

ALTERMAGNETISM

Altermagnetism revealed by polarized neutrons in MnF_2

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Altermagnets are compensated magnets with symmetry-enforced spin split bands. Direct spectroscopic evidence for their existence has been reported in several materials. For example, inelastic neutron scattering (INS) has recently been used to witness a splitting of magnon bands in MnTe that is compatible with altermagnetism. Here, we present polarized INS results on MnF_2 showing that it is altermagnetic. Our data reveal a splitting coming from the long-range dipolar coupling. Furthermore, using a domain-biased sample, the data reveal a nonzero chirality in the neutron scattering cross section that reverses the sign between the two magnon modes. It is this feature that clearly demonstrates altermagnetism in MnF_2 . This finding highlights the potential for polarized INS to characterize altermagnets through its sensitivity to fine structure in magnon spectra.

Although uncompensated magnetism has been known since antiquity, most famously in the mineral magnetite, the existence of antiferromagnets was irrefutably demonstrated only after the invention of neutron scattering techniques in the 1950s. Indeed, many states of matter that are staples of modern quantum condensed matter research rely on sophisticated spectroscopic tools for their characterization. Recent progress on altermagnetism, too, exemplifies the importance of modern spectroscopic probes (1–5). Altermagnets have an underlying antiferromagnetic order parameter, and all of the materials currently studied in this context have long been identified as mere antiferromagnets. However, the feature that sets them apart from their more conventional cousins is a set of so-called spin symmetries (3, 4) that are tied to antiferromagnetism, particularly local multipolar order parameters (6, 7) and, crucially, to a characteristic spin splitting of the band structure (3, 4). The first spectroscopic evidence for altermagnetism appeared only recently (8–19) and considerable effort is being invested in finding new altermagnets and better ways to identify them. These efforts are being driven both to advance our fundamental understanding of magnetism and because altermagnets are promising candidate systems for future spintronics applications.

To date, the most well-established altermagnetic materials are MnTe (8–14) and CrSb (15, 16, 17–21). In both, there is evidence of an altermagnetic spin splitting of the band structures from angle resolved photoemission spectroscopy data, including direct evidence in MnTe coming from a spin-resolved study (10). In other words, altermagnetism has been visualized through electronic degrees of freedom. However, altermagnetism is tied to the underlying magnetocrystalline symmetries and should therefore appear in all degrees of freedom coupled to the magnetism, including the magnetic structure itself and the spin wave excitations. Altermagnetism is most cleanly defined in

the limit of zero spin-orbit coupling, in which the collinear magnetism is invariant under spin symmetries (22–29) including a residual $U(1)$ spin rotation symmetry. In this limit, the magnons exhibit a pattern of chirality splitting analogous to the spin splitting of the electronic band structure first noted in (30) and explored further in (31–33). Although departures from the ideal limit break strict spin or chirality conservation, the energy splittings associated with altermagnetism imply that the phenomenon has a robustness that extends to real materials. Thus, a splitting of the magnon bands consistent with altermagnetism has been observed in MnTe using inelastic neutron scattering (9). However, to make a definitive identification of altermagnetism based on magnons, it is important to visualize the remnant chirality itself. Such a measurement has been carried out using resonant inelastic x-ray scattering in CrSb (21). However, the energy resolution of RIXS is on the order of 10 meV and was insufficient to resolve any magnon splitting.

To resolve both small splittings and the chirality, inelastic neutron scattering with polarization analysis is the method of choice (34). When unpolarized neutrons scatter from magnons in an altermagnet, they acquire a polarization along the scattering wavevector with a sign that directly reflects the chirality of the magnon mode (34). Then, through both the dispersion relations and polarization analysis, one may explore departures, originating from various possible magnetic anisotropies, from the limit where the magnon chirality is conserved.

Here, we report inelastic neutron scattering data on a candidate altermagnetic material MnF_2 . The magneto-crystalline symmetries are compatible with altermagnetism in this material and the magnetic manganese has an essentially pure spin 5/2 magnetic moment, and so spin-orbital effects are very weak, placing MnF_2 among the more likely systems where magnons may carry a (weakly broken) chirality. MnF_2 is well-known in the field of magnetism. It was among the first materials in which an antiferromagnetic order parameter was identified in the early days of neutron scattering and has been used in neutron training courses to teach students about canonical antiferromagnetism (35–42)—a small irony that is indicative of the difficulty of detecting altermagnetism in this material.

