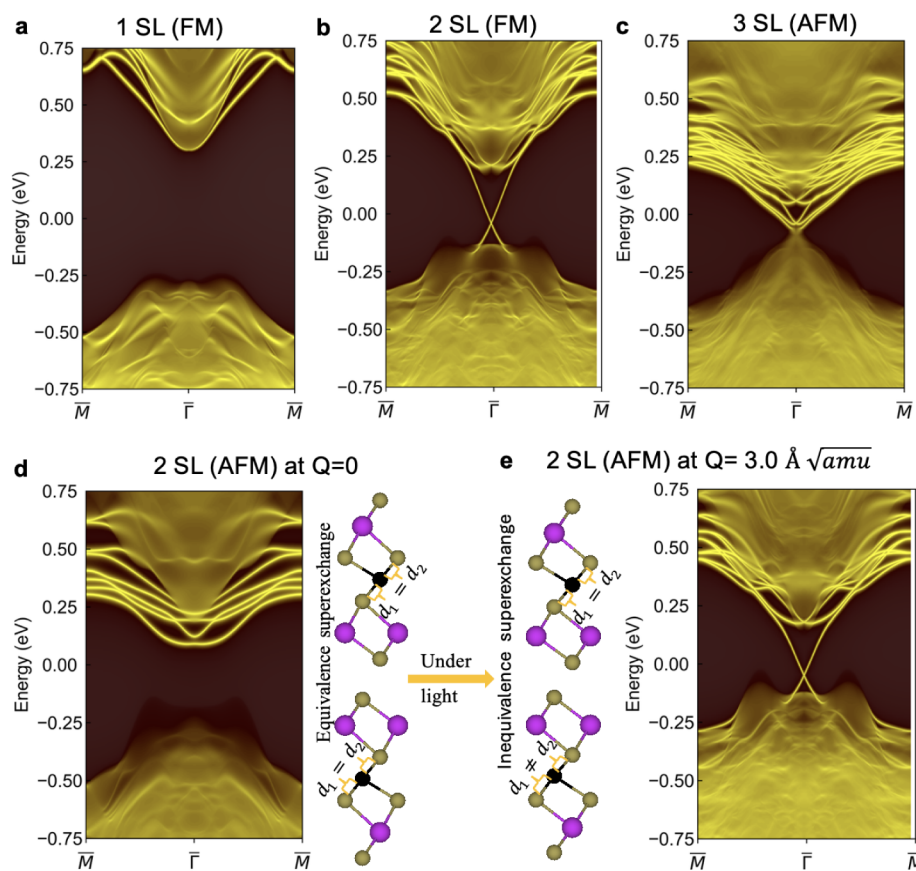


Figure 5. Topological edge states of 2D MnBi_2Te_4 for different magnetic orders, thicknesses, and under light. (a–c) Edge energy spectrum along a defined momentum path of 1 SL FM, 2 SL artificial FM, and 3 SL AFM MnBi_2Te_4 , respectively. Nanoribbons used in these calculations are shown in Figure S1 of the SI. The zero energy indicates the Fermi level. (d) Trivial edge spectrum of 2 SL AFM in the absence of light (i.e., $Q = 0$, no light-induced displacement). The right panel of this figure highlights the equivalence superexchange mediator interbond lengths among Te–Mn–Te. (e) The left panel shows how this equivalence superexchange becomes inequivalence due to the light-induced displacement (the second polaritonic mode drives the change with assistance from breathing mode A_{1g} (12th mode)). The right panel demonstrates how such inequivalence superexchange-induced FM order leads to nontrivial edge states around the Fermi level in the presence of light (i.e., at $Q = 3.0 \text{ \AA}\sqrt{\text{amu}}$). In all figures, the brightest lines represent the contributions from edge modes.

$\mathbf{E}(t) = E_0 e^{-t^2/(2\tau^2)} (\sin \Omega t, \cos \Omega t)$. Eventually, we calculate the dynamic exchange coupling by using the equation:⁴²

$$\langle J(t) \rangle_{\text{inter}} = J_{0(\text{inter})} + \delta J \hat{\delta} \cdot \langle Q(t) \rangle + \delta J^{(2)} [\hat{\delta} \cdot \langle Q(t) \rangle]^2 + O^3 \quad (10)$$

where $J_{0(\text{inter})}$ is the equilibrium interlayer exchange, $Q(t)$ is the corresponding mode ($A_{1g}^{(2)}$ and E_g) displacement coordinate, δJ is the coupling strength between the polaritonic displacement and the exchange, and $\hat{\delta}$ determines the direction of the displacement. Here we calculate the interlayer exchange coupling by using the magnetic force theorem (MFT) to capture the dynamic effect correctly. Readers are suggested to see Note S7 and Note S8 in the SI for details of all these calculations including the cubic-coupling anharmonicities (listed in Table S4).

