

Random singlet physics in the $S = \frac{1}{2}$ pyrochlore antiferromagnet $\text{NaCdCu}_2\text{F}_7$

A. Kancko¹, H. Sakai², C. A. Corrêa³, P. Proschke¹, J. Prokleška¹, T. Haidamak¹, M. Uhlarz⁴, A. Berlie⁵, Y. Tokunaga² and R. H. Colman^{1,*}

¹*Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University, Ke Karlovu 5, 121 16 Prague, Czech Republic*

²*Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan*

³*Institute of Physics, Czech Academy of Sciences, Na Slovance 2, 182 00 Prague, Czech Republic*

⁴*Hochfeld-Magnetlabor Dresden (HLD-EMFL), Helmholtz-Zentrum Dresden-Rossendorf (HZDR), 01328 Dresden, Germany*

⁵*ISIS Neutron and Muon Source, Rutherford Appleton Laboratory, Science and Technology Facilities Council, Chilton, Oxfordshire OX11 0QX, United Kingdom*



(Received 24 September 2025; accepted 9 March 2026; published 31 March 2026)

We report a random singlet ground state in the $S = \frac{1}{2}$ Heisenberg pyrochlore antiferromagnet $\text{NaCdCu}_2\text{F}_7$. Cationic $\text{Na}^+/\text{Cd}^{2+}$ disorder on the pyrochlore A site generates a broad distribution of $\text{Cu}^{2+}-\text{F}^--\text{Cu}^{2+}$ exchange couplings, introducing intrinsic magnetic bond disorder. Despite strong antiferromagnetic interactions ($\theta_{\text{CW}} = -72$ K), no magnetic order or global spin freezing is observed in dc and ac susceptibility, specific heat, or ^{23}Na nuclear magnetic resonance to 120 mK, with muon spin relaxation experiments confirming persistent spin dynamics to 58 mK. T -linear specific heat, a Curie-like susceptibility tail, and power-law scaling with data collapse in $\chi(T)$, $M(H)$, C_{mag}/T , ^{23}Na ($1/T_1T$), and the muon spin polarization $P(t)$ reveal a disorder-driven network of random singlets and orphan spins. Scaling across multiple bulk and local probes is consistent with a broad distribution of exchange energies, $P[\mathcal{J}] \sim \mathcal{J}^{-\alpha}$. This behavior contrasts with previously studied $\text{NaA}''\text{B}_2\text{F}_7$ pyrochlore fluorides, where magnetic bond disorder precipitates spin-glass freezing, underscoring the crucial role of strong $S = \frac{1}{2}$ quantum fluctuations in $\text{NaCdCu}_2\text{F}_7$.

DOI: [10.1103/rw5c-491w](https://doi.org/10.1103/rw5c-491w)

I. INTRODUCTION

Quantum spin liquids (QSLs)—highly entangled dynamic ground states without long-range magnetic order down to $T = 0$ —remain a central focus of frustrated magnetism due to their exotic fractionalized excitations and nontrivial topology [1–4], with possible applications in quantum technologies [5–8]. The $S = \frac{1}{2}$ Heisenberg antiferromagnet on the pyrochlore lattice is a canonical model [9]: Corner-sharing tetrahedra produce strong geometrical frustration, suppressing semiclassical order and promoting competing ground states, with theoretical predictions spanning chiral or dimerized phases and QSLs with short-range correlations [10–17].

Experimental realizations of ideal $S = \frac{1}{2}$ Heisenberg pyrochlore antiferromagnets are exceedingly rare. Multiple rare-earth $J_{\text{eff}} = \frac{1}{2}$ pyrochlore oxides show QSL-like behavior, but suffer from strong anisotropy due to spin-orbit coupling and crystal field effects [18–20], while $J_{\text{eff}} = \frac{1}{2}$ Co^{2+} -based pyrochlore fluorides freeze into glassy states due to magnetic bond disorder [21–23]. $\text{Lu}_2\text{Mo}_2\text{O}_5\text{N}_2$ [24] and

$\text{Hg}_2\text{Cu}_2\text{F}_6\text{S}$ [25] represent the only known realizations of true $S = \frac{1}{2}$ pyrochlores. $\text{Lu}_2\text{Mo}_2\text{O}_5\text{N}_2$ exhibits a dynamic, QSL-like ground state without spin freezing despite significant disorder, in stark contrast to its spin-glass $S = 1$ counterpart $\text{Lu}_2\text{Mo}_2\text{O}_7$, underscoring the role of strong $S = \frac{1}{2}$ quantum fluctuations. $\text{Hg}_2\text{Cu}_2\text{F}_6\text{S}$ also displays QSL-like behavior, but the lack of comprehensive low-temperature characterization leaves its ground-state nature unresolved.

In real materials, quenched disorder via site mixing, vacancies, or lattice distortions is unavoidable and can instead stabilize disorder-driven phases [26]. One prominent outcome is the random singlet (RS) state [27–37], where a broad distribution of antiferromagnetic (AF) exchange energies produces localized singlets over multiple length scales, while weakly coupled moments form paramagnetic “orphan spins.” The RS state can be distinguished from valence-bond solids [38] and resonating valence-bond manifolds [39] by its disorder-induced power-law distributions of exchange couplings $P[\mathcal{J}] \sim \mathcal{J}^{-\alpha}$ and density of states $\rho(E) \sim E^{-\alpha}$, leading to a characteristic T -linear specific heat, gapless susceptibility with a Curie-like tail, broad features in the dynamic spin structure factor, and power-law scaling of $\chi(T)$, $M(H)$, and C_{mag}/T with data collapse as a function of temperature and magnetic field T/H [40–44]. RS behavior has been documented in one-dimensional chains [45–47], two-dimensional layered lattices [41,48–59], and a few three-dimensional (3D) frustrated magnets [60,61], but has not been established in an $S = \frac{1}{2}$ Heisenberg pyrochlore antiferromagnet.

*Contact author: ross.colman@matfyz.cuni.cz

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

In this work, we provide conclusive evidence for an RS ground state in $\text{NaCdCu}_2\text{F}_7$, making it the first $S = \frac{1}{2}$ Heisenberg pyrochlore where the RS state is confirmed across multiple probes. The inherent structural $\text{Na}^+/\text{Cd}^{2+}$ cationic disorder induces randomness in the $\text{Cu}^{2+}-\text{F}^--\text{Cu}^{2+}$ magnetic superexchange pathways, forming the necessary distribution of exchange energies $\mathcal{J}$. Despite the strong AF interactions ($\theta_{\text{CW}} = -72$ K), no magnetic transition or global spin freezing is seen in dc magnetization, ac susceptibility, specific heat, or ^{23}Na nuclear magnetic resonance (NMR), and we confirm continued dynamics down to 58 mK via zero-field (ZF) and longitudinal field (LF) muon spin relaxation (μSR) measurements. Convincingly, evidence of power-law scaling and data collapse can be found in each of these experimental probes. The strong quantum fluctuations of the $S = \frac{1}{2}$ Cu^{2+} ions suppress the spin-glass freezing observed in related pyrochlore fluoride materials [21–23,62–64], instead giving rise to an RS state. This establishes $\text{NaCdCu}_2\text{F}_7$ as a benchmark platform for testing theoretical predictions of 3D random singlet physics and exploring low-energy excitations, field-tuned phenomena, and exotic quantum spin-liquid behavior in three dimensions.

II. EXPERIMENTAL METHODS

$\text{NaCdCu}_2\text{F}_7$ was synthesized using the optical floating-zone method by melting a stoichiometric fluoride mix in a hollow graphite crucible, yielding a largely polycrystalline rod. Phase purity was verified by powder x-ray diffraction (PXRD) using a Bruker D8 Advance diffractometer with $\text{Cu K}\alpha_1$ and $\text{Cu K}\alpha_2$ radiation ($\lambda = 1.5418$ Å). Upon crushing the as-grown rod, small crystallites were isolated and used for structure solution using single-crystal x-ray diffraction (SCXRD). The resulting structural model was then refined against the PXRD data using Rietveld refinement implemented in Topas Academic V6 [65].

SCXRD was measured at 300 K using a Rigaku Xcalibur Gemini ultradiffractometer, with graphite-monochromated $\text{Mo K}\alpha$ ($\lambda = 0.71073$ Å) radiation, and an Atlas S2 charge-coupled device detector. Diffraction data were integrated using CrysAlis Pro [66] with an analytical numeric absorption correction combined with a multifaceted crystal model and an empirical absorption correction using spherical harmonics. The structure was solved by charge flipping using Superflip [67] and refined by full-matrix least squares in Jana2020 [68]. Structural graphics were created using Vesta [69].

Dc magnetization measurements on powdered $\text{NaCdCu}_2\text{F}_7$ were performed in a Quantum Design Magnetic Property Measurement System (model MPMS-XL 7 T), in the temperature range $T = 1.8$ –300 K and the magnetic field range $\mu_0 H = 0$ –7 T, using the reciprocating sample option.

High-field isothermal magnetization was measured in the High Magnetic Field Laboratory (Hochfeld-Magnetlabor Dresden, HLD) in the Helmholtz-Zentrum Dresden-Rossendorf (HZDR) in Dresden, Germany, using a pulsed 60 T magnet with a cryostat setup allowing the temperature range $T = 1.35$ –50 K.

Ac susceptibility was measured in the frequency range $\nu_{\text{AC}} = 215$ –10 000 Hz between $T = 0.4$ and 4 K, using a

custom-made coil for the ^3He option in a Quantum Design Physical Property Measurement System (PPMS).

Specific heat measurements were performed via the heat relaxation method in the PPMS between $T = 1.8$ and 300 K, with the addition of a ^3He low-temperature insert for temperatures $T = 0.4$ –20 K, in fields of $\mu_0 H = 0$ –9 T. The lattice specific heat was removed by subtracting the specific heat of the nonmagnetic analog $\text{NaCdZn}_2\text{F}_7$ from the total specific heat [21,62]. An Oxford Instruments Triton dilution refrigerator (DR) was employed for zero-field and in-field heat capacity measurements between $T = 150$ and 650 mK.

ZF- μSR and LF- μSR measurements were performed at the ISIS Neutron and Muon Source at the Rutherford Appleton Laboratory in Harwell, Oxford, United Kingdom [70]. The MuSR instrument was used, utilizing a dilution refrigerator cryostat setup for temperatures between $T = 0.05$ and 4 K, allowing applied longitudinal fields between $\mu_0 H_{\text{LF}} = 0$ and 3000 G. The obtained spectra were analyzed using WiMDA [71] and Mantid [72].

