Spin liquid states are often described as the antithesis of magnetic order. Recently, however, it has been proposed that in certain frustrated magnets the magnetic degrees of freedom may “fragment” in such a way as to give rise to a coexistence of spin liquid and ordered phases. Recent neutron scattering results [S. Petit et al., Nature Phys., Advance online publication, (2016)] suggest that this scenario may be realized in the pyrochlore magnet Nd$_2$Zr$_2$O$_7$. These observations show the characteristic pinch point features of a Coulombic spin liquid occurring alongside the Bragg peaks of an “all-in-all-out” ordered state. Here we explain the quantum origins of this apparent magnetic moment fragmentation, within the framework of a quantum model of nearest neighbour exchange, appropriate to Nd$_2$Zr$_2$O$_7$. This model is able to capture both the ground state order and the pinch points observed at finite energy. The observed fragmentation arises due to the combination of the unusual symmetry properties of the Nd$^{3+}$ ionic wavefunctions and the structure of equations of motion of the magnetic degrees of freedom. The results of our analysis suggest that Nd$_2$Zr$_2$O$_7$ is proximate to a $U(1)$ spin liquid phase and is a promising candidate for the observation of a Higgs transition in a magnetic system.

The study of frustrated magnets has uncovered many experimental systems in which conventional magnetic order is suppressed— or avoided entirely— opening the door to novel quantum ground states [1][4]. A particularly enticing possibility is to realize a spin-liquid ground state possessing emergent gauge fields and excitations with fractional quantum numbers [7]. As such, spin liquids provide beautiful examples of the emergence of new and unexpected degrees of freedom, out of the collective behaviour of a strongly interacting system [8][13].

While a spin liquid ground state is usually discussed as an alternative to magnetic order, it has been proposed that spin liquid physics can coexist with a conventional magnetic order parameter [16][21]. One way that this can occur [17] is through a “fragmentation” of the magnetisation field into two quasi-independent sets of degrees of freedom, one of which orders and the other of which remains fluctuating in a spin-liquid-like manner. In a spin-ice system, such as that considered in Ref. [17], such a state would be revealed in neutron scattering experiments by the coexistence of magnetic Bragg peaks, indicating long range order, with pinch-point singularities. Pinch-points in neutron scattering measurements are known to be the characteristic of a Coulomb phase [25][24], which hosts an emergent “magnetic flux” obeying Gauss’ law and an associated “electromagnetic” gauge field.

In a remarkable experimental development the signatures of this fragmentation have recently been observed in the pyrochlore magnet Nd$_2$Zr$_2$O$_7$ [25]. Nd$_2$Zr$_2$O$_7$ is known to undergo magnetic ordering at a temperature $T_N \approx 0.3K$ [26][28], with the formation of magnetic Bragg peaks consistent with an antiferromagnetic, “all-in-all-out”, ordered state shown in Fig. (1a). Alongside these Bragg peaks, the authors of Ref. [25] observed pinch point singularities, like those shown in Fig. (1b). These pinch points occur as part of a flat band at finite energy $\Delta \approx 0.07$ meV.

The observation of pinch points, signifying the physics of a Coulomb phase, against the background of these Bragg peaks would seem to provide compelling evidence for the realization of the fragmentation scenario proposed in Ref. [17]. While calculations using the random phase approximation (RPA) presented in Ref. [25] were able to capture the pinch points, the parameters of the fitted model were not compatible with an all-in-all-out ground state. More broadly than this, the question of the mechanism of the moment fragmentation in Nd$_2$Zr$_2$O$_7$ remains open.

Here, we explain the quantum origins of the moment fragmentation observed in Nd$_2$Zr$_2$O$_7$. This fragmentation is the combined consequence of the “dipolar-octupolar” [29] nature of the Nd$^{3+}$ Kramers doublets and of the structure of the equations of motion for the pseudospin operators describing these doublets. Our theory goes beyond previous work by reconciling the magnetic ground state of Nd$_2$Zr$_2$O$_7$ with its observed spectrum in inelastic neutron scattering, including the presence of pinch points. Through this theory we are able to reveal the true nature of the moment fragmentation and find that Nd$_2$Zr$_2$O$_7$ is proximate to a $U(1)$ spin liquid phase.

Model—To model the magnetism of Nd$_2$Zr$_2$O$_7$ we must begin with the physics of Nd$^{3+}$ ions in their local crystal field. The ground state of the crystal field is a Kramers doublet, separated by a gap of $\Delta_{CF} \approx 23$ meV from the lowest excited doublet [27]. This doublet has dipolar-octupolar character [27][30], a fact which will have important consequences in our discussion.

