Evidence for a three-dimensional quantum spin liquid in PbCuTe$_2$O$_6$

Shravani Chillal$^{1,2}$, Yasir Iqbal$^2$, Harald O. Jeschke$^3$, Jose A. Rodriguez-Rivera$^{4,5}$, Robert Bewley$^6$, Pascal Manuel$^6$, Dmitry Khalyavin$^6$, Paul Steffens$^7$, Ronny Thomale$^8$, A. T. M. Nazmul Islam$^1$, Johannes Reuther$^{1,9}$ & Bella Lake$^{1,10}$

The quantum spin liquid is a highly entangled magnetic state characterized by the absence of static magnetism in its ground state. Instead, the spins fluctuate in a highly correlated way down to the lowest temperatures. Quantum spin liquids are very rare and are confined to a few specific cases where the interactions between the magnetic ions cannot be simultaneously satisfied (known as frustration). Lattices with magnetic ions in triangular or tetrahedral arrangements, which interact via isotropic antiferromagnetic interactions, can generate such a frustration. Three-dimensional isotropic spin liquids have mostly been sought in materials where the magnetic ions form pyrochlore or hyperkagome lattices. Here we present a three-dimensional lattice called the hyper-hyperkagome that enables spin liquid behaviour and manifests in the compound PbCuTe$_2$O$_6$. Using a combination of experiment and theory, we show that this system exhibits signs of being a quantum spin liquid with no detectable static magnetism together with the presence of diffuse continua in the magnetic spectrum suggestive of fractional spinon excitations.
Fractionalisation is one of the most fascinating phenomena in modern condensed matter physics. In the context of spin systems, a paradigmatic example of fractionalisation occurs for the one-dimensional (1D) magnet formed from half-integer spin magnetic ions coupled by isotropic antiferromagnetic interactions. In this system, the spins cannot order at any temperature $T > 0$ K, and they exhibit exotic excitation spectra. Even though the spin operators in the Hamiltonian can only flip the spins by an integer unit of Planck’s constant, the actual excitations carry spin $S = \frac{1}{2}$ and are called spinons. Since any experimental technique can only change the angular momentum by an integer unit, spinons cannot be created singly but only in multiple pairs, leading to the characteristic multi-particle excitation continua observed in neutron scattering experiments\(^4\)\(^-\)\(^6\). The concept of fractionalisation and spinons can be extended to two and higher dimensions. In this case, spinons occur as excitations of quantum spin liquids - which have no static magnetism even at $T = 0$ K, i.e. they require an environment of strong quantum fluctuations as realised by magnets with small spin magnitudes and frustrated interactions\(^7\). Examples of two-dimensional (2D) quantum spin-liquid candidates are the kagome materials Herbertsmithite (ZnCu\(_3\)(OH)\(_6\))Cl\(_2\)\(^5\) and Ca\(_{10}\)Cr\(_7\)O\(_{28}\)\(^6\),\(^7\) where the quantum spin-liquid candidates are the kagome materials Herbertsmithite (ZnCu\(_3\)(OH)\(_6\))Cl\(_2\)\(^5\) and Ca\(_{10}\)Cr\(_7\)O\(_{28}\)\(^6\),\(^7\) where the specific thermal function and would show resolution-limited magnetic Bragg features at temperatures around $T \approx 1$ K\(^2\)\(^4\), they do not reveal a sharp $\lambda$-type anomaly and there is no phase transition to long-range magnetic order. Muon spin relaxation measurements also confirm the absence of any static magnetism down to 0.02 K\(^2\)\(^5\).

They do however, reveal enhanced magnetic correlations at low temperatures which onset below $T < 1$ K implying the presence of persistent spin dynamics in the ground state as is expected of a quantum spin liquid.

In the following, we provide strong experimental and theoretical indications for the quantum spin liquid state in PbCuTe\(_2\)O\(_6\). We perform neutron diffraction and inelastic neutron scattering measurements and show that the absence of long-range magnetic order in the powder sample of this compound is accompanied by diffuse spheres of dispersionless inelastic scattering consistent with a multi-particle continuum of spinons. We also determine the exchange interactions using density functional theory and establish that a three-dimensional frustrated motif called the hyper-hyperkagome is responsible for this behaviour. Finally, we reproduce the observed ground state and dynamics using pseudofermion functional renormalisation group calculations and show that this Hamiltonian generates the behaviours associated with a quantum spin liquid. The strength of our work lies at the excellent agreement between the experiment and theory that has not been observed until now for quantum spin liquids arising in such complex three-dimensional systems.