Neutron scattering in MnF_2

Previous inelastic neutron scattering data revealed a single magnon branch with a gap at the zone center that was attributed to the long-range magnetostatic dipolar coupling (35–42). Although it was recognized that this coupling should also induce a small splitting between the two magnon modes (35–37), this was below the resolution of the instruments in previous experiments. Our experimental data reveal the presence of a well-resolved magnon splitting with a maximum located at the zone boundary of ~ 0.2 meV, compared with a 7-meV total bandwidth. This originates primarily from the long-range dipolar coupling.

Before describing the details of the experiment on MnF_2 , we briefly summarize the results expected from a polarized inelastic neutron scattering experiment on an idealized altermagnet—that is, defined to be an altermagnet with an exact $U(1)$ spin rotational symmetry in the magnetically ordered state. We further assume, for simplicity, that there are two magnetic sublattices. Then, the Neel vector $\mathbf{N}$ is the difference of the magnetizations on the two oppositely aligned sublattices. To be altermagnetic, $\mathbf{N}$ must transform as a nontrivial one-dimensional (1D) irrep of the point group of the crystal. For the case of MnF_2 , the two sublattices are related by a fourfold rotation about the tetragonal c axis and a translation by half a unit in each direction. In the ideal altermagnet, the magnon bands are split over most of the Brillouin zone and the bands may be assigned chiralities ± 1 that reverse under the same fourfold symmetry that connects the two sublattices. Now suppose we prepare the sample in a single magnetic domain and carry out an inelastic neutron scattering experiment in a half-polarized setting where we control the in-going

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neutron polarization $\mathbf{P}_{in}$ and measure the overall scattered intensity. The result within linear spin wave theory is

$$I(\mathbf{k}, \omega) \propto \sum_n \left[\frac{1}{2} \left(1 + (\hat{\mathbf{k}} \cdot \hat{\mathbf{N}})^2 \right) + (-)^n (\mathbf{P}_{in} \cdot \hat{\mathbf{k}}) (\hat{\mathbf{k}} \cdot \hat{\mathbf{N}}) \right] C_k \delta(\omega - \epsilon_k^{(n)}) \quad (1)$$

with $\mathbf{k}$ as the wave vector and C_k the 1-magnon band intensity (34). The first term in square brackets is the usual unpolarized neutron scattering intensity, which is the same in the two split modes. The second (chiral) term in square brackets has a polarization dependence and a sign that depends on the mode index n . This term is present whenever the scattering wavevector has a component along the incoming neutron polarization and along the staggered magnetization. It is this term that provides a direct measurement of the altermagnetic features. In practice one may make a measurement of the intensity for $+\mathbf{P}_{in}$ and for $-\mathbf{P}_{in}$, and subtract the two to isolate the chiral term in the cross section.

Experimental setup

Now we can describe the experimental details. The work was carried out on a single crystal of MnF_2 , which has tetragonal space group $\text{P4}_2/\text{mmn}$ (#136) with pure spin $S = 5/2$ manganese ions on $2a$ sites and nominally nonmagnetic fluoride ions on $4f$ sites. The material magnetically orders below $T_N \approx 67$ K into a simple collinear compensated structure with moments oriented parallel and antiparallel to the c axis (Fig. 1A). As mentioned previously, the altermagnetic features of MnF_2 originate

from the fourfold rotation (combined with time-reversal and a half-translation) connecting the two magnetic sublattices. This directly leads to the prediction of d-wave altermagnetism with a pattern of chirality splitting indicated in Fig. 1C. We expect to witness the chirality through polarized inelastic neutron scattering, provided the sample orders into a majority single domain configuration.

