Magnons and magnetic fluctuations in atomically thin MnBi$_2$Te$_4$

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Electron band topology is combined with intrinsic magnetic orders in MnBi$_2$Te$_4$, leading to novel quantum phases. Here we investigate collective spin excitations (i.e. magnons) and spin fluctuations in atomically thin MnBi$_2$Te$_4$ flakes using Raman spectroscopy. In a two-septuple layer with non-trivial topology, magnon characteristics evolve as an external magnetic field tunes the ground state through three ordered phases: antiferromagnet, canted antiferromagnet, and ferromagnet. The Raman selection rules are determined by both the crystal symmetry and magnetic order while the magnon energy is determined by different interaction terms. Using non-interacting spin-wave theory, we extract the spin-wave gap at zero magnetic field, an anisotropy energy, and interlayer exchange in bilayers. We also find magnetic fluctuations increase with reduced thickness, which may contribute to a less robust magnetic order in single layers.
The emergence of van der Waals (vdW) magnetic materials has provided rich opportunities to explore magnetic orders in systems that continuously approach the true two-dimensional (2D) limit \(^1\). While it is widely recognized that no long-range magnetic order can exist in 2D for an exchange isotropic system \(^2\), real materials need to be taken into account to predict or detect magnetic order in vdW magnets, known to be highly anisotropic. For example, long-range magnetic order is found to persist down to the monolayer limit in FePS\(_3\) \(^6\) and CrI\(_3\) \(^7\), but breaks down in NiPS\(_3\) monolayers \(^8\). The detection of magnons, collective spin excitations above a magnetically ordered ground state, serves to confirm the presence of long-range magnetic order. Furthermore, by analyzing magnetic-field-dependent magnon resonances, critical information on various energetic terms as well as the magnon selection rules suggest that MBTs are sequences of Te-Bi-Te-Mn-Te-Bi-Te stacked along the c-axis. The long-range spin ordering is found to increase with decreasing layer thickness. The quantum anomalous Hall effect and correlated insulating states have been reported in thin flakes, although the nature of the insulating states is still under debate \(^19\)-\(^22\). While the electronic topological properties originate from the Bi and Te layers, the long-range spin ordering is attributed to the magnetic Mn\(^{2+}\) ion layers. The interplay between magnetism and electronic topology has been shown to be thickness-dependent \(^23\)-\(^25\). Although single septuple layer (SL) MBT is topologically trivial, two or more SLs present distinct topological phases according to their magnetic order and number of layers \(^9\). At low temperature, the Mn moments in each SL are ferromagnetically (FM) ordered while the Mn moments in the adjacent septuple layer exhibit an antiferromagnetic (AFM) order, leading to a vanishing net magnetic moment in samples with an even number of SLs.

**Results**

Here we investigate magnons and magnetic fluctuations in MBT in the ultrathin thickness limit using Raman spectroscopy measurements. We focus on a 2-SL partially because it represents the thinnest sample in which MBT exhibits all three possible magnetic orders: AFM, canted-AFM (c-AFM), and FM when an external magnetic field perpendicular to the 2D plane is tuned. We observe a long-wavelength magnon mode at low temperature under both zero and finite external magnetic fields and in all three magnetic phases, revealing the formation of long-range magnetic order in MBT 2-SLs. In the FM phase, the unusual selection rule and the lack of magnetic field-dependent frequency shift suggest that two-magnon scattering leads to the observed mode. Using spin-wave theory, we extract a spin-wave gap at zero magnetic field \(-0.2\) meV with an anisotropy energy of \(-0.02\) meV and interlayer exchange \(-0.14\) meV in the bilayer. The energetic terms as well as the magnon selection rules suggest that MBTs are in a different class of vdW magnets from the widely studied CrI\(_3\). Magnetic fluctuations in the few-layer samples and a bulk crystal are quantified by analyzing the quasi-elastic scattering (QES) mode and found to increase with decreasing layer thickness.

