This is the accepted manuscript made available via CHORUS. The article has been published as:

**Order out of a Coulomb Phase and Higgs Transition: Frustrated Transverse Interactions of Nd\(_2\)Zr\(_2\)O\(_7\)**

J. Xu, Owen Benton, A. T. M. N. Islam, T. Guidi, G. Ehlers, and B. Lake

Phys. Rev. Lett. **124**, 097203 — Published 6 March 2020

DOI: [10.1103/PhysRevLett.124.097203](https://doi.org/10.1103/PhysRevLett.124.097203)
Order out of a Coulomb phase and Higgs transition: frustrated transverse interactions of Nd$_2$Zr$_2$O$_7$

J. Xu,$^{1,2,*}$ Owen Benton,$^3,†$ A. T. M. N. Islam,$^1$ T. Guidi,$^4$ G. Ehlers,$^5$ and B. Lake$^{1,2,‡}$

$^1$Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Hahn-Meitner-Platz 1, D-14109 Berlin, Germany
$^2$Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstraße 36, D-10623 Berlin, Germany
$^3$RIKEN Center for Emergent Matter Science (CEMS), Wako, Saitama, 351-0198, Japan
$^4$ISIS facility, Rutherford Appleton Laboratory, Didcot, OX11 0QX, UK
$^5$Oak Ridge National Laboratory, Oak Ridge, P.O. Box 2008, Tennessee 37831, USA

(Dated: January 14, 2020)

The pyrochlore material Nd$_2$Zr$_2$O$_7$ with an “all-in-all-out” (AIAO) magnetic order shows novel quantum moment fragmentation with gapped flat dynamical spin ice modes. The parameterized spin Hamiltonian with a dominant frustrated ferromagnetic transverse term reveals a proximity to a U(1) spin liquid. Here we study the magnetic excitations of Nd$_2$Zr$_2$O$_7$ above the ordering temperature ($T_N$) using high-energy-resolution inelastic neutron scattering. We find strong spin ice correlations at zero energy with the disappearance of gapped magnon excitations of the AIAO order. It seems that the gap to the dynamical spin ice closes above $T_N$ and the system enters a quantum spin ice state competing with and suppressing the AIAO order. Classical Monte Carlo, molecular dynamics and quantum boson calculations support the existence of a Coulombic phase above $T_N$. Our findings relate the magnetic ordering of Nd$_2$Zr$_2$O$_7$ with the Higgs mechanism and provide explanations for several previously reported experimental features.

Competing interactions and geometrical frustration support highly degenerate states which suppress conventional magnetic order and lead to novel emergent states [1]. Classical spin ice (CSI) is a prominent example realized in (Dy/Ho)$_2$Ti$_2$O$_7$ pyrochlores consisting of a network of corner-sharing tetrahedra [2–6]. In the CSI, the single-ion Ising anisotropy due to the crystal electric field (CEF) interactions frustrates the ferromagnetic (FM) spin interactions. This creates the “2-in-2-out” (2I2O) ice rule on the spin configuration leading to infinite degeneracy [5]. In contrast, an antiferromagnetic (AFM) interaction is not frustrated resulting in a long-range “all-in-all-out” (AIAO) order [1].

Introducing transverse spin couplings, CSI transforms to quantum spin ice (QSI) allowing quantum tunnelling between the degenerate ice-rule states which realizes a U(1) quantum spin liquid [7–15]. Recently, there has been a tremendous effort aimed at finding materials supporting QSI [16]. Several materials have been examined, e.g., Tb$_2$Ti$_2$O$_7$ [17], Pr$_2$(Zr/Hf)$_2$O$_7$ [18, 19], and Tb$_2$Ti$_2$O$_7$ [20], but the evidence so far is ambiguous, complicated by multi-phase competitions, structural defects and low-lying crystal field levels [21–26].

As a QSI candidate, Nd$_2$Zr$_2$O$_7$ is an ideal material for modelling, having well-isolated CEF ground state Kramers doublet with Ising anisotropic, dipolar-octupolar character as well as a clean, well-ordered crystal structure [27–37]. Although it has an AIAO order below $T_N \sim 0.4$ K [29, 30], it shows remarkably persistent spin dynamics [31], quantum moment fragmentation, gapped dynamical spin ice [32, 35], gapped kagome spin ice in (1,1,1) fields [36] and quantum spin-1/2 chains in (1,1,0) fields [37]. The parameterized anisotropic spin-1/2 XYZ Hamiltonian based on the pseudospin-1/2 operators $\tau^x$, $\tau^y$ and $\tau^z$ describing the CEF doublet (Eq. 1) indicates that the un-frustrated AFM $J_\tau \approx -0.46$ meV induces the AIAO order, although the FM transverse $J_\tau \approx 0.09$ meV is approximately twice as strong as $|J_\tau|$ [27, 32, 33, 35, 36]. This is a result of the frustration for the FM $J_\tau$ term.