The calculated time evolution profile shows (Figure 4e) that the polaritonic coordinate (Q_{IR}) exhibits coherent oscillations at the polaritonic mode frequency, whose displacement amplitude is modulated by the electric field pulse. When the field pulse reaches its maximum intensity around the center of the time window, the polaritonic oscillations gain their largest displacement amplitude, which leads to a dense and high-

frequency oscillatory pattern. These oscillations persist over several cycles as the system has no significant damping on the timescale of the pulse, which is consistent with the long phonon lifetime reported in a recent experimental study (13.3 ps²⁹). However, the value near $5 \text{ \AA}\sqrt{\text{amu}}$ does not indicate a transition threshold, as the oscillation remains approximately centered around zero, i.e., $\langle Q_{\text{IR}}(t) \rangle \approx 0$. The magnetic transition can be accommodated only by the induced, time-averaged Raman breathing displacement.

Intriguingly, the variations in the time average of the displacement amplitude $\langle Q \rangle_{\text{RMS}}$ of the breathing and shear modes with the peak electric field E_0 reflect the nonlinear excitation pathway (Figure 4f), which is driven by the oscillatory character of Q_{IR} (Figure 4e). The polaritonic mode couples linearly to the electric field. As a result, the amplitude exhibits an approximately linear relationship with E_0 at low electric fields. On the other hand, E_g and $A_{1g}^{(2)}$ modes are not directly coupled to the field but are indirectly assisted through cubic anharmonic couplings to the driven E_u mode. Consequently, their effective driving forces change quadratically with the polaritonic mode amplitude, and hence, these forces increase nonlinearly with E_0 . This behavior leads to a superlinear increase of $\langle Q \rangle_{\text{RMS}}$ with electric field, where damping and anharmonic couplings modify the trend at

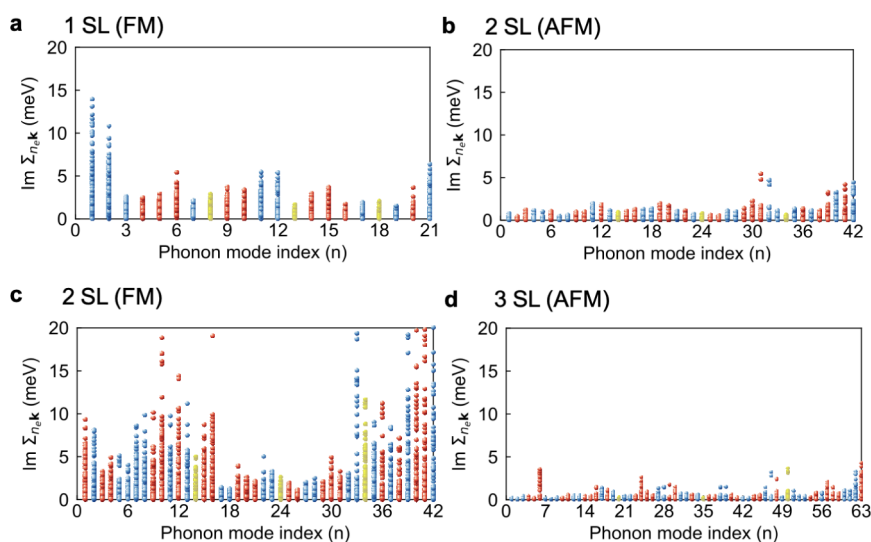


Figure 6. Phonon-mode-resolved electron self-energy of 2D MnBi_2Te_4 from electron–phonon scattering, with nonanalytical corrections, at 20 K. Panel (a) shows imaginary part of the electron self-energy for different phonon modes (along x -axis) of single septuple-layer (SL) MnBi_2Te_4 , where a single SL MnBi_2Te_4 contains 7 atoms per unit cell and thus has 21 phonon modes, panel (b) corresponds to the two SL MnBi_2Te_4 , which naturally has 42 phonon modes due to 14 atoms per unit cell. The imaginary part of the electron self-energy is resolved for each phonon mode with the highest occupied band and lowest unoccupied band of artificial 2 SL FM (c) and 3 SL MnBi_2Te_4 (d). In all cases, the electron self-energy for all of phonon modes has been included in the figure. Therefore, all phonon modes for each system are shown in the respective panels, and no modes are omitted. Pink spheres stand for $\text{Im } \Sigma_{nk}$ of the infrared (IR)-active modes, red-colored spheres stand for $\text{Im } \Sigma_{nk}$ of the Raman-active modes, and yellow-colored spheres stand for polaritonic IR modes.

higher electric fields. As expected, the shear E_g mode has a small effect on the variations of $\langle Q \rangle_{\text{RMS}}$, suggesting that it is barely assisted in the driving process.

