

Dynamic Magnetic Ground State in a Dimer Based Compound $\text{Yb}_2\text{Te}_5\text{O}_{13}$

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In this letter, we study the magnetic and thermodynamic properties of a Yb^{3+} dimer-based compound $\text{Yb}_2\text{Te}_5\text{O}_{13}$ down to sub-Kelvin temperatures using various characterization techniques. Our magnetisation measurements, conducted down to 0.4 K, reveal the absence of long-range magnetic ordering and the emergence of magnetic correlations below 1 K. Additionally, heat capacity measurements performed down to 0.37 K confirm the presence of a finite internal magnetic field of the order of 0.55 K and validate the presence of magnetic correlations with a Kramers doublet ground state ($J_{\text{eff}} = 1/2$). Further insight is provided by zero field muon spin relaxation (μSR) measurements, which show a gradual slowdown of Yb^{3+} spin fluctuations from 30 K down 44 mK, indicating the presence of the Orbach process mediated through crystal electric field (CEF) levels. Furthermore, longitudinal field μSR spectra collected at 70 mK demonstrate that the spins are dynamic even in the presence of $H = 3200$ Oe. Our theoretical investigation unveils the competing exchange interaction and, hence, the origin of the dynamic state. By combining macroscopic and microscopic measurements with theoretical calculations, we propose a quantum spin liquid state within a dimer lattice of $\text{Yb}_2\text{Te}_5\text{O}_{13}$, presenting exciting avenues for future research in quantum magnetism.

The discovery and characterization of novel states of matter have been a driving force in condensed matter physics, offering insights into the fundamental principles governing quantum systems and opening doors to potential technological applications. Among these exotic phases, quantum spin liquids (QSLs) [1–3] are particularly intriguing due to their defiance of conventional magnetic order and the emergence of exotic quantum phenomena. The study of quantum spin liquids can answer various fundamental questions in physics, ranging from the nature of quantum entanglement [4, 5] and high-temperature superconductivity [6] to the existence of fractionalized excitations [7, 8] and topological order [9]. Moreover, QSLs hold promise for applications in quantum information processing, where the robustness of their entangled states could be harnessed for fault-tolerant quantum computing. Various spin lattices, including the triangular [10], kagome [11], hyperkagome [12], pyrochlore [13], square lattices [14] and dimer lattices with frustration [17], can realize spin liquid states. Recently, rare-earth-based frustrated systems, particularly Yb^{3+} based, have been widely considered among the leading candidates for realizing novel quantum states owing to the presence of strong spin-orbit coupling (SOC) and CEF. A combination of a low-dimensional frustrated magnetic lattice and $J_{\text{eff}} = 1/2$ ground state in Yb com-

pounds proves to be an ideal platform for a quantum spin-liquid state.

Herein, we report the discovery of a magnetically dynamic state in $\text{Yb}_2\text{Te}_5\text{O}_{13}$, in which Yb^{3+} ions form dimers in the bc -plane and stack along the a -axis. A single-crystal and polycrystalline sample of $\text{Yb}_2\text{Te}_5\text{O}_{13}$ were prepared using the synthesis routes mentioned in sections 1 and 2 of the supplemental material [16]. The refinement of the X-ray diffraction (XRD) of both the samples confirms that $\text{Yb}_2\text{Te}_5\text{O}_{13}$ crystallizes in a triclinic space group ($P\bar{1}$). Also, the simulated XRD pattern from the single-crystal perfectly agrees with the XRD pattern of the polycrystalline sample, as shown in Fig. S2 of the supplemental material [16]. The refined parameters, which include crystallographic positions, thermal parameters, and occupancies of each atom, are tabulated in Table S1 of the supplemental material [16]. A careful investigation of the crystal structure reveals that Yb^{3+} ions have two distinct Wyckoff positions with two different chemical environments. Yb1 atoms are surrounded by eight oxygen atoms in a bicapped trigonal-prismatic manner, while Yb2 atoms are situated in a pentagonal bipyramid environment of seven oxygen atoms. The Yb atoms form a zig-zag chain along the a axis and form dimers comprising of edge-sharing Yb1O_8 and Yb2O_7 units having an intra-dimer length of 3.81 Å. The inter-dimer distance along the b and c axis is more than 6 Å, confirming that the dimers along the a -axis are responsible for the magnetic properties of the compound. It has been established previously that competition be-