According to linear spin wave theory, the gap to the flat spin ice modes closes at $J_\tau/|J_\tau| = 3$ [33] where spin ice configurations and AIAO order have the same energy. Classically, this signals the formation of an extensive ground-state manifold with icelike character for $\tau^x$ and the mixing of these states by quantum fluctuations caused by $J_\tau$ and $J_\tau$ stabilizes a U(1) spin liquid [33]. If there is a gapless Coulomb phase above $T_N$, the ordering transition would be a candidate for a Higgs transition [33]. From the viewpoint of gauge theory, QSI can be regarded as a magnetic Coulomb phase (based on $\tau^z$ for Nd$_2$Zr$_2$O$_7$) with fractionalized bosonic monopole excitations which hosts fluctuating U(1) gauge fields responsible for quantum electrodynamics [13]. QSI can exhibit a long-range magnetic order with respect to the transverse spin components (the AIAO order for $\tau^z$ for Nd$_2$Zr$_2$O$_7$) via a Bose-Einstein condensation of monopoles [13]. This condensation of matter (monopole) fields coupled to gauge fields occurs through the Higgs mechanism which gaps out all soft spin excitations in the ordered state [38, 39].

In this paper, we study the magnetic excitations of Nd$_2$Zr$_2$O$_7$ above $T_N$ using high-energy-resolution inelastic neutron scattering. We find that the gapped magnon excitations disappear and the pinch point pattern characteristic for spin ice correlations is still present but at zero energy which points to a QSI above $T_N$, supporting

* jianhui.xu@helmholtz-berlin.de
† john.benton@riken.jp
‡ bella.lake@helmholtz-berlin.de
The Nd$_2$Zr$_2$O$_7$ single crystal (~2.5 gram) was grown by using an optical floating zone furnace in the Core Laboratory for Quantum Materials in Helmholtz-Zentrum Berlin (HZB) and characterized using X-ray powder diffraction and Laue diffraction [35]. Inelastic neutron scattering experiments were conducted on the direct-geometry time-of-flight (tof) spectrometer CNCS at SNS in Oak Ridge National Lab and on the indirect-geometry tof spectrometer Osiris at the ISIS Neutron Source in Rutherford Appleton Lab [40, 41]. For the CNCS measurement, the sample was mounted on a $^3$He insert and neutrons of incident wavelength 4.98 Å (3.315 meV) were used in the high-flux mode (energy resolution $\sim$ 0.1 meV). Data were collected at 240 mK, 450 mK and 20 K with a 360-degree sample rotation at a step of one degree. The spin dynamics are resolved better in the Osiris experiment with a energy resolution 25 µeV (PG(200) analyser analysing scattered neutrons of energy 1.84 meV). Data were collected at 30 mK, 450 mK and 20 K using a dilution refrigerator. For both experiments, the 20 K data were used as the background. The software packages Dave [42], Mantid [43] and Horace [44] were used for data processing. Some data were averaged based on the symmetry of the scattering plane in order to improve the statistics. The MC simulations and molecular dynamics calculations were performed using the MATLAB package SPINW [45, 46].

Figure 1(a) shows the constant energy slice at 0.025 meV of the CNCS data at 450 mK. It has a well-formed strong pinch point pattern but at a much lower energy than in the ordered state ($\sim$ 0.075 meV) [46]. Fig. 1(b) presents the background-subtracted data measured on Osiris at 30 mK and 450 mK with a much higher energy resolution 25 µeV. Integrating over [-0.02, 0.02] meV, we see clearly a strong scattering arm along the (1, 1, 1) of the pinch point pattern at 450 mK, whereas the data at 30 mK does not show any signal except for the (2, 2, 0) magnetic Bragg peak. Conversely, in the data with integration over [0.06, 0.08] meV, the 450 mK data does not show a clear pattern while the data at 30 mK shows the expected pinch-point spinwave modes from the AIAO order [32, 35]. In Fig. 1(c), the $E - Q$ slices along the (2, 2, L) direction show that the gapped magnon excitations vanish above $T_N$ and strong scattering appears around the elastic line. In addition, a weak continuum at finite energy around (2, 2, 0) at 450 mK is also a new feature.