A natural choice of basis $\{ | \uparrow_z \rangle , | \downarrow_z \rangle \}$ for this doublet is the one which diagonalizes the $z$-component of the angular momentum operator $\vec{J}$, where the $z$-axis is defined locally as the $C_3$ symmetry axis pointing from a magnetic site through the centres of the two pyrochlore tetrahedra which share it [cf. Fig. (1a)]. While $J_z$ has finite matrix elements within the doublet, the planar components $\{ J_x, J_y \}$ both vanish [27][28].

To describe the interactions of these dipolar-octupolar doublets we introduce on each site $i$ a vector of pseudospin-1/2 operators $\vec{\tau}_i = (\tau^{+}_i, \tau^{0}_i, \tau^{-}_i)$. Since $\langle J_x \rangle = \langle J_y \rangle = 0$ within the doublet, the magnetisation on site $i$ is given by

$$\mathbf{m}_i = g_i \mu_B \tau^{+}_i \hat{z}_i$$

where $\hat{z}_i$ is a unit vector in the local $z$-direction and $g_i$ is the $z$-component of the g-tensor.
As discussed in Ref. [29], the symmetry properties of $\tilde{\tau}$ are somewhat counter-intuitive: both $\tau_i^x$ and $\tau_i^z$ transform like the $z$-component of a magnetic dipole moment. The operator $\tau_i^y$ meanwhile, transforms like an element of the magnetic octupole tensor. From these symmetry properties one can deduce the most general form of nearest-neighbour interactions between the operators $\tau_i^\alpha$ allowed by the symmetries of the system [24, 31] (time reversal $\otimes$ lattice symmetries):

$$\mathcal{H}_{ex}^{DO} = \sum_{\langle ij \rangle} \left[ J_x \tau_i^x \tau_j^x + J_y \tau_i^y \tau_j^y + J_z \tau_i^z \tau_j^z \right. + \left. J_{xz} (\tau_i^x \tau_j^z + \tau_i^z \tau_j^x) \right] \tag{2}$$

The interaction $J_{xz}$ may be removed by a global pseudospin rotation $\tau_i^\alpha \rightarrow \tilde{\tau}_i^\alpha$ where

$$\tilde{\tau}_i^x = \cos(\vartheta) \tau_i^x + \sin(\vartheta) \tau_i^z; \quad \tilde{\tau}_i^y = \tau_i^y; \quad \tilde{\tau}_i^z = \cos(\vartheta) \tau_i^x - \sin(\vartheta) \tau_i^z; \quad \tan(2\vartheta) = \frac{2J_{xz}}{J_x - J_z} \tag{3}$$

This leaves us with an “$XYZ$” Hamiltonian for the rotated pseudospins $\tilde{\tau}_i^\alpha$:

$$\mathcal{H}_{XYZ}^{DO} = \sum_{\langle ij \rangle} \left[ J_x \tilde{\tau}_i^x \tilde{\tau}_j^x + J_y \tilde{\tau}_i^y \tilde{\tau}_j^y + J_z \tilde{\tau}_i^z \tilde{\tau}_j^z \right] \tag{4}$$

The phase diagram of $\mathcal{H}_{XYZ}^{DO}$ is then a function of the three parameters $J_x, J_z$ and does not depend on the angle $\vartheta$.

This does not mean, however, that $\vartheta$ plays no further role in the physics of the system. The magnetisation on each site, in terms of the rotated pseudospins $\tilde{\tau}_i^\alpha$ is

$$m_i = g_x \mu_B (\cos(\vartheta) \tilde{\tau}_i^x + \sin(\vartheta) \tilde{\tau}_i^z) \mathbf{z}_i \tag{5}$$

The angle $\vartheta$ thus controls how the pseudospins $\tilde{\tau}_i^\alpha$ couple to an external probe which scatters off the internal magnetic fields—such as a neutron.

The peculiar moment fragmentation observed in Nd$_2$Zr$_2$O$_7$ stems from Eq. (5). Eq. (5) can be split up in terms of its contributions from $\tilde{\tau}_i^x$ and $\tilde{\tau}_i^z$:

$$m_i = g_x \mu_B \cos(\vartheta) m_i^{(2)} + g_x \mu_B \sin(\vartheta) m_i^{(3)} \tag{6}$$

$$m_i^{(a)} = \tilde{\tau}_i^a \mathbf{z}_i \tag{7}$$

In essence, the origin of the fragmentation is that $m_i^{(3)}$ orders, forming the all-in-all-out order which is responsible for the observed magnetic Bragg peaks, while fluctuations of $m_i^{(2)}$ get shifted to finite energy. These fluctuations of $m_i^{(2)}$ themselves decouple dynamically into a flat band obeying $\nabla \cdot m_i^{(2)} = 0$ and therefore exhibiting the correlations of a Coulomb phase, and two higher energy dispersive bands.