To assess the magnetic domain population in the sample, we make use of the fact that the elastic neutron cross section at reflection $\mathbf{G}$ takes the form

$$I_{\mathbf{P}_{in}}(\mathbf{G}) \propto |\mathbf{F}_{nuc}|^2 + |\mathbf{F}_{mag}|^2 + (\mathbf{P}_{in}^\perp + \hat{\mathbf{N}}) \text{Re}[\mathbf{F}_{nuc} \mathbf{F}_{mag}^*],$$

where $\mathbf{F}_{nuc}$ and $\mathbf{F}_{mag}$ are, respectively, the nuclear and magnetic structure factors and $\mathbf{P}_{in}^\perp$ is the component of the in-going neutron polarization perpendicular to the scattering wavevector (43). The time-reversal odd nuclear-magnetic interference term has nonvanishing amplitude at (H, K, L) reflections for $H + K + L$ odd. To isolate this term, the flipping ratio at the (210) Bragg position, $I_{\mathbf{P}_{in}}/I_{-\mathbf{P}_{in}}$ was determined with $\mathbf{P}_{in}$ parallel to the c axis. The single crystal was found to have an overall mosaicity of $\approx 3^\circ$, arising from three distinct grains. From flipping ratio measurements, the domain population of each grain was determined (43) at 2 K after cooling slowly through the magnetic transition. We found that this annealing protocol was sufficient on our samples resulting in a magnetic configuration with $85 \pm 5\%$ of the volume in a single domain. The presence of domain imbalance observed in the studied MnF_2 crystal is consistent with earlier findings (44, 45).

