Multiphase magnetism in Yb$_2$Ti$_2$O$_7$

Allen Scheie$^{a,b,c}$, Jonas Kindervater$^{a,b}$, Shu Zhang$^{a,b,d}$, Hitesh J. Changlani$^{a,e}$, Gabrielle Sala$^{a}$, Georg Ehlers$^{g}$, Andre Heinemann$^{a}$, Gregory S. Tucker$^{a,l}$, Seyed M. Koohpayeh$^{a,b}$, and Collin Broholm$^{a,b,k}$

$^a$Institute for Quantum Matter, Johns Hopkins University, Baltimore, MD 21218; $^b$Department of Physics and Astronomy, Johns Hopkins University, Baltimore, MD 21218; $^c$Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831; $^d$Department of Physics and Astronomy, University of California, Los Angeles, CA 90095; $^e$Department of Physics, Florida State University, Tallahassee, Florida 32306; $^f$Condensed Matter Science, National High Magnetic Field Laboratory, Tallahassee, FL 32304; $^g$Quantum Condensed Matter Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831; $^h$German Engineering Materials Science Centre at Heinz Maier-Leibnitz-Zentrum, Helmholtz-Zentrum Geesthacht GmbH, D-85748 Garching, Germany; $^i$Laboratory for Neutron Scattering, Paul Scherrer Institut, CH-5232 Villigen, Switzerland; $^j$Laboratory for Quantum Magnetism, Institute of Physics, Ecole Polytechnique Federale de Lausanne, CH-1015 Lausanne, Switzerland; $^k$Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, MD 20899; and $^l$Department of Materials Science and Engineering, Johns Hopkins University, Baltimore, MD 21218

Edited by B. D. Gaulin, McMaster University, Hamilton, ON, Canada, and accepted by Editorial Board Member Zachary Fisk September 9, 2020 (received for review May 4, 2020)

We use neutron scattering to show that ferromagnetism and antiferromagnetism coexist in the low T state of the pyrochlore quantum magnet Yb$_2$Ti$_2$O$_7$. While magnetic Bragg peaks evidence long-range static ferromagnetic order, inelastic scattering shows that short-range correlated antiferromagnetism is also present. Small-angle neutron scattering provides direct evidence for mesoscale magnetic structure that we associate with metastable antiferromagnetism. Classical Monte Carlo simulations based on exchange interactions inferred from (111)-oriented high-field spin wave measurements confirm that antiferromagnetism is metastable within the otherwise ferromagnetic ground state. The apparent lack of coherent spin wave excitations and strong sensitivity to quenched disorder characterize Yb$_2$Ti$_2$O$_7$ as a consequence of this multiphase magnetism.

Significance

Quantum materials have properties that defy conventional theories of solids. Explaining these unusual properties is a frontier in physics, which promises both technological applications and fundamentally new states of matter. Yb$_2$Ti$_2$O$_7$ is a center of attention in this work. While it becomes ferromagnetic at very low temperature, its excitation spectrum resembles that of a quantum spin liquid. We show using neutron scattering on high-quality crystals that the unusual spectra may arise as a superposition of ferromagnetic and antiferromagnetic spin waves. This indicates that these disparate forms of magnetism actually coexist in Yb$_2$Ti$_2$O$_7$, which is consistent with near-perfect degeneracy. The unconventional properties of Yb$_2$Ti$_2$O$_7$ thus appear to arise from the atomic-scale interplay between two conventional types of order.

Author contributions: C.B. designed research; A.S., J.K., S.Z., H.J.C., G.S., G.E., A.H., and C.B. wrote the paper; and S.M.K. provided samples for the experiments.

Published under the PNAS license.

See online for related content such as Commentaries.

1To whom correspondence may be addressed. Email: scheieao@ornl.gov.

This article contains supporting information online at https://www.pnas.org/lookup/suppl/doi:10.1073/pnas.2008791117/-/DCSupplemental.


www.pnas.org/cgi/doi/10.1073/pnas.2008791117

PNAS | November 3, 2020 | vol. 117 | no. 44 | 27245-27254
scattering and find streaks of scattering indicating an unusual lamellar domain structure with a definite orientation relative to the underlying Bravais lattice. Fourth, we show using classical Monte Carlo and semiclassical spin wave theory that antiferromagnetism coexists in metastable state of Yb$_2$Ti$_2$O$_7$. These findings support the hypothesis that ferromagnetism and antiferromagnetism coexists in Yb$_2$Ti$_2$O$_7$ single crystals, forming anisotropic domains that disrupt spin wave propagation.

