Magnons and Magnetic Fluctuations in Atomically Thin $MnBi_2Te_4$

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Electron band topology is combined with intrinsic magnetic orders in $MnBi_2Te_4$, leading to novel quantum phases. Here we investigate collective spin excitations (i.e. magnons) and spin fluctuations in atomically thin $MnBi_2Te_4$ flakes using Raman spectroscopy. Using non-interacting spin-wave theory, we extract the spin-wave gap at zero magnetic field, an anisotropy energy, and interlayer exchange in bilayers.

Van der Waals Materials and $MnBi_2Te_4$

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[2]. $MnBi_2Te_4$ (MBT) is a vdW material and the first synthesized material with both electronic topology and intrinsic magnetic orders[3]. An MBT crystal consists of seven atomic blocks or septuple layers (SLs) in the sequence of Te-Bi-Te-Mn-Te-Bi-Te stacked along the *c*-axis (Fig. 1).

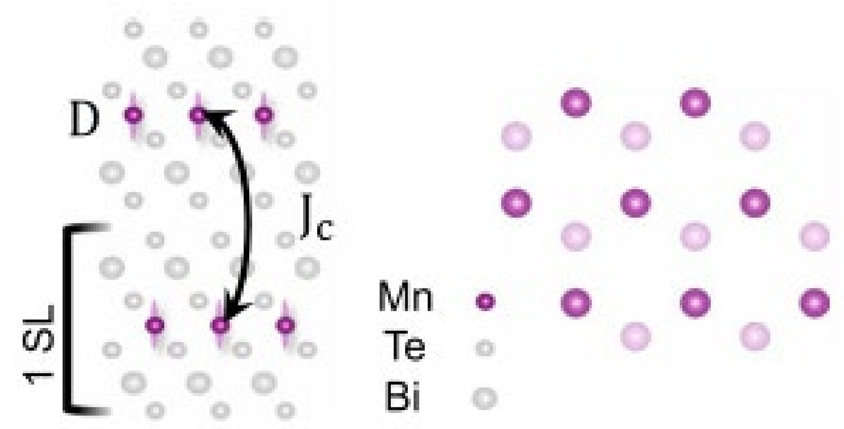

Fig. 1 Illustration of two triangular magnetic lattices formed by Mn ions in the top (bottom) layer indicated by dark (light) purple spheres. The layers follow an ABC stacking order. D and J_c represent anisotropy and interlayer exchange coupling.

We focus on the 2-SL MBT since it is the thinnest sample that can demonstrate the three magnetic phases: antiferromagnetic (AFM), canted-AFM (c-AFM), and ferromagnetic (FM).

Magnons: Proof of Long-range Magnetic Orders

Magnetic properties of materials come from the electron spin. When a material with magnetic orders (eg. FM or AFM material) is excited from the ground state to a low-energy excited state, the inverse spin angular momentum will be coupled and distributed to neighboring atoms' spin angular momentum. The result is the collected spin excitation of the whole material, also known as the spin wave. The quantum of spin wave is the magnon. Therefore, the detection of magnons is the direct proof of long-range magnetic orders.

Magnon Modes

In a 2-SL, the magnetic unit cell contains two Mn atoms, leading to two magnon branches. From the crystal symmetry, the magnetic order, and the spin-wave theory, we establish the Raman selection rules for different magnon modes. The magnon modes of three possible magnetic phases (AFM, c-AFM, and FM) and their invariance under unique symmetry operations are shown in Fig. 2. These selection rules, in conjunction with the energy scale, guide us in identifying magnon modes in different phases.

Experimental Results

The systematic evolution of Raman spectra from the 2-SL sample at low temperature (12 K) as a function of magnetic field in the co-circular polarization configuration is shown in Fig. 3. 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. The discontinuity of the slope indicates the phase transition of magnetic orders. The magnon frequency of c-AFM and FM phases are linear to the magnetic field, while the AFM phase is not due to the two-magnon scattering[4]. 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[1].

