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Competing magnetic interactions in the antiferromagnetic topological insulator MnBi2Te4

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Abstract
The antiferromagnetic (AFM) compound MnBi2Te4 is suggested to be the first realization of an AFM topological insulator. We report on inelastic neutron scattering studies of the magnetic interactions in MnBi2Te4 that possess ferromagnetic triangular layers with AFM interlayer coupling. The spin waves display a large spin gap and pairwise exchange interactions within the triangular layer are long ranged and frustrated by large next-nearest neighbor AFM exchange. The degree of frustration suggests proximity to a variety of magnetic phases, potentially including skyrmion phases, which could be accessed in chemically tuned compounds or upon the application of symmetry-breaking fields.

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Competing Magnetic Interactions in the Antiferromagnetic Topological Insulator MnBi$_2$Te$_4$

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The antiferromagnetic (AFM) compound MnBi$_2$Te$_4$ is suggested to be the first realization of an AFM topological insulator. We report on inelastic neutron scattering studies of the magnetic interactions in MnBi$_2$Te$_4$ that possess ferromagnetic triangular layers with AFM interlayer coupling. The spin waves display a large spin gap and pairwise exchange interactions within the triangular layer are long ranged and frustrated by large next-nearest neighbor AFM exchange. The degree of frustration suggests proximity to a variety of magnetic phases, potentially including skyrmion phases, which could be accessed in chemically tuned compounds or upon the application of symmetry-breaking fields.

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The breaking of time-reversal symmetry by the introduction of magnetism in topological materials is key to unlocking unique topologically protected transport phenomena [1]. For example, the quantum anomalous Hall effect has been demonstrated at low temperatures by inducing bulk ferromagnetism (FM) through the substitution of dilute magnetic ions, such as Cr or V, into (Bi, Sb)$_2$(Se, Te)$_3$ topological insulators [2–4]. While this is an incredibly important discovery, the disorder and inhomogeneity associated with these dilute FM systems present an obstacle to delivering quantum topological transport at routinely accessible temperatures. An alternate route to access these phenomena is to develop a new class of stoichiometric magnetic topological materials. MnBi$_2$Te$_4$ may be the first example of a stoichiometric antiferromagnetic topological insulator (AFTI) [5–11]. AFTI are predicted to provide a platform for novel topological phases, such as quantum anomalous Hall effect [14]. In this Letter, inelastic neutron scattering (INS) measurements on MnBi$_2$Te$_4$ reveal its Ising-like nature, surprisingly strong interlayer exchange interactions, and large lifetime broadening. We find that the next-nearest neighbor AFM interaction ($J_2$) competes with nearest-neighbor FM interaction ($J_1$) within the triangular layer, placing the system close to the classical stability limit for intralayer FM correlations $|J_2/J_1| < 1/3$ [15,16]. In addition, we find that longer-range interactions up to at least the fourth neighbor are necessary to fully describe the intralayer spin dynamics. These experimental observations are supported by first-principles (DFT + $U$) calculations of the magnetic interactions which are long-ranged and frustrated at correlation strengths of $U \approx 4$–$5$ eV. Our classical Monte Carlo simulations show that the system is susceptible to forming long-period magnetic structures. This may allow, for example, the Bi–Te layers containing topological fermions to be subjected to a variety of helimagnetic or topological skyrmionic structures [17] under suitable perturbations, such as chemical substitution or applied magnetic fields.

INS measurements on powder samples of MnBi$_2$Te$_4$ ($T_N = 24$ K) were performed on the Cold Neutron Chopper Spectrometer (CNCS) at the Spallation Neutron Source at Oak Ridge National Laboratory using incident neutron energies of $E_i = 3.3$ and 12 meV. The intensities...
interactions are not negligible, which is surprising given
zone centers. This observation suggests that interlayer
modes, Fig. 2(a) and constant energy Q cuts in Fig. 2(b)
find sharp dispersion minima at momenta of (0, 0, 3/2),
(0, 0, 9/2), and (0, 0, 15/2), corresponding to A-type AFM
zone centers. This observation suggests that interlayer
interactions are not negligible, which is surprising given
the large spacing of 13.6 Å between Mn layers. An energy
cut at the dispersion minimum at (0, 0, 9/2) (Q = 0.7 Å−1)
in Fig. 2(c1) indicates a spin gap with an onset of Δ ≈ 0.5 meV consistent with sizable uniaxial magnetic
anisotropy.