NMR measurements were performed using a phase-coherent pulsed spectrometer with a superconducting NMR magnet. A ^4He variable temperature insert was utilized between $T = 1.8$ and 150 K, and a ^3He - ^4He dilution refrigerator probe head was used between $T = 0.12$ and 2.1 K. A custom-wound excitation coil from silver wire was used. Frequency-swept spectra at different constant fields were collected, with the radio-frequency (rf) circuit tuned and matched at each point. Nuclear spin echoes were generated using a standard 90° – 180° pulse sequence, with a first-pulse duration of 2–3 μs , where the rf power for nuclear spin excitation was optimized at each NMR spectral peak. The separation τ between the first and second pulses was typically 20–30 μs . Echoes were accumulated and Fourier transformed to obtain the spectra.

Details of the synthesis process, extended measurement details, and additional results of powder and single-crystal x-ray diffraction, magnetization, ac susceptibility, heat capacity, ^{23}Na NMR, and μSR measurements can be found in the Supplemental Material [73].

III. RESULTS

A. Structural details

Structural studies confirmed the expected cubic pyrochlore structure [space group $Fd\bar{3}m$, $a = 10.3038(15)$ Å] for $\text{NaCdCu}_2\text{F}_7$, depicted in Fig. 1(a), agreeing with the previously studied $\text{NaA}''\text{B}_2\text{F}_7$ fluorides [21–23,62–64,74–77]. Structural randomness is inherent due to a mixed occupancy of $\text{Na}^+/\text{Cd}^{2+}$ cations fully disordered on the pyrochlore (16d) A site. Inspection of the reciprocal space map cuts along the $(hk0)$, $(h0l)$, and $(0kl)$ planes reveals no superstructure reflections or obvious diffuse scattering, indicating the absence of short- or long-range occupational ordering of Na^+ and Cd^{2+} (see Fig. S2 in the Supplemental Material [73]). The magnetic Cu^{2+} ions on the pyrochlore (16c) B site are octahedrally coordinated by F(2) fluorine on the 48f site and are surrounded by six $\text{Na}^+/\text{Cd}^{2+}$ ions creating a hexagon in the (111) plane [see Fig. 1(b)]. The difference in ionic radii of Na^+ (1.18 Å) and Cd^{2+} (1.10 Å) [78], in combination with

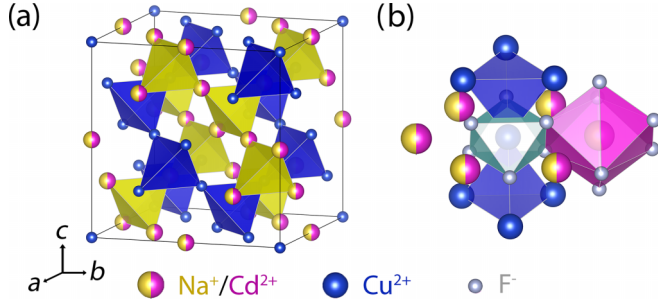


FIG. 1. (a) Cubic unit cell of $\text{NaCdCu}_2\text{F}_7$ with highlighted magnetic (blue) and nonmagnetic (yellow) pyrochlore sublattices. Fluorine ions are omitted for clarity. (b) Local environment around the magnetic Cu^{2+} site—distorted CuF_6 octahedron (light blue), connected to a distorted $(\text{Na/Cd})\text{F}_8$ cube (pink dodecahedron).

their random occupation of the A site, induces small trigonal distortions of the CuF_6 octahedra along the local $[111]$ direction, which leads to a distribution of $\text{Cu}^{2+}\text{--F--Cu}^{2+}$ superexchange pathways—intrinsic magnetic bond disorder. The only free coordinate in the pyrochlore $Fd\bar{3}m$ space group, the $\text{F}(2)$ fluorine x -coordinate $x_{\text{F}(2)}$, dictates the size of the distortion of the CuF_6 octahedron as well as the $(\text{Na/Cd})\text{F}_8$ dodecahedron [21]. The refined value, $x_{\text{F}(2)} = 0.3316(4)$, results in a deformed CuF_6 octahedron with six equal $\text{Cu--F}(2)$ bond lengths $d_{\text{Cu--F}(2)} = 2.0060(17)$ Å, while the $\text{F}(2)\text{--Cu--F}(2)$ angle deviates from the ideal 90° value by $\pm\delta = 7.22(10)^\circ$.

Refinement of the Cd occupancy, constrained with the Na occupancy by $\text{occ}(\text{Na}) + \text{occ}(\text{Cd}) = 1$, leads to a better solution than the ideal stoichiometric $0.5/0.5$ case. A small Cd deficiency of $\text{occ}(\text{Cd}) = 0.462(4)$ was obtained. To preserve charge balance, a refinement of the $\text{F}(2)$ occupancy as a function of the Cd occupancy was attempted in the form of $\text{Na}_{1+x}\text{Cd}_{1-x}\text{Cu}_2\text{F}_{7-x}$, leading to $\text{occ}(\text{F}2) = 0.924(7)$ with slightly better residue values, giving the final refined composition $\text{Na}_{1.076}\text{Cd}_{0.924}\text{Cu}_2\text{F}_{6.924}$. However, this slightly off-stoichiometric structure changes neither the bond lengths (see Fig. S3 in the Supplemental Material [73]) nor the angles defining the CuF_6 and $(\text{Na/Cd})\text{F}_8$ polyhedra, within the 3σ limit.

Atomic positions and thermal displacement parameters are summarized in Tables I and II. Detailed SCXRD structure solution results and additional details of the refinement process can be found in the Supplemental Material [73].

TABLE I. Atomic positions of $\text{Na}_{1.076}\text{Cd}_{0.924}\text{Cu}_2\text{F}_{6.924}$.

Atom	Site	x	y	z	Occupancy	Site symmetry
Na	16d	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	0.538(4)	$\bar{3}m$
Cd	16d	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	0.462(4)	$\bar{3}m$
Cu	16c	0	0	0	1	$\bar{3}m$
F(1)	8b	$\frac{3}{8}$	$\frac{3}{8}$	$\frac{3}{8}$	0.924(7)	$\bar{4}3m$
F(2)	48f	0.3316(4)	$\frac{1}{8}$	$\frac{1}{8}$	1	$2mm$

TABLE II. Anisotropic thermal displacement parameters of $\text{Na}_{1.076}\text{Cd}_{0.924}\text{Cu}_2\text{F}_{6.924}$ at $T = 300$ K.

Atom	$U_{ij} (\times 10^{-4} \text{ \AA}^2)$						U_{ani}
	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}	
Na	258(7)	U_{11}	U_{11}	$-51(3)$	U_{12}	U_{12}	258(4)
Cd	258(7)	U_{11}	U_{11}	$-51(3)$	U_{12}	U_{12}	258(4)
Cu	165(6)	U_{11}	U_{11}	17(3)	$-U_{12}$	$-U_{12}$	165(3)
F(1)	151(18)	U_{11}	U_{11}	0	0	0	151(10)
F(2)	470(20)	585(18)	U_{22}	0	0	60(20)	546(12)

B. Magnetic properties

Temperature-dependent magnetic susceptibility, $\chi(T) = M(T)/H$, shows no magnetic transition down to 1.8 K, seen in Fig. 2(a). Instead, a diverging Curie-like tail is noted below 10 K, a typical sign of paramagnetic “orphan” spins dominating the low-temperature bulk magnetic properties. The orphan-spin response, $n_{\text{orp}} \sim 15\%$, was identified and subtracted by fitting the isothermal magnetization $M(H)$ at

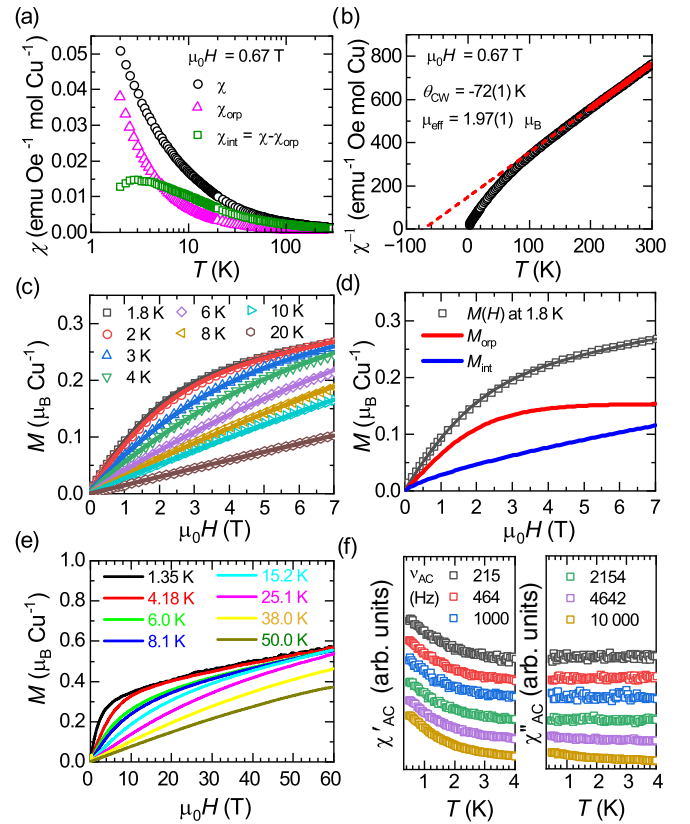


FIG. 2. (a) Magnetic susceptibility $\chi(T)$ at $\mu_0 H = 0.67$ T, decomposed into an orphan-spin contribution χ_{orp} and intrinsic susceptibility $\chi_{\text{int}} = \chi - \chi_{\text{orp}}$. (b) The inverse total susceptibility $\chi^{-1}(T)$, fitted to a Curie-Weiss law between 200 and 300 K. (c) Isothermal magnetization $M(H)$ between 1.8 and 20 K, with fits to Eq. (1) shown as solid lines. (d) Decomposition of the 1.8 K $M(H)$ curve into the fitted orphan spin M_{orp} and intrinsic M_{int} terms. (e) High-field magnetization between 1.35 and 50 K. (f) Real part (left) and imaginary part (right) of ac magnetic susceptibility, vertically offset for clarity.