**Experiments**

**Elastic Scattering.** We collected Yb$_2$Ti$_2$O$_7$ single-crystal elastic scattering data on the Triple Axis Spectrometer for Polarized Neutrons (TASP) at the Paul Scherrer Institute. The sample was a 0.4-g sphere cut from a high-quality Yb$_2$Ti$_2$O$_7$ single crystal (the same sample as in ref. 8). The sphere was mounted on an oxygen-free copper finger and loaded in a dilution refrigerator insert with a (110) direction vertical. We collected elastic scattering ($\hbar\omega = 0.0(1)\) meV) data on the (111), (002), (220), (113), and (004) Bragg peaks, tracking the integrated intensity of rocking curves as a function of temperature. We discovered significant multiple scattering (particularly on the (002) peak), so we collected data using both 5- and 4.5-meV neutrons to isolate single-event diffraction (see SI Appendix for details). We converted the peak intensity to absolute units by also measuring the integrated intensity of the accompanying nuclear peak with structure factors that we calculated from the structure reported in ref. 14. For (002) we interpolated the normalization constant inferred from other Bragg peaks as (002) has zero nuclear intensity. The magnetic structure factor data were fitted to spin structure models assuming an equal population of domains. The results are shown in Fig. 1.

**Spin Wave Spectrum.** We mapped the field- and temperature-dependent energy-resolved neutron-scattering cross-section of Yb$_2$Ti$_2$O$_7$ on the Cold Neutron Chopper Spectrometer (CNCS) at Oak Ridge National Laboratory. We aligned three stoichiometric Yb$_2$Ti$_2$O$_7$ crystals totaling 7.2 g oriented with a (111) direction along a vertical magnetic field and mounted in a dilution refrigerator. These crystals were as described in ref. 8. We collected neutron-scattering data with two instrument configurations. We did a high-neutron-flux measurement with $E_i = 3.32$ meV neutrons, the Fermi chopper at 60 Hz, and the disk chopper at 300 Hz for an energy resolution of 0.15 meV full width at half maximum (FWHM). We also performed a high-resolution measurement with $E_i = 3.32$ meV neutrons, a Fermi chopper at 180 Hz, and the disk chopper at 240 Hz for a FWHM energy resolution of 0.08 meV.

We collected data at 0 (both from a field-cooled and a zero-field–cooled state), 0.35, 0.7, and 1.5 T at base temperature (100 mK) in the high-flux configuration and then zero-field–cooled 0-T data in the high-resolution configuration. We also collected scattering data at 20 K for use as a background. [The elastic line for the in-field data appears oversubtracted—this is due to the field-dependent extinction in Yb$_2$Ti$_2$O$_7$ (31).] We subtracted this background from all datasets, divided by the squared Yb$^{3+}$ magnetic form factor, symmetrized the data using Mantid (32), and made cuts along high-symmetry directions $\Gamma (000) \rightarrow K (220) \rightarrow UL (211) \rightarrow \Gamma (000)$. All high-symmetry cuts plotted in this paper (such as Fig. 2) are symmetrized, but all plotted constant-energy slices (see Fig. SE) are unsymmetrized.

We used the SpinW package (33) to simulate the inelastic scattering cross-section for spin waves calculated for the spin Hamiltonians in refs. 20, 25, and 26 through linear spin wave theory. Each simulation started from the lowest-energy 2-in-2-out spin configuration (optimized separately for each Hamiltonian). We fitted the 1.5 T data (where the spin wave modes are most clearly defined) to a linear spin wave model and extracted a new set of parameters. The data compared with previous theoretical studies are plotted in Fig. 2, while the best-fit spin wave model is plotted in Fig. 3. While consistent with previous scattering data, the revised exchange constants are needed to account for our new data acquired in a different reciprocal lattice plane.

**Small-Angle Neutron Scattering.** We performed two small-angle neutron scattering (SANS) experiments on the SANS-1 instrument at the Heinz Maier-Leibnitz Zentrum. Both experiments used the same 0.4-g spherical sample of Yb$_2$Ti$_2$O$_7$ in a dilution refrigerator, oriented with a (110) direction vertical and a (111) direction along a vertical magnetic field and mounted in a dilution refrigerator, oriented with a (110) direction vertical and a (111)
direction along a horizontal magnetic field oriented perpendicular to the neutron beam. This spherical sample was grown as described in ref. 14 and is the same as used in ref. 8. We collected data with both 4.6- and 17-Å neutrons, to access wave vector transfer from 0.001 to 0.023 Å⁻¹.

We acquired SANS data for temperatures between 800 and 70 mK and fields from 0 to 1 T. The basic scattering pattern is shown (see Fig. 8), where the y axis gives the vertical ⟨110⟩ direction, and the x axis gives the horizontal ⟨111⟩ magnetic-field direction. The field and temperature dependence of SANS features are plotted (see Fig. 9), and the Q dependence is presented (see Fig. 10).

Long-Range Magnetic Order
The measured magnetic Bragg peak intensities are compared to three models in Fig. 1: a ferromagnetic all-in-all-out (AIAO) structure (proposed in refs. 12 and 13), a 2-in-2-out canted ferromagnetic structure (proposed in refs. 11 and 28), and a combination of a ferromagnetic (FM) phase and an antiferromagnetic phase (proposed in refs. 11 and 28). (The two states ψ₂₃ and ψ₂ within Γ₅ have equivalent Bragg intensities for unpolarized neutrons and averaging over all domains, so we are not able to distinguish between them here.) The best fit is the FM+Γ₅, with an ordered moment of 1.3(1) μB, a canting angle of 5.3(8)°, and an effective antiferromagnetic moment of 0.1(2) μB. The large uncertainty on the antiferromagnetic phase is consistent with there being no long-range Γ₅ order. The Γ₅ phase has large (220) Bragg intensity but the observed (220) peak is quite small, which—assuming equal moment sizes for FM and Γ₅—constrains the long-range ordered Γ₅ antiferromagnetic (AFM) phase to occupy less than 10% of the crystal based on the measured Bragg intensities. Extinction is quite strong in Y₂Ti₂O₇ (31), which may account for the less than perfect fit. Note that the Bragg diffraction is insensitive to short-range or quasi-two-dimensional (quasi-2D) order, which we probed through the diffuse scattering as we shall shortly describe.