The quantitative details of the magnetic interactions
become more apparent based on fitting the data to a
local-moment Heisenberg model,

\[ H = - \sum_{ij} J_{ij} S_i \cdot S_j - J_c \sum_{(ij)_1} S_i \cdot S_j - D \sum_i S_i^2, \]

where \( J_{ij} \) describe pairwise interactions between the
hth neighbors within a single triangular layer, \( J_c \) corre-
sponds to an AFM nearest-neighbor (NN) interlayer
coupling, and \( D > 0 \) is the uniaxial anisotropy. Here
\( J > 0 \) corresponds to FM coupling. As we describe below,
the sharpness of interlayer modes at the gap edge and the
broad, high energy intralayer modes necessitate a stepwise
approach to determine all model parameters.

Analysis of the magnetization data provides preliminary
estimates of the interlayer interaction \( J_c \) and uniaxial
anisotropy \( D \) parameters (\( J_c-D \) model). The magnetization,
measured at \( T = 2 \) K on single-crystal specimens shown in
Fig. 2(e), reveals spin-flop and saturation fields \( H_{SF} = 3.4 \) T and \( H_{Sat} = 7.9 \) T with \( |H|/c \) and \( H_{Sat} = 10.3 \) T with
\( |H|/ab \), consistent with previous reports [8,9,11]. Within
the Heisenberg model and starting from A-type order
with moments along \( c \), these critical fields are given by
the expressions \( g \mu_B H_{SF} = 2S \sqrt{D(6|J_c| - D)} \), \( g \mu_B H_{Sat} =
2S(6|J_c| - D) \), and \( g \mu_B H_{Sat} = 2S(6|J_c| + D) \) (where \( g \approx 2 \)
and \( S \approx 5/2 \)) and provide a range of values for
\( SD \approx 0.07-0.1 \) meV and \( -SJ_c \approx 0.08-0.09 \) meV.

The magnetization data provide an estimate for \( \Delta =
2S \sqrt{D(6|J_c| + D)} = 0.4-0.5 \) meV that is consistent with
the INS data in Fig. 2(c1).
FIG. 2. (a) Inelastic neutron scattering intensities of MnBi$_3$Te$_4$ measured at $T = 7.8$ K focused on the low energy gap edge with $E_i = 3.3$ meV. (b) Several constant energy $Q$ cuts at the gap edge from the data (circles) and from Model cD (lines). Plots are vertically offset for clarity. (c1) Low energy magnetic energy spectrum showing the spin gap ($\Delta$) near $(0, 0, 9/2)$ ($Q = 0.7$ Å$^{-1}$, blue circles) and an estimate of the bandwidth ($W$) using a cut near the interlayer AFM zone boundary ($Q = 0.6$ Å$^{-1}$, red circles). Green squares are estimates of the incoherent background originating from intralayer spin wave modes. (c2) Same cuts as in (c1) obtained from the $J_{c}$-D model. (d) Numerical calculations of the INS intensity from the $J_{c}$-D model. In (a) and (d), the red line shows the dispersion of spin wave modes along $c$ from the $J_{c}$-D model. (e) Magnetization data from a single crystal of MnBi$_3$Te$_4$ highlighting spin-flop and saturation fields.

We also analyze the $J_{c}$-D model parameters by comparing the gap edge INS data to calculations of the powder-averaged spin wave intensities following the procedure outlined in Ref. [20]. We assume resolution-limited features (FWHM = 0.15 meV) and fix $SJ_X$ to a nominal value since the energies are too low to effectively fit the intralayer exchange parameters. We then vary $SJ_c$ and $SD$ and compare the calculated spin wave intensities to a series of constant-energy $Q$-cuts from 0.4–0.8 meV, as shown in Fig. 2(b). Much better agreement with the data is obtained by the addition of incoherent background contributions that presumably originate from the broad, intralayer excitations described below. The resulting $\chi^2$ goodness-of-fit displays a rather shallow minimum that does not allow precise determination of $SD$ and $SJ_c$ (see Fig. S4 in the Supplemental Material [18]) and deviates somewhat from the values determined from the magnetization data.