1.8 K (see detailed analysis below), resulting in the intrinsic susceptibility, $\chi_{\text{int}} = \chi - \chi_{\text{orp}}$, also shown in Fig. 2(a). The χ_{int} displays a broad maximum centered around 3 K. A Curie-Weiss fit of the inverse total susceptibility $\chi^{-1}(T)$ between 200 and 300 K, shown in Fig. 2(b), gives $C = 0.486(1)$ emu K mol⁻¹ Oe⁻¹ and $\theta_{\text{CW}} = -72(1)$ K, resulting in a $\mu_{\text{eff}} = 1.97(1)\mu_B$ [79]. The large negative θ_{CW} confirms dominant AF interactions, and using $\theta_{\text{CW}} = 2zS(S+1)\mathcal{J}/3$ with $z = 6$ the nearest-neighbor coordination number of the pyrochlore lattice, we estimate the exchange coupling constant $\mathcal{J} = -24$ K. χ_{int} qualitatively follows recent theoretical predictions, although the observed broad maximum at $T \approx 0.12\mathcal{J}$ is notably lower in temperature than that predicted by numerical linked cluster expansion results for the disorder-free system, at $T = 0.54\mathcal{J}$ [11]. The moment size of $\mu_{\text{eff}} = 1.97(1)\mu_B$ is consistent with other Cu²⁺ systems [47,48,54], and close to the theoretical spin-only moment $\mu_{\text{eff}}^{\text{theo}} = g\sqrt{J(J+1)}\mu_B = 1.73\mu_B$, for $J = S = \frac{1}{2}$ and $g = 2$ with quenched orbital momentum.

Isothermal magnetization $M(H)$, seen in Fig. 2(c), shows a linear AF response at 20 K and begins to show a Brillouin-like curvature for $T \leq 10$ K at fields below 5 T, superimposed on a linear response visible above 5 T. The $M(H)$ data are well fitted by a superposition of a modified $S = \frac{1}{2}$ Brillouin function corresponding to weakly interacting orphan spins $M_{\text{orp}} = n_{\text{orp}}B_{J=1/2}(x')$ and an intrinsic power-law term $M_{\text{int}} = A(\mu_0 H)^\alpha$ [56,59,61]:

$$M(H) = \frac{n_{\text{orp}}g\mu_B}{2} \tanh\left(\frac{g\mu_B\mu_0 H}{2k_B(T - T^*)}\right) + A(\mu_0 H)^\alpha, \quad (1)$$

where n_{orp} is the orphan-spins fraction, g is the orphan-spin g factor fixed to $g = 2$, T^* is the interaction temperature between the orphan spins, and α is the power-law exponent. From the $M(H)$ fit at 1.8 K, seen in Fig. 2(d), we extract $n_{\text{orp}} = 0.154(2)$, $T^* = 0.30(2)$ K, and $\alpha = 0.73(1)$, corresponding to $\sim 15\%$ of weakly ferromagnetically coupled orphan spins. This n_{orp} value was used in the $\chi_{\text{int}} = \chi - \chi_{\text{orp}}$ subtraction in Fig. 2(a). From the 1.8 K $M(H)$ decomposition into M_{orp} and M_{int} shown in Fig. 2(d), we observe a full saturation of the orphan-spin component by $\mu_0 H \sim 5$ T. The sublinear behavior $\alpha < 1$ of the intrinsic AF response comes about as a result of the disorder-induced distribution of exchange energies. With this orphan-spin fraction n_{orp} and the interaction temperature T^* fixed to that from the lowest-temperature dataset where it is best defined, we performed a global fit for $M(H)$ between 2 and 20 K. The fits are shown as solid lines in Fig. 2(c). The power-law exponent α varied between 0.68 and 0.99, indicating a crossover from Brillouin-like to purely linear ($\alpha \sim 1$) magnetization at high temperature (see Table S2 in the Supplemental Material [73]).

High-field isothermal magnetization $M(H)$ in pulsed field up to 60 T, seen in Fig. 2(e) at temperatures between 1.35 and 50 K, exhibits no plateaus or field-induced transitions predicted for an ideal, disorder-free $S = \frac{1}{2}$ Heisenberg pyrochlore antiferromagnet [80,81]. Instead, a rapid low-field ($\mu_0 H < 5$ T) polarization of weakly coupled orphan spins is observed at the lowest temperatures, followed by a slow linear polarization of dynamically fluctuating strongly coupled singlets up to 60 T. This behavior highlights the broad distribution of

exchange couplings expected from the RS state and is consistent with previously studied RS materials [45,59,61]. Even at 60 T we reach only $\sim 0.6\mu_B/\text{Cu}$ at 1.35 K, remaining far below full saturation expected for a spin-only Cu²⁺ ion ($\mu_s^{\text{theo}} = 2S\mu_B = 1\mu_B$), underscoring the robustness of strongly bound singlets and persistent fluctuations.

No bifurcation of zero-field-cooled, χ_{ZFC} , and field-cooled, χ_{FC} , magnetic susceptibility measurements was observed down to 1.8 K, indicating no history dependence, in accordance with a dynamic state (see Fig. S5 in the Supplemental Material [73]). AC susceptibility, measured with an alternating field amplitude $H_{\text{AC}} = 0.05$ Oe at frequencies between 215 and 10 000 Hz, shows an increase with decreasing temperature without reaching a maximum in the real part, $\chi'_{\text{AC}}(\nu, T)$, while the imaginary part $\chi''_{\text{AC}}(\nu, T)$ remains flat without signs of energy-dissipative processes [see Fig. 2(f)]. No frequency dependence was seen in either the real or imaginary parts, ruling out any global collective spin freezing into a static spin-glass ground state down to 0.4 K, despite the sizable magnetic bond disorder present in this material. This is in direct contrast to all previously studied bond-disordered NaA''M₂F₇ pyrochlore fluorides (A'' = Cd, Ca, Sr; M = Co, Ni, Mn, Fe), which all reported a glassy ground state and a frequency-dependent peak in $\chi'_{\text{AC}}(\nu, T)$ at $T_f \approx 2\text{--}4$ K [21–23,62–64].

C. Specific heat

Specific heat, $C_p(T)$, in zero field shows no sharp first- or second-order transitions down to 150 mK, ruling out the development of long-range order or global structural changes; however, a broad hump is observed at around 1 K, seen in Figs. 3(a) and 3(b). This feature shifts to higher temperature with applied field, reminiscent of a Schottky anomaly. After subtraction of the phonon contribution by subtracting the specific heat of the nonmagnetic analog NaCdZn₂F₇ [21,62] [black solid line in Figs. 3(a) and 3(b)] from the total C_p , the magnetic specific heat $C_{\text{mag}} = C_p - C_{\text{lat}} \approx C_p[\text{NaCdCu}_2\text{F}_7] - C_p[\text{NaCdZn}_2\text{F}_7]$ and its evolution in applied fields is shown in Fig. 2(c). The magnetic entropy change ΔS_{mag} , calculated by integration of C_{mag}/T and shown in Fig. 3(d), reaches only $\sim 35\%$ of the expected value of $R \ln(2)$, by 100 K. The large missing entropy is a signature of nonmagnetic singlet formation combined with residual dynamics at low temperature.

$C_{\text{mag}}(T, H)$ was fitted using a combination of a two-level Schottky anomaly C_{Sch} associated with weakly coupled $S = \frac{1}{2}$ orphan spins and a power-law term $C_{\text{power law}} = A_{\text{power law}} T^{1-\alpha_{\text{Cmag}}}$ corresponding to the RS phase [40,58,60]:

$$\frac{C_{\text{mag}}}{T} = A_{\text{Sch}} R \frac{\Delta^2}{T^3} \frac{\exp(\frac{\Delta}{T})}{(1 + \exp(\frac{\Delta}{T}))^2} + A_{\text{power law}} T^{-\alpha_{\text{Cmag}}}. \quad (2)$$

The fits to Eq. (2) are summarized in Fig. 3(e), with the individual contributions shown in Fig. 3(f). We find that α_{Cmag} decreases from 0.87(2) in zero field to 0.28(2) at 9 T, and the extracted Schottky gap Δ in zero field is 1.34(5) K, suggesting a nonzero splitting of the Cu²⁺ $S = \frac{1}{2}$ energy levels due to the local internal fields. From the Zeeman formula $\Delta(H) = gS\mu_B\mu_0 H$, we obtain the slope $gS = 1.32(6)$, giving the orphan-spin g factor $g = 2.6(1)$ (see Fig. S8 in the Supplemental Material [73]). The orphan-spins fraction n_{oph}

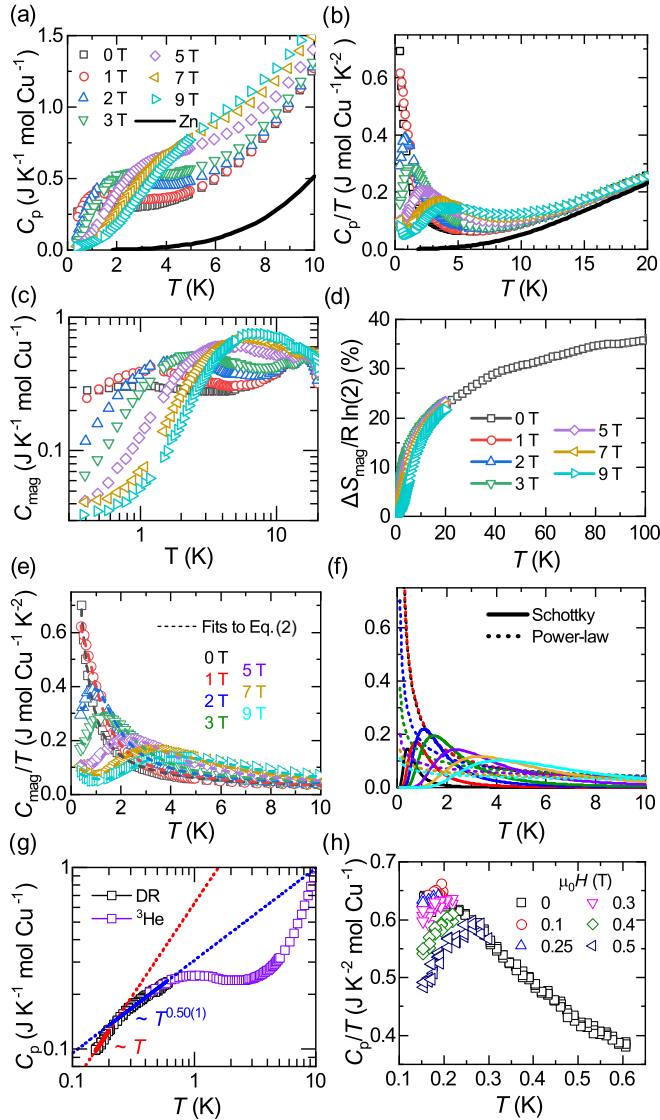


FIG. 3. (a) Specific heat C_p and (b) C_p/T of $\text{NaCdCu}_2\text{F}_7$ in fields of 0–9 T, with the nonmagnetic analog $\text{NaCdZn}_2\text{F}_7$ shown as a black solid line. (c) Magnetic specific heat C_{mag} after subtraction of phonon contribution. (d) Magnetic entropy change ΔS_{mag} in % of $R \ln(2)$. (e) Fits of C_{mag}/T to Eq. (2), with (f) a decomposition into the Schottky and power-law terms. (g) Zero-field DR and ^3He specific heat C_p with a power-law ($T = 200$ – 650 mK) and a linear ($T < 200$ mK) regime. (h) DR C_p/T between 0 and 0.5 T.