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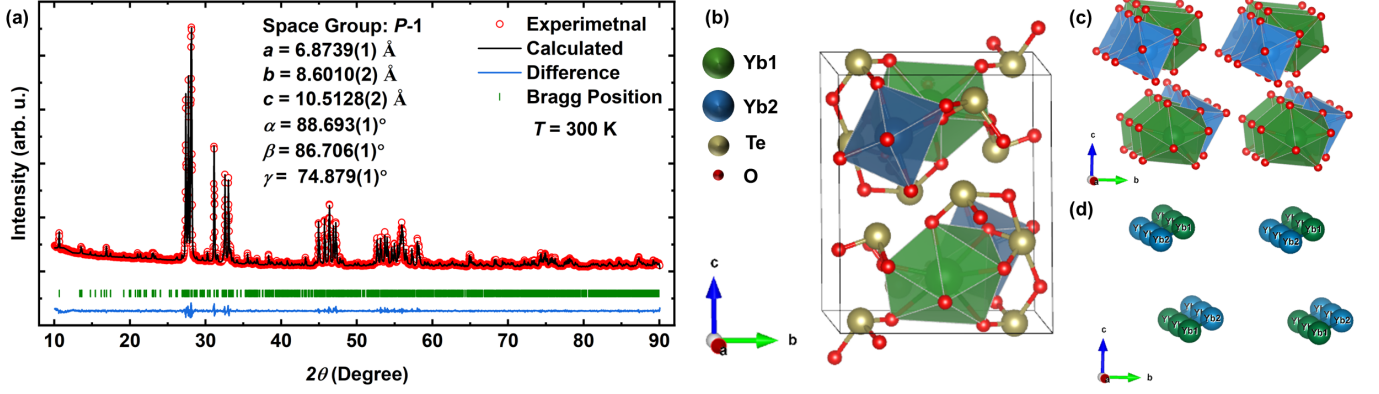


FIG. 1. (a) Refinement of the x-ray diffraction pattern of $\text{Yb}_2\text{Te}_5\text{O}_{13}$. (b) Single unit cell of $\text{Yb}_2\text{Te}_5\text{O}_{13}$. (c) Stacking of the Yb-based two types of polyhedra in the bc plane. (d) Stacking of Yb1-Yb2 dimers along the a -axis.

tween inter- and intra-dimer interactions can lead to a magnetically dynamic state [17]. Given these intriguing properties of $\text{Yb}_2\text{Te}_5\text{O}_{13}$, there is a strong motivation for its low-temperature magnetic behaviour to be characterised.

The dc magnetic susceptibility measurements have been carried out down to 0.4 K, following zero-field-cooled (ZFC) and field-cooled (FC) protocols. We did not observe any magnetic anomaly, indicating the absence of magnetic long-range ordering. **The inverse susceptibility data show deviation from the simple Curie-Weiss model below 125 K, as shown in Fig. S3(a) [16], which is due to splitting the eightfold ground state of Yb^{3+} ($4f^{13}$) ions as a result of CEF. At low temperatures, the ground state can be described as a Kramers doublet with $J_{eff} = 1/2$.** As J is no longer temperature independent, the higher Kramers doublets can also contribute over the temperature range of the measurement (0.4-300 K). So, the inverse susceptibility data can be fitted with a two-level approximation of the Curie-Weiss model, which can be expressed as [18, 19]:

$$1/\chi = 8 \cdot (T - \theta_{CW}) \cdot \left(\frac{1 + \exp^{-\frac{E_{10}}{k_B T}}}{\mu_{eff,0}^2 + \mu_{eff,1}^2 \cdot \exp^{-\frac{E_{10}}{k_B T}}} \right) \quad (1)$$

where θ_{CW} is Curie-Weiss temperature, $\mu_{eff,0}$ and $\mu_{eff,1}$ stand for effective moments in the crystal field ground and first excited states, respectively, and E_{10} denotes the energy difference between them. The fitting of the inverse susceptibility data, presented in the inset of Fig. 2(a), results in $E_{10} \sim 173$ K, further confirming the Kramers doublet ground state at lower temperatures. **The θ_{CW} is found to be -1.73(5) K, suggesting dominating antiferromagnetic interactions. Moreover, the effective moment $\mu_{eff} = [(3k_B/N_A\mu_B^2)\chi T]^{1/2} \sim 2.828\sqrt{\chi T}$ approaches a free ion value corresponding to two Yb^{3+} ions at 300 K ($6.36 \mu_B$), as shown in Fig. S3(b) [16]. The effective moment gradually decreases from 300 to 1.7 K and then shows a sharp decrease below 1.7 K, suggesting the emergence of the magnetic correlations [20].**

