Signatures of a spin-1/2 cooperative paramagnet in the diluted triangular lattice of \( Y_2CuTiO_6 \)

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We present a combination of thermodynamic and dynamic experimental signatures of a disordered dynamic cooperative paramagnet in a 50% site diluted triangular lattice spin-\( \frac{1}{2} \) system, \( Y_2CuTiO_6 \). Magnetic ordering and spin freezing are absent down to 50 mK, far below the Curie Weiss scale of \( \sim -134 \) K. We observe scaling collapses of the magnetic field- and temperature-dependent magnetic heat capacity and magnetisation data, respectively, in conformity with expectations from the random singlet physics. Our experiments establish the suppression of any freezing scale, if at all present, by more than three orders of magnitude, opening a plethora of interesting possibilities such as disorder-stabilized long range quantum entangled ground states.

Conventional wisdom suggests that structural disorder in magnetic insulators usually leads to random spin-exchange interactions, which, in turn, promotes spin freezing at low temperatures [1–3]. There is, however, an interesting alternative where quenched randomness may promote competing magnetic interactions and quantum fluctuations, thereby enhancing the possibility of realizing quantum spin liquids (QSLs) [4], as recently suggested for \( Ba_3CuSb_2O_9 \) [5] and \( Pr_2Zr_2O_7 \) [6].

This raises several interesting and experimentally relevant questions — can structural disorder in magnetic insulators enhance quantum fluctuations and drive a magnetically ordered state (in clean limit) to a quantum paramagnet? What then is the nature of such a paramagnet? Can such a state support non-trivial many-body entanglement and realise a disorder driven QSL? [7, 8] This possibility of realizing QSLs [9–11] and associated novel superconductors [12] arising from the interplay of disorder and interactions near the metal-insulator transition were explored theoretically in the context of doped semiconductors and boron doped diamond. On the experimental front, low temperature dynamic paramagnetism was observed in irradiation induced disordered organic magnet \( \kappa-(ET)_2Cu[N(CN)_2]Cl \) [8]. However, the question of a disorder driven QSL is different from that of the effect of disorder in a clean QSL. In this latter case, the QSL is often stable to at least dilute local impurities albeit with interesting defect states [13–18].

These issues are particularly pertinent for two dimensional spin-\( \frac{1}{2} \) frustrated magnets where reduced dimensionality enhances quantum fluctuations and suppresses ordering tendencies. Thus, they serve as natural platforms to realize QSLs as in candidate materials – Herbertsmithite (\( Cu_3(OH)_6 Cl_2 \)) [19–24] and \( \kappa-(ET)_2Cu_2(NCn)_3 \) [25]. Notably, in Herbertsmithite, QSL is suggested to be stable to off-plane \( Cu^{2+} \) magnetic disorder [26, 27]. Similarly, in three dimensional hyperkagome QSL candidate \( Na_4Ir_3O_8 \) [28, 29], the coexistence of slow time-scales and signatures of quantum fluctuations have been observed [30].

In this paper, we report an experimental realization of a dynamic cooperative paramagnet in a spin-\( \frac{1}{2} \) magnet on a site diluted triangular lattice \( Y_2CuTiO_6 \) (YCTO) [31, 32]. This is established by the absence of any ordered or frozen magnetism down to the lowest accessible temperature (50 mK) despite substantial magnetic interactions, indicated by a large Curie-Weiss temperature (\( \theta_{CW} \sim -134 \) K) and disorder in the system. YCTO, therefore, is an example of a disordered triangular lattice magnet with 50:50 random mixture of spin-\( \frac{1}{2} \) \( Cu^{2+} \) atoms and non-magnetic \( Ti^{4+} \) atoms, as shown in the left frame of Fig. 1. We observe a specific scaling behavior of thermodynamic quantities [1, 33–36] indicating a low temperature dynamic spin-\( \frac{1}{2} \) paramagnet with possible formation of random singlets that survive down to the lowest accessible temperature [37].