can be estimated from the ratio of the Schottky entropy and total magnetic entropy $n_{\text{oph}} = \Delta S_{\text{Sch}}/\Delta S_{\text{mag}}$, yielding an apparent increase from $\sim 10\%$ in zero-field, to $\sim 54\%$ at 9 T (see Table S4 in the Supplemental Material [73]). This increase does not reflect the creation of new orphan spins. Rather, it arises from the field-induced polarization of weakly bound random singlet pairs, which contribute to the low-temperature Schottky-like feature only once the applied field overcomes their singlet binding energy. In zero field these pairs remain largely inactive, so the intrinsic orphan-spin density is field independent and best represented by the zero-field value.

At temperatures between 200 and 650 mK, the specific heat shows a power-law dependence $C_p \sim T^{1-\alpha_{C_p}}$ with

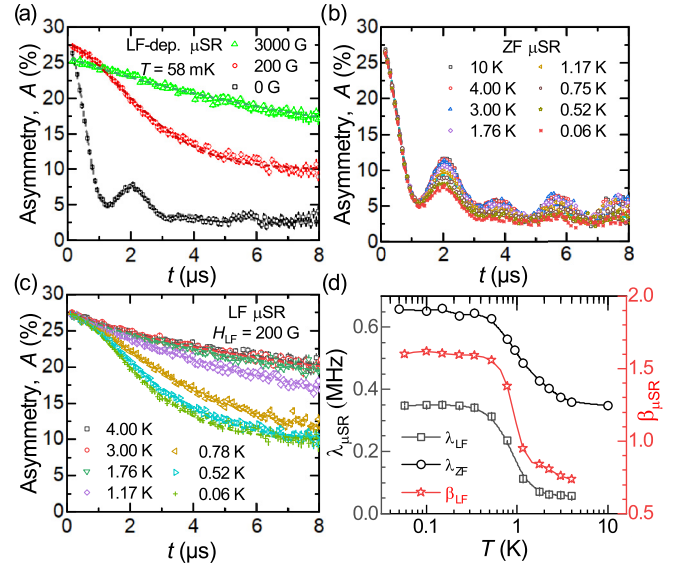


FIG. 4. (a) LF dependence of the $T = 58$ mK spectra, showing the ZF F– μ –F oscillations (black), stretched exponential relaxation of asymmetry at 200 G (red), and a purely exponential relaxation at 3000 G (green). Temperature dependence of (b) the ZF spectra, fitted to Eq. (3), and (c) the LF 200 G spectra, fitted to Eq. (5). (d) Temperature dependence of the fitted ZF and LF muon relaxation rates λ_{LF} and λ_{ZF} (left axis), and the stretching exponent β_{LF} (right axis).

$\alpha_{C_p} = 0.50(1)$, followed by a T -linear specific heat $C_p = \gamma T$ below 200 mK, with $\gamma = 0.64(2)$ J K $^{-2}$ mol Cu $^{-1}$ [see Fig. 3(g)]. The crossover to the linear regime at the lowest temperatures is a characteristic thermodynamic signature of the RS state [27–32]. In close analogy to the Sommerfeld coefficient in metals, the linear specific heat coefficient γ in QSL/RS Mott insulators is proportional to the density of low-energy magnetic excitations in the zero-energy limit, consistent with a gapless excitation spectrum. In Fig. 3(h), we observe a constant C_p/T below 200 mK in zero field corresponding to the $C_p/T = \gamma$ regime, and an emerging sharp peak in applied 0.1–0.5 T fields, which shifts to higher temperature with increasing field. The emergence of a sharp low-temperature peak in C_p/T under small applied magnetic fields may indicate the development of short-range correlations involving orphan spins, stabilized by the suppression of low-energy random singlet fluctuations.

D. Muon spin relaxation

Spin dynamics were characterized down to 58 mK using local-probe μSR experiments. Zero-field measurements revealed oscillations coming from the F^- – μ^+ – F^- bound complex, commonly seen in fluoride materials [82–84]. The base-temperature ZF muon relaxation can be seen in Fig. 4(a), and the temperature dependence of ZF- μSR is shown Fig. 4(b). The ZF spectra were fitted using a simple exponential function multiplied by a modified, phase-shifted ZF F– μ –F function $G_{\text{F-}\mu\text{-F}}(t)$:

$$A_{\text{ZF}}(t) = A_0 G_{\text{F-}\mu\text{-F}}(t) \exp(-\lambda_{\text{ZF}} t) + A_{\text{base}}, \quad (3)$$

where A_0 is the relaxing amplitude, λ_{ZF} is the zero-field muon relaxation rate, A_{base} is a nonrelaxing constant baseline, and the ZF $F-\mu-F$ function $G_{F-\mu-F}$ has the form [82]

$$G_{F-\mu-F}(t) = \frac{1}{6} \left[3 + \cos(\sqrt{3} \omega_d t + \phi) + \left(1 - \frac{1}{\sqrt{3}}\right) \cos\left(\frac{3 - \sqrt{3}}{2} \omega_d t + \phi\right) + \left(1 + \frac{1}{\sqrt{3}}\right) \cos\left(\frac{3 + \sqrt{3}}{2} \omega_d t + \phi\right) \right]. \quad (4)$$

Here, ϕ is the phase shift and ω_d is the dipolar interaction frequency. On cooling from 10 K in zero field, the $F^--\mu^+-F^-$ oscillations are damped by electronic fluctuations slowing into the time window of the muon. This exponential damping increases significantly below ~ 3 K, indicating a rapid slowing of the spin fluctuation rate, but plateaus below ~ 0.7 K—a hallmark of continued quantum fluctuations [85,86] [see Fig. 4(d)]. This plateau is also seen in measurements in an applied 200 G longitudinal field, used to decouple the muons from the static dipolar fields of the $F^--\mu^+-F^-$ state, seen in Fig. 4(d). However, the cleaner spectra, shown in Fig. 4(c), highlight a decay of asymmetry at low temperatures that is not purely exponential, but instead show a Gaussian shoulder at $t < 3$ μs , and are better fitted by a stretched exponential function

$$A_{\text{LF}}(t) = A_0 \exp(-(\lambda_{\text{LF}} t)^{\beta_{\text{LF}}}) + A_{\text{base}}, \quad (5)$$

where A_0 is the relaxing amplitude, λ_{LF} is the LF muon relaxation rate, β_{LF} is the stretching exponent, and A_{base} is a nonrelaxing constant baseline. The stretched exponential relaxation indicates an inhomogeneous mixture of both dynamic and partially frozen local fields. This can be explained by a slowing of dynamics of the weakly interacting orphan spins below 1 K within the RS matrix [61,87,88]. The inhomogeneous distribution of local fields is tracked using the stretching exponent, β_{LF} , in Fig. 4(d). At higher longitudinal fields, these weakly interacting frozen orphan spins are fully polarized and the spectra return to a purely exponential relaxation, shown in Fig. 4(a), confirming continued spin dynamics even at 58 mK.

E. Nuclear magnetic resonance

To complement the μSR results, ^{23}Na NMR measurements were performed between 120 mK and 150 K in fields of 0.67 and 3 T. ^{23}Na occupies the axially symmetric A site (point group $\bar{3}m$), where it is statistically distributed with Cd^{2+} with equal probability. As shown in Figs. 5(a) and 5(b), the spectra exhibit a single broad peak with broad tails on both sides. The spectral shape likely reflects a broadened $I = \frac{3}{2}$ powder pattern, in which the Na/Cd site distribution merges the central and quadrupolar satellite transitions. In the related compound $\text{NaCaCo}_2\text{F}_7$, the ^{23}Na nuclear quadrupole frequency was reported to be on the order of $\nu_Q(^{23}\text{Na}) \sim 1$ MHz [89]. At every temperature, the nuclear spin-lattice relaxation rate $1/T_1$ was measured at the position of the resonance peak by fitting the

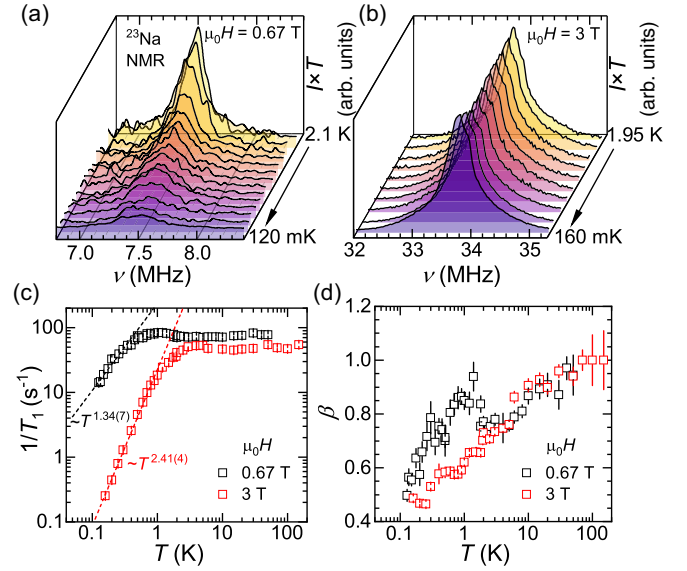


FIG. 5. Temperature dependence of the ^{23}Na NMR spectral intensity scaled by temperature $I \times T$ (a) at $\mu_0 H = 0.67$ T and (b) at $\mu_0 H = 3$ T. Temperature dependence of (c) the ^{23}Na spin-lattice relaxation rate $1/T_1$ and (d) the stretching exponent β_{NMR} .

recovery curves with a stretched exponential function:

$$\frac{M(\infty) - M(t)}{M(\infty)} = 0.4 \exp\{-(t/T_1)^{\beta_{\text{NMR}}}\} + 0.6 \exp\{-(6t/T_1)^{\beta_{\text{NMR}}}\}, \quad (6)$$

assuming that only the central transition was saturated while the populations of satellite transitions remain unaffected. The temperature dependence of the extracted $1/T_1$ and β_{NMR} values is summarized in Figs. 5(c) and 5(d).