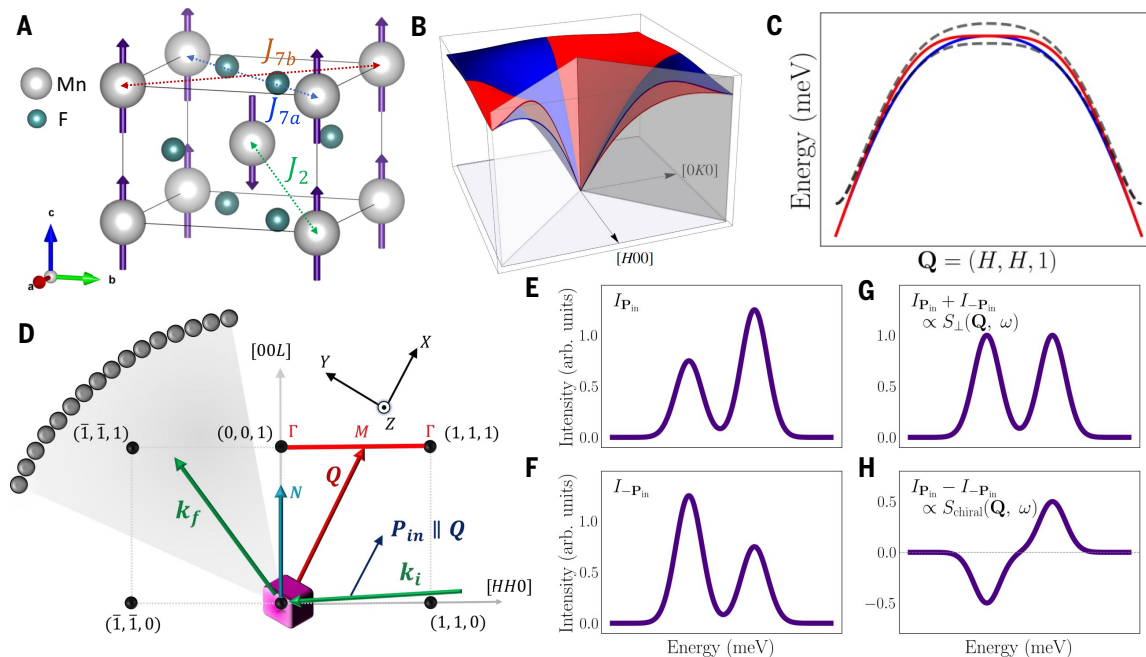


Fig. 1. Origin and detection of altermagnetism in the spin waves of MnF_2 . (A) The crystal and magnetic structure of MnF_2 with magnetic Mn^{2+} ions on the sites of a body-centered tetragonal lattice and decorating fluoride ions that are crucial for the altermagnetism by breaking the symmetry connecting the magnetic sublattices down to a C_4 along the c axis, a $\mathbf{Q} = (1/2, 1/2, 1/2)$ translation and time reversal. (B) This symmetry allows a magnon band splitting in models with zero spin-orbit coupling. The pattern of magnon chiralities $\pm$ is enforced by symmetry and is shown through colors red (+) and blue (-). Polarized neutron scattering is sensitive to this chirality. (C) The maximal splitting is along $[H00]$ in the middle of the zone (solid lines). Dipolar couplings, in the absence of an altermagnetic splitting, do not lead to a net chiral term but they do split the bands even at the zone boundary (dashed lines). (D) Scattering geometry for the polarized inelastic neutron scattering experiment. The MnF_2 crystal is oriented in the (HHL) plane, with its magnetic moments aligned along the Néel vector $\mathbf{N}$. An incident neutron beam, characterized by wavevector $\mathbf{k}_i$ and polarization $\mathbf{P}_{in}$, arrives at the sample under the condition $\mathbf{P}_{in} \parallel \mathbf{Q} \parallel \mathbf{X}$, where $\mathbf{X}$ defines the (X, Y, Z) Blume–Maleev coordinate system for polarized neutrons. The neutrons are then scattered into a final wavevector $\mathbf{k}_f$ and detected by position-sensitive detectors. Identification of altermagnetism means detecting a non-vanishing chiral term in the neutron scattering that reverses sign between the two magnon branches. To investigate this: the scattering intensity is measured at constant momentum with (E) ingoing polarization $\mathbf{P}_{in}$ and (F) $-\mathbf{P}_{in}$. The usual unpolarized neutron cross section is (G) the sum of these contributions and (H) the chiral contribution is isolated from their difference.

Unpolarized neutron results

We now turn to the inelastic neutron scattering results obtained on the HYSPEC spectrometer at SNS (ORNL) with measurements made in the (*HHL*) scattering plane. Figure 2A shows unpolarized scattering intensity along various high symmetry line segments in momentum space (the paths indicated in the inset). The main feature visible in this plot is a dispersive branch of excitations with modulated intensity extending from about 1 meV at the zone center up to about 7 meV at the zone corners, consistent with previous measurements (35–42). The HYSPEC spectrometer at 9 meV incident energy provides an energy resolution of about 100 μ eV at an energy transfer of 6 meV. Close inspection of Fig. 2A shows that the main dispersive branch actually consists of two weakly split modes. The splitting is most clearly visible at (1/2, 1/2, 1). Fig. 2, C and E show the experimental dispersion relations passing through this point, respectively along (*H, H, L*) and (1/2, 1/2, *L*) showing clearly two modes with a maximum splitting of about 178(3) μ eV.

As MnF₂ has two magnetic sublattices, two spin wave modes are expected. Moreover, the magneto-crystalline symmetries do not protect their degeneracy. However, as the manganese ions carry pure spin 5/2, any exchange anisotropies should be very weak. Even so, the gap Δ at the zone center (Fig. 2A) points to departures from the pure Heisenberg limit. Previous work recognized that this gap originates mainly from the long-range magnetostatic dipolar coupling (35–37). The magnitude of the dipolar coupling, D_{nn} , between nearest neighbor moments, is fixed by the moment size and is of the order of a few tens of μ eV. The magnitude of Δ is compatible with the small

dipolar coupling as $\Delta \sim \sqrt{JD_{nn}}$. However, the maximum splitting between the magnon branches induced by the dipolar coupling is roughly on the scale of D_{nn} , which is therefore a very small energy scale compared with the magnon bandwidth. Indeed, a calculation (43) reveals that the dipolar splitting is maximum at the zone boundary with magnitude 179 μ eV compatible with the HYSPEC data (Fig. 2D).

We extracted dispersion points from the data by fitting cuts at constant energy (for steep dispersions) and constant momentum (otherwise) (43). Fig. 2A shows a selection of these points overlaid onto the scattering intensity plots showing that the extracted points faithfully capture the measured intensities. We parameterized the data by carrying out a multiboson (or flavor wave) calculation based on a Hamiltonian $H = H_{ex} + H_{dip}$, in which H_{ex} consists of Heisenberg exchange couplings J_n between neighboring Mn ions at shell *n* for $n = 1, \dots, 7$. This is equivalent to linear spin wave theory up to a scale depending on the spin length. The cutoff was fixed at 7 because for shorter range Heisenberg couplings, the symmetry of the model is higher than that of the lattice. Only when J_7 is included do the interactions capture the correct space group symmetry of MnF₂. This feature of the model is crucial as altermagnetic splittings in the zero spin-orbit coupled limit may arise only once the model has this symmetry. In fact, the symmetry lowering at the 7th neighbor is correlated with the fact that at this range there are two distinct couplings denoted J_{7a} and J_{7b} (Fig. 1A). For altermagnetic splittings to appear, the fourfold symmetry of the body-centered tetragonal sublattice