**Results**

**Magnetic interactions in PbCuTe\(_2\)O\(_6\).** The positions of the magnetic Cu\(^{2+}\) ions in PbCuTe\(_2\)O\(_6\) are shown in Fig. 1a where the green and red bonds represent the 1st and 2nd nearest-neighbour interactions $J_1$ and $J_2$, respectively. All Cu\(^{2+}\) ions are crystallographically equivalent. On its own, $J_1$ couples the Cu\(^{2+}\) moments into isolated triangles, while $J_2$ forms a three-dimensional network of corner-sharing triangles known as the hyperkagome lattice. Further neighbour interactions are also shown in Fig. 1b where the 3rd neighbour interaction $J_3$ forms isolated chains running parallel to the crystalline a, b, c axes and the 4th neighbour interaction $J_4$ is responsible for chains parallel to the body diagonals. The complete Hamiltonian is

$$\mathcal{H} = \sum_{k=1,2,3} J_k \mathbf{S}_i \cdot \mathbf{S}_j \tag{1},$$

where the interactions are assumed to be spin-isotropic, thus allowing the spins to point in any direction. This is justified because, as a light transition metal ion with only one hole in the 3d shell, the orbital moment of Cu\(^{2+}\) is quenched by the strong square-planar crystal field due to the surrounding O\(^2-\) ions. DC susceptibility confirms that the interactions are spin-isotropic, since it is almost independent of the direction of the applied magnetic field (Supplementary Note 1).

The ground state. In agreement with previous specific heat\(^24\) and muon spin relaxation\(^25\) (Supplementary Note 2) measurements we did not find any evidence for long-range magnetic order or static magnetism in powder samples of PbCuTe\(_2\)O\(_6\) down to 20 mK. We also performed neutron diffraction which directly measures the spatial Fourier transform of the spin–spin correlation function and would show resolution-limited magnetic Bragg peaks in the case of long-range magnetic order. Figure 2a shows the neutron powder diffraction patterns of PbCuTe\(_2\)O\(_6\) measured at temperatures $T = 2$ K and 0.1 K above and below $T = 1$ K respectively where features are observed in the specific heat and muon spin relaxation. Both patterns can be described entirely by considering only the known crystal structure of PbCuTe\(_2\)O\(_6\).\(^{26}\)

The absence of any additional Bragg peaks that could correspond to long-range magnetic order is further revealed by taking the difference between the diffraction patterns at these two temperatures as shown by the lower green curve. To establish an upper limit on the maximum size of any possible static ordered

ARTICLE NATURE COMMUNICATIONS | https://doi.org/10.1038/s41467-020-15594-1
$T \sim 1$ K whose origin is not yet understood. It is, however, well
known that these single crystals suffer from impurities with
5–10% of the chemical composition being Pb$_2$Te$_2$O$_6$ (Supple-
mental Note 3). This is in contrast to the higher quality powder
samples which do not show evidence of any transition around 1
K, hence, supporting the view that the low temperature transition
in single crystals results from disorder and strain effects. While
this transition could, in principle, mark the onset of magnetic
order, our analysis (Supplementary Note 4) at least rules out the
most obvious types of order which prompts us to speculate that
the transition is of structural type. In this work, however, we will
not focus on possible effects of impurities but rather discuss the
physics of the cleaner powder samples and single crystals above
the temperature/energy scale of 1 K.

Diffuse continuum of excitations. To explore the magnetic
excitations of PbCuTe$_2$O$_6$, we performed inelastic neutron
scattering. This technique directly measures the dynamical structure
factor $S(Q, E)$, which is the Fourier transform in space and time
of the spin-spin correlation function and allows the magnetic
excitation spectrum to be mapped out as a function of energy $E$
and momentum (or wavevector) transfer $Q$. Figure 3a shows
the excitation spectrum of a powder sample measured at $T = 0.1$ K. A
dispersionless, broad diffuse band of magnetic signal is clearly
visible around momentum transfer $|Q| = 0.8$ Å$^{-1}$. The magnetic
excitations extend up to 3 meV and are much broader than the
instrumental resolution. Figure 3c shows the magnetic signal at
$|Q| = 0.8$ Å$^{-1}$ plotted as a function of energy. The intensity is
greatest at $E = 0.5$ meV and weakens gradually with increasing
energy. The intensity also decreases rapidly with decreasing
energy and the presence of an energy gap smaller than 0.15 meV
is possible, but cannot be confirmed within the experimental
uncertainty.

To obtain a more detailed picture, inelastic neutron scattering
was performed on a single crystal sample which also shows the
continuous magnetic excitations at $|Q| = 0.8$ Å$^{-1}$ extending up to
3 meV (see Fig. 3b, d), in agreement with the powder data.
Figure 3e–g show the momentum-resolved excitations in the [h, k, 0]-plane measured at the constant-energy transfers $E = 0.75,$
1.5 and 2 meV, respectively, while Fig. 4b gives the scattering at
$E = 0.5$ meV. For all energy transfers, the excitations form a
diffuse ring at $|Q| = 0.8$ Å$^{-1}$, while additional weaker branches of
scattering extend outwards to higher wavevectors. At low-energy
transfers ($E < 1$ meV) the diffuse ring has double maxima at
wavevectors (1.69, $\pm 0.3$, 0) and ($\pm 0.3$, 1.69, 0), etc. (see Fig. 4h)
while at higher energies it broadens and becomes weaker. The
ring can also be observed in the [h, k, l]-plane where its intensity
also shows a modulation (as shown in Fig. 4a for $E = 0.5$ meV).
Together, these results indicate that the excitations in fact form a
diffuse sphere in reciprocal space with a radius of $|Q| \approx 0.8$ Å$^{-1}$. The
excitations of PbCuTe$_2$O$_6$ are clearly very different from the
sharp and dispersive spin-wave excitations expected in conven-
tional magnets with long-range magnetically ordered ground
states or from the gapped and dispersive magnon excitations of
dimer magnets$^{27,28}$. The possibility of a multimagon continuum
can also be excluded since in this case sharp excitations due
to single magnons would still be expected at low energies below the
continuum which are not observed (Supplementary Note 5).
Additionally, the stoichiometric nature of the compound rules
out disorder as the origin of the diffuse inelastic spectrum. The
diffuse scattering features observed in PbCuTe$_2$O$_6$ may indicate a
multi-spinon continuum of excitations as has been well
documented in one-dimensional antiferromagnets formed from
half-integer spin magnetic ions$^{1–3}$. They have also been observed in
several two-dimensional quantum spin liquids where similar