Although it is key to distinguishing between the AIAO and the 2-in-2-out splayed cubic FM structures, conclusions about the presence or absence of an (002) magnetic Bragg peak have been disputed between different diffraction studies (12, 13, 34). As shown in Fig. 1, Inset we definitively detect a nonzero magnetic intensity at (002) in the absence of any Bragg diffraction for T > Tc, and we can exclude multiple scattering as its origin (SI Appendix). This rules out the AIAO structure as the majority phase in the present sample. This is consistent with the famous pyrochlore phase diagram in ref. 28 (which does not contain the proposed AIAO order) and the associated model Hamiltonian (the refined structure is shown in SI Appendix).

Spin Dynamics and Short-Range Correlations
The inelastic magnetic neutron-scattering cross-section (plotted in Figs. 2 and 3) contains broadened spin-wave–like ridges in zero field and sharper spin-wave–like modes in a field of 1.5 T applied along ⟨111⟩. This result is significant because previous studies in crystals grown at higher temperatures without the benefit of a traveling solvent did not detect spin-wave–like scattering in zero field (11, 20, 21, 25); presumably the higher-quality crystal in this experiment makes the difference. There is no visual distinction between the field-cooled and zero-field-cooled spectrum that we detect (Fig. 2A and B) except a slight enhancement of intensity in the field-cooled (FC) case. In zero field, the modes appear to come down to the elastic line at the (220) Bragg peak.

Having observed zero-field spin waves, albeit broadened, we can compare the data to the magnetic-scattering cross-section associated with the Hamiltonians reported by Ross et al. (25), Robert et al. (26), and Thompson et al. (20) (hereafter referred to as the “Ross,” “Robert,” and “Thompson” Hamiltonians, with the same g tensors as used in each respective study) based on linear spin wave theory. We used the SpinW package (33) to carry out these calculations. As shown in Fig. 2D, F, and H, the only Hamiltonian which predicts a soft (nearly gapless) mode at (220) is Robert; the rest predict a gapped spectrum. At 1.5 T, we again find that the Robert Hamiltonian yields the best match to the data. In particular, the shapes of the modes at (1.5, 1.5, 0) do not match calculations based on the Thompson and Ross Hamiltonians well, but they do match the Robert predictions well (Fig. 2C and G). The agreement with the Robert Hamiltonian spin waves is not perfect—for example, the highest Γ point mode at 1.5 T is at 0.9 meV instead of 1.0 meV as observed—but these comparisons make it clear that the Robert Hamiltonian treated with lowest-order spin wave theory is close to describing the zero-field broadened spin wave modes in Y₂Ti₂O₇. As an aside, this comparison highlights that high-field fits to the spin wave-neutron-scattering cross-section can be underconstrained.
To further refine the magnetic Hamiltonians, we fit the nearest-neighbor exchange matrix to the corresponding intensity of the 1.5-T neutron spectrum where the spin wave modes are sharpest. For Yb on the pyrochlore lattice the exchange matrix takes the form

$$J = \begin{pmatrix} J_2 & J_3 & J_4 \\ -J_2 & J_1 & J_3 \\ -J_3 & -J_3 & J_1 \end{pmatrix},$$

where $J_1$ (XY), $J_2$ (Ising), $J_3$ (pseudodipolar), and $J_4$ (Dzyaloshinskii–Moriya) are the four symmetry-allowed exchange variables (25). The reduced $\chi^2$ is defined as a sum over the $Q - \omega$ pixels spanning Fig. 2C so that least-squares fitting takes account of both the energy and intensity of the modes. (We used the $g$ tensor from ref. 20. The $g$ tensors from refs. 35 and 36 are slightly different but they give values for the exchange constants that are generally within the statistical error bars quoted; see SI Appendix for details.) Just like Robert et al.’s refinement of Ross et al.’s data (26), we find that there is a best-fit line through parameter space ($J_1, J_2, J_3, J_4$) where $\chi^2$ is approximately constant. This is depicted in Fig. 3A, with $\chi^2$ in Fig. 3C. Our line is nearly the same as that in Robert et al. (26). The fit can be constrained to a point along this line by considering the (220) spin wave gap.