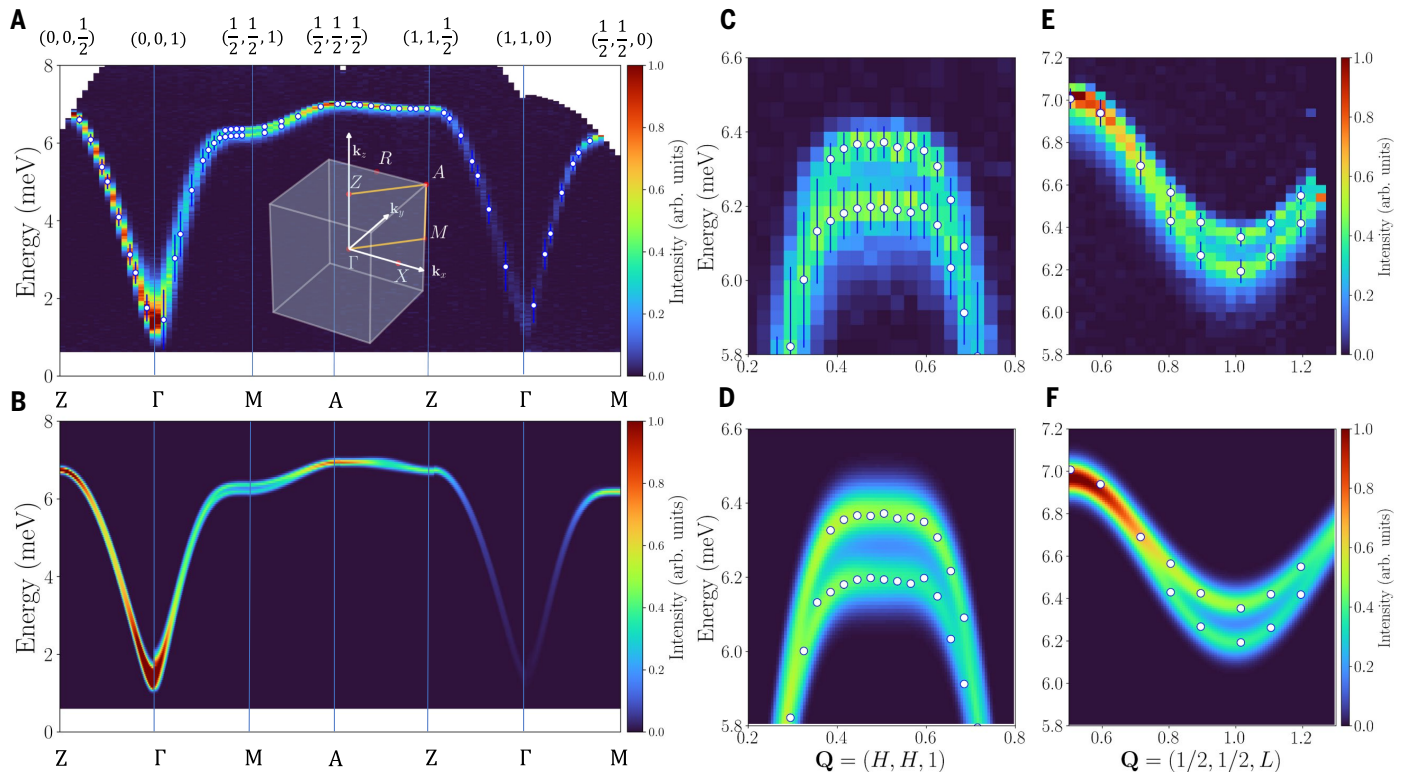


Fig. 2. Unpolarized inelastic neutron spectra of MnF₂. (A) Inelastic neutron spectra along high-symmetry directions within the Brillouin zone (inset) obtained with an incident energy of $E_i = 9$ meV. Blue circles show energies of the magnetic modes obtained from fits of the data using a sum of Gaussians [“Unpolarized inelastic scattering measurements” in (43)]. (B) Calculated spin-wave spectra obtained from parameters obtained from global fits of the data (and listed below). (C and E) Zoom onto the spin wave spectrum of MnF₂ along $\mathbf{Q} = (H, H, 1)$ and $\mathbf{Q} = (1/2, 1/2, L)$ where the dipolar splitting is revealed. (D and F) Corresponding calculated spin-wave spectra. All data are integrated over 0.04 meV steps along energy and 0.03 r.l.u. steps in $\mathbf{Q}$ along the plotted axis. The transverse- $\mathbf{Q}$ averaging window is 0.1 r.l.u. in all cases. Calculations are based on a Heisenberg exchange model with long-range dipolar coupling (43). The exchange couplings obtained by fitting the experimental data are: $J_1 = -0.075(2)$ meV, $J_2 = 0.287(3)$ meV, $J_3 = -0.012(1)$ meV, $J_5 = 0.008(2)$ meV, $J_6 = -0.001(2)$ meV, $J_{7a} = -0.006(3)$ meV, $J_{7b} = -0.002(3)$ meV.

occupied by Mn ions must be broken by setting $J_{7a} \neq J_{7b}$. In addition to the exchange, we include also the long-range dipolar interaction H_{dip} which has a fixed coupling strength. As we have no a priori knowledge of the exchanges we carry out a least squared fit to the data including eight free parameters (corresponding to the Heisenberg couplings just described), assuming spin-orbital anisotropies to be negligible. Further details of the data processing and fitting procedure may be found in the supplementary materials (43).

Figure 2, B, D and F shows the results of calculations of the unpolarized neutron scattering intensity based on parameters extracted from the fitting procedure. The dispersion relations, including the splitting of the modes, are all very well captured by the fit and the calculation. The intensities are broadly in agreement between the experiment and the calculation, for example the modulation along $(1/2, 1/2, L)$ and the strong and weak intensities at the zone centers at $(0, 0, 1)$ and $(1, 1, 0)$ respectively. There is an apparent qualitative difference along the ΓM path where the calculation has monotonically decreasing intensities away from the zone center, whereas the experiment has intensities that increase again toward $(1/2, 1/2, 1)$. Additional comparisons between the experiment and calculations are given in the supplementary section (43) (fig. S9).