In Fig. 2, we show the one-dimensional energy cuts through the Osiris data measured at 30 mK, 450 mK and 20 K with integrating the arm of the pinch point pattern along (1, 1, 1) direction and at $Q = (2, 2, 0)$. Comparing with the 20 K background data, we see pronounced gapped inelastic signals at 30 mK as well as the magnetic (2, 2, 0) Bragg peak which nearly disappear at 450 mK [Fig. 2(a) and (c)]. After background subtraction as shown in Fig. 2(b) and (d), we see that most of the spectral weight becomes elastic at 450 mK. Additionally, there is an extended tail on the high energy side of the elastic peak in the cut with $Q = (1, 1, 1)$ at 450 mK [Fig. 2(b)] which could be attributed to gapped excitations. The strong sharp intensity at (2, 2, 0) at finite energy in the 30 mK Osiris data is an instrumental spurion resulted from leakage beyond the elastic channel.
are integrated along $Q_{20K}$ and $450mK-30mK$ [only for (b)] (lower panels). The data $20K$ (upper panels) and subtracted data $30mK-20K$, $450mK-30mK$ define the pinchoff point from $\sim Q$ excitation from short-range AIAO order. This is consistent with the zeros of the axes.

$r_{\alpha}^{i}$ ($\alpha = x, y, z$) is the $\alpha$ component of the pseudospin-1/2 at site $i$ defined in the rotated local frames and $J_{\alpha}$ is the corresponding nearest-neighbor exchange constant [27, 33]. The calculations were done using the exchange interaction parameters given in Ref. [35] with a supercell of $6 \times 6 \times 6$ cubic unit cells (3456 spins) [46]. The simulated specific heat indicates that the system enters the AIAO phase at a theoretical Neel temperature $T_{N} \approx 0.18$ K (lower than the experimental one possibly due to quantum effects stabilizing the order). Above $T_{N}$ at 0.25 K, the calculated neutron scattering structure factor [Fig. 3(a)] shows a pinch point pattern co-existing with broad signals around the AIAO Bragg peak wavevectors $[2, 2, 0]$ and $[1, 1, 3]$, which is quite consistent with the experiment [Fig. 1(a)].

To identify the spin correlations responsible for this novel scattering pattern, we calculated the thermal average of the amplitudes of the pseudospin components, $\langle |r_{\alpha}^{i}| \rangle$ ($\alpha = x, y, z$) and the probability distribution functions (pdfs) of $r_{\alpha}$ and the average over tetrahedra $1/4 \sum_{\alpha} r_{\alpha}^{i}$. Fig. 3(b) shows the temperature dependence of $\langle |r_{\alpha}^{i}| \rangle$. At base temperature, only $\langle |r_{x}^{i}| \rangle$ is significant consistent with the AIAO order. With increasing temperature, $\langle |r_{x}^{i}| \rangle$ decreases while $\langle |r_{y}^{i}| \rangle$ and $\langle |r_{z}^{i}| \rangle$ increase due to enhanced thermal fluctuations and approach $\tau/2 = 0.25$ ($\tau = 1/2$ is the amplitude of pseudospin) as expected for a completely random spin configuration. Remarkably, above $T_{N}^{*}$, $\langle |r_{x}^{i}| \rangle$ is greater than both the thermal average $\tau/2$ and $\langle |r_{y}^{i}| \rangle$, and then decays slowly with increasing temperature exhibiting a maximum around $T_{N}^{*}$. By contrast, $\langle |r_{z}^{i}| \rangle$ decreases quickly and even becomes slightly lower than $\tau/2$, revealing the breakdown of the AIAO correlations.