In YCTO, each \( Cu/Ti \) site is surrounded by a triangular pyramid of five oxygen atoms with three in the basal (a, b) plane and two along the c-axis. The resultant \( Cu/Ti \) units arranged in a triangular lattice in the (a,b) plane are well-separated by an intervening layer of non-magnetic \( Y^{3+} \) ions along the c-axis with a large
interlayer separation of \( \sim 5.7 \) Å. The exchange interaction strengths between nearest neighbor Cu atoms in the (a,b) plane \((J_{nn})\) and the interlayer magnetic coupling \((J_c)\) were calculated using density functional theory (see Supplemental Material (SM) [38] for calculation details). \( J_{nn}/k_B \) and \( J_c/k_B \) were determined to be \( \sim -33.6 \) K and \( \sim -1.0 \) K, respectively. These estimates lead to a calculated \( \theta_{CW} \) of \( \sim -104 \) K in reasonable agreement with the experimental value of \(-134 \) K. Larger than an order of magnitude anisotropy in magnetic interactions indicates two-dimensional triangular lattices of corner-shared \((\text{Cu/Ti})\text{O}_5\) units, as shown in the right frame of Fig. 1, are coupled only weakly along the c-axis. Thus, we may think of the system approximately as one spin-\( \frac{1}{2} \) at each Cu\(^{2+}\) site sitting on an isotropic triangular lattice which is 50% diluted with non-magnetic Ti\(^{4+}\) ions. Experimentally no superlattice formation is observed in spite of the charge difference of the Cu\(^{2+}\) and Ti\(^{4+}\) ions with the structural uniformity achieved only at a statistical level. We also note that superlattice formations on a triangular motif is geometrically frustrated at 50% dilution [39]. Thus, YCTO, to a very good approximation, is a randomly 50% diluted spin-rotation invariant spin-\( \frac{1}{2} \) magnet on a triangular lattice. Note that the random in-plane dilution and the spin-rotation symmetry are the two key differences between YCTO and the recently much investigated YbMgGaO\(_4\) [7, 40].

Polycrystalline samples of YCTO were synthesized by standard solid state reaction techniques. Details of the sample preparation and measurements (magnetization, heat capacity, \(^{89}\)Y nuclear magnetic resonance (NMR), and muon spin relaxation (\(\mu\)SR)) are presented in Supplemental Material [38]. From Rietveld refinements of x-ray diffraction data (see SM [38] for details), we confirmed that YCTO crystallizes in the non-centrosymmetric hexagonal structure with the space group \(P6_3\)cm [31, 32], isostructural to LuMnO\(_3\) [41].

The dc susceptibility, \(\chi(T) = M(T)/H\), as a function of \(T\), is shown in Fig. 2, where no signature of any magnetic ordering is seen down to 2 K. The divergence of \(\chi\) at lowest temperatures is well-known to generally arise from the presence of a minor fraction \((\sim 2\% \) in the present case) of free spins in many such systems, as detailed in SM [38]. A fitting of the high temperature (200-400 K) dc susceptibility to a Curie-Weiss form \(\chi = \chi_0 + C/(T - \theta_{CW})\); where \(\chi_0\), \(C\), and \(\theta_{CW}\) are temperature independent Van Vleck paramagnetic and core diamagnetic susceptibilities, Curie constant, and the Curie-Weiss temperature, respectively, yields \(\chi_0 = 3.48 \times 10^{-4}\) emu mol\(^{-1}\) Oe\(^{-1}\), \(C = 0.47\) emu K \(\text{mol}^{-1}\) Oe\(^{-1}\) and the \(\theta_{CW} = -134\) K. The value of \(\theta_{CW}\) is in agreement with earlier reports [31, 42] and the inferred moment of \(\mu_{\text{eff}} = 1.94\mu_B\) is consistent with the expected value of \(\sim 1.9\mu_B\) for a spin \(S = \frac{1}{2}\) Cu\(^{2+}\) system with \(g = 2.2\), as reported for many cuprates. Large and negative \(\theta_{CW}\) suggests substantial antiferromagnetic spin-spin interactions within each triangular layer.

The real and the imaginary parts of the ac susceptibility (see inset-I of Fig. 2) did not show any indication of magnetic ordering. Further, the lack of frequency dependence of the ac susceptibility over a range of frequencies indicates the absence of spin-freezing down to 2 K. The lack of freezing despite extensive disorder is re-emphasized by the absence of any irreversibility between the FC and ZFC magnetisation data at a low-field (50 Oe) (see inset-II of Fig. 2), also ruling out any magnetic ordering down to 500 mK. Thus, this system with substantial antiferromagnetic interactions on an essentially two-dimensional triangular lattice with spin-\( \frac{1}{2}\)S and a relatively small spin-orbit coupling strength with no evidence of any magnetic ordering/freezing down to 500 mK offers a unique opportunity to probe a dynamic low temperature correlated paramagnetic phase that can possibly harbor intricate interplay of quantum fluctuations and disorder.
We extend the limit of the low temperature probe down to 50 mK with \( \mu \)SR experiments. Fig. 3a shows smooth exponential depolarisation of muon spins without any oscillations at all temperatures in absence of an external magnetic field. This shows that the fluctuation frequency of the local fields is much greater than the muSR frequency, ruling out any static magnetic ordering down to 50 mK, though it cannot rule out a dynamic magnetic order \([43]\). We fit the muon spin polarization data to \( e^{-\lambda t} \), as detailed in SM \([38]\), and obtain the muon spin relaxation rate, \( \lambda \), plotted as a function of the temperature in the inset of Fig. 3a. We note that for a random internal static magnetic field with Gaussian distribution and zero mean, one expects a muon signal to decay as \( e^{-\mu^2} \) with \( \mu \) being a constant which is different from the \( e^{-\lambda t} \) dependence seen in our experiments \([44]\).