To understand the low-temperature magnetic properties further, we have performed isothermal magnetization measurements at 0.4, 0.7, 1.0, and 1.2 K by constantly sweeping the magnetic field from +7 T to -7 T in five quadrants. Figure 2(b) depicts the field dependence of the isothermal magnetization curves in the first quadrant, which seem to saturate above 3 T after rapidly increasing in the low-field region. The slope of magnetization curves at higher magnetic fields correspond to the Van Vleck contribution produced by higher crystal electric field (CEF) levels [21]. After subtracting the Van Vleck contribution, the saturation magnetization is $3 \mu_B/\text{f.u.}$, which is in agreement with the saturation magnetization values observed for other Yb-based polycrystalline compounds [21, 22]. Furthermore, the paramagnetic state of a compound can be described by $M(H) = \chi_{\nu\nu}H + gJ_{eff}N_A\mu_B B_{J_{eff}}(H)$, where $B_{J_{eff}}(H)$ is the Brillouin function [23]. We observed that the magnetization data slightly deviates from the Brillouin function fit below 1 K, as can be seen from the inset of Fig. 2(b). This deviation suggests the emergence of magnetic correlations below 1 K, which requires further exploration.

To explore the thermodynamic properties at low temperatures, we conducted specific heat measurements on a polycrystalline sample of $\text{Yb}_2\text{Te}_5\text{O}_{13}$ down to 370 mK. No lambda anomaly corresponding to the main phase has been observed. A small anomaly was observed at 2.3 K, which can be attributed to a tiny amount of Yb_2O_3 ($\approx 0.5\%$) impurity phase, as depicted in Fig. 3. However, the presence of this impurity phase does not affect the main phase properties. The specific heat data have been fitted with the Debye-Einstein model to separate out the magnetic contribution (see inset of Fig. 3(a)), as discussed in section 5 [16]. Despite the absence of long-range magnetic ordering, we observed a Schottky anomaly at low temperatures for $H \geq 1$ T. The Schottky anomaly originates from the splitting of the Kramers doublets in the presence of the magnetic field. The maximum in the Schottky anomaly, which corresponds to the energy at which both the levels are equally degenerate,

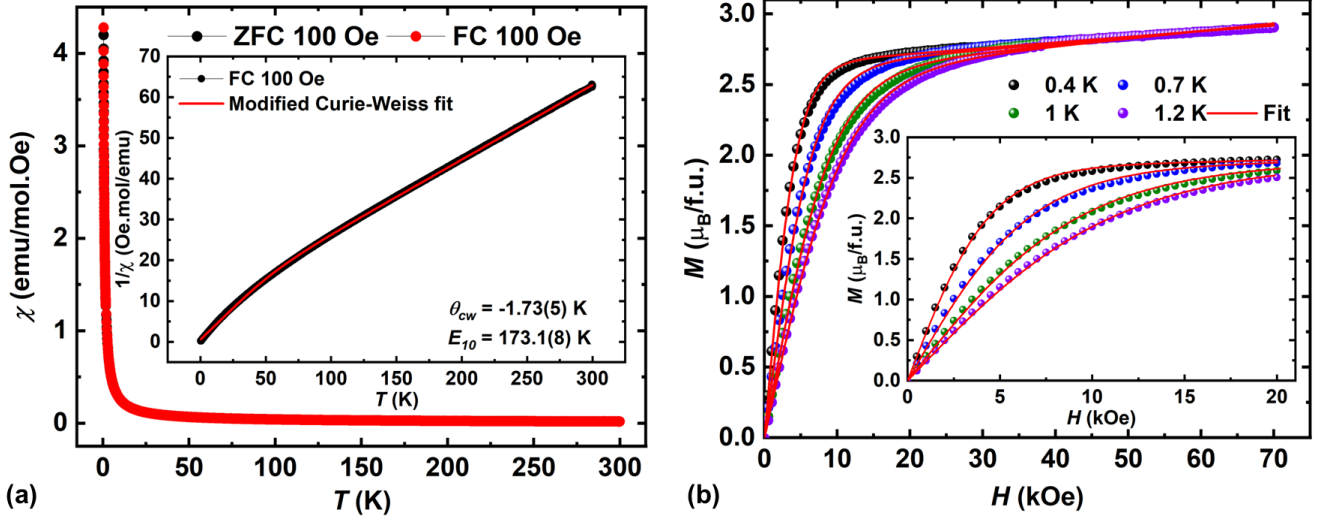


FIG. 2. (a) Temperature dependence of dc magnetic susceptibility performed under ZFC and FC protocol down to 0.4 K. Inset shows the fitting of the inverse susceptibility data with the modified Curie-Weiss model. (b) Isothermal magnetization curves measured at different temperatures along with the Brillouin fits represented by solid lines. Inset shows the zoomed version of the fitting in the low-field region.