$\mathcal{H}_{XYZ} = \sum_{(ij)} \left[ J_{x}^{i,j} x_{i}^{z} x_{j}^{z} + J_{y}^{i,j} y_{i}^{z} y_{j}^{z} + J_{z}^{i,j} z_{i}^{z} z_{j}^{z} \right]$, 

where $r_{\alpha}^{i}$ ($\alpha = x, y, z$) is the $\alpha$ component of the pseudospin-1/2 at site $i$ defined in the rotated local frames and $J_{\alpha}$ is the corresponding nearest-neighbor exchange constant [27, 33]. The calculations were done using the exchange interaction parameters given in Ref. [35] with a supercell of $6 \times 6 \times 6$ cubic unit cells (3456 spins) [46]. The simulated specific heat indicates that the system enters the AIAO phase at a theoretical Neel temperature $T_{N} \approx 0.18$ K (lower than the experimental one possibly due to quantum effects stabilizing the order). Above $T_{N}$ at 0.25 K, the calculated neutron scattering structure factor [Fig. 3(a)] shows a pinch point pattern co-existing with broad signals around the AIAO Bragg peak wavevectors $[2, 2, 0]$ and $[1, 1, 3]$, which is quite consistent with the experiment [Fig. 1(a)].

To identify the spin correlations responsible for this novel scattering pattern, we calculated the thermal average of the amplitudes of the pseudospin components, $\langle |r_{\alpha}^{i}| \rangle$ ($\alpha = x, y, z$) and the probability distribution functions (pdfs) of $r_{\alpha}$ and the average over tetrahedra $1/4 \sum_{\alpha} r_{\alpha}^{i}$. Fig. 3(b) shows the temperature dependence of $\langle |r_{\alpha}^{i}| \rangle$. At base temperature, only $\langle |r_{x}^{i}| \rangle$ is significant consistent with the AIAO order. With increasing temperature, $\langle |r_{x}^{i}| \rangle$ decreases while $\langle |r_{y}^{i}| \rangle$ and $\langle |r_{z}^{i}| \rangle$ increase due to enhanced thermal fluctuations and all approach $\tau/2 = 0.25$ ($\tau = 1/2$ is the amplitude of pseudospin) as expected for a completely random spin configuration. Remarkably, above $T_{N}^{*}$, $\langle |r_{x}^{i}| \rangle$ is greater than both the thermal average $\tau/2$ and $\langle |r_{y}^{i}| \rangle$, and then decays slowly with increasing temperature exhibiting a maximum around $T_{N}^{*}$. By contrast, $\langle |r_{z}^{i}| \rangle$ decreases quickly and even becomes slightly lower than $\tau/2$, revealing the breakdown of the AIAO correlations.

$\mathcal{H}_{XYZ} = \sum_{(ij)} \left[ J_{x}^{i,j} x_{i}^{z} x_{j}^{z} + J_{y}^{i,j} y_{i}^{z} y_{j}^{z} + J_{z}^{i,j} z_{i}^{z} z_{j}^{z} \right]$, 

where $r_{\alpha}^{i}$ ($\alpha = x, y, z$) is the $\alpha$ component of the pseudospin-1/2 at site $i$ defined in the rotated local frames and $J_{\alpha}$ is the corresponding nearest-neighbor exchange constant [27, 33]. The calculations were done using the exchange interaction parameters given in Ref. [35] with a supercell of $6 \times 6 \times 6$ cubic unit cells (3456 spins) [46]. The simulated specific heat indicates that the system enters the AIAO phase at a theoretical Neel temperature $T_{N} \approx 0.18$ K (lower than the experimental one possibly due to quantum effects stabilizing the order). Above $T_{N}$ at 0.25 K, the calculated neutron scattering structure factor [Fig. 3(a)] shows a pinch point pattern co-existing with broad signals around the AIAO Bragg peak wavevectors $[2, 2, 0]$ and $[1, 1, 3]$, which is quite consistent with the experiment [Fig. 1(a)].

To identify the spin correlations responsible for this novel scattering pattern, we calculated the thermal average of the amplitudes of the pseudospin components, $\langle |r_{\alpha}^{i}| \rangle$ ($\alpha = x, y, z$) and the probability distribution functions (pdfs) of $r_{\alpha}$ and the average over tetrahedra $1/4 \sum_{\alpha} r_{\alpha}^{i}$. Fig. 3(b) shows the temperature dependence of $\langle |r_{\alpha}^{i}| \rangle$. At base temperature, only $\langle |r_{x}^{i}| \rangle$ is significant consistent with the AIAO order. With increasing temperature, $\langle |r_{x}^{i}| \rangle$ decreases while $\langle |r_{y}^{i}| \rangle$ and $\langle |r_{z}^{i}| \rangle$ increase due to enhanced thermal fluctuations and all approach $\tau/2 = 0.25$ ($\tau = 1/2$ is the amplitude of pseudospin) as expected for a completely random spin configuration. Remarkably, above $T_{N}^{*}$, $\langle |r_{x}^{i}| \rangle$ is greater than both the thermal average $\tau/2$ and $\langle |r_{y}^{i}| \rangle$, and then decays slowly with increasing temperature exhibiting a maximum around $T_{N}^{*}$. By contrast, $\langle |r_{z}^{i}| \rangle$ decreases quickly and even becomes slightly lower than $\tau/2$, revealing the breakdown of the AIAO correlations.