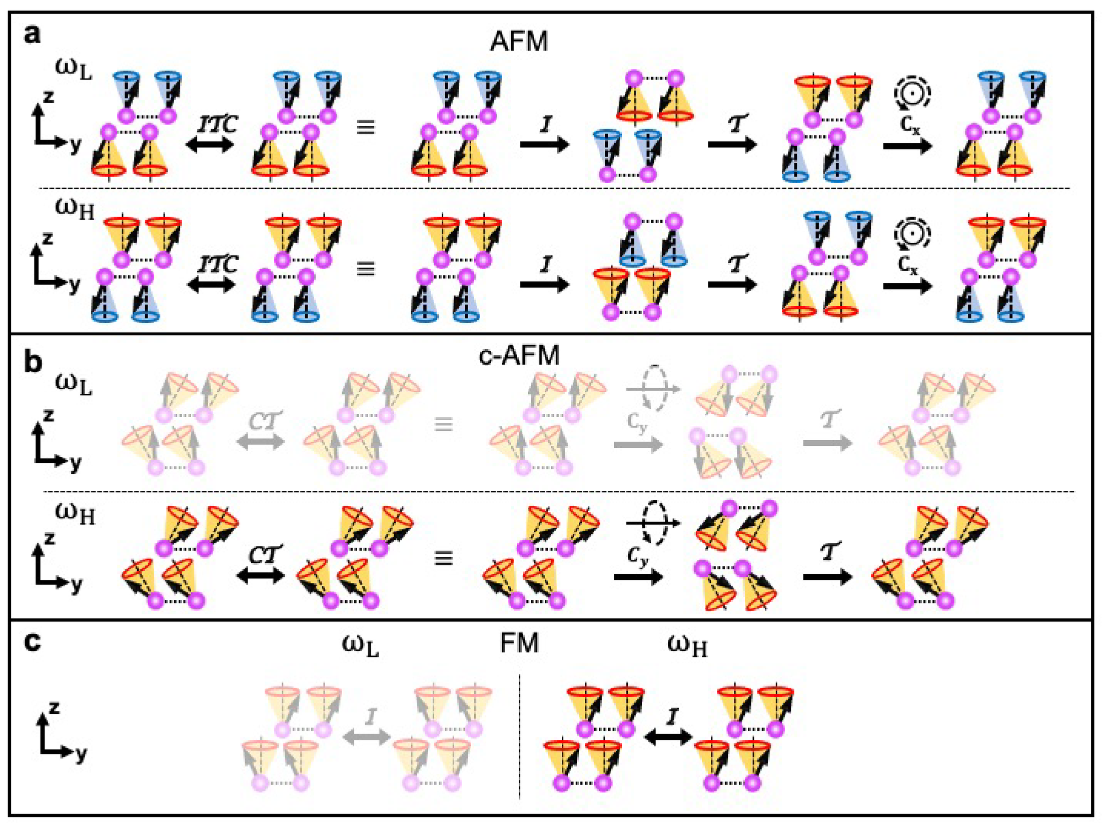

Fig. 2. Three possible magnetic phases of 2-SL MBT. (a) AFM, (b) c-AFM, and (c) FM magnon modes and their corresponding invariance under symmetrical operations (*I*: inversion, *T*: time reversal, *C*: 2-fold rotation).