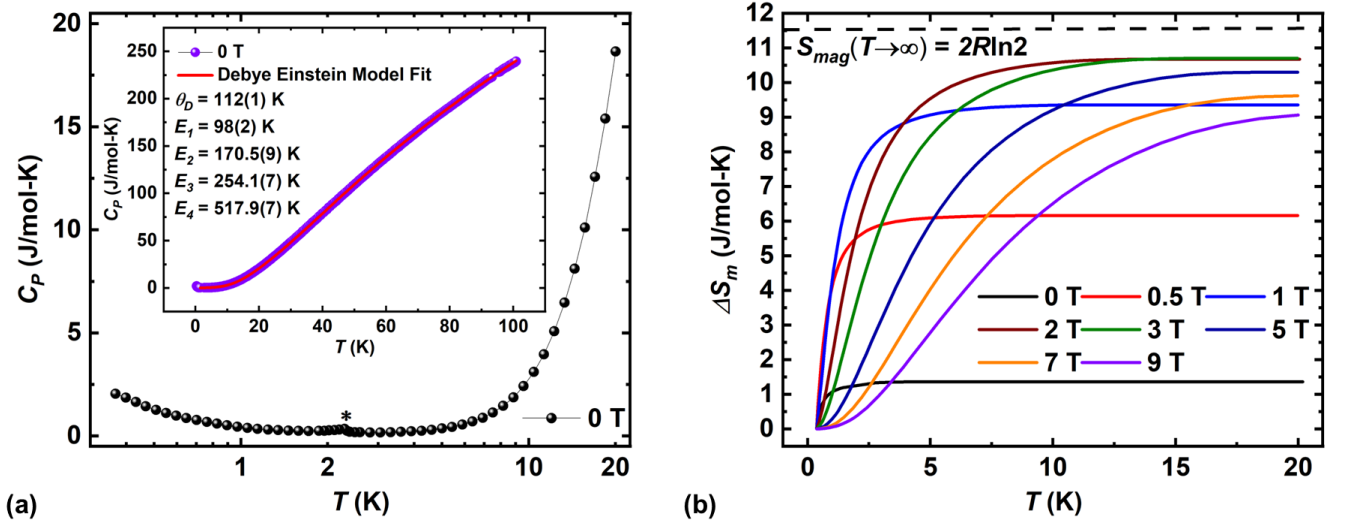


FIG. 3. (a) Temperature dependence of the specific heat in the absence of an applied magnetic field. The asterisk indicates an anomaly corresponding to Yb_2O_3 ($T_N = 2.3$ K). Inset: Fitting of zero field specific heat data using the Debye-Einstein model. (b) Temperature dependence of the magnetic entropy, derived from the specific heat data, is depicted under different magnetic fields.

can be used to estimate the CEF ground state magnetic moment by using the following expression [32–34]: $\Delta = T_{\text{max}} \cdot k_B / 0.417$. The Schottky anomaly has a maxima around 1.8 K for 2 T, which results in $1.6 \mu_B$ per Yb^{3+} ion. This agrees well with the saturation magnetization per Yb^{3+} at low temperatures. Moreover, analysis of the Schottky anomaly reveals the presence of Yb^{3+} - Yb^{3+} correlations of the order of 0.5 K, as described in the last para of section 5 [16].

The magnetic entropy, obtained by subtracting the lattice contribution, is illustrated in Fig. 3(b). A compar-

ison with the entropy of a two-level system (with an effective spin $J_{\text{eff}} = 1/2$) of two Yb^{3+} ions reveals that approximately 88% of the magnetic entropy embedded in the system below 360 mK. Furthermore, the temperature dependence of the magnetic entropy under various magnetic fields reveals that saturation magnetic entropy continues to rise with an increase in the applied magnetic field and attains a value of 10.75 J/mol-K for $H \geq 2 \text{ T}$, which is approximately 93 % of $2R\ln 2$, further corroborating the $J_{\text{eff}} = 1/2$ ground state.