The initial increase in \( \lambda \) (inset of Fig. 3a) on cooling indicates the expected slowing down of spin dynamics of Cu moments with lowering of temperature down to about 2 K below which it essentially levels off at a value of \( \lambda \approx 0.1 \, (\mu s)^{-1} \) down to \( \sim 50 \) mK. This indicates a dynamic low temperature state \([45, 46]\). If the observed muon depolarization arises from any static magnetic fields, \( \lambda \approx 0.1 \, (\mu s)^{-1} \) would suggest an estimate of the field (\( \sim 2\pi \lambda / \gamma \mu \), with \( \gamma \mu \) being the muon gyromagnetic ratio) to be about 7.4 Oe. Then the muon spins can be decoupled from the static moments with the application of a longitudinal field about ten times this internal field \([44]\). We have measured the muon polarization as a function of time at various temperatures and and applied longitudinal fields up to 2048 Oe—about 270 times larger than 7.4 Oe, yet we did not see the total suppression of muon depolarization indicating existence of strong fluctuating local magnetic field in the system. A set of representative data obtained at 500 mK is shown in Fig. 3b. The corresponding decay constants, \( \lambda \), are shown in the inset to Fig. 3b as a function of the applied field for different temperatures. At all the measured temperatures \( \lambda \) is nearly constant apart from a peak around 10 Oe which is presumably due to a quadrupolar level crossing resonance \([47]\) coming from the neighboring copper nuclei. This temperature independence suggests rapid spin fluctuations and the absence of any spin ordering or freezing down to at least 50 mK, despite sizable magnetic interactions.

This observation of the absence of spin-freezing or magnetic ordering down to 50 mK then raises the interesting possibility of realizing disorder driven quantum paramagnetism in YCTO. The lack of spin ordering/freezing till a very low temperature is also emphasized in the total heat capacity, \( C_p(T) \), (see SM \([38]\)) measured down to 350 mK for various magnetic fields. The total contribution, \( C_n \), of all spins to the specific heat is estimated by subtracting the lattice contribution, \( C_{lat} \), from \( C_p \). We find \( C_n/T \approx 0 \) by 20 K (inset to Fig. 4a). Spin entropy by integrating \( C_n/T \) vs. \( T \) up to 20 K is shown in Fig. 4a for different applied magnetic fields. Clearly, this procedure accounts for only about 15% of the total spin-entropy per mole of spin-\( \frac{1}{2} \) Cu ions, evidencing a huge unquenched spin entropic content down to lowest temperatures (350 mK) complementing the muon relaxation data. We can further remove the contribution of free-spins from \( C_n \) by subtracting the Schottky terms as detailed in SM \([38]\), thereby defining \( C_m \). Inset to Fig. 4b shows the \( T \)-dependence of \( C_m \) vs. \( T \) at different fields on a log-log scale for better clarity in the low temperature region. The data clearly show that above \( \sim 2 \) K, \( C_m \) is independent of the applied magnetic field. Below 2 K, \( C_m \) follows a power law (\( C_m \approx \beta T^{1.4} \)) with temperature indicating the presence of a large number of low energy excitations. The regime of power-law behavior of \( C_m \) shrinks with a decreasing magnetic field to a fraction of a Kelvin for a field of 1 T suggesting that the low energy excitations directly couple to the magnetic field which is characteristic of random singlets with a distribution of bond energies \([7, 27]\).