---

**Fig. 1 The magnetic interactions and Hamiltonian of PbCuTe$_2$O$_6$.** a The
magnetic structure drawn from the crystallographically equivalent magnetic
chains due to triangles and 10-spin loops in the hyperkagome lattice.

---

moment, several magnetic structures were simulated and com-
pared to the data. Figure 2b shows a modelled magnetic Bragg
peak compatible with the magnetic structure of the iso-
structural compound SrCuTe$_2$O$_6$ for different sizes of the ordered moment.
The ordered moment if present must be smaller than $=0.05 \mu_B$/
Cu$^{2+}$ which is much less than the total spin moment of the Cu$^{2+}$
ion of $1 \mu_B$, indicating that static magnetism is suppressed. It
should be mentioned that single crystals of PbCuTe$_2$O$_6$ do show
signs of a phase transition in their specific heat at temperature

diffuse ring-like features have been found\textsuperscript{3,6}. In three dimensions, most spin liquid candidates are based on the pyrochlore structure and their scattering forms a distinctive pinch-point pattern\textsuperscript{32}.

**Magnetic Hamiltonian.** Having confirmed that PbCuTe\textsubscript{2}O\textsubscript{6} exhibits features characteristic of a quantum spin liquid, we now investigate the origins of this behaviour by deriving the exchange interactions. For this purpose, we employ density functional theory (DFT). The resulting values of the interaction strengths are plotted as a function of the onsite interaction \( U \) in Fig. 1c for \( U = 5.5–8\ eV \) as this range spans the usual values for Cu\textsuperscript{2+}. We find that all the interactions are antiferromagnetic. In contrast to previous perturbation theory based DFT calculations where the hyperkagome interaction \( J_2 \) was found to be much stronger than the other interactions\textsuperscript{24}, our significantly better approach of energy mapping within DFT reveals that the two frustrated interactions \( J_1 \) and \( J_2 \) are of almost equal strength and are significantly stronger than the chain interactions \( J_3 \) and \( J_4 \). The combined effect of \( J_1 \) and \( J_2 \) is to couple the Cu\textsuperscript{2+} ions into a highly frustrated three-dimensional network of corner-sharing triangles similar to the hyperkagome lattice (\( J_2 \) only) but with a higher density of triangles. In the hyperkagome lattice each magnetic ion participates in two corner-sharing triangles, while in PbCuTe\textsubscript{2}O\textsubscript{6} each Cu\textsuperscript{2+} ion participates in three triangles resulting in a higher connectivity - we name this lattice the hyper-hyperkagome. An important difference between these two lattices is the size of the smallest possible closed loops (beyond the triangles) around which the spins can resonate. The hyperkagome lattice consists of interconnected loops of 10 spins. The hyper-hyperkagome can also be viewed as interconnected loops, however, with the smallest connecting 4 spins and another consisting of 6 spins. For comparison, the 2D kagome has smallest loops of 6 spins (see Fig. 1a). As shown in Fig. 1c, the values of the exchange interactions decrease as the value of \( U \) increases. For each value of \( U \) the resulting set of interaction strengths can be used to calculate the Curie-Weiss temperature \( \theta_{\text{CW}} \). Since DC susceptibility measurements yield \( \theta_{\text{CW}} = -22\ K \)\textsuperscript{24,25}, we use \( U = 7.5\ eV \) (corresponding to \( \theta_{\text{CW}} = -23\ K \)) giving interaction sizes \( J_1 = 1.13\ \text{meV}, \ J_2 = 1.07\ \text{meV}, \ J_3 = 0.59\ \text{meV} \) and \( J_4 = 0.12\ \text{meV} \) \((J_1 : J_2 : J_3 : J_4 \approx 1 : 1 : 0.5 : 0.1)\). These values are significantly different from the reported ratio \( J_1 : J_2 : J_3 = 0.54 : 1 : 0.77 \) given in ref. 24 (Supplementary Note 6).