The zero-field gap at (220) is a measure of proximity to the $\Gamma_5$ AFM phase. Spin wave simulations show that as the Hamiltonian approaches the AFM ordered phase, soft spin wave modes come down in energy close to the (220), (111), and (113) Bragg peaks (which are the magnetic Bragg peaks for $\Gamma_5$), until they touch zero energy right at the phase boundary (see SI Appendix for details). So the smaller the gap is, the closer to the FM + AFM phase boundary. High-resolution spin wave measurements (see Fig. 7A) show a gap around 0.11(3) meV (Discussion), which is slightly smaller than 0.136 meV, obtained from Robert’s Hamiltonian. We can use this knowledge of the gap size to constrain the fit along the best-fit line, as shown in Fig. 3B. This yields the following optimized parameters:

$$J_1 = -0.026(2) \text{ meV} \quad J_2 = -0.307(3) \text{ meV}$$

This is approximately the same as that in Robert et al. (26). The fit can be constrained to a point along this line by considering the (220) spin wave gap.

The zero-field gap at (220) is a measure of proximity to the $\Gamma_5$ AFM phase. Spin wave simulations show that as the Hamiltonian approaches the AFM ordered phase, soft spin wave modes come down in energy close to the (220), (111), and (113) Bragg peaks (which are the magnetic Bragg peaks for $\Gamma_5$), until they touch zero energy right at the phase boundary (see SI Appendix for details). So the smaller the gap is, the closer to the FM + AFM phase boundary. High-resolution spin wave measurements (see Fig. 7A) show a gap around 0.11(3) meV (Discussion), which is slightly smaller than 0.136 meV, obtained from Robert’s Hamiltonian. We can use this knowledge of the gap size to constrain the fit along the best-fit line, as shown in Fig. 3B. This yields the following optimized parameters:

$$J_1 = -0.026(2) \text{ meV} \quad J_2 = -0.307(3) \text{ meV}$$

For a comparison to previously published results see SI Appendix, Table S2.

To show how close this Hamiltonian is to the phase boundary, we built the phase diagrams in Fig. 4 by mapping the calculated (220) gap as a function of $J_1/|J_1|$ and $J_2/|J_2|$ for the four different Hamiltonians (holding $g$ and $J_4$ constant). This study puts Yb$_2$Ti$_2$O$_7$ closer to the FM + AFM phase boundary than any previous experimental study, although the Robert Hamiltonian also comes close. In passing, we note that $J_3$, although neglected in the plotted phase diagram of ref. 28, noticeably shifts the phase boundary between the FM and AFM states.

**AFM + FM Phase Coexistence.** Although linear spin wave theory based on the fitted Hamiltonian successfully reproduces many features in the Yb$_2$Ti$_2$O$_7$ neutron spectrum, there are some features which it does not reproduce, most notably the broadened excitations at low magnetic fields. We shall argue that these features arise from FM + AFM phase coexistence.

Within the broadened zero-field excitations [which may be broadened by multimagnon decay (20) or by a heterogeneous ground state], a feature not captured by our fitted Hamiltonian is that the zero-field spectrum at ($\pm \frac{1}{2}, \frac{1}{2}, \frac{1}{2}$) (between UL and $\Gamma$) appears to come close to zero energy (Fig. 3J and K). The same is true of the low-energy scattering near $\Gamma$, the ferromagnetic spectrum does not reproduce these features. However, the Hamiltonian can be “nudged” into the AFM phase (by setting $J_2 = -0.313$ meV), and then the simulated scattering spectrum does reproduce these features (Fig. 5). If one takes the average of the FM and AFM spectra (50% weight on each), the simulated spectrum resembles the experimental data more closely (Fig. 5C). Fitting the ratio between FM and AFM scattering with the high-symmetry cuts gives 43(3)% AFM and 57(3)% FM (see SI Appendix for details). The improvement over the FM spectrum becomes especially evident when comparing constant energy slices, as shown in Fig. 5 E–G. The lobe of scattering at ($\pm \frac{1}{2}, \frac{3}{2}, \frac{1}{2}$) observed in the experimental data is absent in the FM calculated spectrum, but it is clearly present in the FM + AFM spectrum. In addition, the FM + AFM spectrum reproduces the elongated spin wave dispersions above the K point (Fig. 5D; see further details in Fig. 7). These features indicate the presence of the $\Gamma_5$ AFM phase in the zero-field ground state of Yb$_2$Ti$_2$O$_7$.

If this line of reasoning is correct, the observed broadening is partly due to the overlap of many spin wave modes (Fig. 5C). However, it seems to apply only to the zero-field state: At nonzero magnetic fields the FM + AFM spectrum does not match the scattering data (SI Appendix). Thus, it appears that the FM + AFM phases coexist only at very low fields ($|B| < 0.1$ T).

One may object that the above analysis is inconsistent with the elastic scattering, which shows long-range antiferromagnetism constitues $<10\%$ of the ground-state order. This is, however, not the case because the Bragg peaks are exclusively associated with three-dimensional long-range order. If the AFM order were present in smaller regions (for instance, within domain walls) or on short timescales, it would not produce sharp Bragg peaks and would only weakly influence the refinement of the Bragg diffraction data.