We characterize the correlations of $\tau_{x}$ with the probability distribution functions mentioned above. As shown in Fig. 3(c), at temperatures below $T_{N}^{*}$, pdf($\tau_{x}$) is a Gaussian function centered at zero and its width increases with raising temperature. Above $T_{N}^{*}$, it surprisingly develops...
two maxima at $\pm \tau$ decaying with further increasing temperature, which means that $\tau^x$ points into the tetrahedron for half of the spins and out of the tetrahedron for the other half. Pdf$(1/4 \sum_{\tau^x})$ shows how the in/out $\tau^x$ is distributed on the tetrahedron which is always a Gaussian function at zero (shown in Ref. [46]). The above two statistical quantities indicate that ice-rule correlations appear for $\tau^x$ on the tetrahedron consistent with the FM $J_x$. On the other hand, pdf$(\tau^x)$ and pdf$(1/4 \sum_{\tau^x})$ change from peaks at either $\tau$ or $-\tau$ (depending on the AIAO domain type) to be a broad peak at zero with increasing temperature indicating the loss of the AIAO correlations of $\tau^x$ [46].

Both experiments and classical MC simulations reveal strong spin ice correlations above $T_N$. Assuming the existence of a Coulomb phase with respect to $\tau^x$ above $T_N$, we have calculated the correlations based on the bosonic many-body theory of QSI [46, 47]. As shown in Fig. 4, the calculated dynamical structure factor at 450 mK (integrated over [0, 0.05] meV) exhibits a pinch point pattern with additional broad scattering around $[2, 2, 0]$ and $[1, 1, 3]$ in good agreement with experimental data. Besides the pinch point pattern at zero energy due to the scattering of the Coulomb phase, monopole creation and hopping cause broad scattering around $[2, 2, 0]$ and $[1, 1, 3]$. Neutrons can be scattered by monopoles via two different processes: (i) the incoming neutron flips a spin belonging to an ice-rule tetrahedron creating a pair of monopoles, which gives a continuum at finite energy above a small gap; (ii) at finite temperature where there is a finite density of monopoles already in the system, the incoming neutron can flip a spin belonging to a monopole tetrahedron causing this monopole to hop which gives a continuum of scattering around zero energy. These scattering features are shown in Fig. 4(b). This agrees with our data which exhibits strong broad signals around $[2, 2, 0]$ and $[1, 1, 3]$ at zero energy and a continuum at finite energy. However, the gapped feature expected around $[2, 2, 0]$ and $[1, 1, 3]$ is not clear in the data which could be attributed to possible fast decay of the coherent monopole excitations due to, for example, strong thermal fluctuations. The signal also may be contaminated by the scattering from possible short-range AIAO correlations.

The existence of strong gapless spin-ice correlations for $\tau^x$ above $T_N$ which become gapped flat dynamical spin-ice modes in the $\tau^x$-ordered state below $T_N$ [32, 35] makes the ordering a candidate for a Higgs transition where the emergent gauge field of the Coulomb phase above $T_N$ is gapped by the condensation of emergent gauge charges (monopoles) [39, 48]. Our semiclassical molecular dynamics calculations (Fig. 3(d) and Ref. [46]) show that the gap closes with raising temperature which supports this picture. A Higgs transition was also proposed for pyrochlore Yb$_2$Ti$_2$O$_7$ by demonstrating the first order nature of the ordering transition and the sudden suppression of the intensity of the pinch point pattern below the transition [48]. The gapped and gapless pinch point patterns below and above $T_N$ is further important evidence for a Higgs transition.