**Comparison to theory.** To gain further insight into the magnetic behaviour of PbCuTe\textsubscript{2}O\textsubscript{6}, the static susceptibility expected from this set of interactions was calculated using the theoretical technique of pseudo-fermion functional renormalisation group (PFFRG). This method calculates the real part of the static spin susceptibility which corresponds to the energy-integrated neutron scattering cross-section as discussed in the methods section. In agreement with the experimental observations for the powder sample, the susceptibility does not show any sign of long-range magnetic order even down to the lowest temperatures, confirming that static magnetism is suppressed by this Hamiltonian. The momentum resolved susceptibility calculated at \( T = 0.2\ K \) is shown in Fig. 4c, d for the \([h, k, l] \)- and \([h, h, l] \)-planes respectively. It predicts a diffuse sphere of scattering at the same wavevectors and with similar intensity modulations as those observed experimentally (Figs. 3e–g and 4a, b), and is even able to reproduce the weaker features. The accuracy of the calculations can be further demonstrated by comparing cuts through the data and simulations. As shown in Fig. 4g, h, the theory reproduces the double maxima as well as the structure of the slopes of these peaks to high precision. We emphasise that this level of agreement has hardly ever been achieved for such a material with many competing interactions on a complicated three-dimensional lattice and in the extreme quantum (spin-\( \frac{1}{2} \)) limit. From a more general viewpoint, it demonstrates that the combination of DFT and PFFRG provides a powerful and flexible numerical framework for the investigation of real quantum magnetic materials. The PFFRG method was also used to test the robustness of the spin liquid state to variations in the Hamiltonian. We find that the ground state shows no tendency toward long-range magnetic order when the ratio of interactions are varied over 0.975 \( \leq J_1/J_2 \leq 1.08 \) (corresponding to \( -37 \leq \theta_{\text{CW}} \leq -21\ K \)) while the momentum-resolved susceptibility changes only slightly (Fig. 4g, h).

**Discussion**

In total, the neutron data and numerical simulations, together with the small spin\( \frac{1}{2} \) moments and the isotropic interactions point to the presence of strong quantum fluctuations that destroy long-range magnetic order or any static magnetism in the ground state of PbCuTe\textsubscript{2}O\textsubscript{6}. This is in stark contrast to the previously studied 3D pyrochlore classical spin ice materials with large moments and highly anisotropic interactions, where the magnetic

---

\textbf{Fig. 2 Powder neutron diffraction patterns of PbCuTe\textsubscript{2}O\textsubscript{6}.} a Measured at 0.1 K and 2 K using the WISH high-flux diffractometer and plotted as a function of d-spacing. Both patterns are refined in the established cubic space group, \( P4_3_2 \)\textsuperscript{26} where the lattice constant at 0.1 K is 12.4454(3) Å. The difference between the patterns at these two temperatures is plotted below in green and clearly shows that no magnetic Bragg peaks appear at the base temperature. b Assuming a magnetic structure compatible with the long-range magnetic order found in SrCuTe\textsubscript{2}O\textsubscript{6}, a magnetic Bragg peak is estimated at the \((1, 0, 0)\) reflection (12.4454 Å in d-spacing). The expected Bragg peak amplitude is shown for different values of ordered moment per Cu\textsuperscript{2+} ion by the curves. Clearly, if present, the maximum ordered moment can be no greater than 0.05 \( \mu_B \).
moments are static in the ground state\textsuperscript{14,15}. A fluctuating ground state as observed for PbCuTe\textsubscript{2}O\textsubscript{6} is known to provide the right physical environment for spin fractionalisation associated with deconfined spinon excitations. Such particles are generally observed as a multi-spinon spectrum that is broad and diffuse in momentum and energy. This, in turn, is the type of signal which we independently observed in both inelastic neutron experiments and PFFRG calculations making our quantum spin-liquid interpretation plausible. The issue of whether this is a gapped or gapless quantum spin liquid remains unresolved, however a clear depletion of magnetic states at low-energy suggests that a gap $<0.15$ meV could exist.

An important remaining question is why the complex model we propose for PbCuTe\textsubscript{2}O\textsubscript{6} induces sufficiently strong quantum
fluctuations for quantum spin liquid formation. According to common understanding, quantum effects for small spins are particularly strong when the corresponding classical (large spin) model exhibits an infinite ground state degeneracy, as is the case for the kagome or pyrochlore models with isotropic antiferromagnetic interactions. Performing a classical Monte Carlo analysis of our system, we found that the full $J_1-J_2-J_3-J_4$ model in fact does not exhibit infinite degeneracy for large spin but instead shows long-range magnetic order. However, we have identified an infinite degeneracy in the classical model with only the $J_1$ and $J_2$ interactions. From this perspective, the weaker $J_3$ and $J_4$ couplings act as perturbations inducing a small energy splitting in the degenerate classical $J_1-J_2$-only system. We, therefore, propose that the strong quantum fluctuations of the full $J_1-J_2-J_3-J_4$ model with quantum spin-$\frac{1}{2}$ originate from the degeneracy of the classical $J_1-J_2$ model. This is supported by PFFRG.
Fig. 4 The low-energy magnetic excitation spectrum of PbCuTe2O6 compared to theory. The colour maps in a, b show the excitations measured at energy transfer \( E = 0.5 \text{ meV} \) and temperature \( T < 0.1 \text{ K} \) in the \([h, h, j]-\) and \([h, k, 0]-\) planes respectively. The \([h, j]-\) map was measured using the LET spectrometer with an incident energy of \( E_i = 5.46 \text{ meV} \). The data has been integrated over energy transfer 0.4 \( \leq E \leq 0.6 \text{ meV} \), and out-of-plane wavevector transfer \(-0.1 \leq h, -0.1 \leq 0.1 \text{ r.l.u.} \). The \([h, k, 0]\) spectrum was measured on the ThALES spectrometer with an energy resolution of 0.097 meV. c, d Static (real valued) spin susceptibility calculated using the pseudo-fermion functional renormalisation group (PFFRG) method in the quantum limit (\( S = 1/2 \)) for \( J_1 = 1.13 \text{ meV} \), \( J_2 = 1.07 \text{ meV} \), \( J_3 = 0.59 \text{ meV} \) and \( J_4 = 0.12 \text{ meV} \) (corresponding to a Curie-Weiss temperature \( \theta_{\text{CW}} = -23 \text{ K} \)). The dashed black lines indicate the positions of strong scattering in the classical \( J_1 - J_2 \) only model [see e, f]. e, f Corresponding classical PFFRG results obtained in the limit of large spin magnitude for a model with only \( J_1 \) and \( J_2 \) couplings of equal strength. g, h The experimental and theoretical magnetic intensity as a function of wavevector transfer along \([h, -1, -1] \) and \([h, -1, -1] \) respectively. The data points (blue circles) were measured at an energy transfer \( E = 0.5 \text{ meV} \) and temperature \( T < 0.1 \text{ K} \) and were obtained by integrating the data shown in a, b over the respective shaded regions. The solid lines show the theoretical intensity obtained by performing the same integration through the same theoretical simulations shown in c, d. The theoretical intensity distribution is also shown for another set of exchange interactions represented by their corresponding Curie-Weiss temperature \( \theta_{\text{CW}} = -37 \text{ K} \) (see Fig. 1c).