**Theoretical Simulations.** To better understand the nature of the coexisting AFM and FM phases, we simulated the neutron spectrum of Yb$_2$Ti$_2$O$_7$ by first performing classical Monte Carlo (MC) simulation based on our spin Hamiltonian in Eq. 2, followed by linear spin wave theory on the corresponding optimized spin configuration. The MC simulations were conducted for $T = 0.2$ K using single-spin updates...
for continuous spin on pyrochlore lattices (with 16 site cubic unit cells) of size $N = 16L^3$ for $L = 8$ (8,192 total spins) for a total of $2 \times 10^{8}$ steps. Further iterative minimization (37) was performed on the last configuration encountered in the run to obtain the classical spin configuration that corresponds to the nearest local energy minimum. This entire process was performed 400 times for different starting random seeds. Such a simulation cell is found to not be large enough to capture domain effects, but it is large enough to probe the stability of AFM and FM states. We found that each of these configurations became trapped either in the canted ferromagnetic phase or in the $\psi_2$ ($\Gamma_2$) manifold with a preference for $\psi_2$ (SI Appendix, Fig. S10). In all 400 cases, the configurations correspond to a $q = 0$ state; i.e., all tetrahedra were alike. This lends credence to the idea of phase coexistence: Classically, pockets of the system are kinetically trapped in the $\psi_2$ AFM phase.

The preference for $\psi_2$ is surprising because the $\text{Yb}_2\text{Ti}_2\text{O}_7$ Hamiltonian is near the $\psi_2$ phase boundary. This occurs because classically, $\psi_2$ and $\psi_3$ are degenerate, and $\psi_3$ is an unstable saddle point in energy that will rotate into the canted ferromagnetic ground state without increasing the total energy of the spin configuration (28). $\psi_2$, meanwhile, is protected by a finite-energy barrier and becomes favored by fluctuations (see SI Appendix for details). Put a different way, while $\psi_2$ is degenerate, the system has an easier path to canted ferromagnetism from $\psi_3$ than from $\psi_2$, and thus $\psi_2$ can form a metastable phase. Thus we do not expect to see any stable $\psi_3$ states in a finite-size MC simulation without dipole interactions, which is consistent with our MC results.

Calculating and summing the calculated spin wave spectra from all 400 states (computational details are in SI Appendix), we get a remarkable agreement with the observed spin wave spectrum, as shown in Fig. 6. This suggests the broadened zero-field spin wave spectrum arises from admixture of antiferromagnetic regimes into the otherwise ferromagnetic low-$T$ spin configuration.

Although the phase coexistence hypothesis successfully accounts for many features in the measured neutron spectrum, not all features are captured by the MC + linear spin wave theory (LSWT) simulations. Most notable is the low-lying excitation at the $K$ point above the (220) Bragg peak shown in Fig. 7 (circular binning around (220) is shown in Fig. 7 C–E to highlight the dispersion). Spin wave theory, from both the FM and the AFM phase, predicts strongly dispersive modes atop (220). However, the measured spectrum shows an intense flat mode at 0.11 meV that is not accounted for by any of these simulations. A similar flat mode was also observed above the (111) Bragg peak by Antonio et al. (34) at 0.10 meV. These may possibly be associated with tunneling in and out of the AFM phase. Alternatively, standing AFM spin waves within a finite-size AFM domain or domain wall might produce such scattering.

Considering regions of AFM within a FM ground state brings us to larger length scales where dipolar interactions become relevant. To experimentally probe such length scales and explore the role of magnetic domains and dipolar interactions in $\text{Yb}_2\text{Ti}_2\text{O}_7$, we turn to small-angle neutron scattering.

![Inelastic magnetic neutron-scattering data for Yb$_2$Ti$_2$O$_7$ compared to the calculated magnetic neutron-scattering cross-section for FM and AFM and mixed FM + AFM ground states.](image1)

![Monte Carlo LSWT calculated neutron spectrum for Yb$_2$Ti$_2$O$_7$ (A) compared to the field-cooled 70 mK spectrum (B).](image2)

![MC + LSWT simulation better matches the scattering pattern, particularly near ($\frac{1}{4}, \frac{3}{4}, \frac{1}{4}$) at 0.2 meV.](image3)
Mesoscale Magnetic Structure

SANS within the magnetic ordered phase shows highly anisotropic mesoscale spin correlations. The SANS pattern shown in Fig. 8 has a star pattern formed by streaks of scattering extending along the (111), (110), and (201) directions. The (111) streaks have been previously reported (38), but not the streaks extending along the (111), (1¯10) shown in Fig. 8 has a star pattern formed by streaks of scattering along a finite Q vector extending from the origin of reciprocal space to the measured plane. Since [110] is parallel to the vertical rotation axis of the cryostat, we could not measure its rocking curve and cannot say whether the (110) scattering is plane-like or rod-like in three-dimensional Q space.