To parameterize the uncertainty in the exchange couplings, we carried out fits to the experimental dispersion points together with Gaussian noise with variance on each point taken from the experimental peak widths. We focus here on features of the fits to noisy data that relate to altermagnetic couplings J_{7a} and J_{7b} . We find that these couplings are constrained to be much smaller than the nearest neighbor exchange, J_2 ,

between the two magnetic sublattices, as might be expected especially in a magnetic insulator such as MnF_2 . In the fits shown in the figures, we take $J_2 = 0.287$ meV, whereas $J_2 = -0.006$ meV and $J_{7b} = -0.002$ meV. We find that J_{7a} and J_{7b} are strongly anticorrelated. Therefore the data are fully compatible with $J_{7b} - J_{7b}$ nonzero and also on the order of roughly 10 μeV (43).

Polarized neutron results

Having achieved some further understanding of the magnetic couplings in MnF_2 based on the HYSPEC data, we present the results of the polarized inelastic neutron scattering experiment whose goal was to find evidence of a nonzero chiral term in the cross section indicative of altermagnetism. The experiment was carried by fixing $\mathbf{P}_{in}$ parallel to $\mathbf{Q} = (1/2, 1/2, 1)$ and measuring scattering intensity (43). A similar measurement was performed with fixed $-\mathbf{P}_{in}$. The standard inelastic neutron scattering intensity is proportional to the sum of these measurements and the chiral term is isolated from the difference (Fig. 1, E to H). The sum and difference maps along $(H, H, 1)$ are shown in Fig. 3, C and E, respectively, with the former showing a pronounced splitting in the vicinity of the zone boundary. Focusing on Fig. 3E, we observe a clear signal correlated to the magnetic excitations with a sign reversal between the upper and lower modes. Cuts at constant momentum (43) (figs. S5 to S7) reveal that the magnitude of the chiral term has a maximum at about $H = 0.35$ (43) and that it goes to zero, within errors, at the boundary $H = 1/2$. Fig. 3E provides a compelling signature of altermagnetism in MnF_2 . The magnetocrystalline symmetries of MnF_2 guarantee that the sign of the chiral term reverses