However, the $A_{1g}^{(2)}$ can provide enough support in the driving process, leading to a large displacement amplitude $\langle Q \rangle_{\text{RMS}}$ (Figure 4f) that is strongly related to the (time) averaged interlayer exchange ($\langle J \rangle_{\text{inter}}$) coupling (Figure 4g). This strong relationship of $\langle Q \rangle_{\text{RMS}}$ with $\langle J \rangle_{\text{inter}}$ directly reflects magnetophononic coupling between lattice modulation and magnetic exchange, and warrants a name of the polaritons reported here as magnetophononic polaritons. This relationship deviates from linearity, suggesting the intrinsic anharmonic dependence of exchange coupling on displacement amplitude. This further highlights the nonperturbative regime, where optically driven polaritons can strongly modify magnetic interactions. Intriguingly, $\langle J \rangle_{\text{inter}}$ changes its sign at an amplitude of $2.94 \text{ \AA}\sqrt{\text{amu}}$ and enters into the ferromagnetic region.

This displacement amplitude can be achieved at an electric field of 2.09 MV/cm with a pulse width of 0.6 ps (Figure 4f). Some experimental studies obtained high fields $\leq 90 \text{ MV/cm}$ in the frequency range of $15\text{--}50 \text{ THz}$ ^{43–45} with displacement-limited field strengths, while theoretical studies reported that highly confined phonon polaritons in 2D systems (e.g., 2D hBN) can induce local electric fields on the order of 10 MV/cm .^{46–48} Importantly, a recent experimental study indicated that the E_u mode in MnBi_2Te_4 could easily be excited through their electric-field pulses.⁴⁹ Their study further demonstrated a strong light–matter-driven anharmonic interaction, particularly by the excited Raman E_g mode. We therefore anticipate the high feasibility of such a magnetic transition in a 2 SL AFM driven by light through its interactions with magnetophononic polaritons.

To determine whether a magnetic phase transition induced by light–matter coupling through magnetophononic polaritons

leads to a change in electronic topology in 2 SL AFM, we compute edge spectra using Wannier tight-binding models derived from DFT. As a confirmation of the previously discussed topological nature (Figure 1), we first describe the edge states of 1–3 SL MnBi_2Te_4 without any external effect (Figure 5a–d and edge band structures in Figure S11). Across the systems, we find a clear evolution from a topologically trivial 1 SL FM to regimes where the 2D bulk topology becomes tunable by changing SL thickness and magnetic ordering, providing a direct 2D bulk–boundary correspondence. In particular, both 3 SL AFM (Figure 5c) and 2 SL FM (Figure 5b) phases exhibit in-gap edge modes (the calculated edge states of 3 SL AFM are in excellent agreement with the experimentally measured dispersions [ref 33], confirming their topologically nontrivial nature (QAH and zero-plateau QAH phases, respectively), while both 1 SL FM (Figure 5a) and 2 SL AFM (Figure 5d) exhibit no in-gap edge modes, confirming their topologically trivial nature.

In 2 SL AFM, the interlayer Te–Mn–Te distance is symmetrically equivalent (right panel of Figure 5d), inducing equivalence superexchange and leading to doubly degenerate bulk bands (Figure 1b) at the zone center. Under light–matter coupling through the polaritonic mode of 2 SL FM, this interlayer Te–Mn–Te distance becomes symmetrically inequivalent (left panel of Figure 5e), inducing inequivalence superexchange and making the bulk bands nondegenerate at the zone center. Consequently, 2 SL AFM undergoes a topological phase transition, marked by the emergence of symmetry-protected in-gap edge modes (Figure 5e) in 2 SL FM with finite magnetization and inequivalent interlayer Te–Mn–Te distance in the presence of light. This demonstrates that light–lattice coupling through magnetophononic polaritons provides a practical knob to switch edge conduction on and off in 2 SL AFM. Together, these results establish light-controlled lattice displacement as an effective and SL-selective