Fig. 9 shows the temperature and field dependence of the different SANS features (the windows defining the regions of interest are shown in Fig. 10 A–C, Insets). The temperature dependence of the zero-field SANS shows a clear onset at the ordering temperature of 270 mK. At 0.1 and 0.2 T, the ordering transition (shown by the onset of (111) scattering) increases in temperature in accord with the reentrant phase diagram (8). In 0.1- and 0.2-T applied fields, (110) and (201) scattering is absent within the ordered phase, but appears briefly near Tc. (Full colormap plots of paramagnetic, critical, and ordered scattering patterns are shown in SI Appendix, Fig. S9.) Meanwhile, the field-dependent SANSs are shown in Fig. 9 D–J. At 120 mK (within the ordered phase), an applied field initially enhances the (111) scattering and then gradually suppresses it (most clearly in Fig. 9 G–J), while the (110) and (201) scatterings are totally suppressed by 0.1 T [where the internal demagnetizing field becomes nonzero (8)]. Reducing the field from 0.8 T at 120 mK results in a much more gradual increase in (111) SANS, but an anomaly occurs at 0.1 T when (110) and (201) intensity reappears. At 300 mK [above the zero-field Tc and where the ordered phase is reentrant versus field (8)], hysteresis in the field dependence of (111) intensity is less pronounced than at 100 mK, and the (110) and (201) intensity appears only at the lower field phase boundary.

The streaks of scattering follow a power law I = AQ−n (where I is scattering intensity, Q is the scattering vector, and A is a fitted constant), but the power n is different for the three different
visible streaks as shown in Fig. 10. (These data were taken only from the maximum-intensity cryostat rotation angle.) For wave vector transfer along the [111] oriented field direction (which, because we have a cubic sample, is parallel to [111]), the exponent is \( n = 3.37(7) \) in zero field and \( n = 3.04(6) \) at 0.075 T. For the (110) and (201) directions (where the SANS vanishes beyond 0.05 T), \( n = 3.81(7) \) and 4.16(7), respectively, in zero field.

The power \( n \), when measured from randomly oriented surfaces, is called a Porod exponent: It provides insights into the real space structure underlying the SANS (41). However, this theory does not apply to the anisotropic SANS pattern we see here. According to ref. 42, the SANS scattering from a sharp discontinuity in the scattering length density (like a ferromagnetic domain wall) goes as

\[
S(Q) = C \frac{1}{Q^2} \delta(Q_x) \delta(Q_y),
\]

where the discontinuity is normal to \( z \) and the incident beam along \( x \). Integrating over detector pixels in the direction perpendicular to the plane yields

\[
I = I_0 \int \sin(2\theta) d(2\theta) S(Q) \propto \frac{1}{Q^6}.
\]

Thus the (111) rods of small-angle scattering can arise from a lamellar pattern of planar domain walls with normals along (111).

The streaks of scattering extending along (110) and (201) have exponents closer to \( n = 4 \), which is more difficult to explain theoretically. However, \( Q^{-4} \) and \( Q^{-3} \) SANS power-law exponents along different crystallographic directions have been observed from superalloy grain boundaries (43) due to sharp edges and double curvature. Similar effects could be at play here in the surfaces of the domain walls. This may also explain why the (201) streak intensity is independent of \( Q \) (Fig. 8D): If they are due to domain wall edges (not lamellar flat domain walls), they will not have a rod shape.

The extreme anisotropy of the SANS features is unusual for ferromagnets (44). It is important to ask why the magnetic domains are so anisotropic, and the explanation may lie in the domain walls’ internal structure. Because the magnetic Hamiltonian for \( \text{Yb}_2\text{Ti}_2\text{O}_7 \) is on the boundary between a canted ferromagnetic and \( \Gamma_5 \) antiferromagnetic phase, then the lowest-energy domain wall includes a slab of \( \Gamma_2 \) antiferromagnetic phase, as depicted in Fig. 11. For a domain wall separating \( \hat{z} \) from a \( –\hat{z} \)-oriented magnetization domain, the rotation is a simple transition through a \( \psi_3 \) phase. But if the rotation is to a domain at 90°, the domain wall rotates through a \( \psi_3 \) phase, then rotates within the \( \psi_3 – \psi_2 \) manifold [which is \( U_1 \) degenerate at the mean-field level (45), and then rotates to the ferromagnetic phase again. On the FM + AFM phase boundary, such a rotation costs zero energy, but slightly away from the phase boundary it is still the lowest-energy way to connect two FM states (SI Appendix).

This observation may explain the dramatic anisotropy in the domain wall structure. The \( \text{Yb}_2\text{Ti}_2\text{O}_7 \) FM domains can point in \( \pm[100], \pm[010], \) or \( \pm[001] \) directions. Dipolar interactions favor domain walls with zero net magnetic charge (i.e., the dot product of the magnetization vector and the domain wall normal pointing into each domain are identical). Thus, if two domains are magnetized along \( [100] \) and \( [010] \), then the domain wall separating them can have surface normals along the (110) or (111) directions, which may explain the streaks of scattering extending in those directions. The (201) streaks of scattering extending along (201) and (021) are independent of sample rotation around the vertical \([1,–1,0]\) axis and have a larger power-law exponent. These could be associated with intersections between (110)- and (111)-oriented domain walls as in the growth habit for a cubic crystal with a preference for (111) and (110) surfaces. Rather than sharp edges where (111) and (110) surfaces meet, smaller (201)- and (021)-oriented surfaces are anticipated. The lack of

---

**Fig. 9.** Field- and temperature-dependent SANS data for \( \text{Yb}_2\text{Ti}_2\text{O}_7 \). (A–C) Temperature dependence of the (111), (100), and (201) rods at 0, 0.1, and 0.2 T. (D–F) Field dependence of the (111), (110), and (201) rods at 120 and 300 mK. The (111), (110), and (201) directions are defined in the insets to Fig. 10. Insets. (G–J) Colormap images of the \( \text{Yb}_2\text{Ti}_2\text{O}_7 \) SANS pattern as a function of field at 100 mK.