Considering the extreme sensitivity of muons to very

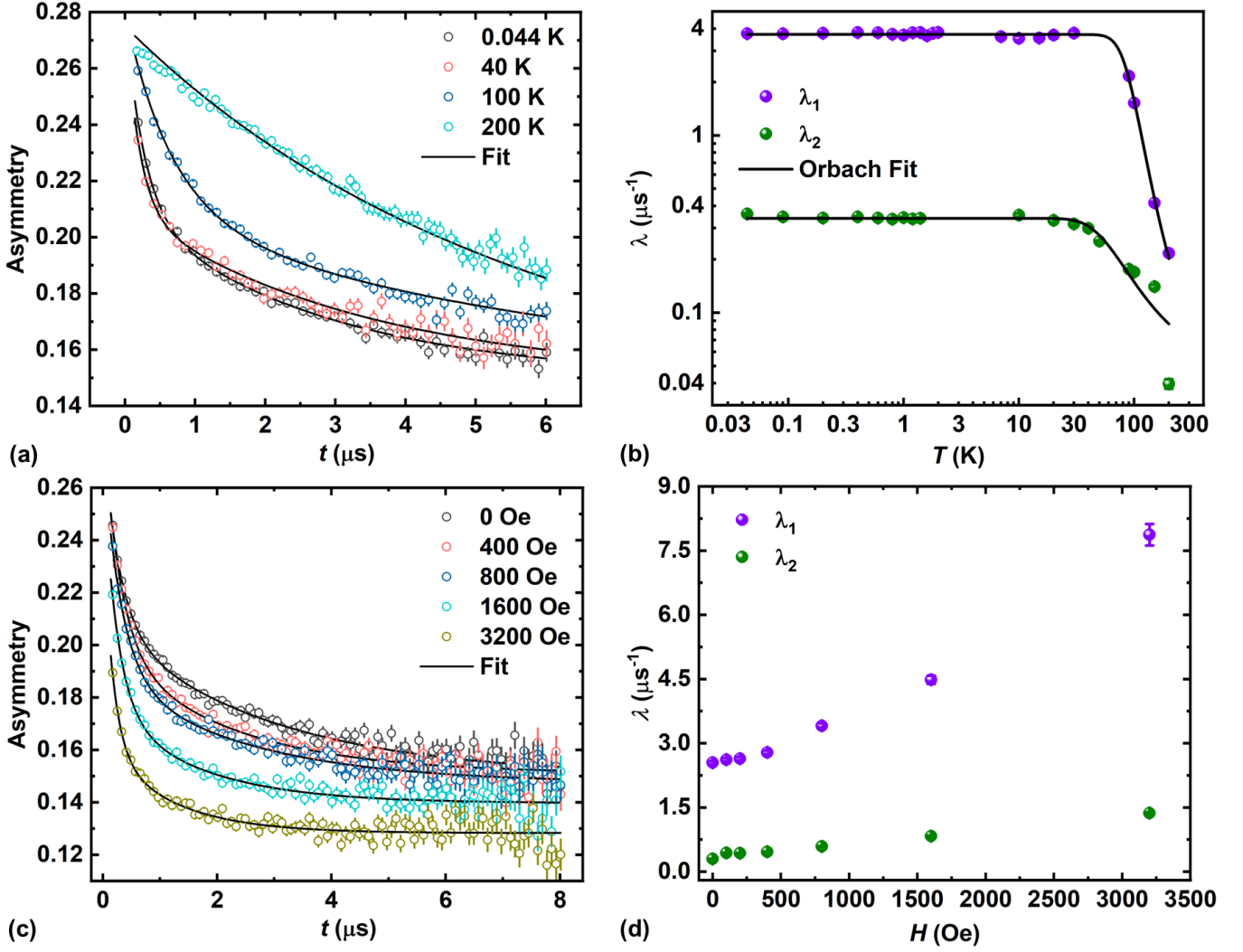


FIG. 4. (a) Zero field muon spin relaxation data measured at different temperatures. The solid lines show fits to the data using equation 2. (b) The temperature dependence of the relaxation rates obtained from the fitting. The solid line depicts their fitting with the Orbach process (equation 3). (c) Longitudinal Field measurements are carried out at 70 mK in the presence of the different applied fields. (d) Field dependence of both the relaxation rates.

small local magnetic fields, we have performed zero field (zF) μ SR measurements down to 44 mK. Representative zF μ SR spectra, taken at several different temperatures, are shown in figure 4(a). The absence of oscillations, along with no loss in the initial asymmetry over the whole temperature range, confirms that there is no long-range magnetic order or freezing of the spins [24]. Moreover, the asymmetry relaxes to the same zero baseline for all the temperatures and does not recover to 1/3 of the initial asymmetry, ruling out a random distribution of static fields [25]. These observations confirm the presence of a dynamic magnetic ground state.