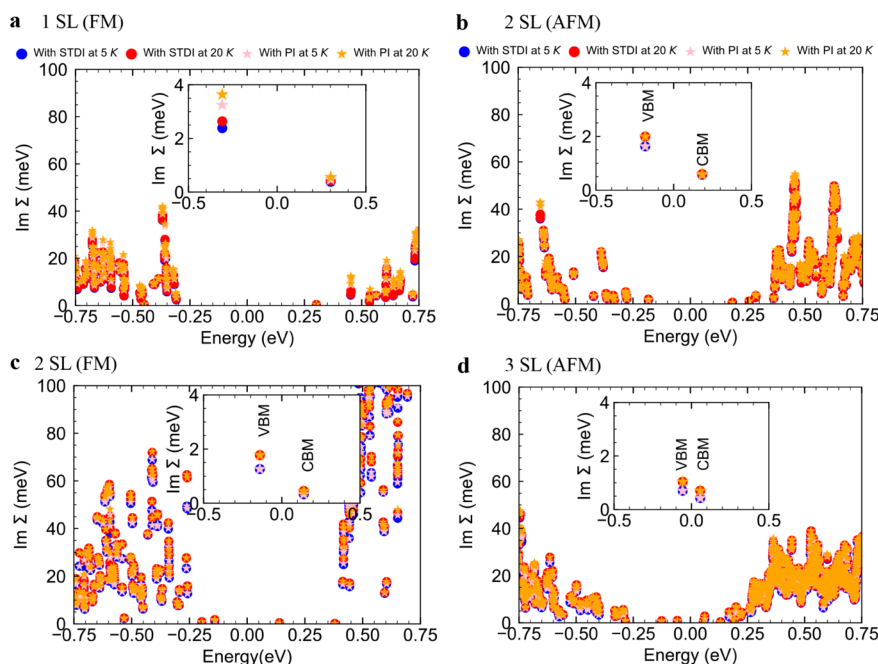


Figure 7. Layer-dependent electron self-energy (proportional to carrier scattering rates) from electron–phonon scattering in 2D MnBi_2Te_4 . Imaginary part of the electron self-energy ($\text{Im } \Sigma$) of 1 SL (a), 2 SL AFM (b), artificial 2 SL FM (c), and 3 SL AFM MnBi_2Te_4 (d) at 5 K (blue or gray symbols) and 20 K (red or yellow symbols). The filled circles stand for the imaginary part of the electron self-energy without long-range interactions, standard 2D interpolation (STDI), i.e., no nonanalytical corrections, while star symbols stand for the imaginary part of the electron self-energy with long-range interactions, long-range 2D interpolation (PI), i.e., with 2D nonanalytical corrections. Inset figures in each subfigure show $\text{Im } \Sigma$ at the conduction band minima (CBM) and valence band maxima (VBM). We set the Fermi level at zero energy in all cases. In these calculations, we considered Hubbard corrections for energy eigenvalues and wave functions, but no Hubbard corrections were explicitly included in the interpolation.

process for switching topologically nontrivial electronic edge modes, providing a coherent 2D bulk–boundary picture of topology activation via light-magnetophononic polariton coupling in 2D MnBi_2Te_4 . These results demonstrate the significant role of the polariton modes, even if photonic energy contributions might be smaller than elastic energy due to a large deceleration factor. Importantly, the long-wavelength magnetophononic polaritons discussed herein should be regarded as a coherently driven near-zone-center mode across the illuminated region, rather than as a point-like polariton packet traversing the sample. At each spatial coordinate, the IR-active ionic coordinate behaves as a driven, damped oscillator that can persist beyond the passage after the electromagnetic pulse envelope has passed. Its nonlinear coupling to the Raman-assisted mode can thus generate a rectified lattice displacement, whose duration is governed by the pulse characteristics and phonon damping times. The present calculations establish the local dependence of magnetic exchange interactions on these ionic distortions; however, they do not explicitly model the spatial propagation of polaritons, the evolution of magnetic domains, or persistent switching phenomena throughout the entire sample (see Note 4 in the SI for further details).

Due to such evolution of topology with magnetic ordering, it can be interesting to explore how these polaritonic modes interact with the topologically trivial and nontrivial electrons. The strength of their interaction can be determined by the e-ph scattering rates (τ^{-1}), which are directly proportional to the imaginary part of the electron self-energy ($\text{Im } \Sigma_{nk}$), i.e., $\tau^{-1} = 2\text{Im } \Sigma_{nk}$. We therefore calculate the e-ph scattering rates resolved for separate phonon modes (Figure 6) including long-

range interactions. As can be seen from Figure 6, the polaritonic modes (yellow-colored) generally have relatively weaker e-ph scattering rates (smaller $\text{Im } \Sigma_{nk}$) than almost all other modes. The scattering rates exhibit clear dependencies on phonon mode, thickness, and magnetic order. In particular, the scattering rates of IR-active (pink-colored) and Raman-active (red-colored) modes significantly differ with layer thickness and magnetic ordering (Figure 6). For example, in the case of the A_{1g} Raman-active mode, the e-ph scattering is reduced with the layer thickness (from 1 SL to 3 SL) as well as with the magnetic ordering (from AFM to FM in 2 SL). This observation is well consistent with a recent experimental study on e-ph scattering and spin–phonon coupling in MnBi_2Te_4 .²⁹