With the above results, we now turn to investigate the nature of the low temperature dynamic, correlated...
paramagnet. The primary in-plane super-exchange between two nearest neighbour Cu$^{2+}$ atoms is mediated by the intermediate in-plane oxygen as is evident in Fig. 1. Further neighbour exchanges either involve more intermediate O$^{2−}$ and in-plane Cu$^{2+}$/Ti$^{4+}$ or out-of-plane Y$^{3+}$—hence expected to be suppressed similar to the inter-plane magnetic exchanges. This indicates that the magnetic physics of YCTO can be understood within a minimal model of diluted short-range antiferromagnet on a triangular lattice with a Hamiltonian: $H = \sum J_{rr′} \eta_r \eta_{r′} S_r \cdot S_{r′}$ where $S_r$ are spin-$\frac{1}{2}$ operators on the triangular lattice sites, $r$, and $\eta_r = 0(1)$ for a Ti$^{4+}$ (Cu$^{2+}$) site.\textsuperscript{[48, 49]} The distribution of Ti$^{4+}$ and Cu$^{2+}$ in 1:1 ratio is given by a probability distribution function, $P[\{\eta_r\}]$. $J_{rr′}$ denotes short range antiferromagnetic interactions between the Cu$^{2+}$ ions and the randomness of the position of Cu$^{2+}$ lead to a distribution in $J_{rr′}$ given by $P[\{J_{rr′}\}]$ which is correlated with (but not necessarily same as) $P[\{\eta_r\}]$. The absence of signatures of formation of a super-lattice or any other structural anomalies in diffraction experiments suggests $P[\{\eta_r\}]$ to have weak correlations among different sites. Therefore, we expect that $P[\{J_{rr′}\}]$ has a relatively small width.

Due to the site-dilution, YCTO is similar to doped semiconductors \textsuperscript{[1, 9, 10]} than the recently discovered YbMgGaO$_4$ \textsuperscript{[40]}. However, in contrast to the low density of magnetic moments in doped semiconductors, YCTO has a dense (50%) concentration of spins. In the absence of any magnetic order, as established experimentally, the natural option for the system in such circumstances is to locally minimize energy of the antiferromagnetic exchanges by forming singlets. In the process of formation of these singlets, some spins, fewer in number, are left over due to lack of partners. However these spins, sitting on a random network, are not isolated because of the high density of the magnetic ions with which they interact. Indeed, if the background network of the dimers is dynamic, the positions of such unpaired spins are not even static \textsuperscript{[26]}. These unpaired dynamic quasispins \textsuperscript{[51]} then interact with each other with effective exchange interactions of the form $H_{eff} = \sum_{i,j} J_{ij} S_i \cdot S_j$ where the effective couplings are expected to be $|J_{ij}| \sim e^{-|r_i−r_j|/\xi}$ where $\xi$ is the underlying spin correlation length \textsuperscript{[7]}. Thus, $J_{ij}$ are weak and random and the fate of the system crucially depends on their distribution as well as the sign structure. Owing to the lack of bipartite structure of the underlying triangular motif, these exchanges are expected to be a mixture of ferromagnetic and antiferromagnetic interactions eventually leading to a spin-glass state at a much lower temperature \textsuperscript{[1, 7, 52]} depending on the magnitude and distribution of $J_{ij}$. A power law distribution $\sim J^{−\gamma}$ leads to a magnetic specific heat scaling, at finite magnetic field of $C_m \sim T H^{−\gamma}$ for $T/H < 1$.

The heat capacity measurements reveal a power-law behavior with the universal scaling of the $H^{\gamma} C_m/T$ with $T/H$ and $\gamma \simeq 0.7$ (Fig. 4b). Indeed, recently such a a scaling has been argued to result from an intermediate temperature random singlet phase in a bond disordered system \textsuperscript{[27,53]}. The above conclusions are consistent with the finite dynamic rate observed in muon spin-relaxation experiments where the rate is fairly insensitive to the applied small external magnetic fields.

Further evidence of the correlated nature of the low temperature paramagnet comes from the dependence of the magnetization with magnetic field at low tempera-
tures (inset of Fig. 4c) which cannot be described by a free spin Brillouin function. Indeed, the magnetization after the removal of the contribution of the free spin magnetization, as estimated in the analysis of the specific heat, shows a scaling collapse of the form $MT^{-\alpha}$ (with $\alpha = 0.34 \pm 0.02$) as a function of $H/T$ (see Fig. 4c) as expected for the above-mentioned power-law distribution of the effective exchanges with $\alpha = 1 - \gamma$ [26, 27].