Our results provide an explanation for the seemingly contradictory temperature dependences of the intensities of the AIAO Bragg peak and the pinch point pattern in the energy-integrated polarized neutron scattering data reported for Nd$_2$Zr$_2$O$_7$ [32] and very recently for Nd$_2$ScNbO$_7$ [49]. It was shown that with increasing temperature, the magnetic Bragg peak weakens and disappears at $T_N$ while the pinch point intensity maximizes around $T_N$ and persists to much higher temperatures ($\sim 1$ K) for both compounds. This cannot be rationalized if the pinch point scattering is only a feature of the magnons in the ordered state. We argue that at $T_N$ the gapped pinch point pattern is replaced by a pinch point pattern at zero energy due to the Coulomb phase built on $\tau^x$ which has the strongest ice correlations around $T_N$ and gets weaker slowly with increasing temperature, similar to the temperature evolution of $\langle |\tau^x| \rangle$ in the MC simulations [Fig. 3(b)]. The temperature range where the pinch point pattern presents is also comparable with the strength of $J_z$. In addition, the Coulomb phase with strongly correlated spins could induce slow spin dynamics which supports the observed anomalously slow paramagnetic spin dynamics in the muon spin relaxation experiments [31].

Furthermore, it was reported that a spin ice model with frustrated transverse terms exhibits competing phases and nematicity [50, 51]. Our results provide a experimental and theoretical example of an Ising antiferromagnet with frustrated transverse terms which also shows interesting physics. Our further MC simulations [46] show that the ordering temperature is strongly suppressed with increasing $J_z/|J_x|$ which should be constant in the mean field theory and the ordered phase invades the spin ice phase at finite temperature for $J_z/|J_x| > 3$ similar to the phase diagram in Ref. [50] which is surprising because the spin ice phase should be more stable due to the
higher entropy. Further theoretical study is needed.

In summary, we used inelastic neutron scattering, classical MC simulations, semi-classical molecular dynamics simulations and a bosonic theory to show that Nd$_2$Zr$_2$O$_7$ has a non-trivial paramagnetic state with spin–ice correlations, which is possibly a magnetic Coulomb phase, despite an A1IO ordered ground state. We attributed to the dominant frustrated transverse $J_z$ term in the spin Hamiltonian and related the ordering transition to the Higgs mechanism. Our results indicate that the paramagnetic phase of an ordered system may host unconventional spin correlations different from the ground-state order in nature due to competition and frustration among different terms of anisotropic exchange interactions. This expands the field for searching for QSI to ordered systems with frustrated terms in the spin Hamiltonian and makes it interesting to examine several similar QSI candidates with A1IO order, such as Nd$_2$(Hf/Sn/Pb)$_2$O$_7$, Nd$_2$ScNbO$_7$ and Sm$_2$(Ti/Sn)$_2$O$_7$ [49, 52–56].

ACKNOWLEDGMENTS

We thank K. Siemensmeyer, Y.-P. Huang, M. Hermele, S. T. Bramwell and A. T. Boothroyd for helpful discussions on the related theory. This research was supported by the DFG through project B06 of SFB 1143 (project-id 247310070). This research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory. Experiments at the ISIS Neutron and Muon Source were supported by a beamtime allocation RB1810504 from the Science and Technology Facilities Council (DOI: 10.5286/ISIS.E.92924095).

[1] C. Lacroix, P. Mendels, and F. Mila, Introduction to frustrated magnetism: materials, experiments, theory (Springer Science and Business Media, 2011), Vol. 164.
[2] J. S. Gardner, M. J. P. Gingras, and J. E. Greedan, Rev. Mod. Phys. 82, 53 (2010).
[3] T. Fennell, P. P. Deen, A. R. Wildes, K. Schmalzl, D. Prabhakaran, A. T. Boothroyd, R. J. Aldus, D. F. McMorrow, and S. T. Bramwell, Science 326, 415 (2009).
[4] D. J. P. Morris, D. A. Tennant, S. A. Grigera, B. Klemke, C. Castelnovo, R. Moessner, C. Z绅ernasty, M. Meissner, K. C. Rule, J.-U. Hoffmann, K. Kiefer, S. Gerischer, D. Slobinsky, R. S. Perry, Science 326, 411 (2009).
[5] M. J. Harris, S. T. Bramwell, D. F. McMorrow, T. Zeiske, and K. W. Godfrey, Phys. Rev. Lett. 79, 2554 (1997).
[6] C. L. Henley, Phys. Rev. B 71, 014424 (2005).
[7] M. Hermele, Matthew P. A. Fisher, and L. Balents, Phys. Rev. B 69, 064404 (2004).
[8] Nic Shannon, Olga Sikora, Frank Pollmann, Karlo Penc, and Peter Fulde, Phys. Rev. Lett. 108 067204 (2012).
[9] L. Savary and L. Balents, Phys. Rev. Lett. 108, 037202 (2012).
[10] S. Onoda and Y. Tanaka, Phys. Rev. Lett. 105, 047201 (2010).
[11] S. Onoda and Y. Tanaka, Phys. Rev. B 83, 094411 (2011).
[12] O. Benton, O. Sikora and N. Shannon, Phys. Rev. B 86, 075154 (2012).
[13] S. B. Lee, S. Onoda, and L. Balents, Phys Rev B 86, 104412 (2012).
[14] Y. Kato and S. Onoda, Phys. Rev. Lett. 115, 077202 (2015).
[15] C. J. Huang, Y. Deng, Y. Wan and Z. Y. Meng, Phys. Rev. Lett. 120, 167202, (2018).
[16] M. J. P. Gingras and P. A. McClarty, Rep. Prog. Phys. 77, 056501 (2014).
[17] Kate A. Ross, Lucile Savary, Bruce D. Gaulin, and Leon Balents, Phys. Rev. X 1, 021002 (2011).
[18] K. Kimura, S. Nakatsuji, J-J. Wen, C. Broholm, M. B. Stone, E. Nishibori and H. Sawa, Nat. Commu. 4, 1934 (2013).
[19] R. Sibille, N. Gauthier, Han Yan, Monica Ciomaga Hat-