As the e-ph scattering rates change with layer thickness which determines the magnetism and topology in MnBi_2Te_4 , the scattering rates are expected to relate to electronic topology. To explore how the e-ph scattering rates relate to the electronic topology, we also calculate the e-ph scattering rates resolved for separate electronic bands (Figure 7) by using Wannier interpolation with and without long-range interactions. In the subsequent sections, we refer to calculations of $\text{Im } \Sigma$ with long-range interactions as long-range interpolation (PI), while calculations of $\text{Im } \Sigma$ without long-range interactions as the standard 2D interpolation (STDI). However, PI has a negligible effect on $\text{Im } \Sigma$ (Figure 7); only a small change (yellow star symbols) is noticeable for some bands at 20 K. This is expected, as at such low temperatures, the long-range interaction is very weak.⁵⁰

Here, we consider low temperatures (5 and 20 K) because 2D MnBi_2Te_4 becomes paramagnetic above $T_N \approx 20 - 25$ K²⁸ and the calculations of $\text{Im } \Sigma$ is not possible for the

paramagnetic phase based on the current method. In the analysis of $\Sigma_{n,k}$, we mainly focus on the value of $\text{Im } \Sigma$ in the VBM and CBM (inset figures in Figure 7). In all cases, the e-ph scattering rates in the CBM (0.55–0.70 meV for 1 SL–3 SL) are weaker than in the VBM (3.6–1.0 meV for 1 SL–3 SL). Intriguingly, the e-ph scattering rates depend not only on the layer thickness but also on the magnetic ordering. For example, the e-ph scattering rates get 1.15 times weaker when turning AFM into FM order in 2 SL MnBi_2Te_4 (Figure 7c). This also suggests that the e-ph scattering rates can be tuned by changing the orientation of electron spins, i.e., spin–phonon coupling via magnetic ordering, which would notably reduce energy dissipation from the VBM to CBM.

Even though the change in electron self-energy at the VBM and CBM is not large on the numerical scale ($\sim 15\%$), this magnitude is significant on the scale of magnetophonon polaritonic energies (a few meV). As a result, the renormalization strongly influences the phonon resonance energy and damping factor (due to spin–phonon coupling), the oscillator strength, the band edges, and the topological character of the low-energy states. However, the intrinsic magnetophonon polaritonic modes and topology are only relevant to the renormalization through the electron self-energy of band-edge states. We thus concentrate only on the electron self-energy at the VBM and CBM.

Compared to 1 SL and 2 SL (AFM), the e-ph scattering rates are weaker in both the VBM and CBM of 3 SL MnBi_2Te_4 (Figure 7d), as the VBM and CBM are topologically protected in 3 SL (i.e., the QAH phase). As temperatures from 5 to 20 K are too low to resolve distinguishable changes in e-ph scattering with magnetic order and topology, we also calculate the carrier lifetime and electrical conductivity at 20 and 300 K by artificially keeping the same magnetic ordering at 300 K. Note that the lifetime and electrical conductivity at 20 K contain significant uncertainty, and we illustrate this only to show the trend. The electrical conductivity (Figure S12) shows a significant increase with magnetic ordering (as well as topology), as the scattering rates, or inverse carrier lifetime, are reduced when turning AFM to FM in 2 SL MnBi_2Te_4 .

To analyze how changes in e-ph scattering rates with topology manifest in MnBi_2Te_4 , we further calculate electron dispersions considering e-ph scattering at 0 and 300 K (Figure S7). A clear evolution of e-ph scattering rates with topology and magnetic ordering is demonstrated in the electronic band structure at 300 K, depicting the weakest e-ph scattering rates in 3 SL MnBi_2Te_4 (QAH) compared to other phases (see Note S9 in the SI for details). These results suggest the tunable nature of e-ph interactions by layer thickness and magnetic and topological orderings. Importantly, we find that the topologically trivial 2 SL AFM (and potentially higher even SLs) is one of the most suitable systems for topological switching, which has stronger e-ph scattering than other topological phases. As a result, the reduced electrical conductivity in this system leads to a weak screening of the electric field of light by trivial electrons. It is worth to mention that we present electrical conductivity here as an indicator of underlying e-ph scattering strength and its relationship to topology and magnetic order. It is not presented here as a parameter directly intended as a source of polaritonic formation. We study all these properties at the intrinsic band edges, not at the extrinsic level (doping level).