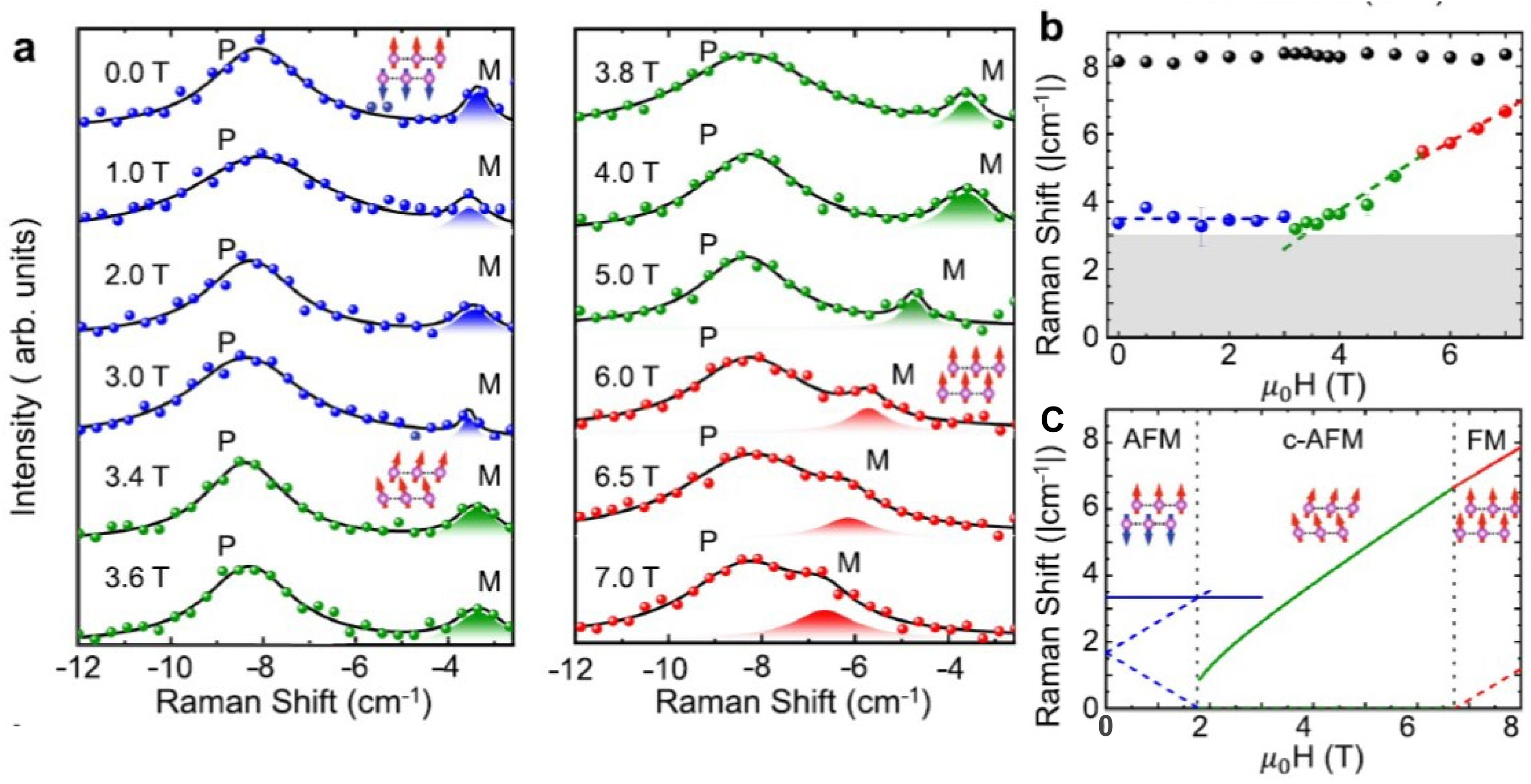

Fig. 3. (a) Raman spectra (co-circular polarization) at 12 K under different magnetic fields. P and M represent the phonon and magnon, respectively. (b) Extracted central frequency for both the phonon and magnon vs. B field. Shaded area is below the experimental detection range. (c) Calculated magnon modes in the AFM, c-AFM, and FM phases, which are matched to the measured results.

In all displayed Raman spectra, we removed a quasi-elastic scattering (QES) background. Magnetic fluctuations in the few-layer samples and a bulk crystal are quantified by analyzing the QES mode and found to increase with decreasing layer thickness.

Outlook

A recent scanning tunneling microscopy experiment suggests that electronic structures fluctuate at the atomic scale on the surfaces[5] although it is likely related to disorders that have yet to be better controlled for realizing interesting quantum phases[6]. 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.

References

[1]Lujan, D. et al, Magnons and magnetic fluctuations in atomically thin $MnBi_2Te_4$, *Nat. Commun.* **13**, 2527 (2022, this work).
[2]See reference 1-4 in ref. 1. [3]See reference 9-18 in ref. 1. [4]See reference 30-33 in ref. 1. [5]See reference 45 in ref. 1. [6]See reference 46 in ref. 1.

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