nean, J. Ollivier, B. Winn, U. Filges, G. Balakrishnan, M. Kenzelmann, Nic Shannon and Tom Fennell, Nat. Phys. 14, 711 (2018).
[20] 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, Phys. Rev. Lett. 82, 1012 (1999).
[21] L. D. C. Jaubert, Owen Benton, Jeffrey G. Rau, J. Oitmaa, R. P. Singh, Nic Shannon and Michel J. P. Gingras, Phys. Rev. Lett. 155, 267208 (2015).
[22] Han Yan, Owen Benton, Ludovic Jaubert, and Nic Shannon, Phys. Rev. B 95, 094422 (2017).
[23] N. Martin, P. Bonville, E. Lhotel, S. Guitteny, A. Wildes, C. Decorse, M. Ciomaga Hatanean, G. Balakrishnan, I. Mirebeau, and S. Petit Phys. Rev. X 7, 041028 (2017).
[24] J. J. Wen, S. M. Kooiphayeh, K. A. Ross, B. A. Trump, T. M. McQueen, K. Kimura, S. Nakatsuji, Y. Qiu, D. M. Pajerowski, J. R. D. Copley and C. L. Broholm, Phys. Rev. Lett. 118, 107206 (2017).
[25] Hamid R. Molavian, Michel J. P. Gingras, and Benjamín Canals Phys. Rev. Lett. 98, 157204 (2007).
[26] A. J. Princep, H. C. Walker, D. T. Adroja, D. Prabhakaran, and A. T. Boothroyd, Phys. Rev. B 91, 224430 (2015).
[27] Y.-P. Huang, G. Chen, and M. Hermele, Phys. Rev. Lett. 112, 167203 (2014).
[28] M. C. Hatanean, M. R. Lees, O. A. Petrenko, D. S. Keeble, G. Balakrishnan, M. J. Gutmann, V. V. Klekovkina, and B. Z. Malkin, Phys. Rev. B 91, 174416 (2015).
[29] E. Lhotel, S. Petit, S. Guitteny, O. Florea, M. Ciomaga Hatanean, C. Colin, E. Ressouche, M. R. Lees, and G. Balakrishnan, Phys. Rev. Lett. 115, 197202 (2015).
[30] J. Xu, V. K. Anand, A. K. Bera, M. Frontzek, D. L. Abernathy, N. Casati, K. Siemensmeyer, and B. Lake, Phys. Rev. B 92, 224430 (2015).
[31] J. Xu, C. Balz, C. Baines, H. Luetkens, and B. Lake, Phys. Rev. B 94, 064425 (2016).
[32] S. Petit, E. Lhotel, B. Canals, M. Ciomaga Hatanean, J. Ollivier, H. Mutka, E. Ressouche, A. R. Wildes, M. R. Lees, and G. Balakrishnan, Nat. Phys. 12, 746 (2016).
[33] O. Benton, Phys. Rev. B 94, 104430 (2016).
[34] L. Opherden, J. Hornung, T. Herrmannsdörfer, J. Xu, A. T. M. N. Islam, B. Lake, and J. Wosnitza, Phys. Rev. B 95, 184418 (2017).
[35] J. Xu, Owen Benton, V. K. Anand, A. T. M. N. Islam, T. Guidi, G. Ehlers, E. Feng, Y. Su, A. Sakai, P. Gegenwart, and B. Lake Phys. Rev. B 99, 144420 (2019).
[36] E. Lhotel, S. Petit, M. Ciomaga Hatznean, J. Ollivier, H. Mutka, E. Ressouche, M. R. Lees, and G. Balakrishnan, Nat. Comm. 9, 3786 (2018).