Within the shallow minimum in $\chi^2$, a representative set of parameters can be ascertained from INS data by considering the spin gap and the bandwidth of interlayer excitations ($W$) shown in Fig. 2(c1). The bandwidth is determined by the energy at the AFM zone boundary at $Q = 0.6$ Å$^{-1}$, where $W = 6|J_c| + 2SD - \Delta \approx 0.1$ meV. $\Delta$ and $W$ provide rough estimates of $SD \approx 0.12$ and $SJ_c \approx -0.055$ meV that sit within the minimum in $\chi^2$ and Figs. 2(a)–2(d) shows these parameters provide a good representation of the gap edge data.

We now turn to the determination of the intralayer model parameters. This fitting was performed by fixing the $J_{c}$-D model values and sampling intralayer $J_Y$ values over a regular mesh and calculating the powder averaged magnetic scattering. The calculated spectrum was convoluted with the instrumental resolution function and compared to the measured magnetic spectrum of the $E_i = 12$ meV data summed over the momentum range from $Q = 0.8$–1.9 Å$^{-1}$. A satisfactory accounting of all features in the magnetic spectrum requires the introduction of intralayer pairwise exchange interactions up to the 4th neighbor.

For NN coupling ($J_1$) only, the best-fit model spectrum consists of a single sharp peak near the top of the spin wave band with $SJ_1 = 0.26$ meV. Reasonable fitting to the $J_1$ model requires the introduction of a substantial Gaussian lifetime broadening FWHM of $\Gamma = 1.5$ meV to the calculated spectra. Figure 1(c) shows that the $J_1$ model is clearly an unsatisfactory description of the experimental spectrum. The introduction of a frustrating AFM next-nearest-neighbor (NNN) interaction ($J_2$) improves the fit by shifting magnetic spectral weight from high to low energies. As Fig. 1(c) shows, the resulting fit to the $J_1$-$J_2$ model with optimized values of $SJ_1 = 0.31$ and $SJ_2$ = $-0.06$ meV is better, but even this model requires sizable damping of 1.1 meV. Neither the $J_1$ nor the $J_1$-$J_2$ models capture the broad, low energy peak between 1–1.5 meV which led us to consider even longer-range interactions.
Based on analysis of the spin wave density-of-states (see Supplemental Material [18]), an additional van Hove singularity is introduced to the magnetic spectrum for 4th neighbor interactions \( J_4 \), but not for \( J_3 \). This lead us to finally consider the refinement of a \( J_1-J_2-J_4 \) model for the intralayer spin dynamics. As shown in Fig. 1(c), this model captures the two-peaked spectrum with optimal values of \( SJ_1 = 0.3 \), \( SJ_2 = -0.083 \), and \( SJ_4 = 0.023 \) meV and \( \Gamma = 0.7 \) meV. In all models, the value obtained for \( SJ_1 \) is consistent with that obtained from single-crystal INS studies of hexagonal MnTe [21], whose structure contains similarly stacked Te-Mn-Tc triangular layers. All fitting parameters are reported in Table I.

In all models of the intralayer exchange constants, best fits are obtained when we introduce substantial Gaussian broadening to the calculated spectra beyond the instrumental resolution, suggesting significant lifetime broadening of the intralayer spin waves. Nonetheless, gap edge data representing the interlayer interactions and strong Ising anisotropy will stabilize FM layers, we expect that chemical doping or other perturbation, such as strain, can possibly induce noncollinear phases. To quantify this expectation, we have calculated the magnetic phase diagram (including a magnetic field) using classical Monte Carlo (MC) simulations for a single layer with interactions up to NNN. In Fig. 3(a), we show the low-\( T \) phase diagram close to the experimentally found anisotropy value \( D/J_1 = 0.4 \) as a function of \( J_2/J_1 \) and magnetic field \( h/J_1 \) along the z direction. Vertical spiral, skyrmion, and up-up-down-down stripe phases [see Figs. 3(b)–3(d)] appear at larger frustration ratios of \( J_2/J_1 \geq 0.5 \). MC simulations also find skyrmion phases appear for smaller anisotropy values \( D/J_1 \approx 0.1 \) at \( J_2/J_1 = 0.4 \) (see Supplemental Material [18]). This raises the possibility for spiral or skyrmion phases to appear, for example, in Sb-substituted Mn(Bi,Sb)\(_2\)Te\(_4\), where \( D \) is found to be significantly smaller [22].