We are aware of recent work on (nonequilibrium) light-driven switching of magnetic and topological states in 2 SL

MnBi_2Te_4 and MnSb_2Te_4 , where the effect of resonant phonons under low-frequency light was explored with some serious limitations (see Note S10 in the SI for detailed information).⁵¹ However, here we have explored how magnetophononic polaritons in 1–3 SL (2D) MnBi_2Te_4 lead to intriguing physics and applications, as well as their relationship with magnetism, topology, and electron–phonon coupling, indicating a completely different perspective compared to the published work.

CONCLUSIONS

Based on density functional perturbation theory, we systematically study the thickness- and magnetic-order-dependent lattice dynamics, nonanalytic long-range interactions, and e-ph scattering for all phonon modes in 2D MnBi_2Te_4 , a prototypical magnetic topological insulator. Our results demonstrate that MnBi_2Te_4 is an intriguing material for hosting three magnetophononic polaritons with high confinement, long propagation, and tunability. These polaritonic features of 2D MnBi_2Te_4 with light–matter coupling-driven topological switching discovered in this study will strongly motivate further theoretical and experimental investigation in nanoelectronics and photonic applications. Also, their relationships with magnetism- and topology-induced polaritonic responses, potential polaritonic-induced recombination processes, and enhanced carrier dynamics have many potential technological applications in photonic and phononic devices. We provide a more comprehensive discussion in the SI (see Note S11).

Our calculated features of magnetophononic polaritons in 2D MnBi_2Te_4 suggest that an EELS experiment can be suitable for accessing these features.^{52–54} Also, these polaritonic modes can potentially lead to the observation of a strong- to ultrastrong-coupled multimode mechanism of quantum electrodynamics in a 2D MnBi_2Te_4 system.⁷ We find that these polaritons parabolically modulate exchange coupling and weaken intrinsic magnetic coupling. Even strong light–matter coupling through the second magnetophononic polariton in 2 SL MnBi_2Te_4 drives an antiferromagnetic-to-ferromagnetic phase transition. Consequently, such a transition leads to the observation of light-induced topological phase transitions, opening a pathway to achieve read-write functionality in topological insulators using light.

We also show that these intriguing polaritons have relatively weak e-ph scattering rates and exhibit thickness-, topology-, and magnetic-order-dependent e-ph scattering rates due to spin–phonon coupling (the magnetoelastic effect). Unlike the magnetoelastic effect in 2D MnBi_2Te_4 , ordinary magnetoelastic effects can typically shift phonon energies due to changes in magnetic ordering.^{55–57} However, the ordinary magnetoelastic effects have yet to report whether they are strongly related to magnetic and topological ordering. Therefore, the observed magnetophononic polariton in 2D MnBi_2Te_4 is not a trivial magnetoelastic effect, but a hybrid quasiparticle that couples phonons, magnetism, and topology, providing a strategy for experimentally realizing light–matter-driven topological and magnetic transitions. This study therefore provides a significant insight into how we may minimize the impact of e-ph scattering on the topologically protected carrier conduction in 2D MnBi_2Te_4 at relatively high temperatures. We believe the research presented here can lead to several intriguing research directions such as experimental measurements of these magnetophononic polaritons, calculations of the lifetimes of

these polaritonic modes from three-phonon scattering processes, and implementation of these in real quantum optical devices.

METHODS

The electronic and phononic calculations based on density functional theory (DFT) and density functional perturbation theory (DFPT)⁵⁸ were performed using Quantum Espresso,^{59,60} which uses the plane wave (PW) pseudopotential method. Before performing the calculations, we added an 18 Å vacuum along the *z* direction to avoid artificial interactions between the periodic layers. We then fully relaxed the structures by minimizing the energy and forces. In all calculations, we considered spin–orbit coupling effect explicitly and used ultrasoft pseudopotentials with the following valence configurations: Mn-3s² 3p⁶ 4s² 4d⁵ 4p⁰; Bi-5d¹⁰ 6s² 6p³; Te-5s² 5p⁴. In the self-consistent calculations, we chose the Perdew–Burke–Ernzerhof (PBE) functional⁶¹ scheme of the generalized gradient approximation (GGA), a 65 Ry cutoff energy for wave functions, 325 Ry for charge density, 12 × 12 × 1 *k*-point, and a 0.001 Ry width of Gaussian smearing. We considered an on-site Hubbard parameter *U* = 4 eV for Mn and the Engel–Vosko functional³⁴ for accurate electronic structure calculations. In electron–phonon calculations, we used 12 × 12 × 1, 8 × 8 × 1 *k*-point grids and 661 and 441 *q*-point grids for 1 SL and 2–3 SL, respectively, after testing convergence, as these calculations are computationally too expensive, and a 441 *q*-point grid gives accurate enough phonons. To demonstrate topological phonons, we constructed the force constants from the dynamical matrix⁶² obtained in DFPT-based electron–phonon calculations. Then, the topological invariant *Z*_{2ph} and edge dispersions were calculated using a Green's function approach implemented in WannierTools.⁶³