Spin wave theory—To see this we will first use a spin wave expansion around the all-in-all-out ground state. An all-in-all-out ground state, with $\langle \tilde{\tau}_i^{x/z} \rangle \neq 0$ and $\langle \tilde{\tau}_i^y \rangle = \langle \tilde{\tau}_i^{\pm} \rangle = 0$ is a classical ground state of $\mathcal{H}_{DO}^{DO}$ [Eq. (4)] when

$$\tilde{J}_z < 0, \quad -|\tilde{J}_z| < \tilde{J}_x, \tilde{J}_y < 3|\tilde{J}_z| \tag{8}$$

FIG. 1: (a) All-in-all-out configuration of magnetic moments on the pyrochlore lattice. The blue dashed line indicates the local $z$-axis on the central site. (b) Pinch point singularities in the neutron scattering structure factor $S(q, \omega = \Delta_{\text{eq}})$ [Eq. (14)] at finite energy above the all-in-all-out ground state in dipolar-octupole magnets. These pinch points are reminiscent of those predicted in the “Coulomb phase” of spin ice [22, 24]. Pinch points of this form were observed at energy $\Delta \approx 0.07\text{meV}$ in recent experiments on the pyrochlore material Nd$_2$Zr$_2$O$_7$ [25], along with Bragg peaks signifying an all-in-all-out ordered phase. This suggests that Nd$_2$Zr$_2$O$_7$ exhibits the phenomenon of “moment fragmentation”, proposed in Ref. [17]. Calculation of the structure factor in the flat band was performed using a linear spin wave treatment of the exchange Hamiltonian $\mathcal{H}_{DO}^{DO}$ [Eq. (4)] and the exchange parameters in Eq. (18).

The spin wave expansion proceeds by introducing Holstein-Primakoff bosons $\{a_i, a_i^\dagger\} = \delta_{ij}$ and writing

$$\tilde{\tau}_i^x = S - a_i^\dagger a_i \tag{9}$$

$$\tilde{\tau}_i^z = i\tilde{\tau}_i^x + i\tilde{\tau}_i^y = \sqrt{2S} - a_i^\dagger a_i \approx \sqrt{2S}a_i \tag{10}$$

$$\tilde{\tau}_i^+ = \tilde{\tau}_i^x + i\tilde{\tau}_i^y = \sqrt{2S} - a_i^\dagger a_i \approx \sqrt{2S}a_i \tag{11}$$

where $S = \frac{1}{2}$ since we are dealing with pseudospin-$\frac{1}{2}$ operators.

Inserting Eqs. (9)–(11) into Eq. (4) and keeping terms only up to bilinear order in $a_i, a_i^\dagger$ yields the linear spin wave Hamiltonian:

$$\mathcal{H}_{LSW}^{DO} = -3N|\tilde{J}_z|S^2 + 6|\tilde{J}_z|S \sum_i a_i^\dagger a_i$$

$$+ \frac{S}{2} \sum_{\langle ij \rangle} (a_i^\dagger, a_i) \left( \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \right) \left( \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{1}{2} \right) \tag{12}$$

After Fourier transformation into momentum space $\{q, \omega\}$ and a subsequent Bogoliubov transformation [22, 33] to a new set of bosonic operators $\{b^\dagger_{\lambda q}, b_{\lambda q}\} = \delta_{qq'} \delta_{\lambda \lambda'}$ we arrive at a diagonalized Hamiltonian for four bands $\lambda$ of bosons with dispersion $\omega_{\lambda}(q)$:

$$\mathcal{H}_{LSW}^{DO} = -3N|\tilde{J}_z|S(S + 1)$$

$$+ \sum_{\lambda} \omega_{\lambda}(q) \left( \frac{b_{\lambda q}^\dagger b_{\lambda q} + 1}{2} \right) \tag{13}$$
These four bands \( \omega_m(q) \) consist of two degenerate flat bands at energy \( \Delta_{\text{flat}} = \sqrt{3|J_z| - J_x}(3|J_z| - J_y) \) and two dispersive bands.

We can invert the transformations used in obtaining Eq. (13) to calculate the dynamical correlations accessible in neutron scattering, in terms of the expectation values of bosonic bilinears. The structure factor for magnetic neutron scattering is

\[
\mathcal{S}(q, \omega) = \int dt e^{-i\omega t} \sum_{\mu\nu} \left( \delta_{\mu\nu} - \frac{q_\mu q_\nu}{q^2} \right) \times \langle m^\mu(-q,0)m^\nu(q, t) \rangle
\]

where \( m(q, t) \) is the lattice Fourier transform of the site magnetisation [Eq. (5)] at time \( t \).