The stretching exponent β_{NMR} relates to the spatial distribution of T_1 , with $\beta_{\text{NMR}} \approx 1$ above 100 K implying uniform relaxation and a decreasing $\beta_{\text{NMR}} < 1$ below 100 K signaling inhomogeneous dynamics. At 0.67 T, both β_{NMR} and $1/T_1$ exhibit a broad ~ 1 K hump [seen in Figs. 5(c) and 5(d)], consistent with the broad anomaly in C_p and the increases of $\lambda_{\mu\text{SR}}$ and $\beta_{\mu\text{SR}}$ near 1 K. Moreover, for 0.67 T the spectral intensity multiplied by temperature $I \times T$ drops below ~ 1 K without a shape change [seen in Fig. 5(a)], pointing to an inhomogeneous distribution of static internal fields as also seen by μSR . The intensity loss likely reflects a subset of ^{23}Na sites experiencing static fields that broaden or shift the resonance, leaving the low- T signal from ^{23}Na near Cu moments that remain dynamic. In contrast, at 3 T the $I \times T$ is nearly temperature independent because the orphan spins that were responsible for the static-field broadening at 0.67 T are now mostly polarized, as can be inferred from the orphan-spin Brillouin function in Fig. 2(d). Correspondingly, the ~ 1 K anomaly in β_{NMR} is suppressed at 3 T, and the $1/T_1$ hump is smoothed and shifts to higher temperature.

F. Scaling behavior

We now turn to the scaling behavior and data collapse of measured physical quantities $\chi(T)$, C_{mag}/T , and $^{23}(1/T_1 T)$

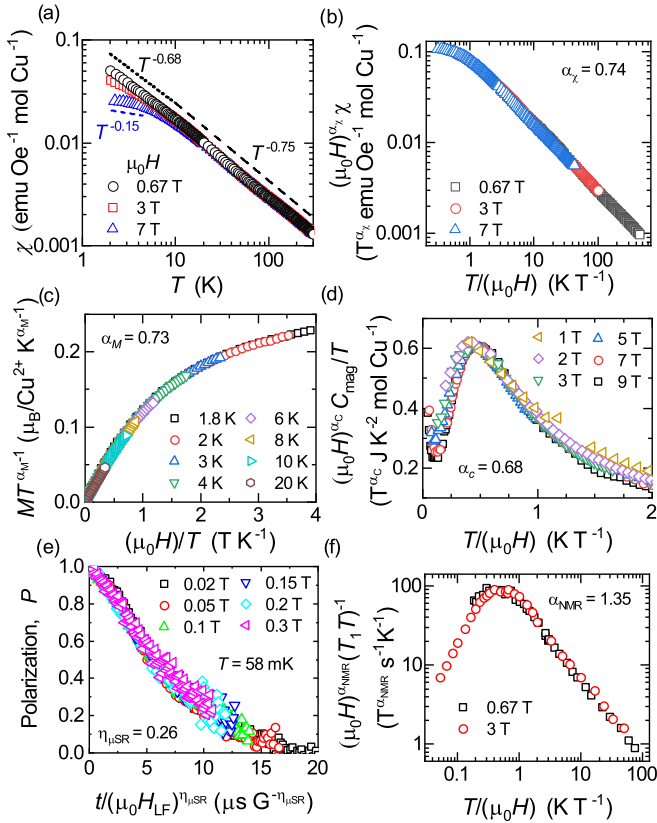


FIG. 6. (a) Power-law scaling of dc magnetic susceptibility $\chi(T) \sim T^{-\alpha_\chi}$ at 0.67, 3, and 7 T fields, with (b) a data collapse of $(\mu_0 H)^{\alpha_\chi} \chi$ vs $T/(\mu_0 H)$ for $\alpha_\chi = 0.74$. Data collapse of (c) isothermal magnetization scaled by temperature MT^{α_M-1} vs $(\mu_0 H)/T$ for $\alpha_M = 0.73$, (d) magnetic specific heat scaled by field and temperature $(\mu_0 H)^{\alpha_C} C_{\text{mag}}/T$ vs $T/(\mu_0 H)$ for $\alpha_C = 0.68$, (e) muon polarization $P(t)$ as a function of $t/(\mu_0 H_{\text{LF}})^{\eta_{\mu\text{SR}}}$ for $\eta_{\mu\text{SR}} = 1 - \alpha_{\mu\text{SR}} = 0.26$, and (f) the spin-lattice relaxation rate scaled by field and temperature $(\mu_0 H)^{\alpha_{\text{NMR}}} (T_1 T)^{-1}$ vs $T/(\mu_0 H)$ for $\alpha_{\text{NMR}} = 1.35$.

in $T/(\mu_0 H)$, as well as $M(H)$ in $(\mu_0 H)/T$ and $P(t)$ in $t/(\mu_0 H)^{\eta_{\mu\text{SR}}}$. This behavior provides the smoking gun evidence for the RS ground state in NaCdCu₂F₇, in agreement with the scaling theory for $S = \frac{1}{2}$ frustrated quantum magnets with quenched disorder [40–42].

In Fig. 6(a), we show the temperature-dependent dc susceptibility in multiple fields, on a log-log scale. $\chi(T)$ exhibits two distinct power-law regimes as $\chi(T) \sim T^{-\alpha_\chi}$: a field-independent regime above 10 K with $\alpha_\chi = 0.75(1)$, as well as a field-dependent regime below 10 K with varying $\alpha_\chi = 0.15(1)–0.68(1)$, decreasing with applied field as $\chi(T)$ flattens upon polarization of the orphan spins. In addition, the susceptibility data in different fields collapse onto a single curve after scaling by field $(\mu_0 H)^{\alpha_\chi} \chi(T)$ as a function of $T/(\mu_0 H)$, with $\alpha_\chi = 0.74$ [see Fig. 6(b)]. Figure 6(c) shows the data collapse of isothermal magnetization at temperatures between 1.8 and 20 K, onto a single curve after scaling by temperature MT^{α_M-1} as a function of $\mu_0 H/T$ [40]. This displays a perfect data collapse for $\alpha_M = 0.73$, in agreement with the fitted exponent in the power-law term in Eq. (1). Further-

more, in Fig. 6(d), we observe a data collapse of magnetic specific heat scaled by field $(\mu_0 H)^{\alpha_C} C_{\text{mag}}/T$ as a function of $T/(\mu_0 H)$ for $\alpha_C = 0.68$. Strikingly, the power-law scaling also applies to the muon relaxation rate $\lambda_{\text{LF}} \sim (\mu_0 H_{\text{LF}})^{-\eta_{\mu\text{SR}}}$ acquired from fitting the field-dependent spectra between 200 and 3000 G by a stretched exponential function (see Figs. S9 and S10 in the Supplemental Material [73]). Here, $\eta_{\mu\text{SR}} = 1 - \alpha_{\mu\text{SR}} = 0.26(2)$, giving $\alpha_{\mu\text{SR}} = 0.74(2)$. This leads to a data collapse of the field-dependent muon polarization spectra $P(t) = (A_{\text{LF}}(t) - A_{\text{base}})/A_0$ when plotted as a function of $t/(\mu_0 H)^{\eta_{\mu\text{SR}}}$ [see Fig. 6(e)]. The observed power-law behavior is a result of a power-law decaying spin-spin correlation function $S(t) \sim t^{-(1-\alpha)}$ governed by a power-law spectral density $S(\omega) \sim \omega^{-\alpha}$ [90], which has been reported in several other QSL candidates with spin chain, pyrochlore, kagome, and triangular lattices [86,90–93]. The power-law scaling is also evident in the ²³Na NMR as $1/T_1 \sim T^{\eta_{\text{NMR}}}$, exhibiting $\eta_{\text{NMR}} = 1.34(7)$ at 0.67 T below 400 mK and $\eta_{\text{NMR}} = 2.41(4)$ at 3 T below 1 K [see Fig. 5(c)], consistent with other RS and QSL candidates [50,94]. This leads to a data collapse of $^{23}(1/T_1 T)$ scaled by field $(\mu_0 H)^{\alpha_{\text{NMR}}} (1/T_1 T)$ as a function of $T/(\mu_0 H)$ for $\alpha_{\text{NMR}} = 1.35$ [see Fig. 6(f)]. We note that the exponent $\alpha_{\text{NMR}} = 1.35$ is approximately double in value compared to α_χ , α_M , α_C , or $\alpha_{\mu\text{SR}}$. While the bulk probes $\chi(T)$, $M(H)$, or C_{mag}/T reflect the static density of low-energy excitations, the quantity $(1/T_1 T) \sim \sum_{\mathbf{q}} |A_{\mathbf{q}}|^2 (\chi''(\mathbf{q}, \omega_0)/\omega_0)_{\omega_0 \rightarrow 0}$ is proportional to the momentum $\mathbf{q}$ -averaged imaginary part of the local spin susceptibility, reflecting the low-frequency spectral weight of spin fluctuations. Although LF- μSR also probes the low-frequency limit of the dynamical susceptibility as $\lambda_{\text{LF}} \sim \sum_{\mathbf{q}} |A_{\mathbf{q}}|^2 (\chi''(\mathbf{q}, \omega_\mu)/\omega_\mu)_{\omega_\mu \rightarrow 0}$, the $\alpha_{\mu\text{SR}} = 0.74(2)$ exponent agrees more with the bulk-probe exponents rather than α_{NMR} . We attribute this to the different frequency windows in which these probes operate: LF- μSR is sensitive to fluctuations in the $\sim 10^5$ – 10^7 Hz range set by the muon gyromagnetic ratio ($\omega_\mu = \gamma_\mu H_{\text{LF}}$), whereas ²³Na NMR probes dynamics at the Larmor frequency at $\sim 10^7$ – 10^8 Hz ($\omega_0 = \gamma_{\text{Na}} H_{\text{LF}}$). In a random singlet system with a broad distribution of excitation energies, the ultra-low-frequency window accessed by LF- μSR emphasizes the same low-energy excitations that dominate bulk thermodynamics, while NMR probes higher-frequency dynamics, resulting in different scaling exponents.