In summary, through a complementary set of experiments on the randomly 50% depleted triangular lattice $S = 1/2$ magnet $\text{Y}_2\text{CuTiO}_6$, we establish absence of magnetic order and/or spin freezing down to 50 mK. This is 0.037% of the Curie-Weiss scale of about $-134$ K, the latter implying strong antiferromagnetic couplings between the magnetic Cu$^{2+}$ spins. While, at even lower temperatures spin-freezing may occur, such drastic suppression of freezing compared to the Curie-Weiss scale opens up a cooperative paramagnetic regime at least between 50 mK and 2 K. In the cooperative paramagnet, a scaling collapse, consistent with the random singlet phenomenology [27], is observed in the magnetic field dependent specific heat and magnetisation. An exciting question, fuelled by our experimental observation of dynamical signatures, pertains to the role of quantum coherence in the cooperative paramagnet and in particular whether it can support non-trivial entanglement expected in a QSL. While our existing understanding in clean frustrated magnets indicates that cooperative paramagnets provide the right background to look for QSLs, search for such quantum coherence in $\text{Y}_2\text{CuTiO}_6$ is clearly a very interesting future step in exploring disorder driven QSLs. The issue of doping away from the 50% dilution, or with carrier doping forms similar sets of interesting open questions.

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[38] See Supplemental material, which includes Refs. [56–72], for more details.
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Supplemental Material

Signatures of a spin-$\frac{1}{2}$ cooperative paramagnet in the diluted triangular lattice of $Y_2CuTiO_6$

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A. Details of sample preparation and crystal structure of YCTO

At first, the intimate stoichiometric mixture of \( \text{Y}_2\text{O}_3 \) (which was preheated at \( 1150^\circ\text{C} \) for 6 hours to remove carbonate contamination and moisture), CuO (99.9%) and TiO\(_2\) (99.9%) was prepared. Then this mixture was pressed into a pellet and placed in an alumina crucible and heated in air at 900\(^\circ\text{C}\) for 24 hours in a tubular furnace. Further, the sample was again ground, pelletized and reheated in air at 1050\(^\circ\text{C}\) for 150 hours with three intermediate stages of regrinding for better homogeneity. In the next step, the sample was fired again in air at 1300\(^\circ\text{C}\) for 30 hours to form the final product.

The powder x-ray diffraction data (PXRD) of polycrystalline YCTO sample was collected at room temperature with Cu-K\(_\alpha\) as the radiation source (\( \lambda = 1.54056 \) Å) using Panalytical X-ray diffractometer (Fig. 1). The PXRD pattern confirms the formation of single phase YCTO. After refinement using FullProf software, we found that the prepared YCTO crystallizes in the hexagonal non-centrosymmetric space group \( P6_3cm \), as reported earlier [1], with lattice parameters \( a = b = 6.1951(1) \) Å, \( c = 11.4713(2) \) Å, \( \alpha = \beta = 90^\circ \), \( \gamma = 120^\circ \) and is isostructural with LuMnO\(_3\) [1, 2]. The atomic coordinates of different elements after refinement are shown in Table I. The goodness of the Rietveld refinement as defined by the following parameters are \( R_p: 12.7\%; \) \( R_{wp}: 8.79\%; \) \( R_{exp}: 4.19\%; \) \( \chi^2: 4.40 \).

| Atom | Wyckoff position | x     | y     | z     | Occupancy |
|------|------------------|-------|-------|-------|-----------|
| Cu   | 6c               | 0.3270| 0.0000| -0.0196| 0.50      |
| Ti   | 6c               | 0.3270| 0.0000| -0.0196| 0.50      |
| Y1   | 4b               | 0.3333| 0.6667| 0.2196| 1.00      |
| Y2   | 2a               | 0.0000| 0.0000| 0.2455| 1.00      |
| O1   | 6c               | 0.3156| 0.0000| 0.1489| 1.00      |
| O2   | 6c               | 0.6541| 0.0000| 0.3203| 1.00      |
| O3   | 2a               | 0.0000| 0.0000| -0.0229| 1.00     |
| O4   | 4b               | 0.3333| 0.6667| 0.4922| 1.00      |
Figure 1. XRD refinement of YCTO (color online). Black circle is observed data, red line is theoretically calculated, blue line is the difference between the observed and calculated data and the vertical marks are the Bragg’s positions. The plane of reflection (hkl) is marked for each corresponding Bragg peak.

B. Experimental details

Magnetization, $M$, measurements as a function of applied field $H$ (0 to 5 T) and temperature $T$ (in the range 1.8 K to 400 K)) were performed using a Quantum Design SQUID VSM. Zero-field cooled (ZFC) and field cooled (FC) magnetization measurements in a low field of 50 Oe were performed down to 500 mK. The heat capacity $C_p(T)$ was measured with a Quantum Design PPMS in various applied fields down to 0.35 K. Local probe NMR measurements were performed on the $^{89}$Y nucleus in a fixed field of 93.95 kOe. Muon spin relaxation ($\mu$SR) measurements down to 50 mK were performed on the MuSR instrument at ISIS, RAL in UK (for data, see Ref. [3]) and PSI in Switzerland.