[37] J. Xu, A. T. M. N. Islam, I. N. Glavatskyy, M. Reehuis, J.-U. Hoffmann, and B. Lake, Phys. Rev. B 98, 060408(R) (2018).
[38] E. Fradkin. and S.H. Shenker, Phys. Rev. D. 19, 3682 (1979).
[39] S. Powell, Phys. Rev. B 84, 094437 (2011).
[40] G. Ehlers, A. A. Podlesnyak, and A. I. Kolesnikov, Rev Sci Instrum 87, 093902 (2016).
[41] M. T. F. Telling and K. H. Andersen, Phys. Chem. Chem. Phys. 7, 1255 (2004).
[42] C.M. Brown, J.R.D. Copley, and R.M. Dimeo, J. Res. Natl. Inst. Stan. Technol. 114, 341 (2009).
[43] O. Arnold et al., Nucl. Instrum. Methods Phys. Res. Sect. A 764, 156166 (2014).
[44] R. A. Ewings, A. Buts, M. D. Le, J. van Duijn, I. Bustin-duy, and T. G. Perring, Nuc. Ins. Methods Phys. Res. Sec. A: Accelerators, Spectrometers, Detectors and Associated Equipment, 834, 132 (2016).
[45] S. Toth and B. Lake, J. Phys.: Condens. Matter 27, 166002 (2015).
[46] Supplementary Materials.
[47] Z. Hao, A. G. R. Day and M. J. P. Gingras, Phys. Rev. B 90, 214430 (2014).
[48] L.-J. Chang, S. Onoda, Y. Su, Y.-J. Kao, K.-D. Tsuei, Y. Yasui, K. Kakurai, and M. R. Lees, Nat. Commun. 3, 992 (2012).
[49] C. Mauws, N. Hiebert, M. Rutherford, H. D. Zhou, Q. Huang, M. B. Stone, N. P. Butch, Y. Su, E. S. Choi, Z. Yamani, and C. R. Wiebe, arXiv:1906.10763 [cond-mat.str-el] (2019).
[50] Mathieu Taillefumier, Owen Benton, Han Yan, L. D. C. Jaubert, and Nie Shannon Phys. Rev. X 7, 041057 (2017).
[51] Owen Benton, L. D. C. Jaubert, Rajiv R. P. Singh, Jaan Oitmaa, and Nie Shannon Phys. Rev. Lett. 121, 067201 (2018).
[52] V. K. Anand, A. K. Bera, J. Xu, T. Herrmannsdörfer, C. Ritter, and B. Lake, Phys. Rev. B 92, 184418 (2015).
[53] A. Bertin, P. Dalmas de Réotier, B. Fäk, C. Marin, A. Yaouanc, A. Forget, D. Sheptyakov, B. Frick, C. Ritter, A. Amato, C. Baines, and P. J. C. King, Phys. Rev. B 92, 144423 (2015).
[54] A. M. Hallas, A. M. Arevalo-Lopez, A. Z. Sharma, T. Munsie, J. P. Attfield, C. R. Wiebe, and G. M. Luke, Phys. Rev. B 91, 104417 (2015).
[55] C. Mauws, A. M. Hallas, G. Sala, A. A. Aczel, P. M. Sarte, J. Gaudet, D. Ziat, J. A. Quilliam, J. A. Lussier, M. Bieringer, H. D. Zhou, A. Wildes, M. B. Stone, D. Abernathy, G. M. Luke, B. D. Gaulin, and C. R. Wiebe, Phys. Rev. B 98, 100401(R) (2018).
[56] Viviane Peçanha-Antonio, Erxi Feng, Xiao Sun, Devashibhai Adroja, Helen C. Walker, Alexandra S. Gibbs, Fabio Orlandi, Yixi Su, and Thomas Breckel, Phys. Rev. B 99, 134415 (2019).