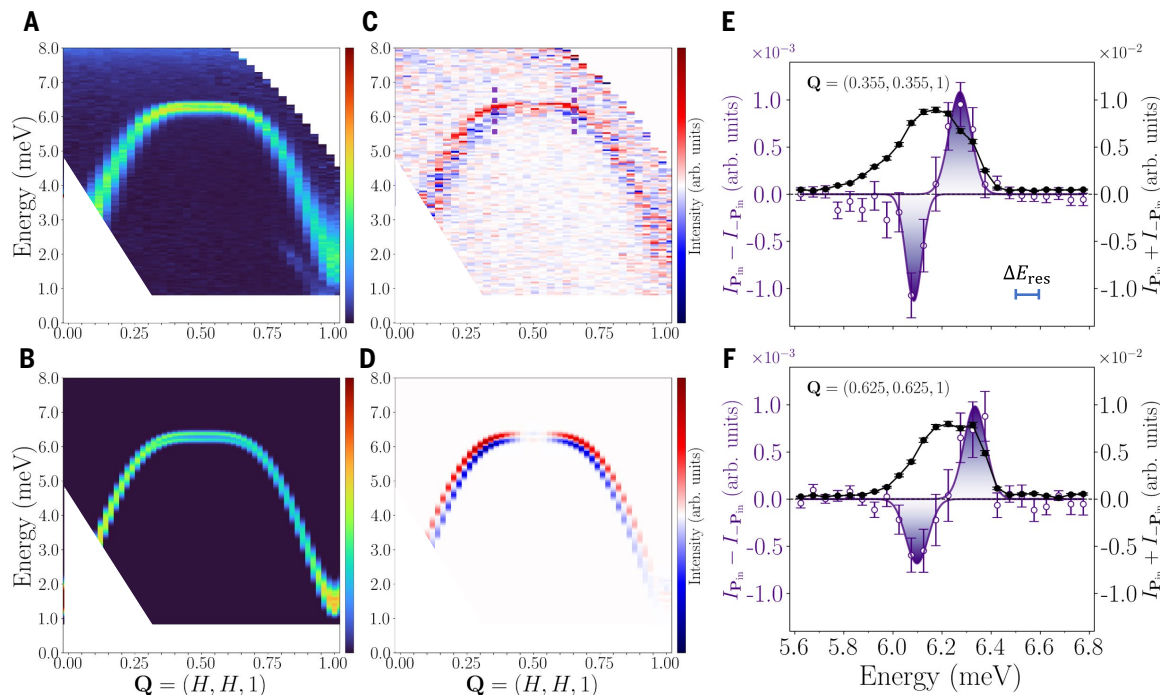


Fig. 3. Chirality of magnon bands in MnF_2 revealed by polarized neutrons. (A) Sum of spin wave spectra measured along $\mathbf{Q} = (H, H, 1)$ with polarized neutrons $+\mathbf{P}_{in}$ and $-\mathbf{P}_{in}$. The sum is only relative to the unpolarized cross section. (B) Corresponding calculated spin-wave spectra obtained from fitted parameter of unpolarized neutrons spectra (See (43) for details). (C) The difference of spectra, relative only to the chiral part of the cross section. Dashed vertical purple lines represent constant- $\mathbf{Q}$ energy slices shown in (E and F). (D) Calculated chiralities of the magnon bands obtained from fitted parameters (43). The pronounced broadening of the magnon bands observed experimentally, compared to the calculations—particularly in regions of steep dispersion (0–0.3 r.l.u. and 0.7–1 r.l.u.) is likely caused by a combination of data integration along perpendicular directions and the mosaic spread of the sample ($\approx 3^\circ$). (E and F) Constant- $\mathbf{Q}$ energy scans obtained from experimental sum (black points) and difference (purple points) at $\mathbf{Q} = (0.355, 0.355, 1)$ (E) and $\mathbf{Q} = (0.625, 0.625, 1)$ (F). Black curves are a guide to the eye. Purple curves are results of a double Gaussian fit. All data herein are integrated over 0.05 meV steps along energy and 0.03 r.l.u. steps in $\mathbf{Q}$ along the plotted axis. The transverse- $\mathbf{Q}$ averaging window is 0.1 r.l.u. in both cases. The blue line in (E) denotes the energy resolution ΔE_{res} which is calculated to be around 0.1 meV at 6 meV. This value approximately corresponds to the full width at half maximum (FWHM) of each fitted Gaussian; in the fits, the minimum allowed FWHM was constrained by ΔE_{res} .

when passing from $(H, H, 1)$ to $(H, -H, 1)$. Furthermore, measurements with $\pm \mathbf{P}_{in}$ were performed consecutively, with polarization reversal independently achieved through the XYZ coils or the Mezei flipper, yielding consistent cross sections within uncertainty and confirming that the observed difference signal is intrinsic and of altermagnetic origin (43).