An MBT crystal consists of seven atomic blocks or SLs in the sequence of Te-Bi-Te-Mn-Te-Bi-Te stacked along the c-axis. Upon cleaving along the \((001)\) surface, the termination layer is typically a Te layer followed by a Bi layer. Mn atoms form a triangular lattice in each SL with an ABC stacking order between the layers, i.e., a Mn atom of the top layer (dark purple in Fig. 1a) is aligned with the center of a triangle in the bottom layer (lighter purple). The magnetic phase diagram for the 2-SL sample as a function of temperature and magnetic field is illustrated in Fig. 1b, which is informed by recent magnetic circular dichroism spectroscopy measurements on few-layer thick MBT \(^21\)-\(^22\). At low temperature and small field, MBT exhibits an AFM phase. At zero magnetic field, the Néel temperature is \(T_N = 17\) K, slightly lower than the bulk value \(T_N = 24\) K as determined from transport measurements \(^18\)-\(^21\). When a magnetic field is applied perpendicular to the 2D plane, the spin-flop transition occurs at \(H_{sf} = 2\) T and the FM transition at \(H_{FM} = 6\) T for a 2-SL. We caution that the precise B-field range over which the c-AFM phase exists in ultrathin samples is still under debate \(^26\)-\(^27\), and the transition field can differ depending on the magnetic defect density \(^18\)-\(^28\).

In a 2-SL, the magnetic unit cell contains two Mn atoms, leading to two magnon branches. In each of the three phases, symmetry operations and Raman tensors derived from the magnetic point groups are different (more details in SI). Spin wave theory calculations (analogous to DFT calculations for phonons) are then performed. The calculated magnon wavefunctions carry further symmetry constraints and predict the polarization configurations of the Raman signal associated with a magnon mode (more details in SI).

We illustrate the possible magnon modes in each phase and their invariance under unique symmetry operations in Fig. 1c-e. When the magnetic structure is (not) inversion-symmetric, a magnetic mode is Raman active (silent). In the simplest case of the FM phase, only the highfrequency branch \(\omega_{IT}\) of the two calculated magnon modes is invariant under inversion \(I\) operation, and thus, Raman active (right panel in Fig. 1e). One expects to observe this magnon mode in the \((a_{\sigma}/a_{\bar{\sigma}})\) configuration, i.e., the incident and scattered photons have the same helicity. In the AFM phase, magnons oscillate with different amplitudes in each layer (Fig. 1c). While these modes break inversion symmetry, they are symmetric following the combined operation of inversion, time-reversal, and a two-fold rotation \((JT)\). One expects to observe these magnons in the cross-circularly polarized channel \((a_{\sigma}/a_{\bar{\sigma}})\). In the c-AFM phase shown in Fig. 1d, the canted moments remain symmetric under the combined operations of a two-fold rotation and time reversal \((CT)\). In this phase, the high-frequency branch is observable in the co-circular polarization configuration. The low-frequency branch has a vanishing frequency at the Brillouin zone center even in the presence of single-ion anisotropy, and thus, is not observable in Raman experiments. These selection rules, in conjunction with the energy scale, guide us in identifying magnon modes in different phases. However, we caution that they are only applicable to one-magnon scattering process.

We first report magneto-Raman measurements of the FM phase on the 1-SL, 2-SL, and 4-SL samples at 11 K with a 6 T B-field (Fig. 2a). The spectra taken with co-circular (red points) and cross-circular (blue points) polarization are compared to the photoluminescence (PL) background. The spectra, which will be discussed later in this paper. After this background removal, Raman spectra from the 1-SL sample exhibit little signal in the frequency range from \(-3\) cm\(^{-1}\) to \(-12\) cm\(^{-1}\). We only show the anti-Stokes side of each spectrum because our instrument has an artifact in the ultralow frequency Raman line which obscures real signals from samples in the Stokes side. In both the 2- and 4-SL samples, we observe a phonon mode that persists above the magnetic transition temperature. The co-circular selection rule is in good agreement with that of an \(A_g\) phonon mode with a crystalline symmetry point group \(D_{3d}\). We attribute this phonon to the layer breathing mode, often observed in vdW materials \(^29\).

In the 2-SL, an additional peak with a central frequency \(\sim 5.7\) cm\(^{-1}\) is observed in the co-circular configuration. To
elucidate the nature of this mode, we present another Raman spectrum taken at low temperature and zero external B-field, which reveals a mode at ~3.4 cm$^{-1}$ (see Fig. 2b). The intensity of this zero-field mode decreases with temperature and disappears above the Néel temperature of ~17.5 K, suggesting its dependence on the magnetic order. The temperature-dependent intensity can be fitted with a function of $\sqrt{1 - T/T_N}$ (solid line in Fig. 2c), consistent with the mean-field theory of magnetic moments. The co-circular selection rule, however, contradicts that expected for one-magnon scattering in the AFM phase.