Recent first-principles electronic structure calculations with \( U = 5.34 \) eV predict that \( J_2/J_1 < 0.03 \) [8,10], which is ten times less than that obtained from our INS data. Here, we extract the Heisenberg parameters by performing an analysis of the energies of six ordered spin states [23] based on DFT + U calculations [24] including spin-orbit coupling. Figure 4(a) reveals that the DFT results support the presence of strong long-range intralayer interactions at small values of \( U \). Larger values of \( U \) suppress long-range interactions. While the DFT + U generated exchange values are generally larger than the experimental values, the ratios of \( J_2/J_1 \) and \( J_4/J_1 \) at values of \( U \approx 4–5 \) eV are consistent with the INS data. Further details of the computational methods in full can be found in the Supplemental Material [18], which includes Refs. [25–29].

Overall, our findings indicate that AFTI MnBi\(_2\)Te\(_4\) shows elements of frustration \((J_2/J_1 \approx 0.3)\), Ising anisotropy \((D/J_1 \approx 0.4)\), metamagnetism \((J_z/D \approx 0.45)\), and long-range intralayer exchange interactions \((J_4/J_1 \approx 0.1)\). The presence of low-field metamagnetism in MnBi\(_2\)Te\(_4\) is similar to that found in \( MX_2 \) transition metal halide triangular lattice antiferromagnets [30]. Compounds such as FeCl\(_2\) [31] and FeBr\(_2\) [32] also display

\[ TABLE I. \text{Heisenberg model parameters obtained from low energy INS data (}\epsilon - D\text{ model), high energy INS data (}\epsilon_1, \epsilon_{1.2}, \text{ and } J_1-J_2-J_4 \text{ models), and DFT + U + SOC calculations. The broadening parameter (}\Gamma\text{) is also provided. All values are in meV.}\]

| \( SJ_1 \) | \( SJ_2 \) | \( SJ_3 \) | \( SJ_4 \) | \( SD \) | \( \Gamma \) | \( \chi^2 \) |
|---|---|---|---|---|---|---|
| \( J_1 - D \) | 0.23 | ⋯ | ⋯ | ⋯ | ⋯ | ⋯ |
| \( J_1 \) | 0.26(1) | ⋯ | ⋯ | ⋯ | ⋯ | ⋯ |
| \( J_1 - J_2 \) | 0.31(2) | -0.06(2) | ⋯ | ⋯ | ⋯ | ⋯ |
| \( J_1 - J_2 - J_4 \) | 0.30(2) | -0.083(9) | 0.023(8) | ⋯ | ⋯ | ⋯ |
| DFT | 0.81 | -0.30 | 0.13 | -0.09 | 0.15 | ⋯ |

[FIG. 3. (a) Low-temperature magnetic phase diagram as a function of \( J_2/J_1 \) and magnetic field \( h/J_1 \) for fixed anisotropy \( D/J_1 = 0.4 \) and temperature \( T = 0.08/J_1 \). Different phases are polarized paramagnet (blue), vertical spiral (red), multi-q (skyrmion) phase (yellow), and up-up-down-down (orange). (b)–(d) Real-space spin configurations of vertical spiral (b), multi-q (skyrmion) crystal (c), and up-up-down-down phases (d). Color denotes \( S' \) component (scale bar shown) and arrows denote the in-plane components (\( S', S'' \)). Panels show a 20 \times 20 part of the full 52 \times 52 lattice with lattice constant \( a = 1 \).]

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strongly competing $J_1$-$J_2$ interactions within the triangular layer and can host multi-$q$ magnetic structures [30]. $MX_2$ compounds have therefore been proposed to host skyrmion phases in applied fields [17]. In MnBi$_2$Te$_4$, similar frustration could lead to skyrmion phases and may also result in complex spin textures near the sample surface, where magnetic interactions may be modified by strain or surface termination effects. This could explain recent ARPES [33,34] and thin film magnetization data [13] that are not consistent with uniformly FM layers near the surface. Even from our powder samples, we find evidence for strongly $\mathbf{Q}$-dependent broadening, which should be investigated in INS studies of single-crystal samples. Such lifetime broadening could be related to frustration or to coupling between magnetic fluctuations and charge carriers, as inferred from magnetotransport measurements [22].

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FIG. 4. First-principles calculations of (a) Heisenberg parameters and (b) ratios of key exchange interactions versus $U$ including spin-orbit coupling. In (b), the red and purple horizontal dashed lines correspond to experimental values for $J_2/J_1$ and $J_3/J_1$ and the vertical black dashed line shows the best value of $U \approx 4.5$ eV.
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