C. Calculation of the magnetic exchange interaction strengths

Ab-initio electronic structure calculations were carried out within a plane-wave projected augmented wave [4] implementation of density functional theory in the Vienna Ab-initio
Table II. Energetics for different spin configurations.

| State  | ∆E (meV) |
|--------|-----------|
| AAFM   | -0.70     |
| AFM    | -17.74    |

Simulation package (VASP) [5, 6]. The GGA approximation [7] to the exchange correlation functional was used, in addition to treating electron-electron interactions at the Cu site with a $U$ of 8 eV [8–10] within the Dudarev [11] implementation. A cutoff energy of 600 eV was used to determine the number of plane waves included in the basis. All $k$-space integrations were carried out using a $\Gamma$ centered $5\times5\times3$ Monkhorst-Pack $k$-point mesh. A unit cell of YCTO consisting of 3 formula units as shown in Fig. 1 (main manuscript) with a total of 30 atoms was used for the calculations. The calculations were carried out for the ferromagnetic (FM) configuration, an inter-planar antiferromagnetic configuration (AAFM) where the spins in the plane are ferromagnetic while the coupling between spins in different planes is antiferromagnetic, as well as AFM where the spins are antiferromagnetically coupled in a plane. Changing the cutoff energy to 750 meV, changed the energy difference between $E_{FM}$ and $E_{AFM}$ by 0.11 meV ($\sim$0.6% change), while increasing the k-points to $9\times9\times5$ changed it by 0.9 meV ($\sim$5% change), indicating that the results were well converged. The magnetic part of the total energy, determined for each configuration, was mapped onto a Heisenberg model [12] given by

$$H_{mag} = -\sum_{i,j} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j$$

where $i, j$ are the site indices, $J_{ij}$ denotes the exchange coupling strength between the spins and $\mathbf{e}_{ij}$ represents the unit vectors along the corresponding spin directions. The energy difference ($\Delta E$) in table II is defined as $E_{AAFM} - E_{FM}$ and $E_{AFM} - E_{FM}$ for AAFM and AFM, respectively. Using the energy differences as shown in the table II we have estimated the nearest neighbour exchange coupling, $J_{nn}$ and inter-planar exchange coupling, $J_c$ to be $\sim -2.90$ meV ($\equiv -33.6$ K) and $\sim -0.09$ meV ($\equiv -1.0$ K), respectively. A comparison with iso-structural YMnO$_3$ reveals that the ratio of $J_{nn}$ to $J_c$ ($= 33.0$) in the present case represents an even more two-dimensional nature than that ($= 14.2$) [12] for YMnO$_3$.

The exchange interaction strengths were used to determine the Curie-Weiss temperature
which was found to be \( \approx -104 \) K using the method outlined in [13, 14], in reasonable agreement with the experimentally obtained value of \( \approx -134 \) K.

D. Details of analyses

\(^{89}\text{Y} \text{NMR spectra:}\)

A local probe of the nature of the Cu moments down to 4 K, specifically pertaining to their dynamical/static nature is obtained from the nuclear magnetic resonance of the nearby Y sites, making use of the natural abundance of the \(^{89}\text{Y}\) nuclei with a spin \( I = 1/2 \) and gyromagnetic ratio \( \gamma / 4\pi = 2.08583 \text{ MHz/Tesla} \). The variation of the lineshape with \( T \) is shown in Fig. 2a. While the spectra are asymmetric due to the presence of different local magnetic environments in this disordered system, the line-width of the spectrum increases monotonically with decreasing temperature, essentially following the Curie-Weiss susceptibility of the Cu moments and showing no sign of any divergence down to 4 K. In order to quantify the line-width and the peak position as a function of the temperature, we minimally fit the \(^{89}\text{Y}\) spectra at various temperatures to a sum of two Gaussians. We find that one of the two peaks remains nearly at the reference position, while the other shifts gradually to lower frequencies as temperature decreases, as also evident in the spectra shown in Fig. 2a. Specifically, the absence of any divergence/peak in the observed \(^{89}\text{Y}\) NMR shift and width indicates that the Cu moments remain dynamic down to 4 K.