In magnetophononic polariton calculations, we use the method developed by Rivera et al.,⁷ where three distinct LO modes directly generate three polariton spectra, and this is fully consistent with the more general mode-sum conductivity formalism used in the ref 7. The conductivity expression used here is not a phenomenological Lorentz formula, rather the result of a microscopic derivation including all phonon modes (*ω*) and momenta. Each distinct LO phonon branch in the material therefore gives rise to its own solution of the equation (*ω*_{LO}(*q*)) and to its own magnetophononic polariton response.

We use the light-driven lattice response in terms of a minimal set of coupled and mass-weighted mode displacements—a magnetophononic polaritonic *E_u* mode (*Q_u*, *Q_y*), a Raman-active *E_g* mode (*Q_x*, *Q_y*), and *A_{1g}* mode (*Q_u*). The polaritonic mode acts as a driven mode with a frequency of 2.74 THz and the far-infrared electric field through polaritonic mode charges *Z_u*, *Z_y*, while the Raman modes are indirectly excited through symmetry-allowed anharmonic couplings to the driven polaritonic mode. Here, the pump field is taken as a carrier at (the polaritonic) frequency *ω_p* with a Gaussian field profile with circular polarization. We solve the classical equations of motion, considering the real-time propagation of all these mode coordinates and the phonon damping *γ* factor. This method therefore captures not only the transiently induced response during the pulse but also the subsequent coherent oscillations of the coupled magnetophononic polariton system.

After solving the equations of motion (see Note S7 in the SI), we quantify polaritonic excitation by the root-mean-square (rms) displacement amplitude over a postpulse window from stimulation trajectories as

$$\langle Q \rangle_{\text{RMS}} = \sqrt{\langle Q(t)^2 \rangle_{t \in [t_1, t_2]}} \quad (11)$$

where $\langle Q \rangle_{\text{RMS}} \geq 0$, i.e., it measures the magnitude of oscillation. However, we add the actual sign of the displacement in Figure 4f to show both directions of motion. To relate lattice displacement to magnetism, we calculate the exchange coupling parameters (from the magnetic force theorem) as instantaneous functions of the displacement amplitude via precomputed mappings *J_{inter}*(*Q_u*) for *A_{1g}*⁽²⁾ mode and *J_{inter}*(*Q_R*) for the doubly degenerate *E_g* mode. See Note S7 and S8 in the SI for further detailed computational information.

Then, the imaginary part of the electron-self-energy (*ImΣ_{n,k}*) was calculated by using 2D electron–phonon Wannier interpolation in EPW^{50,64,65} based on DFPT data. In the calculations, we considered Hubbard corrections for eigenvalues and wavefunction generation and employed a twice-as-dense *q*-point grid in the interpolation. In this method, the *ImΣ_{n,k}* can be obtained by performing an analytical integration of the following equation:^{66,67}

$$\begin{aligned} \text{Im}\Sigma_{n,k} = \pi \sum_{mn_e} \int \frac{d\mathbf{q}}{\Omega_{\text{BZ}}} |g_{mn_e,n}^{\text{SE}}(\mathbf{k}, \mathbf{q})|^2 [n(\omega_{qn}, T) \\ + f(\epsilon_{mk+q}, T) \delta(\epsilon_{n,k} - (\epsilon_{mk+q} - \epsilon_F) + \omega_{qn}) \\ + (n(\omega_{qn}, T) + 1 - f(\epsilon_{mk+q}, T)) \\ \delta(\epsilon_{n,k} - (\epsilon_{mk+q} - \epsilon_F) - \omega_{qn})] \end{aligned} \quad (12)$$

where the integration is performed over the 2D BZ (Figure S1b) and *g_{mn_e,n}^{SE}*(*k*, *q*) is the electron–phonon scattering matrix is defined by

$$g_{mn_e,n}^{\text{SE}}(\mathbf{k}, \mathbf{q}) = \left(\frac{\hbar}{2m_0\omega_{qn}} \right)^{1/2} \langle \psi_{mk+q} | \partial_{qn} V | \psi_{n,k} \rangle \quad (13)$$