The calculations showing that the correlation profiles of both systems resemble each other (see Fig. 4e, where the degeneracy of the \( J_1 - J_2 \) only classical model manifests as streaks in the \([1, 1, 1] \) direction). Finally, the degeneracy in the classical \( J_1 - J_2 \) model can be understood from the fact that the hyper-hyperkagome lattice forms a network of corner-sharing triangles. As for the classical anti-ferromagnetic kagome and pyrochlore lattices, the ground states in such corner-sharing geometries must obey the local constraint that the vector sum of the spins in each triangle or tetrahedron is zero. The large ground-state degeneracy then follows from the fact that there are infinitely many states which fulfill all constraints.

In conclusion, while no experimental technique or theoretical method is able to conclusively prove the existence of a quantum spin liquid we show using a combination of theory and experiment that PbCuTe2O6 exhibits the measureable features expected of a quantum spin liquid including no detectable static magnetism and the presence of diffuse dispersionless spinon-like excitations. Although PbCuTe2O6 has a complex Hamiltonian it is clear that the frustration arises from the network of corner-sharing triangles due to the dominant \( J_1 \) and \( J_2 \) interactions. While this has been explored in the hyperkagome lattice where each spin participates in two corner-sharing triangles giving closed loops of 10 spins18-20, there has until now been little experimental or theoretical exploration of this more highly connected hyper-hyperkagome lattice where each spin participates in three corner-sharing triangles resulting in smaller closed loops of 4 spins. The weaker interaction \( J_3 \) which reduces the classical ground state degeneracy has the tendency to bring the quantum system closer to long-range magnetic order and may be the reason why order might be present in the single crystal samples with more impurities where impurities and additional defects could disrupt the frustration. The hyper-hyperkagome lattice has also been found in Co doped \( \beta \)-Mn30. Here the spin is effectively classical and the metallic nature of this material promotes long-range interactions where the ferromagnetic 6th neighbour interaction has a similar strength to the antiferromagnetic \( J_1 \) and \( J_2 \). The wavevector-dependent scattering has some similarities to PbCuTe2O6 although with sharper features and can be explained by a model where \( J_3 \) ferromagnetically couples the spins into rods which then form competing triangular lattices.

In summary, three-dimensional spin liquids are very rare and current examples are confined mostly to the pyrochlore and hyperkagome lattices, thus our experimental and theoretical results are of high importance because they reveal a distinctly different type of three-dimensional lattice capable of supporting spin liquid behaviour.

Methods
Neutron scattering measurements. Powder neutron diffraction was performed on the time-of-flight diffractometer WISH at the ISIS Facility, Didcot, U.K. The sample (weight 13 g) was placed into a copper can and the diffraction patterns were collected at \( T = 2 \text{ K} \) and 0.1 K. The powder inelastic neutron scattering data was obtained at the time-of-flight spectrometer LET also located at the ISIS facility. For these measurements the same powder sample (weight 13 g) was placed between two coaxial copper cans to achieve a cylindrical sample shape, and Helium gas exchange was used for better temperature stability. The measurements were performed at \( T = 0.1 \text{ K} \) with incident energies: \( E_i = 18.2 \text{ meV} \), 5.64 meV, 2.72 meV and 1.59 meV. Single crystal inelastic neutron measurements in the \([h, 0, 0]\)–plane were obtained at the ThALES triple-axis spectrometer using the flatcone detector at the ILL, Grenoble, France, and also at the MACS triple-axis spectrometer at NIST, Gaithersburg, USA. Wavevector maps at constant energy were measured on ThALES at \( T = 0.05 \text{ K} \) while rotating the crystal in 0.5 deg steps with a fixed final energy of \( E_f = 4.06 \text{ meV} \) giving an energy resolution 0.097 meV. The wavevector resolution in the plots is 0.05 r.l.u \times 0.05 r.l.u. At MACS, the initial energy was set to \( E_i = 4 \text{ meV} \) for energy transfer of \( E = 0.75 \text{ meV} \) (giving energy resolution of 0.24 meV) and \( E_i = 5 \text{ meV} \) for \( E = 1.5 \text{ meV} \) and 2 meV (energy resolution 0.35 meV). The wavevector maps were obtained by rotating the crystal with a step size of 1 deg and the data were plotted by rebinning to 0.04 r.l.u \times 0.04 r.l.u pixels. The maps in the \([h, 0, 0]\)–plane were obtained at the LET spectrometer in ISIS at \( T = 0.03 \text{ K} \) with incident energies of \( E_i = 26.24 \text{ meV} \), 5.46 meV, 2.29 meV, 1.25 meV and 0.79 meV. For \( E_i = 5.46 \text{ meV} \) this gives an energy resolution of 0.18 meV.