The \(^{89}\text{Y}\) NMR spin-lattice relaxation rate, \( 1/T_1 \), was obtained by the standard saturation recovery pulse sequence (with a \( \pi/2 \) pulse of duration \( 6 \mu s \)) in a field of 93.95 kOe. The recovery of the longitudinal magnetization was fitted to a stretched exponential \( (A \exp\left(-\frac{4\pi}{T_1}\right)^{\beta}) \). The stretching exponent \( \beta \) becomes essentially 1 above 100 K, thereby leading to the expected exponential form for \( I = 1/2 \) nuclei. Significantly, a stretched exponential recovery of the nuclear magnetization has been predicted [15] for the random singlet phase due to the distribution of magnetic environments that the nuclei sense. \( 1/T_1 \), shown in Fig. 2b, is found to decrease continuously with the temperature until about 30 K and below 30 K, it is observed to be nearly constant. Once again, the absence of any peak, indicating a divergence, in the \( 1/T_1 \) vs \( T \) data suggests the absence of a phase transition to an ordered state with the spins remaining dynamic down to 4 K [16].
Figure 2. (a) $^{89}$Y NMR spectra at different temperatures (the vertical dashed line is the reference frequency for $^{89}$Y NMR). A representative two-Gaussian fit is shown for the spectrum at 4 K, (b) Temperature dependence of the $^{89}$Y spin-lattice relaxation rate $1/T_1$ is shown. Inset shows the variation of the stretching exponent with temperature.

$\mu$SR:

The time-evolution of the muon decay asymmetry has been (see Fig. 3 of main manuscript) modeled by the equation $A(t) = A_r e^{-\lambda(T)t} + A_{bg}$, where $A_{bg}$ takes into account the muons that stop outside the sample and do not get depolarized, and the 1st term, which reflects the contribution from the muons that are stopped inside the sample, contains the relaxation rate ($\lambda$) and the relaxing asymmetry ($A_r$). We observed difference in background contributions between helium cryostat ($A_{bg} \sim 0.085$) and dilution refrigerator ($A_{bg} \sim 0.050$), which were kept fixed during the data analysis. It is important to mention that the given background constants do not evolve with time. During the analysis of the muon depolarization at applied longitudinal field (see Fig. 3 of main manuscript), we have not put any constraint in the magnitude of relaxing asymmetry and the background contributions, as the given two quantities vary with applied magnetic field presumably because of the deviation of amount of muons that are hitting the sample as well as the change in the direction of the positrons that are coming out of the sample and going towards the detector.
with the change in applied magnetic field.

Heat capacity:

In zero-field, we measured $C_p(T)$ in the $T$-range 0.35 - 290 K (see Fig. 3a). It is seen that below about 10 K the $C_p(T)$ data is dependent on the applied magnetic field (shown in inset-I of Fig. 3a). For clarity we plotted $C_p/T$ vs. $T$ in inset-II of Fig. 3a. The low temperature anomaly in $C_p/T$ data gradually shifts towards the higher temperatures as field increases. This is nothing but the so called Schottky anomaly. This Schottky contribution ($C_{Sch}$) to the specific heat most likely arises from the isolated paramagnetic Cu$^{2+}$ spins or orphan spins within the system. These spins give rise to a two-level energy system in the presence of an applied magnetic field. As our specific heat $C_p(T)$ is weakly field dependent at low temperature, we can write the total specific heat of the system as:

$$C_p(T, H) = C_{\text{lat}}(T) + C_{\text{Sch}}(T, H) + C_m(T, H)$$  \hspace{1cm} (2)

Here, $C_{\text{lat}}$ is the lattice specific heat, $C_{\text{Sch}}$ is the Schottky contribution due to isolated paramagnetic spins within the system and $C_m$ is the intrinsic magnetic contribution to the specific heat. While the total spin contribution, $C_s$, from all spins to the total specific heat, $C_p$, can be easily estimated from $C_p - C_{\text{lat}}$, our prime intention is to find the intrinsic magnetic contribution $C_m$. To obtain $C_m$, we first have to evaluate $C_{\text{lat}}$ and $C_{\text{Sch}}$ and then subtract these from the total specific heat $C_p$.