IV. CONCLUSION

In summary, using a plethora of complementary bulk and local-probe techniques, we have shown that the frustrated, magnetic-bond-disordered $S = \frac{1}{2}$ Heisenberg pyrochlore antiferromagnet NaCdCu₂F₇ does not exhibit long-range magnetic order or global spin freezing down to 58 mK. Instead, the observed power-law scaling behavior and data collapse across multiple experimental probes are consistent with a random singlet ground state characterized by a broad distribution of exchange couplings, noted by the coexistence of singlet dimers and weakly coupled orphan spins. This behavior originates from intrinsic Na⁺/Cd²⁺ disorder on the pyrochlore A site, which modifies the Cu²⁺–F[–]–Cu²⁺ superexchange pathways on the magnetic, pyrochlore B-site sublattice. The

absence of spin-glass freezing contrasts with previously studied $\text{NaA}''\text{B}_2\text{F}_7$ pyrochlore fluorides [21–23,62–64], where comparable magnetic bond disorder leads to glassy ground states, suggesting that strong $S = \frac{1}{2}$ quantum fluctuations play an important role in suppressing spin freezing in $\text{NaCdCu}_2\text{F}_7$ and stabilizing a quantum-disordered state.

The present study establishes $\text{NaCdCu}_2\text{F}_7$ as a well-characterized example of disorder-driven quantum magnetism, but several important questions remain. All measurements reported here were performed on polycrystalline samples, and the growth of high-quality single crystals would enable momentum-resolved and anisotropic measurements. Thermal transport experiments would be particularly valuable for probing low-energy excitations and further distinguishing random singlet behavior from other quantum-disordered states. Neutron scattering studies would provide direct access to the dynamic spin structure factor; however, such experiments are presently hindered by the strong neutron absorption of Cd, motivating isotopic enrichment or the investigation of chemically related pyrochlore fluorides such as $\text{NaCaCu}_2\text{F}_7$ or $\text{NaSrCu}_2\text{F}_7$.

Finally, recent theoretical work has shown that entanglement-sensitive quantities such as quantum Fisher information, extracted from experimentally accessible correlation functions [37], may distinguish random singlet states from quantum spin liquids even when thermodynamic signatures are similar. Applying such approaches to $\text{NaCdCu}_2\text{F}_7$ and related materials represents an interesting direction for future experimental and theoretical studies.

ACKNOWLEDGMENTS

We acknowledge funding from Charles University in Prague, within Grant Agency Univerzita Karlova program (GAUK 48924). Crystal growth, structural analysis, and magnetic properties measurements were carried out in the MGML [95] which is supported within the program of Czech Research Infrastructures (Project No. LM2023065). This work was supported by the Ministry of Education, Youth and Sports of the Czech Republic through the INTER-EXCELLENCE II program (LUABA24056), by the Grant Agency of the Czech Republic (26-23051S), by the HLD at HZDR, member of the European Magnetic Field Laboratory (EMFL), and by EPSRC (UK) via its membership to the EMFL (Grant No. EP/N01085X/1). Muon spectroscopy measurements were supported by the ISIS neutron and muon source [70]. H.S. and Y.T. acknowledge support from Japan Society for the Promotion of Science (JSPS) through KAKENHI (JP24KK0062 and JP23K25829). Work at the Japan Atomic Energy Agency (JAEA) was partially supported by the JAEA REIMEI Research Program. The authors additionally thank Gael Bastien for insightful discussions and Martin Misek for performing complementary magnetization measurements.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

- [1] L. Balents, Spin liquids in frustrated magnets, *Nature (London)* **464**, 199 (2010).
- [2] L. Savary and L. Balents, Quantum spin liquids: A review, *Rep. Prog. Phys.* **80**, 016502 (2017).
- [3] Y. Zhou, K. Kanoda, and T.-K. Ng, Quantum spin liquid states, *Rev. Mod. Phys.* **89**, 025003 (2017).
- [4] T. Lancaster, Quantum spin liquids, *Contemp. Phys.* **64**, 127 (2023).
- [5] A. Kitaev, Fault-tolerant quantum computation by anyons, *Ann. Phys.* **303**, 2 (2003).
- [6] C. Nayak, S. H. Simon, A. Stern, M. Freedman, and S. Das Sarma, Non-Abelian anyons and topological quantum computation, *Rev. Mod. Phys.* **80**, 1083 (2008).
- [7] F. Yang, L. Henriet, A. Soret, and K. Le Hur, Engineering quantum spin liquids and many-body Majorana states with a driven superconducting box circuit, *Phys. Rev. B* **98**, 035431 (2018).
- [8] Y. Tokiwa, S. Bachus, K. Kavita, A. Jesche, A. A. Tsirlin, and P. Gegenwart, Frustrated magnet for adiabatic demagnetization cooling to milli-Kelvin temperatures, *Commun. Mater.* **2**, 42 (2021).
- [9] B. Canals and C. Lacroix, Quantum spin liquid: The Heisenberg antiferromagnet on the three-dimensional pyrochlore lattice, *Phys. Rev. B* **61**, 1149 (2000).
- [10] A. B. Harris, A. J. Berlinsky, and C. Bruder, Ordering by quantum fluctuations in a strongly frustrated Heisenberg antiferromagnet, *J. Appl. Phys.* **69**, 5200 (1991).
- [11] R. Schäfer, I. Hagymási, R. Moessner, and D. J. Luitz, Pyrochlore $S = \frac{1}{2}$ Heisenberg antiferromagnet at finite temperature, *Phys. Rev. B* **102**, 054408 (2020).
- [12] I. Hagymási, R. Schäfer, R. Moessner, and D. J. Luitz, Possible inversion symmetry breaking in the $S = \frac{1}{2}$ pyrochlore Heisenberg magnet, *Phys. Rev. Lett.* **126**, 117204 (2021).
- [13] J. H. Kim and J. H. Han, Chiral spin states in the pyrochlore Heisenberg magnet: Fermionic mean-field theory and variational Monte Carlo calculations, *Phys. Rev. B* **78**, 180410(R) (2008).
- [14] H. Tsunetsugu, Spin-singlet order in a pyrochlore antiferromagnet, *Phys. Rev. B* **65**, 024415 (2001).
- [15] N. Astrakhantsev, T. Westerhout, A. Tiwari, K. Choo, A. Chen, M. H. Fischer, G. Carleo, and T. Neupert, Broken-symmetry ground states of the Heisenberg model on the pyrochlore lattice, *Phys. Rev. X* **11**, 041021 (2021).
- [16] B. Schneider, J. C. Halimeh, and M. Punk, Projective symmetry group classification of chiral $\mathbb{Z}_2$ spin liquids on the pyrochlore lattice: Application to the spin- $\frac{1}{2}$ XXZ Heisenberg model, *Phys. Rev. B* **105**, 125122 (2022).
- [17] V. R. Chandra and J. Sahoo, Spin- $\frac{1}{2}$ Heisenberg antiferromagnet on the pyrochlore lattice: An exact diagonalization study, *Phys. Rev. B* **97**, 144407 (2018).
- [18] J. S. Gardner, S. R. Dunsiger, B. D. Gaulin, M. J. P. Gingras, J. E. Greedan, R. F. Kiefl, M. D. Lumsden, W. A. MacFarlane, N. P. Raju, J. E. Sonier, I. Swainson, and Z. Tun,