We further study the systematic evolution of Raman spectra from the 2-SL sample at low temperature (12 K) as a function of magnetic field (Fig. 3) in the co-circular polarization configuration. All spectra in Fig. 3a are fitted with two Lorentzian functions.

Fig. 1 Magnetic lattice, phase diagram, and magnon modes in a 2-SL MBT. a) Illustration of two triangular magnetic lattices formed by Mn ions in the top (bottom) layer indicated by dark (light) purple spheres. The layers follow an ABC stacking order. D and $J_c$ represent anisotropy and interlayer exchange coupling. b) Phase diagram as a function of temperature and an applied magnetic field perpendicular to the 2D plane. c) Magnons in antiferromagnetic (AFM) phase: spins from two layers oscillate with different amplitudes, corresponding to two magnon modes. Illustrations of spins before and after applying a time-reversal ($T$), inversion ($I$) and two-fold rotational (C w.r.t x-axis) operator ($ITC$). d) Magnons in canted AFM (c-AFM) phase: the spins are canted at an angle with respect to the applied magnetic field. Illustrations of spins before and after time-reversal and two-fold rotational (y-axis) operator ($CY$). e) Magnons in ferromagnetic (FM) phase: spins in the two layers oscillate in-phase or out of phase. Illustrations of spins before and after inversion operator ($I$). Shaded modes are either Raman silent or near zero frequency. $\omega_L$ ($\omega_H$) refers to the magnon branch with a higher (lower) frequency in the left (right) column.

Fig. 2 Raman spectra of magnon in different setuple layers, magnetic fields, and its temperature dependence at zero field. a) Raman spectra taken at 12 K and 6 T field in 1-SL (bottom), 2-SL (middle), and 4-SL (top) MBT taken with co-circular $\sigma_+/\sigma_+$ (red points) and cross-circular $\sigma_+/\sigma_-$ (blue points) polarized incident and scattered photons. Black solid line is fitting with the sum of two Lorentzian functions, corresponding to a phonon (P) and a magnon (M) mode, respectively. b) Raman spectrum from the 2-SL taken with co-circular polarizations at 12 K and 0 T. c) Temperature dependence of the integrated Raman intensity of the ~3.4 cm$^{-1}$ magnon peak from the 2-SL at 0 T. The intensity diminishes as the 2-SL goes through the antiferromagnetic (AFM) to paramagnetic (PM) transition.
and the extracted central frequencies of the modes are summarized in Fig. 3b. The phonon frequency (black dots) does not show any magnetic field dependence, while the magnon mode shows distinct B-field dependence in different magnetic phases. At the low field, the AFM magnon mode shows a frequency of ~3.4 cm$^{-1}$ independent of the applied magnetic field. Both the magnetic-field independence of the energy and the selection rules (i.e. co-circular polarization) contradict the expectations for a one-magnon process. At 3 T, we observe a discontinuity in the frequency, signaling a transition into the c-AFM phase. The transition between the c-AFM to FM phases at ~5.5 T causes a subtle change in the slope of magnon frequency vs. B field. Error bars displayed are smaller than the data points.

In the AFM phase, the theory predicts two Raman active magnon modes with opposite field-dependent frequency shifts (Zeeman shifts). The calculated AFM one-magnon frequency is below our experimental detection range ≥3 cm$^{-1}$ (shaded area). Summing the energies of these two magnon branches leads to a field-independent two-magnon resonance, close to the observed resonance at ~3.4 cm$^{-1}$. Furthermore, the Loudon–Fleury scattering theory model$^{30–33}$ adopted in our calculation predicts that two-magnon scattering is observable in the co-circular polarized configuration, consistent with the experimental observation but in contrast with the selection rule expected for one-magnon scattering (Fig. 1c). The transition between the AFM to c-AFM
Magnetic fluctuations quantified by the QES peak in the paramagnetic phase at 300 K. a Normalized Raman susceptibility $\chi''/\omega$ for different SLs. The blue dots represent data points. The purple curves are fittings given by Eq. (1). Black solid lines are fitting for several phonon modes using either Lorentzian or Fano functions. b Summary of the fitted $\chi''/\omega$ for several flakes with different thicknesses. c Layer-dependent dynamic Raman susceptibility $\chi''_{\text{dyn}}$ calculated from the integration of $\chi''/\omega$ up to 180 cm$^{-1}$.