In the absence of a suitable non-magnetic analog for YCTO, we used a combination of Debye and Einstein terms as expressed by Eq. 3 to estimate the lattice contribution $C_{\text{lat}}$. Here in Eq. 3, $C_D$ and $C_{E_i}$ indicate the weightage factors corresponding to acoustic and optical modes of atomic vibrations and $\theta_D$, $\theta_{E_i}$ are the corresponding Debye and Einstein temperatures, respectively.

$$C_{\text{Debye}}(T) = C_D \left[ 9R \left( \frac{T}{\theta_D} \right)^3 \int_0^{\infty} \frac{x^4 e^x}{(e^x - 1)^2} dx \right]$$  \hspace{1cm} (3)

$$C_{\text{Einstein}}(T) = \sum C_{E_i} \left[ 3R \left( \frac{\theta_{E_i}}{T} \right)^2 \frac{e^{\theta_{E_i}/T}}{(e^{\theta_{E_i}/T}) - 1)^2} \right]$$  \hspace{1cm} (4)
Figure 3. The main figure in (a) shows the $T$-dependence of the heat capacity of YCTO in zero field and the solid line shows the Debye-Einstein fit (see text) of the zero field data. The red solid line is an extrapolation of the fit to higher and lower temperatures. The inset-I shows an expanded view of the low-$T$ data in variable magnetic field and the inset-II shows $C_p/T$ data in the low-$T$ region. (b) Schottky fitting of low temperature anomaly of YCTO. In the inset, the energy gap ($\Delta/k_B$) as function of applied field is shown and it varies linearly.

We fitted the $C_p(T)$ data at 0 T with Eq. 3 for a limited range 30 - 120 K and then extrapolated the curve to cover the entire temperature range 0.35 - 290 K (as shown in Fig. 3a). We checked several combinations of Debye plus Einstein terms and among them the best fit we obtained is for one Debye plus two Einstein terms with weightage factors in the ratio: $C_D : C_{E_1} : C_{E_2} = 1:3:6$. The sum $C_D + \sum C_{E_i}$ is equal to 10 which is same as the number of atoms per formula unit of YCTO. The fitting also yields the Debye temperature $\theta_D = (144 \pm 1)$ K and Einstein temperatures to be $\theta_{E_1} = (246 \pm 1)$ K, $\theta_{E_2} = (610 \pm 2)$ K. Thus we obtained $C_{lat}$ which will be subtracted later from the total $C_p(T)$.

Now to eliminate the Schottky contribution, we follow a protocol described here. In the absence of an applied field, $C_p(0, T)$, the $C_p$ data at $H = 0$ T, contains lattice ($C_{lat}(T)$) as well as magnetic ($C_m(H, T)$) part but lacks Schottky contribution. So, we consider $C_p(0, T)$ as a reference and subtract it from higher field data $C_p(H, T)$ to obtain $\Delta C_p$ [$\equiv C_p(H, T) - C_p(0, T)$]. Now this $\Delta C_p$ represents the Schottky contribution. In Fig. 3b, we have plotted $\Delta C_p/T$ vs. $T$ and fitted by Eq. 5 which is the generalized heat capacity
Figure 4. The as-measured heat capacity is plotted as a function of T/H in various fields. The data collapse is observed with the scaling parameter $\gamma = 0.5$.

equation for a two level system.

$$C_{Sch} = f \left[ R \left( \frac{\Delta}{k_B T} \right)^2 \left( \frac{g_0}{g_1} \right) \frac{e^{\Delta/k_B T}}{1 + \left( \frac{g_0}{g_1} \right) e^{\Delta/k_B T}} \right]$$

where $f$ is fraction of free spins within the system, $\Delta$ is the Schottky gap, $R$ is the universal gas constant, $k_B$ is the Boltzman constant and $g_0$ and $g_1$ are the degeneracies of the ground and excited states respectively. Here $g_0 = g_1 = 1$. Fig. 3b shows that the Schottky anomalies are well fitted for each field. From the fits, we found the percentage of free $S = \frac{1}{2}$ spins is $\sim 2\%$. The energy gap ($\Delta$) varies linearly with applied fields shown in the inset of Fig. 3b. From the slope of the linear fit, we obtained the spectroscopic splitting factor $g = 2.34$. The linear fit has a non-zero intercept ($\simeq 1.08$ K) at zero field which might indicate the presence of an intrinsic field within the system. The as-measured heat capacity (without any subtraction) is plotted in a scaled manner where the data collapse is evident for $\gamma = 0.5$ as shown in Fig. 4.

**Power law behavior of dc Susceptibility**:

The fraction of free spins which contribute to the heat capacity through the Schottky term will also contribute a Curie term to the magnetic susceptibility, as has been reported in many
Figure 5. The susceptibility as a function of temperature after the subtraction of 2% free spin contribution from the measured $\chi(T)$.

instances of frustrated magnets. [17–21] If we take the approximate fraction of free spins to be 2% and subtract a corresponding Curie term from the measured dc susceptibility $\chi(T)$ we obtain the data shown in Fig. 5. A fit to power law variation at low-$T$ yields $\gamma = 0.68$ which is similar to that obtained from the heat capacity scaling, as expected for an RSS.
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