Discussion and outlook

It is interesting to examine possible microscopic origins of the chiral neutron scattering intensity in MnF_2 . Let us first consider a model where the exchange constants have the constraint that $J_{7a} = J_{7b}$ so that H_{ex} alone leaves the magnon bands degenerate. Introduction of the long-range dipolar coupling lifts the degeneracy over most of the zone. The dipolar interaction in the model couples point-like magnetic moments living on a body-centered tetragonal lattice. This model has a fold-fold rotation symmetry leaving the magnetic sublattices invariant. This symmetry is incompatible with altermagnetism. Indeed, the dipolar coupling maximally mixes chiralities so that the chiral term in the neutron cross section vanishes. Starting from this model, we may now tune $\delta J_7 \equiv |J_{7a} - J_{7b}|$ to be nonzero. For infinitesimal δJ_7 , a finite chiral term is present although the presence of the dipolar coupling means that the magnon chirality is not a good quantum number. This result shows concretely how altermagnetism can persist even into regimes where the altermagnetic splitting is small compared with those caused by chirality breaking anisotropies.

In (43), we further consider possible anisotropic exchange contributions to the magnetic Hamiltonian originating from finite spin-orbit coupling. We expect such contributions to be very weak in MnF_2 . We find that these terms, including symmetry-allowed DMI terms, do not generate a chiral contribution to the neutron cross section (43).

We integrate the chiral contribution to the intensity finding that it is between 5 and 11% of the total intensity depending on the wavevector. We confirm that this ratio is compatible with the fact that the fits to the dispersion relations produce parameters with $\delta J_7 < 5 \mu\text{eV}$. In other words, the polarization signal directly probes a significantly smaller energy scale than is directly measured through the dispersion relations. Therefore one message of this work is that potentially important beyond the scope of altermagnetism, is that polarized neutron intensity may be sensitive to fine structure in chiral magnon spectra to which more standard probes may be insensitive. This is analogous to magnetic circular dichroism, in which a small Zeeman splitting due to an applied magnetic field can be resolved due to their derivative-like spectrum (46).

As we have established, polarized neutron scattering provides a powerful means of detecting altermagnetism (i) by focusing directly on the magnetic degrees of freedom (ii) through its sensitivity to perturbations away from the idealized altermagnetic limit on the sub-meV scale through the dispersion relations (iii) through the fact that the cross section is known precisely in terms of underlying magnetic correlators, and, crucially, (iv) through its ability to measure the key feature of altermagnetism, namely the chirality of the magnons. We have demonstrated these features through measurements that establish altermagnetism in MnF_2 . These results provide a concrete benchmark for understanding altermagnetic spin excitations and will guide the exploration of altermagnetism and magnon chirality across a wide range of condensed matter systems.

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SUPPLEMENTARY MATERIALS

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Materials and Methods; Supplementary Text; Figs. S1 to S11; Tables S1 to S3; References (48–51)

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Altermagnetism revealed by polarized neutrons in MnF_2

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Editor's summary

Altermagnets, a recently identified category of magnetic materials, have been predicted to host spin waves (“magnons”) that have distinct energies depending on their handedness. Using different techniques, two groups are now reporting evidence of these chiral magnons. Jost *et al.* used left- and right-handed circularly polarized light to create magnetic excitations in the material MnTe, a well-established altermagnet candidate (see the Perspective by van den Brink). The resulting magnon intensity was dependent on the sense of light polarization, indicating chirality. Faure *et al.* used polarized neutron scattering to investigate the material MnF_2 . The difference in the scattering intensity of oppositely polarized neutrons likewise indicated magnon chirality, helping build a case for altermagnetism in this material. —Jelena Stajic

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