Density functional theory calculations. We determined the parameters of the Heisenberg Hamiltonian in Eq. (1) for PbCuTe2O6 using density functional theory (DFT) calculations with the all electron full potential local orbital (FLOPT) basis32. We based our calculations on the structure determined via powder X-ray diffraction by Koteswararao et al.24. The exchange couplings were extracted by mapping the total energies of many different spin configurations onto the classical energies of the Heisenberg Hamiltonian. Note that this approach differs from the second order perturbation theory estimates using \( J = \frac{\Delta H}{\theta} \) for the exchange interactions reported in ref. 24 which includes only the antiferromagnetic super-exchange contribution based on one virtual process. In order to increase the number of inequivalent Cu2+ ions from one to six and thus to allow for different spin configurations, we lowered the symmetry of the crystal from \( P4_3 32 \) to \( P2_1_2_1_2 \). We convolved the total energies with 6 \times 6 \times 6 k-meshes and accounted for the strong electronic correlations using a GGA + U exchange correlation functional23. The value of the Hund’s rule coupling was fixed at the typical value \( J_h = 1 \text{ eV} \) and the onsite correlation strength \( U \) varied between 5.5 eV and 8 eV. We determined the most relevant \( U \) by using the constraint that the exchange couplings reproduce the experimentally determined Curie–Weiss temperature of \( \theta_{\text{CW}} = -22 \text{ K} \). This led to a DFT result for the first four exchange couplings of PbCuTe2O6 of \( J_1 = 1.13 \text{ meV} \), \( J_2 = 1.07 \text{ meV} \), \( J_3 = 0.59 \text{ meV} \) and \( J_4 = 0.12 \text{ meV} \). The full results are given in the Supplementary Note 6.

Pseudofermion functional renormalisation group calculations. The microscopic spin model proposed by DFT calculations is treated within the PFFRG approach33,34, which first reformulates the original spin operators in terms of Abrikosov fermions. The resulting fermionic model is then explored within the well-developed FRGG framework35,36. Effectively, the PFFRG method amounts to generating and summing up a large number of fermionic Feynman diagrams, each representing a spin-spin interaction process that contributes to the magnetic susceptibility. In terms of the original spin degrees of freedom, this summation corresponds to a simultaneous expansion in 1/\( S \) and 1/N, where \( S \) is the spin magnitude and \( N \) generalises the symmetry group of the spins from \( S = 2 \) to \( S = N \). The exactness of the PFFRG in the limits \( 1/S \to 0 \) and \( 1/N \to 0 \) ensures that magnetically ordered states (typically obtained at large \( S \)) and non-magnetic spin liquids (favoured at large \( N \)) can both be faithfully described within the same numerical framework. Particularly, due to this property, no bias towards either magnetic order or non-magnetic behaviour is built-in. In principle, the PFFRG treats an infinitely large lattice, however, spin–spin correlations are only taken into account if they are large.
account up to a certain distance while longer range correlations are put to zero. The computation times of the PFFRG scale quadratically with the correlated volume, where the experimental data weighted by the inverse energy. However, it is clear from this equation that the PFFRG results since it has the strongest signal. If a magnetic system develops magnetic order, the spin susceptibility $\chi(\mathbf{Q})$ manifests in a breakdown of the renormalisation group flow, accompanied by distinct peaks. An important advantage of the PFFRG is that it is even strongly fluctuating non-ordered magnetic phases, short-range spin correlations and their momentum profiles can be accurately calculated and compared to neutron scattering results. For consistency, the PFFRG spin susceptibility is corrected for the magnetic form factor of the Cu$^{2+}$ ion in the dipole approximation.

**Classical simulations.** For the numerical treatment of spin systems in the classical limit $S \rightarrow \infty$ we have employed a spin-$S$ generalisation of the PFFRG approach. On a technical level, this requires the introduction of 4S fermionic degrees of freedom per lattice site as discussed in ref. 39. In the classical limit, the PFFRG equations can be solved analytically to obtain the momentum-resolved magnetic susceptibility. It can be shown that the classical wave vector at which the susceptibility is strongly peaked is identical to the one predicted within the Luttinger–Tisza method. The final susceptibility is corrected for the Cu$^{2+}$ magnetic form factor in the dipole approximation.