- Cooperative paramagnetism in the geometrically frustrated pyrochlore antiferromagnet $\text{Tb}_2\text{Ti}_2\text{O}_7$, *Phys. Rev. Lett.* **82**, 1012 (1999).
- [19] S. Nakatsuji, Y. Machida, Y. Maeno, T. Tayama, T. Sakakibara, J. van Duijn, L. Balicas, J. N. Millican, R. T. Macaluso, and J. Y. Chan, Metallic spin-liquid behavior of the geometrically frustrated Kondo lattice $\text{Pr}_2\text{Ir}_2\text{O}_7$, *Phys. Rev. Lett.* **96**, 087204 (2006).
- [20] R. Sibille, E. Lhotel, V. Pomjakushin, C. Baines, T. Fennell, and M. Kenzelmann, Candidate quantum spin liquid in the Ce^{3+} pyrochlore stannate $\text{Ce}_2\text{Sn}_2\text{O}_7$, *Phys. Rev. Lett.* **115**, 097202 (2015).
- [21] A. Kancko, G. Giester, and R. H. Colman, Structural and spin-glass properties of single crystal $J_{\text{eff}} = \frac{1}{2}$ pyrochlore antiferromagnet $\text{NaCdCo}_2\text{F}_7$: Correlating T_f with magnetic-bond-disorder, *Phys. Scr.* **98**, 075947 (2023).
- [22] J. W. Krizan and R. J. Cava, $\text{NaCaCo}_2\text{F}_7$: A single-crystal high-temperature pyrochlore antiferromagnet, *Phys. Rev. B* **89**, 214401 (2014).
- [23] J. W. Krizan and R. J. Cava, $\text{NaSrCo}_2\text{F}_7$, a Co^{2+} pyrochlore antiferromagnet, *J. Phys.: Condens. Matter* **27**, 296002 (2015).
- [24] L. Clark, G. J. Nilsen, E. Kermarrec, G. Ehlers, K. S. Knight, A. Harrison, J. P. Attfield, and B. D. Gaulin, From spin glass to quantum spin liquid ground states in molybdate pyrochlores, *Phys. Rev. Lett.* **113**, 117201 (2014).
- [25] S. Kawabata, Y. Yasui, Y. Kobayashi, and M. Sato, Magnetic behavior of spin- $\frac{1}{2}$ frustrated system $\text{Hg}_2\text{Cu}_2\text{F}_6\text{S}$, *J. Phys. Soc. Jpn.* **76**, 084705 (2007).
- [26] A. P. Ramirez and S. V. Syzranov, Short-range order and hidden energy scale in geometrically frustrated magnets, *Mater. Adv.* **6**, 1213 (2025).
- [27] K. Uematsu and H. Kawamura, Randomness-induced quantum spin liquid behavior in the $S = \frac{1}{2}$ random-bond Heisenberg antiferromagnet on the pyrochlore lattice, *Phys. Rev. Lett.* **123**, 087201 (2019).
- [28] K. Uematsu and H. Kawamura, Randomness-induced quantum spin liquid behavior in the $S = \frac{1}{2}$ random $J_1 - J_2$ Heisenberg antiferromagnet on the honeycomb lattice, *J. Phys. Soc. Jpn.* **86**, 044704 (2017).
- [29] K. Uematsu and H. Kawamura, Randomness-induced quantum spin liquid behavior in the $S = \frac{1}{2}$ random $J_1 - J_2$ Heisenberg antiferromagnet on the square lattice, *Phys. Rev. B* **98**, 134427 (2018).
- [30] K. Uematsu, T. Hikihara, and H. Kawamura, Frustration-induced quantum spin liquid behavior in the $S = \frac{1}{2}$ random-bond Heisenberg antiferromagnet on the zigzag chain, *J. Phys. Soc. Jpn.* **90**, 124703 (2021).
- [31] H. Kawamura, K. Watanabe, and T. Shimokawa, Quantum spin-liquid behavior in the Spin- $\frac{1}{2}$ random-bond Heisenberg antiferromagnet on the kagome lattice, *J. Phys. Soc. Jpn.* **83**, 103704 (2014).
- [32] H. Kawamura and K. Uematsu, Nature of the randomness-induced quantum spin liquids in two dimensions, *J. Phys.: Condens. Matter* **31**, 504003 (2019).
- [33] K. Watanabe, H. Kawamura, H. Nakano, and T. Sakai, Quantum spin-liquid behavior in the spin- $\frac{1}{2}$ random heisenberg antiferromagnet on the triangular lattice, *J. Phys. Soc. Jpn.* **83**, 034714 (2014).
- [34] S. Sanyal, K. Damle, J. T. Chalker, and R. Moessner, Emergent moments and random singlet physics in a Majorana spin liquid, *Phys. Rev. Lett.* **127**, 127201 (2021).
- [35] L. Liu, H. Shao, Y. C. Lin, W. Guo, and A. W. Sandvik, Random-singlet phase in disordered two-dimensional quantum magnets, *Phys. Rev. X* **8**, 041040 (2018).
- [36] L. Savary and L. Balents, Disorder-induced quantum spin liquid in spin ice pyrochlores, *Phys. Rev. Lett.* **118**, 087203 (2017).
- [37] T. Shimokawa, S. Sabharwal, and N. Shannon, Can experimentally-accessible measures of entanglement distinguish quantum spin liquids from disorder-driven “random singlet” phases? [arXiv:2505.11874](https://arxiv.org/abs/2505.11874).
- [38] M. Isoda and S. Mori, Valence-bond crystal and anisotropic excitation spectrum on 3-dimensionally frustrated pyrochlore, *J. Phys. Soc. Jpn.* **67**, 4022 (1998).
- [39] P. Anderson, Resonating valence bonds: A new kind of insulator? *Mater. Res. Bull.* **8**, 153 (1973).
- [40] I. Kimchi, J. P. Sheckelton, T. M. McQueen, and P. A. Lee, Scaling and data collapse from local moments in frustrated disordered quantum spin systems, *Nat. Commun.* **9**, 4367 (2018).
- [41] I. Kimchi, A. Nahum, and T. Senthil, Valence bonds in random quantum magnets: Theory and application to YbMgGaO_4 , *Phys. Rev. X* **8**, 031028 (2018).
- [42] C. Dasgupta and S.-K. Ma, Low-temperature properties of the random Heisenberg antiferromagnetic chain, *Phys. Rev. B* **22**, 1305 (1980).
- [43] D. S. Fisher, Random antiferromagnetic quantum spin chains, *Phys. Rev. B* **50**, 3799 (1994).
- [44] R. N. Bhatt and P. A. Lee, Scaling studies of highly disordered spin- $\frac{1}{2}$ antiferromagnetic systems, *Phys. Rev. Lett.* **48**, 344 (1982).
- [45] P. A. Volkov, C.-J. Won, D. I. Gorbunov, J. Kim, M. Ye, H.-S. Kim, J. H. Pixley, S.-W. Cheong, and G. Blumberg, Random singlet state in $\text{Ba}_5\text{CuIr}_3\text{O}_{12}$ single crystals, *Phys. Rev. B* **101**, 020406(R) (2020).
- [46] T. Shiroka, F. Eggenschwiler, H. R. Ott, and J. Mesot, From order to randomness: Onset and evolution of the random-singlet state in bond-disordered $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ spin-chain compounds, *Phys. Rev. B* **99**, 035116 (2019).
- [47] J. Khatua, M. Gomilšek, J. C. Orain, A. M. Strydom, Z. Jagličić, C. V. Colin, S. Petit, A. Ozarowski, L. Mangin-Thro, K. Sethupathi, M. S. R. Rao, A. Zorko, and P. Khuntia, Signature of a randomness-driven spin-liquid state in a frustrated magnet, *Commun. Phys.* **5**, 99 (2022).
- [48] S. Kundu, A. Hossain, P. K. S., R. Das, M. Baenitz, P. J. Baker, J.-C. Orain, D. C. Joshi, R. Mathieu, P. Mahadevan, S. Pujari, S. Bhattacharjee, A. V. Mahajan, and D. D. Sarma, Signatures of a $S = \frac{1}{2}$ cooperative paramagnet in the diluted triangular lattice of Y_2CuTiO_6 , *Phys. Rev. Lett.* **125**, 117206 (2020).
- [49] K. Kitagawa, T. Takayama, Y. Matsumoto, A. Kato, R. Takano, Y. Kishimoto, S. Bette, R. Dinnebier, G. Jackeli, and H. Takagi, A spin-orbital-entangled quantum liquid on a honeycomb lattice, *Nature (London)* **554**, 341 (2018).
- [50] C. Lee, S. Lee, Y. Choi, C. Wang, H. Luetkens, T. Shiroka, Z. Jang, Y.-G. Yoon, and K.-Y. Choi, Coexistence of random singlets and disordered Kitaev spin liquid in $\text{H}_3\text{LiIr}_2\text{O}_6$, *Phys. Rev. B* **107**, 014424 (2023).
- [51] J. P. Sheckelton, J. R. Neilson, D. G. Soltan, and T. M. McQueen, Possible valence-bond condensation in the frustrated cluster magnet $\text{LiZn}_2\text{Mo}_3\text{O}_8$, *Nat. Mater.* **11**, 493 (2012).