Initially, we used a stretched exponential function to fit the zF spectra, which did not yield a good agreement with our data. As discussed above, Yb ions have two different chemical environments, resulting in two distinct CEFs due to different surrounding charge distributions.

Since μ SR is a local probe, muons stopping at Yb1 and Yb2 will experience different CEFs, as observed earlier [22, 35]. Thus, we used a combination of two exponential functions with a flat background term to analyze this data:

$$A(t) = A_{bg} + A_1 \exp(-\lambda_1/t) + A_2 \exp(-\lambda_2/t) \quad (2)$$

where A_{bg} denotes the constant background, and A_1 and A_2 denote the initial asymmetry of each component. The λ_1 and λ_2 represent muon relaxation rates for the two different sites. Note that A_{bg} , A_1 , and A_2 are kept constant at 0.15, 0.07, and 0.06, respectively, during the data analysis. The values of A_1 and A_2 signify that the muon population implanted at both sites is almost equal [21]. The temperature evolution of both the relaxation rates is shown in Fig. 4(b), which precisely tracks the emergence of the correlations among Yb³⁺ spins. Although λ_1 is

one order higher than λ_2 , both show similar temperature dependence with a plateau-like behavior at lower temperatures. Similar behavior can be seen in the case of muon diffusion, where both relaxation rates should drop at the same temperature. However, λ_2 drops at a lower temperature compared to λ_1 . Also, $\text{Yb}_2\text{Te}_5\text{O}_{13}$ is an insulating oxide, and muon diffusion is not commonly observed in such systems [26]. In fact, the decrease in the λ_1 and λ_2 results from an Orbach process [27], which is a two-phonon scattering process mediated by an excited CEF level. The temperature variation of the relaxation rate can be described using the Orbach equation:

$$\lambda^{-1} = \lambda_0^{-1} + B_{me} \exp(-\Delta_{CEF}/T) \quad (3)$$

where λ stands for the relaxation rates λ_1 and λ_2 , λ_0 is the saturation value of relaxation rates at lower temperatures, B_{me} accounts for the magnetoelastic coupling of the Yb^{3+} spins with the phonon bath, and Δ_{CEF} gives an estimation of the energy difference of the intermediate CEF levels involved in the process. Fitting of the λ_2 variation results in the following parameters: $\lambda_{0,2} = 2.94 \mu\text{s}$, $B_{me,2} = 19.75 \mu\text{s}$ and $\Delta_{CEF,2} = 165.21 \text{ K}$. The value of $\Delta_{CEF,2}$ obtained from the fit agrees very well with the value estimated from the analysis of the inverse susceptibility data analyzed using the modified Curie-Weiss law. This agreement further reaffirms that the ground state is well separated from the higher levels and can be truly described by $J_{eff} = 1/2$. Further, fitting the temperature dependence of λ_1 gives the following parameters: $\lambda_{0,1} = 0.27 \mu\text{s}$, $B_{me,1} = 59.22 \mu\text{s}$ and $\Delta_{CEF,1} = 507.59 \text{ K}$. The higher value of $\Delta_{CEF,1}$ signifies that higher excited states also participate in the relaxation process at site 1. The temperature-independent behavior of λ_1 and λ_2 indicate the presence of a dynamic state at lower temperatures [28].

To further explore the spin dynamics, longitudinal field (LF) μSR measurements have been carried out at 70 mK by changing the applied fields from 0 to 3200 Oe. Representative LF μSR spectra, along with corresponding fits to the data using 2, are shown in Figure 4(c). In an LF- μSR measurement, the external magnetic field is applied along the initial muon spin direction. If the relaxation arises primarily from a static internal field distribution, the application of a field which is a few times larger than the field corresponding to λ_1 (as λ_2 is smaller) will decouple the signal, and full asymmetry will be recovered. However, if the relaxation originates from dynamic fields, the relaxation will not be decoupled even in higher fields. The local field corresponding to λ_1 at low temperatures can be calculated as $B_{loc} = \lambda_1/\gamma_\mu \approx 44 \text{ Oe}$, where γ_μ is the gyromagnetic ratio for muons. The LF measurements clearly show the persistence of relaxation at least up to 3200 Oe (around 70 times higher than the B_{loc}), confirming the dynamic nature of Yb^{3+} spins [21].