**Data availability**

Powder neutron diffraction data were obtained on the time-of-flight diffractometer WISH at the ISIS facility, Didcot, UK. Powder and single-crystal inelastic neutron scattering data were measured on the time-of-flight spectrometer LET also at the ISIS facility. Single-crystal inelastic neutron scattering data were also collected on the triple-axis spectrometers ThALES with the flat cone option at the Institut Laue-Langevin (data available at ref. 41). Grenoble, France, and MAGIS II at the NIST Center for Neutron Research, Gaithersburg, USA. All the raw and derived data that support the findings of this study are available from the authors upon reasonable request.

Received: 12 December 2017; Accepted: 29 February 2020; Published online: 11 May 2020

**References**

1. Lake, B., Tennant, A., Frost, C. D. & Nagler, S. E. Quantum criticality and universal scaling of a quantum antiferromagnet. *Nat. Mater.* 4, 329–334 (2005).
2. Lake, B. et al. Multispinon continua at zero and finite temperature in a near-ideal Heisenberg chain. *Phys. Rev. Lett.* 111, 137205 (2013).
3. Mourigal, M. et al. Fractional spin excitations in the quantum Heisenberg antiferromagnetic chain. *Nat. Phys.* 9, 435–441 (2013).
4. Balents, L. Spin liquids in frustrated magnets. *Nature* 464, 199–208 (2010).
5. Han, T.-H. et al. Fractionalized excitations in the spin-liquid state of a kagome-lattice antiferromagnet. *Nature* 492, 406–410 (2012).
6. Balz, C. et al. Magnetic Hamiltonian and phase diagram of the quantum spin liquid Ca$_2$Cu$_3$O$_8$. *Phys. Rev. B* 95, 174414 (2017).
7. Canals, R. & Lacroix, C. Pyrochlore antiferromagnet: a three-dimensional quantum spin liquid. *Phys. Rev. Lett.* 80, 2923–2926 (1998).
8. Moessner, R. & Chalker, J. T. Properties of a classical spin liquid: the Heisenberg pyrochlore antiferromagnet. *Phys. Rev. Lett.* 80, 2929–2932 (1998).
9. Benton, O., Jaubert Jaubert, L. D. C., Yuan, H. & Shannon, N. A spin liquid with pinch-like singularities on the pyrochlore lattice. *Nat. Commun.* 7, 2923–334 (2016).
10. Savary, L. & Balents, L. Disorder-induced quantum spin liquid in spin ice pyrochlores. *Phys. Rev. Lett.* 118, 087203 (2017).
11. Iqbal, Y. et al. Signatures of a gearwheel quantum spin liquid in a spin$^\frac{1}{2}$ pyrochlore molybdate Heisenberg antiferromagnet. *Phys. Rev. Mater.* 1, 023601 (2017).
12. Iqbal, Y. et al. Quantum and classical phases of the pyrochlore Heisenberg model with competing interactions. *Phys. Rev. X* 9, 011005 (2019).
13. Fennell, T. et al. Magnetic Coulomb phase in the spin ice Ho$_2$Ti$_2$O$_7$. *Science* 326, 415–417 (2009).
14. Morris, D. J. P. et al. Dirac strings and magnetic monopoles in the spin ice pyrochlore *Science* 326, 411–414 (2009).
15. Gingras, M. J. P. & McClarty, P. A. Quantum spin ice: a search for gapless quantum spin liquids in pyrochlore magnets. *Rep. Prog. Phys.* 77, 056501 (2014).
16. Benton, O., Sikora, O. & Shannon, N. Seeing the light: experimental signatures of emergent electromagnetism in a quantum spin ice. *Phys. Rev. B* 86, 075154 (2012).
17. Hopkinson, J. M., Isakov, S. V., Kee, H.-Y. & Kim, Y. B. Classical antiferromagnet on a pyrochlore lattice. *Phys. Rev. Lett.* 99, 037201 (2007).
18. Zhou, Y., Lee, P. A., Ng, T.-K. & Zhang, F.-C. Na$_4$Ir$_3$O$_8$ as a 3D spin liquid with fermionic spinons. *Phys. Rev. Lett.* 101, 197201 (2008).
19. Bergholtz, E. J., Läuchli, A. M. & Moessner, R. Symmetry breaking on the three-dimensional pyrochlore lattice of Na$_4$Ir$_3$O$_8$. *Phys. Rev. Lett.* 105, 237202 (2010).
20. Schiffer, P. et al. Frustration induced spin freezing in a site-ordered magnet: gadolinium gallium garnet. *Phys. Rev. Lett.* 74, 2379–2382 (1995).
21. Quilliam, J. A. et al. Just-in-time spin reorientation and long range order in a series of geometrically frustrated antiferromagnetic gadolinium garnets. *Phys. Rev. B* 87, 174421 (2013).
22. Dally, R. et al. Short-range correlations in the magnetic ground state of Na$_4$Ir$_3$O$_8$. *Phys. Rev. Lett.* 113, 247601 (2014).
23. Koteswararao, B. et al. Magnetic properties and heat capacity of the three-dimensional frustrated PbCuTe$_2$O$_7$. *Phys. Rev. B* 90, 035141 (2014).
24. Khuntia, P. et al. Spin liquid state in the 3D frustrated antiferromagnet PbCuTe$_2$O$_7$. NMR and muon spin relaxation studies. *Phys. Rev. Lett.* 116, 107203 (2016).
25. Wulff, L. & Müller-Buschbaum, H. Planar CuO$_2$ polygons and a one-sided open Te$_{4d}$O$_4$ coordination in Sr$_2$Cu$_2$O$_2$. *Z Naturforsch.* B 52, 1341–1344 (1997).
26. Quintero-Castro, D. L. et al. Magnetic excitations of the gapped quantum spin dimer antiferromagnet Sr$_2$CrO$_4$. *Phys. Rev. B* 81, 014415 (2010).
27. Kofu, M. et al. Magnetic-field-induced phase transitions in a weakly coupled system. *Phys. Rev. B* 49, 197201 (1994).
28. Henley, C. L. Power-law spin correlations in pyrochlore antiferromagnets. *Phys. Rev. B* 71, 044424 (2005).
29. Koppelmek, K. & Eschrig, H. Full-potential nonorthogonal local-orbital minimum-basis band-structure scheme. *Phys. Rev. B* 59, 177204–177209 (1999).
30. Gutering, D., Valenti, R. & Jeschke, H. O. Reduction of magnetic interlayer coupling in barlowite through isoelectronic substitution. *Phys. Rev. B* 94, 125136 (2016).
31. Liechtenstein, A. I., Anisimov, V. I. & Zaanen, J. Density-functional theory and strong interactions: orbital ordering in Mott-Hubbard insulators. *Phys. Rev. B* 52, R5467–R5470 (1995).
32. Reuther, J. & Wölfle, P. J1–J2 frustrated two-dimensional Heisenberg model: random phase approximation and functional renormalization group. *Phys. Rev. B* 81, 144410 (2010).
33. Iqbal, Y., Thomale, R., Toldin, F. P., Rachel, S. & Reuther, J. Functional renormalization group for three-dimensional quantum magnetism. *Phys. Rev. B* 94, 134008 (2016).
34. Hollihoki, J. Renormalization and effective Lagrangians. *Nucl. Phys. B* 231, 269–295 (1984).
35. Metzner, W., Salhofer, M., Honerkamp, C., Meden, V. & Schönhammer, K. Functional renormalization group approach to correlated fermion systems. *Rev. Mod. Phys.* 84, 299–352 (2012).
36. Brown, P. I. Magnetic Form Factors. p. 454–461. (Springer Netherlands, Dordrecht, 2004).
37. Baer, M. L. & Reuther, J. Numerical treatment of spin systems with unrestricted spin length S: A functional renormalization group study. *Phys. Rev. B* 96, 045144 (2017).
38. Luttinger, J. M. & Tisza, L. Theory of dipole interaction in crystals. *Phys. Rev.* 70, 954–964 (1946).
Acknowledgements