- [52] P. Song, K. Zhu, F. Yang, Y. Wei, L. Zhang, H. Yang, X.-L. Sheng, Y. Qi, J. Ni, S. Li, Y. Li, G. Cao, Z. Y. Meng, W. Li, Y. Shi, and S. Li, Evidence for the random singlet phase in the honeycomb iridate SrIr_2O_6 , *Phys. Rev. B* **103**, L241114 (2021).
- [53] S. Yoon, W. Lee, S. Lee, J. Park, C. H. Lee, Y. S. Choi, S.-H. Do, W.-J. Choi, W.-T. Chen, F. Chou, D. I. Gorbunov, Y. Oshima, A. Ali, Y. Singh, A. Berlie, I. Watanabe, and K.-Y. Choi, Quantum disordered state in the J_1 - J_2 square-lattice antiferromagnet $\text{Sr}_2\text{Cu}(\text{Te}_{0.95}\text{W}_{0.05})\text{O}_6$, *Phys. Rev. Mater.* **5**, 014411 (2021).
- [54] O. Mustonen, S. Vasala, E. Sadrollahi, K. P. Schmidt, C. Baines, H. C. Walker, I. Terasaki, F. J. Litterst, E. Baggio-Saitovitch, and M. Karppinen, Spin-liquid-like state in a spin- $\frac{1}{2}$ square-lattice antiferromagnet perovskite induced by $d^{10} - d^0$ cation mixing, *Nat. Commun.* **9**, 1085 (2018).
- [55] O. H. Mustonen, E. Fogh, J. A. Paddison, L. Mangin-Thro, T. Hansen, H. Y. Playford, M. Diaz-Lopez, P. Babkevich, S. Vasala, M. Karppinen, E. J. Cussen, H. M. Ronnow, and H. C. Walker, Structure, spin correlations, and magnetism of the $S = \frac{1}{2}$ square-lattice antiferromagnet $\text{Sr}_2\text{CuTe}_{1-x}\text{W}_x\text{O}_6$ ($0 \leq x \leq 1$), *Chem. Mater.* **36**, 501 (2024).
- [56] W. Lee, S. Yoon, S. Jeon, Y. Cai, K. Kojima, L. T. Nguyen, R. J. Cava, K.-Y. Choi, and S. Lee, Random singlet-like state in the dimer-based triangular antiferromagnet $\text{Ba}_6\text{Y}_2\text{Rh}_2\text{Ti}_2\text{O}_{17-8}$, *Phys. Rev. Res.* **6**, 023225 (2024).
- [57] S. Pal, K. Kumar, R. Sharma, A. Banerjee, S. B. Roy, J.-G. Park, A. K. Nigam, and S.-W. Cheong, Possible glass-like random singlet magnetic state in $1\text{T} - \text{TaS}_2$, *J. Phys.: Condens. Matter* **32**, 035601 (2020).
- [58] H. Murayama, T. Tominaga, T. Asaba, A. d. O. Silva, Y. Sato, H. Suzuki, Y. Ukai, S. Suetsugu, Y. Kasahara, R. Okuma, I. Kimchi, and Y. Matsuda, Universal scaling of specific heat in the $S = \frac{1}{2}$ quantum kagome antiferromagnet herbertsmithite, *Phys. Rev. B* **106**, 174406 (2022).
- [59] S. H. Do, J. Van Tol, H. D. Zhou, and K. Y. Choi, Dynamical spin-orbital correlations versus random singlets in $\text{Ba}_3\text{CuSb}_2\text{O}_9$ investigated by magnetization and electron spin resonance, *Phys. Rev. B* **90**, 104426 (2014).
- [60] S. M. Hossain, S. S. Rahaman, H. Gujrati, D. Bhoi, A. Matsuo, K. Kindo, M. Kumar, and M. Majumder, Evidence of random spin-singlet state in the three-dimensional quantum spin liquid candidate $\text{Sr}_3\text{CuNb}_2\text{O}_9$, *Phys. Rev. B* **110**, L020406 (2024).
- [61] B. Sana, M. Barik, S. Lee, U. Jena, M. Baenitz, J. Sichelschmidt, S. Luther, H. Kühne, K. Sethupathi, M. S. Rao, K. Y. Choi, and P. Khuntia, Possible realization of a randomness-driven quantum disordered state in the $S = \frac{1}{2}$ antiferromagnet $\text{Sr}_3\text{CuTa}_2\text{O}_9$, *Phys. Rev. B* **110**, 134412 (2024).
- [62] A. Kancko, C. A. Corrêa, and R. H. Colman, Glassy disordered ground states in the frustrated pyrochlore and fluorite antiferromagnets NaCdM_2F_7 ($M = \text{Ni}^{2+}, \text{Mn}^{2+}$), *J. Alloys Compd.* **1024**, 180142 (2025).
- [63] J. W. Krizan and R. J. Cava, $\text{NaCaNi}_2\text{F}_7$: A frustrated high temperature pyrochlore antiferromagnet with $S = 1\text{Ni}^{2+}$, *Phys. Rev. B* **92**, 014406 (2015).
- [64] M. B. Sanders, J. W. Krizan, K. W. Plumb, T. M. McQueen, and R. J. Cava, $\text{NaSrMn}_2\text{F}_7$, $\text{NaCaFe}_2\text{F}_7$, and $\text{NaSrFe}_2\text{F}_7$: Novel single crystal pyrochlore antiferromagnets, *J. Phys.: Condens. Matter* **29**, 045801 (2017).
- [65] A. A. Coelho, TOPAS and TOPAS-Academic: An optimization program integrating computer algebra and crystallographic objects written in C++, *J. Appl. Crystallogr.* **51**, 210 (2018).
- [66] R. C. Oxford UK, CrysAlisPro, Oxford Diffraction, Agilent Technologies UK Ltd, England (2022), <https://rigaku.com/products/crystallography/x-ray-diffraction/crystalispro>.
- [67] L. Palatinus and G. Chapuis, SUPERFLIP—A computer program for the solution of crystal structures by charge flipping in arbitrary dimensions, *J. Appl. Crystallogr.* **40**, 786 (2007).
- [68] V. Petříček, L. Palatinus, J. Plášil, and M. Dušek, Jana2020—A new version of the crystallographic computing system Jana, *Z. Kristallogr.-Cryst. Mater.* **238**, 271 (2023).
- [69] K. Momma and F. Izumi, VESTA: A three-dimensional visualization system for electronic and structural analysis, *J. Appl. Crystallogr.* **41**, 653 (2008).
- [70] R. H. Colman, A. Kancko, and A. Berlie, Probing the spin dynamics of a $3d$ -pyrochlore spin-glass, $\text{NaCdCo}_2\text{F}_7$, and quantum spin liquid candidate, $\text{NaCdCu}_2\text{F}_7$, STFC ISIS Neutron and Muon Source (2024), <https://data.isis.stfc.ac.uk/doi/STUDY/123074305/>.
- [71] F. L. Pratt, WIMDA: A muon data analysis program for the Windows PC, *Physica B* **289-290**, 710 (2000).
- [72] O. Arnold, J. C. Bilheux, J. M. Borreguero, A. Buts, S. I. Campbell, L. Chapon, M. Doucet, N. Draper, R. Ferraz Leal, M. A. Gigg, *et al.*, Mantid—Data analysis and visualization package for neutron scattering and μ SR experiments, *Nucl. Instrum. Methods Phys. Res., Sect. A* **764**, 156 (2014).
- [73] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/rw5c-491w> for details on sample synthesis, x-ray diffraction, magnetization, ac susceptibility, heat capacity, NMR, and μ SR results.
- [74] R. Hänsler and W. Rüdorff, $\text{A}_2\text{B}_7\text{F}_7$ -Verbindungen mit Pyrochlor- und Weberitstruktur, *Z. Naturforsch. B* **25**, 1306 (1970).
- [75] F. Kubel and B. Dundjerski, Synthesis and crystal structure analysis of $\text{NaSrMg}_2\text{F}_7$, a fully fluorinated compound of the pyrochlore family, *Z. Anorg. Allg. Chem.* **627**, 1589 (2001).
- [76] E. A. Oliveira, I. Guedes, A. P. Ayala, J. Y. Gesland, J. Ellena, R. L. Moreira, and M. Grimsditch, Crystal structure and vibrational spectrum of the $\text{NaCaMg}_2\text{F}_7$ pyrochlore, *J. Solid State Chem.* **177**, 2943 (2004).
- [77] A. Grzechnik, R. Kaindl, and K. Frieze, Temperature dependent study of the crystal structure of $\text{NaCdZn}_2\text{F}_7$ pyrochlore, *J. Phys. Chem. Solids* **68**, 382 (2007).
- [78] R. D. Shannon, Revised effective ionic radii and systematic studies of interatomic distances in halides and chalcogenides, *Acta Crystallogr. A* **32**, 751 (1976).
- [79] S. Mugiraneza and A. M. Hallas, Tutorial: A beginner's guide to interpreting magnetic susceptibility data with the Curie-Weiss law, *Commun. Phys.* **5**, 95 (2022).
- [80] S. Pal and S. Lal, Magnetization plateaus of the quantum pyrochlore Heisenberg antiferromagnet, *Phys. Rev. B* **100**, 104421 (2019).
- [81] I. Hagymási, R. Schäfer, R. Moessner, and D. J. Luitz, Magnetization process and ordering of the $S = \frac{1}{2}$ pyrochlore Heisenberg antiferromagnet in a magnetic field, *Phys. Rev. B* **106**, L060411 (2022).
- [82] J. H. Brewer, S. R. Kreitzman, D. R. Noakes, E. J. Ansaldo, D. R. Harshman, and R. Keitel, Observation of muon-fluorine

- “hydrogen bonding” in ionic crystals, *Phys. Rev. B* **33**, 7813 (1986).
- [83] Y. Cai, M. N. Wilson, A. M. Hallas, L. Liu, B. A. Frandsen, S. R. Dunsiger, J. W. Krizan, R. J. Cava, O. Rubel, Y. J. Uemura, and G. M. Luke, μ SR study of spin freezing and persistent spin dynamics in NaCaNi₂F₇, *J. Phys.: Condens. Matter* **30**, 385802 (2018).
- [84] J. M. Wilkinson and S. J. Blundell, Information and decoherence in a muon-fluorine coupled system, *Phys. Rev. Lett.* **125**, 087201 (2020).
- [85] Y.-X. Yang, C.-Y. Jiang, L.-L. Huang, Z.-H. Zhu, C.-S. Chen, Q. Wu, Z.-F. Ding, C. Tan, K.-W. Chen, P. K. Biswas, A. D. Hillier, Y.-G. Shi, C. Liu, L. Wang, F. Ye, J.-W. Mei, and L. Shu, Muon spin relaxation study of spin dynamics on a Kitaev honeycomb material H₃LiIr₂O₆, *npj Quantum Mater.* **9**, 77 (2024).
- [86] Y. Li, D. Adroja, P. K. Biswas, P. J. Baker, Q. Zhang, J. Liu, A. A. Tsirlin, P. Gegenwart, and Q. Zhang, Muon spin relaxation evidence for the U(1) quantum spin-liquid ground state in the triangular antiferromagnet YbMgGaO₄, *Phys. Rev. Lett.* **117**, 097201 (2016).
- [87] Y. J. Uemura, A. Keren, K. Kojima, L. P. Le, G. M. Luke, W. D. Wu, Y. Ajiro, T. Asano, Y. Kuriyama, M. Mekata, H. Kikuchi, and K. Kakurai, Spin fluctuations in frustrated Kagomé lattice system SrCr₈Ga₄O₁₉ studied by muon spin relaxation, *Phys. Rev. Lett.* **73**, 3306 (1994).
- [88] M. A. de Vries, J. O. Piatek, M. Misek, J. S. Lord, H. M. Rønnow, and J.-W. G. Bos, Low-temperature spin dynamics of a valence bond glass in Ba₂ YMoO₆, *New J. Phys.* **15**, 043024 (2013).
- [89] R. Sarkar, J. W. Krizan, F. Brückner, E. C. Andrade, S. Rachel, M. Vojta, R. J. Cava, and H. H. Klauss, Spin freezing in the disordered pyrochlore magnet NaCaCo₂F₇: NMR studies and Monte Carlo simulations, *Phys. Rev. B* **96**, 235117 (2017).
- [90] F. L. Pratt, S. J. Blundell, T. Lancaster, C. Baines, and S. Takagi, Low-temperature spin diffusion in a highly ideal $S = \frac{1}{2}$ Heisenberg antiferromagnetic chain studied by muon spin relaxation, *Phys. Rev. Lett.* **96**, 247203 (2006).
- [91] S. Lee, R. Klauer, J. Menten, W. Lee, S. Yoon, H. Luetkens, P. Lemmens, A. Möller, and K. Y. Choi, Unconventional spin excitations in the $S = \frac{3}{2}$ triangular antiferromagnet RbAg₂Cr[VO₄]₂, *Phys. Rev. B* **101**, 224420 (2020).
- [92] A. Keren, J. S. Gardner, G. Ehlers, A. Fukaya, E. Segal, and Y. J. Uemura, Dynamic properties of a diluted pyrochlore cooperative paramagnet (Tb_{*p*}Y_{1-*p*})₂Ti₂O₇, *Phys. Rev. Lett.* **92**, 107204 (2004).
- [93] E. Kermarrec, A. Zorko, F. Bert, R. H. Colman, B. Koteswararao, F. Bouquet, P. Bonville, A. Hillier, A. Amato, J. Van Tol, A. Ozarowski, A. S. Wills, and P. Mendels, Spin dynamics and disorder effects in the $S = \frac{1}{2}$ kagome Heisenberg spin-liquid phase of kapellasite, *Phys. Rev. B* **90**, 205103 (2014).
- [94] Y. Shimizu, K. Miyagawa, K. Kanoda, M. Maesato, and G. Saito, Emergence of inhomogeneous moments from spin liquid in the triangular-lattice Mott insulator κ -(ET)₂Cu₂(CN)₃, *Phys. Rev. B* **73**, 140407 (2006).
- [95] <https://mgml.eu/>.