The field-dependence of the relaxation rates obtained from the LF- μSR data fitting are presented in Figure 4(d). Intriguingly, both relaxation rates increase with an increase in the field, which is unusual as the magnetic

field generally suppresses the dynamic relaxation rate [29, 30]. The increase in relaxation rates can be attributed to the enhancement of the density of spin excitations due to the application of the field. The increase in density of spin excitations competes with the quenching effect of the applied field. A similar behavior has previously been observed for Tb- and Yb-based compounds [22, 31].

Thus, milli-Kelvin magnetization, thermodynamics, and μSR measurements complement each other and affirm the presence of a dynamic state in $\text{Yb}_2\text{Te}_5\text{O}_{13}$ at lower temperatures. To understand the origin of this state, we have performed a combined first principles and Monte Carlo simulation study, as discussed in more detail in the supplemental material [16]. The calculated nearest, next-nearest, and third next-nearest neighbor exchange interactions, J_1 , J_2 , and J_3 , as well as the zero-field splitting parameter, D and local anisotropic parameter, E , are given in Table I.

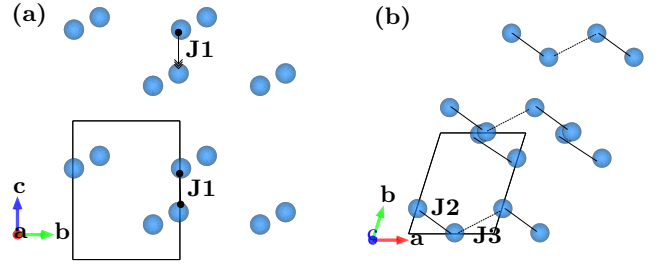


FIG. 5. (a) Spin exchange paths; J_1 shown in (a) and J_2 , J_3 are shown in (b).

TABLE I. The nearest, next nearest, third nearest neighbor exchange couplings and the zero-field splitting parameters calculated from first principles calculations.

J_1 (meV)	J_2 (meV)	J_3 (meV)	D (meV)	E (meV)
10.8	3.9	-7.3	-0.43	-0.009

With the above coupling parameters, we constructed an effective spin Hamiltonian given by,

$$H = J_1 \sum_{\langle ij \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle ij \rangle\rangle} S_i \cdot S_j + J_3 \sum_{\langle\langle\langle ij \rangle\rangle\rangle} S_i \cdot S_j + D \sum_i S_{iz}^2 + E \sum_i (S_{ix}^2 - S_{iy}^2) \quad (4)$$

A Monte Carlo simulation was performed using the above equation. The nearest neighbor interactions, antiferromagnetic in nature, are along the c direction, while J_2 , antiferromagnetic in nature, and J_3 , ferromagnetic in nature, are in the ab plane, and our computational studies confirm that these competing magnetic interactions are the primary inducers of the dynamic state in $\text{Yb}_2\text{Te}_5\text{O}_{13}$.

The paramagnetic phase observed at room temperature remains unchanged down to 125 K, where we see a