**Fig. 10.** Fits to SANS in \( \text{Yb}_2\text{Ti}_2\text{O}_7 \). (A–C) The \( Q \)-dependent scattering at various fields in the (A) (111), (B) (110), and (C) (201) directions. The regions defining these directions are shown in A–C. Insets. (D and E) The results of a Porod exponent fit \( I = AQ^{-n} \).
a rocking curve peak for the corresponding streaks of scattering and the larger exponent, \( n \), are both consistent with those facets having a smaller extent than for the (111)- and (110)-oriented domain walls.

The existence of lamellar domain walls could be a mechanism for FM + AFM phase coexistence. At the FM/AFM boundary, FM and \( \psi_2 \) become degenerate; and at the mean-field level (neglecting thermal and quantum fluctuations), \( \psi_3 \) and \( \psi_2 \) are degenerate. This allows for both \( \psi_3 \) and \( \psi_2 \) to be present and favored as domain walls by dipolar interactions within the ferromagnetic ordered state (Fig. 11). In other words, dipolar interactions require domain walls, which take on an antiferromagnetic character and could occupy a considerable volume fraction because of the near degeneracy between FM and AFM. The quasi-2D nature of the AFM volume fraction means that it gives rise to rods of scattering and not proper Bragg peaks. Indeed, the rods of scattering that we see in SANS may well in part be ascribed to AFM diffraction.

To examine whether the apparent phase coexistence indicated by the inelastic scattering could arise from domain walls, we carried out the MC + LSWT analysis for domain walls: We minimized the energy for a particular AFM domain wall and calculated the spin wave spectrum. However, the levels of AFM scattering were far too weak for this to be a reasonable explanation for the broad low-field spectrum; the domain walls were simply too sparse. The observed scattering pattern requires more extended AFM within the sample, which would perhaps be produced by the influence of magnetic dipolar interactions. (Such a signal may produce a diffuse elastic signal at \( Q \approx \frac{1}{2\pi} \) be produced by the influence of magnetic dipolar interactions.\( Q \approx \frac{1}{2\pi} \)\) and \( \pm \frac{1}{2} \) ferromagnetic domains, and bottom image shows a domain wall between \( +x \) and \( +z \), rotating between two states in the \( \Gamma_5 \) manifold.

Fig. 11. Domain walls in \( \text{Yb}_2\text{Ti}_2\text{O}_7 \) rotating through a \( \Gamma_5 \) antiferromagnetic phase. Top image shows a domain wall between \( -z \) and \( +2 \) ferromagnetic domains, and bottom image shows a domain wall between \( +x \) and \( +z \), rotating between two states in the \( \Gamma_5 \) manifold.

Discussion

This study resolves several long-standing puzzles concerning \( \text{Yb}_2\text{Ti}_2\text{O}_7 \). First, we have shown that the ground-state order is the 2-in-2-out canted ferromagnetism as predicted by theory (28). Second, we have shown that \( \text{Yb}_2\text{Ti}_2\text{O}_7 \) has some surprising properties, albeit broadened, in the zero-field state. Third, we have shown that soft zero-field spin wave modes are those of the AFM \( \Gamma_5 \) phase, which constitutes evidence that \( \text{Yb}_2\text{Ti}_2\text{O}_7 \) is near the phase boundary and contains a significant volume fraction of short-range AFM order within the otherwise FM ground state.

Our SANS data show a highly anisotropic magnetic domain structure, which may be associated with incorporation of AFM slabs as domain walls. We also have demonstrated using Monte Carlo simulation that \( \psi_2 \) is a metastable phase of \( \text{Yb}_2\text{Ti}_2\text{O}_7 \) at low temperatures, and fluctuations may generate the AFM scattering patterns that we observe in low-energy inelastic magnetic neutron scattering.

The nature of the boundary between the FM and AFM phases, as revealed by our spin wave calculations, is quite interesting. The linear spin wave calculation shows that the FM ground states have a tendency to fluctuate into the \( \Gamma_5 \) states. The finite-energy flat band mode at (220) carries the structure factor of the \( \Gamma_5 \) order parameter and the energy scales as the square root of the energy cost of the \( \Gamma_5 \) states. This continuous gap closing suggests a continuous phase boundary between FM and AFM states. However, numerical calculations in ref. 26 show it is a first-order boundary even at finite temperature, allowing for coexistence of phases. These calculations are both approximate, and we leave this apparent contradiction to be resolved in a future study.