Here the superscript indicates that the matrix has units of self-energy (i.e., meV), *ψ_{n,k}* is the electronic wave function for electronic band *n*, momentum *k*, and energy eigenvalue *ε_{n,k}*; *ω_{qn}* is the phonon frequency of a mode *n* with momentum *q*; *∂_{qn}V* is the derivative self-consistent potential; *ε_F* is the Fermi energy; *T* is the temperature; *n*(*ω_{qn}*, *T*) is the Bose occupation factor; and *f*(*ε_{mk+q}*, *T*) is the Fermi occupation factor. As MnBi₂Te₄ is a material with long-range Coulomb interactions, we need to account for the long-range interactions by calculating the long-range part of the electron–phonon scattering matrix as follows:^{50,68}

$$g_{mn_e,n}^l(\mathbf{k}, \mathbf{q}) = i \sum_k \left(\frac{\hbar}{2m_0\omega_{qn}} \right)^{1/2} \times \sum_{\mathbf{G} \neq -\mathbf{q}} \frac{(\mathbf{q} + \mathbf{G}) \cdot \mathbf{Z}_k^* \cdot \mathbf{e}_{k,n}(\mathbf{q})}{(\mathbf{q} + \mathbf{G}) \cdot \mathbf{e}^{\infty} \cdot (\mathbf{q} + \mathbf{G})} \langle \psi_{mk+q} | e^{i(\mathbf{q}+\mathbf{G}) \cdot \mathbf{r}} | \psi_{n,k} \rangle \quad (14)$$

Here *Z_k^{*}* is the Born effective charge, *e_{k,n}* is the eigendisplacement vector for *k*th atom of mode *n*, *G* is the reciprocal lattice vector, and *ε[∞]* is the high-frequency dielectric constant. Then, we add the eqs 13 and 14 and insert it into eq 12 for considering the long-range interactions.

ASSOCIATED CONTENT

Data Availability Statement

All data are included in the manuscript, and the raw data are available upon request to the corresponding author.

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsnano.6c06828>.

Septuple layer (SL) structure of MnBi₂Te₄, its 2D Brillouin zone, 1D zigzag nanoribbon structure, and orbital-decomposed density of states; electronic band structure of 1 SL MnBi₂Te₄ based on DFT, and 2D EPW interpolated; vibrational visualization of six modes of 2 SL MnBi₂Te₄ in the AFM and FM configurations; vibrational visualization of six modes of 3 SL MnBi₂Te₄ in the AFM and FM configurations; magnetic sublattice contributions to a few specific modes; computed phonon modes and their activity of 1–2 SL MnBi₂Te₄; Raman *E_g* mode splitting; computed magnetic order-dependent phonon modes and their activity of 3 SL MnBi₂Te₄; magnetic order-dependent IR and Raman

activity; longitudinal optical (LO) phonon modes in MnBi_2Te_4 and hBN; calculated properties of phonon polaritons in 2D hBN; comparison of phonon dispersions calculated with different methods, topological Z_{2ph} invariant, and mode effective charges; nature of LO phonons at finite q ; description of the topological invariant for phonons; phonon dispersion of zigzag (1D nanoribbon) edge of MnBi_2Te_4 ; details of nonlinear coupling coefficient calculations; comparison of Raman mode-induced interlayer Mn–Mn distance in 2 SL MnBi_2Te_4 (AFM); details of exchange coupling calculations; evolution of electronic topology in 2D MnBi_2Te_4 via magnetic order, thickness, and light; carrier lifetime and electrical conductivity of MnBi_2Te_4 from electron–phonon scattering at 20 and 300 K; temperature-dependent band structures of MnBi_2Te_4 from electron–phonon scattering; description of electronic band structure at 0 and 300 K; detailed information on a recent study on light-driven lattice dynamics in MnBi_2Te_4 ; concluding outlook (PDF)

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Author Contributions

E.H. conceptualized this research, performed all calculations, analyzed all data, wrote the manuscript, revised manuscript, and implemented necessary corrections. Y.Y. and D.C. provided critical reviews and corrections to the manuscript. M.T.E. provided a critical review and suggestions for the

manuscript. N.V.M. conceptualized this research, provided critical reviews and corrections, and supervised the project.

Notes

The authors declare no competing financial interest.

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