We thank K. Siemensmeyer for his help with the susceptibility measurements, D. Voneshen for his help with the inelastic neutron experiments performed on LET at the ISIS facility, Tobias Müller for performing classical Monte Carlo simulations and C. Baines for his help with μSR experiments on LET at S burns facility at PSI. S.C., B.L. and A.T.M.N.I. acknowledge the support of DFG through project B06 of SFB 1143 (ID 247310070). Access to MACS was provided by the Center for High Resolution Neutron Scattering, a partnership between the National Institute of Standards and Technology and the National Science Foundation under Agreement No. DMR-1508269. Y.I. and R.T. gratefully acknowledge the Gauss Centre for Supercomputing e.V. for funding this project by providing computing time on the GCS Supercomputer SuperMUC at the Leibniz Supercomputing Centre (LRZ). J.R. is supported by the Freie Universität Berlin within the Excellence Initiative of the German Research Foundation. Y.I. acknowledges the Science and Engineering Research Board (SERB), India for support through Startup Research Grant No. SRG/2019/000056. This research was supported in part by the International Centre for Theoretical Sciences (ICTS) during a visit to participate in the programme The 2nd Asia Pacific Workshop on Quantum Magnetism (Code: ICTS/apfm2018/11). Y.I. acknowledges the kind hospitality of the Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany where a part of the research was carried out.

Author contributions

A.T.M.N.I. made the powder and single crystal samples. S.C. performed or participated in all neutron measurements, and analysed the data with help from the other authors. J.A.R.-R., R.B. and P.S. supported the INS measurements and D.K. and P.M. supported the neutron diffraction measurements. B.L. participated in most measurements and directed the experimental aspects of the project. The DFT calculations were performed by H.O.J.; Y.I. carried out the quantum PFFRG calculations with the help of J.R. and R.T., while J.R performed the classical simulations and directed the theoretical aspects of the project. S.C. and B.L. wrote the manuscript with contributions from all authors.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information is available for this paper at https://doi.org/10.1038/s41467-020-15594-1.

Correspondence and requests for materials should be addressed to S.C.

Peer review information: Nature Communications thanks Lukas Janssen and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

Reprints and permission information is available at http://www.nature.com/reprints

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2020