The natural question that arises from our calculations and procedure is, Why does averaging over the structure factor of different spin configurations reproduce the neutron spectrum (Fig. 6)? The MC simulations suggest that regions of AFM order are kinetically trapped in a system which has partly lost ergodicity. However, the diffraction results in Fig. 1 show \( \sim 10\% \) of the elastic magnetic scattering is in the form of antiferromagnetic Bragg scattering. This means that the AFM components are localized in space and/or time. The MC simulations do not reveal small AFM regions (smaller than \( 8 \times 8 \times 8 \) unit cells). However, these simulations do not include magnetic dipolar interactions which could produce a dense network of AFM regions in between FM domains as a means of reducing the magnetostatic energy. Under this hypothesis, the flat modes above (220) and (111) may be due to standing spin waves in a finite-sized AFM region favored by dipole interactions. If the antiferromagnetism is restricted to domain walls (extended in two dimensions and constrained in a third), its spin waves orthogonal to the wall will have nodes at the edges of the AFM domain, which leads to standing wave resonance modes at nonzero energies. In other words, the flat modes above (220) and (111) may be spin wave resonance modes within the effective “quantum well” of a domain wall.

Alternatively, it could be that \( \text{Yb}_2\text{Ti}_2\text{O}_7 \) fluctuates in time and out of the FM and AFM phases, such that AFM scattering appears only at finite-energy transfer. Such fluctuations would occur via quantum effects, which are also neglected by the MC simulations. We know from refs. 8 and 24 that quantum effects are important—they can change the energy of a factor of 2 in zero field. Moreover, if one looks at the classical energy histogram, the energy difference between the AFM and the FM manifold is only about 0.0025 meV per site. This means that quantum mechanical tunneling locally between the FM and the AFM will be possible.

The classical order parameters describing the system, FM, \( \psi_2 \), and \( \psi_3 \) are associated with noncommuting operators at the quantum level, indicating tunneling between these classically defined ordered states will occur. If quantum tunneling from one state to another happens coherently in a region on some long timescale (\( \sim 0.1 \) ns), by measuring the classical order parameter, we might get a sense of domain wall sweeping through the region. In addition, the flat modes above (220) and (111) may be low-energy modes of the spins tunneling in and out of the AFM phase. Such a mechanism would yield a neutron-scattering signal, and these flat modes are one of the most glaring features not captured by semiclassical theory. It must also be added that even the static correlations at 50 mK (i.e., in the ordered phase) show signatures of both FM and AFM correlations, as has been discussed in recent work by Pandey et al. (46). These static correlations were modeled by taking a FM/AFM ratio of 2/3 and averaging their individual structure factors similar in spirit to what has been done in our work. Remarkably, the averaging procedure...
Conclusions

This study puts the enigmatic Yb$_2$Ti$_2$O$_7$ order phase in a different light: that of phase coexistence. Interactions in Yb$_2$Ti$_2$O$_7$ create a mixed magnetic state with regions of antiferromagnetism confined within an otherwise ferromagnetic phase. Previous authors have studied phase competition (27, 28) and proposed phase coexistence in the paramagnetic phase (46). We have presented corroborating lines of evidence— theoretical and experimental—that phase coexistence persists even within the ordered state of the highest-quality single crystalline samples of Yb$_2$Ti$_2$O$_7$. Many puzzling and strongly sample-dependent features of the materials that first drew attention to Yb$_2$Ti$_2$O$_7$ persist today.


J. D. Thompson et al., Quasiparticle breakdown and spin Hamiltonian of the frustrated quantum pyrochlore Yb$_2$Ti$_2$O$_7$ in a mixed field. J. Appl. Phys. 120, 094422 (2016).


Y. Han, O. Bentol, J. Jaubert, N. Shannon, Theory of multiple-phase competition in pyrochlore magnets with anisotropic exchange with application to Y$_2$Ti$_2$O$_7$, Sr$_2$Ti$_2$O$_7$, and Er$_2$Sn$_2$O$_7$ (49) in a near phase boundary; it would be interesting to explore evidence for multiphase magnetism in these materials too.

Data Availability. Some study data are available.

ACKNOWLEDGMENTS. This work was supported as part of the Institute for Quantum Matter, an Energy Frontier Research Center funded by the US Department of Energy (DOE), Office of Science, Basic Energy Sciences under Award DE-SC0019331. This research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory, managed by UT-Battelle, LLC, under contract DE-AC05-00OR22725 with the US Department of Energy (DOE). The US government retains the right to publish or reproduce the published form of this article, or allow others to do so, for US government purposes. DOE will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (http://energy.gov/downloads/doe-public-access-plan).


12. A. Aouac, P. D. de R´eotier, L. Keller, B. Roessli, A. Forget, A novel type of splayed domain walls prefer to align normal to the ⟨111⟩ direction. Large-box classical simulations of AFM domain walls based on the Hamiltonian derived above show a mild preference for a domain to be along ⟨100⟩, but not ⟨111⟩ as indicated by the data—but these simulations neglected dipolar interactions and 2) dynamic fluctuations into the AFM phase. The specific mechanism needs to be clarified with quantitative theory and further experimental work for example at very high-resolution energy.


15. R. M. D'Ortenzio et al., Unconventional magnetic ground state of the highest-quality single crystalline samples of Yb$_2$Ti$_2$O$_7$. Many puzzling and strongly sample-dependent features of the materials that first drew attention to Yb$_2$Ti$_2$O$_7$ persist today.