The blue dots represent data points. The purple curves are fittings given by Eq. (1). Black solid lines are fitting for several phonon modes using either Lorentzian or Fano functions.

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magnetic field $2 \sqrt{SD(SD - S_L^2)} \approx 0.2$ meV using anisotropy $SD = 0.024$ meV and interlayer exchange $S_L = -0.138$ meV. Since magnetic anisotropy stabilizes the long-range magnetic order in these 2D materials\(^6\), the weak magnetic anisotropy energy may contribute to a less robust magnetic order in 1-SL MBT. In contrast, the anisotropy energy in CrI\(_3\) is estimated to be 0.27 meV from Raman measurements\(^7\), which is nearly one order of magnitude stronger and sufficient to stabilize the FM order in CrI\(_3\) monolayers. This interplay between anisotropy and interlayer exchange also leads to a new c-AFM phase in MBT.

**Discussion**

In summary, we report direct observations of magnons in a 2-SL MBT as the ground state is tuned through three magnetically ordered phases by an external field. We were not able to observe a systematic evolution in magnon spectra as a function of layer thickness. In 4-SLs for example, the low-frequency phonon mode spectrally overlaps with and masks the anticipated magnon resonances. Magnetic fluctuations increase with reduced thickness due to enhanced spin-lattice coupling, which may contribute to the absence of magnons in the 1-SL MBT. We completely rule out the possibility that the absence of magnons in 1-SL is related to coupling to the substrate. Future experiments on 1-SLs performed under different conditions (e.g. at higher magnetic fields) are needed. MBT provides an interesting comparison with CrI\(_3\) as a strongly coupled exchange system with rich magnetic phases including a c-AFM phase controllable via electric field\(^44\). A recent scanning tunneling microscopy experiment suggests that electronic structures fluctuate at the atomic scale on the surfaces\(^45\) although it is likely related to disorders that have yet to be better controlled for realizing interesting quantum phases\(^46\).

Our finding complements these prior studies by investigating magnetic orders in ultra-thin MBT layers. Our studies will also guide future investigations of coupled collective excitations and searches for exotic phases. For example, the spectral overlap between a magnon and a phonon may lead to hybridization between these modes, which would fundamentally change the topological nature of magnons in MBT\(^47,48\).

**Methods**

**Sample preparation.** High-quality MnBi\(_2\)Te\(_4\) (MBT) crystals were grown by the flux method\(^18\). We obtained thin MBT flakes using the Al\(_2\)O\(_3\)-assisted exfoliation method\(^20,49\). Optical transmission measurements were used to determine the layer thickness based on the Beer–Lambert law\(^50\). Ultrathin MBT layers may degrade in an ambient environment. The samples studied here were prepared in a glove box –7 Torr. An out-of-plane magnetic field\(^7\) was applied using a cryostat to present results from a sample without encapsulation layers. Thus, we choose an ambient environment. The samples studied here were prepared in a glove box.

**Raman spectroscopy.** Raman measurements were performed using a 632.81 nm excitation laser with a full-width-half maximum (FWHM) of 0.85 cm\(^{-1}\). The laser power was kept below ~100 mW to avoid local heating and damage to the samples. The laser beam was focused onto the sample via a ×40 microscope objective to a spot size of about 3 μm in diameter. The Raman signal was collected in the back-scattering geometry and measured with a Horiba LabRAM HR Evolution Raman microscope (1800 grooves/mm grating). All measurements were taken in a closed-cycle helium cryostat from 11 to 300 K with a base pressure lower than 7 × 10\(^{-7}\) Torr. An out-of-plane magnetic field ranging from 0 to 7 T was applied.

**Data availability**

The data sets generated and/or analyzed during the current study are available from the corresponding author upon reasonable request.

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Competing interests
The authors declare no competing interests.

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