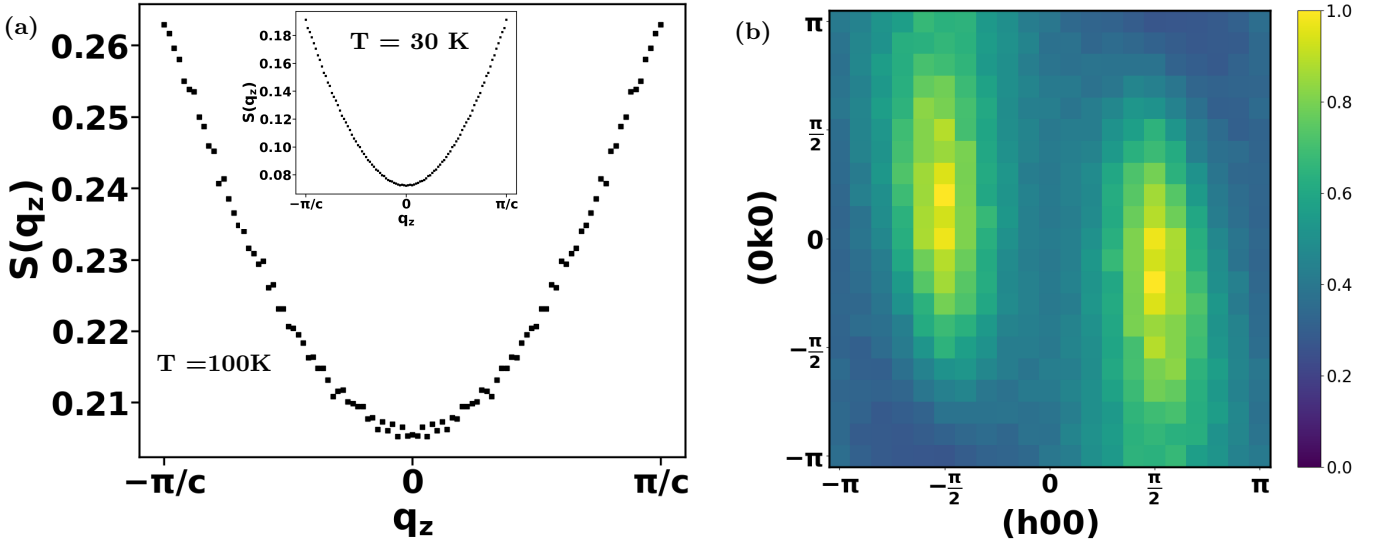


FIG. 6. (a) The Structure Factor along the c -axis in at 100 K and 30 K(inset) (b) the static structure factor at 50 K.

deviation from the Curie-Weiss law. For temperatures above 125 K, the magnetic behavior is predominantly determined by the leading exchange coupling, J_1 (10.8 meV). Since this interaction is along the c -axis, we have plotted the spin structure factor $S(q_z)$ vs q_z , as shown in Figure 6(a). We find that the magnetic structure factor peaks at $|q_z| = \frac{\pi}{c}$, confirming antiferromagnetic interactions along the c -axis. As the temperature is lowered, this nature along the c direction prevails, as seen in the inset of Figure 6(a). However, below 125 K, J_2 and J_3 play a bigger role, thus making the system much more interesting. The spin structure factor along the a and b axes are plotted in Supplementary (Fig. S5), shows a peak in the structure factor at $\frac{\pi}{a}$, which corresponds to antiferromagnetic interactions along $\vec{a}$, but no prominent peak in Sq_y . We calculated the spin-spin correlation function at various temperatures to visualize the nature of spins in the ab plane. We observe that the correlation function decays exponentially along a, b , and c axes with correlations existing beyond nearest neighbors along a and c , but dying down relatively fast along b . The static spin structure factor is calculated as a Fourier Transform of the spin-spin correlation ($\langle \vec{S}_i \cdot \vec{S}_j \rangle$) as follows:

$$S_q = \frac{1}{N} \sum_{ij}^N \langle \vec{S}_i \cdot \vec{S}_j \rangle e^{i\vec{q} \cdot |\vec{r}_i - \vec{r}_j|} \quad (5)$$

where N is the total number of spins in the three-dimensional simulation box. The static structure factor for $T = 50\text{ K}$ is plotted in Fig. 6 (b). We find that the spin structure factor in the ab plane shows diffused peaks in the first Brillouin zone as the temperature is lowered (see the bright spots in Figure 6(b)). The peaks reveal that there is competition of exchange interactions in the ab owing to the spiral nature of the interaction along a

and b . This phase is best described as a spiral non-trivial phase [36, 37], which exists down to a temperature close to where the susceptibility starts rising.

In conclusion, we have performed a comprehensive study of a quantum-dimer-based compound $\text{Yb}_2\text{Te}_5\text{O}_{13}$ using magnetization, heat capacity, and μSR measurements. $z\text{F-}\mu\text{SR}$ measurements have confirmed the absence of long-range ordering down to 44 mK and the presence of a transition among excited CEF levels (Orbach process). Moreover, LF measurements confirm the fluctuating nature of Yb^{3+} spins in the system even up to 3200 Oe and reveal a field-induced enhancement of the density of spin excitations. Theoretical calculations affirm the experimental findings and reveal that competing exchange interactions are responsible for such a dynamic state. This discovery contributes to our comprehension of unusual magnetic phases and paves the way for investigating quantum spin liquids in new materials that have not been explored in this context before.

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