The structure factor \( \mathcal{S}(q, \omega) \) breaks up into two contributions

\[
\mathcal{S}(q, \omega) = \mathcal{S}^{(2)}(q, \omega)\mathcal{S}^{(2)}(q, \omega).
\]

The first contribution \( \mathcal{S}^{(2)}(q, \omega) \) comes from the ordered and static correlations of \( m_i(q) \) and gives rise to magnetic Bragg peaks at \( \omega = 0 \).

The second contribution to \( \mathcal{S}(q, \omega) \) in Eq. (15) comes from the dynamic correlations of \( m_i(q) \), which have the form

\[
\mathcal{S}^{(2)}(q, \omega) \approx \sin^2(\vartheta)q^2 \mu_B^2 \sum_{\lambda} s_\lambda(q)
\]

\[
[(1 + n_B(\omega))\delta(\omega - \omega_\lambda(q)) + n_B(\omega)\delta(\omega + \omega_\lambda(q))]
\]

where \( s_\lambda(q) \) are coefficients calculated from the Bogoliubov transformation.

Due to the flat bands, \( \mathcal{S}^{(2)}(q, \omega) \) exhibits a peak at \( \omega = \Delta_{\text{flat}} \) for all \( q \). The intensity of this flat band peak is plotted in Fig. 1(b). For all choices of exchange parameters \( J_\alpha \) within the all-in-all-out phase it exhibits a pattern of pinch-point singularities, as observed at finite energy in Nd\(_2\)Zr\(_2\)O\(_7\) [25]. Our calculations thus simultaneously reproduce the observation of pinch points and the correct ground state order for Nd\(_2\)Zr\(_2\)O\(_7\), something which was not done previously.

The values of the exchange parameters \( \tilde{J}_\alpha \) can be constrained by considering the inelastic spectrum, which was measured in Ref. [25]. The model used in Ref. [25] is equivalent to taking Eq. (4) with parameters

\[
\tilde{J}_x = 0; \tilde{J}_y = -0.047 \text{ meV}; \tilde{J}_z = 0.103 \text{ meV}.
\]

This parameterisation gives a good description of the inelastic spectrum but incorrectly predicts an octupolar ground state. Here, we take instead

\[
\tilde{J}_x = 0.103 \text{ meV}; \tilde{J}_y = 0; \tilde{J}_z = -0.047 \text{ meV}.
\]

This transformed set of parameters also gives equally good agreement with the inelastic spectrum, while correctly reproducing the experimental ground state [46]. The predicted inelastic scattering calculated from linear spin wave theory for the parameters in Eq. (18) is plotted in Fig. 2 for comparison with Ref. [25].

The theory presented here is also capable of accounting for the positive Curie-Weiss temperature of Nd\(_2\)Zr\(_2\)O\(_7\) [26-28, 30], in spite of the antiferromagnetic ground state. Specifically, the Curie-Weiss temperature for the model Eq. (4) is

\[
T_{\text{CW}} = \frac{1}{2k_B} \left( J_z \cos^2(\vartheta) + \tilde{J}_x \sin^2(\vartheta) \right).
\]

where \( k_B \) is Boltzmann’s constant. If we take \( \vartheta \approx 0.83 \) this reproduces the Curie-Weiss temperature \( T_{\text{CW}} \approx 0.2 \text{K} \) measured in Ref. [27, 28, 30]. As a final consistency check, we can then calculate the magnitude of the ordered moment \( m_{\text{ord}} \) which we expect to find in the ground state. At our level of approximation, the ratio of \( m_{\text{ord}} \) to the full, saturated, moment \( m_{\text{sat}} \) is given by

\[
\frac{m_{\text{ord}}}{m_{\text{sat}}} = \cos(\vartheta) \left( \frac{S - \langle a^\dagger a \rangle \langle a^\dagger a \rangle}{S} \right).
\]
on the Helmholtz decomposition of the magnetisation density $m$ when magnetic order occurs in Ref. [27], but a bit higher than the value $m_{\text{ord}} \approx 59$ obtained in Nd$_2$Zr$_2$O$_7$. This is close to the value $m_{\text{ord}} \approx 0.33$ obtained in Ref. [28]. It is interesting to note that most of this moment reduction comes from the pseudospin rotation $\vartheta$, not from the zero-point fluctuations, as one might typically expect.

The theory presented in this article is thus the first to present a consistent treatment of the ground state and the finite energy spectrum in Nd$_2$Zr$_2$O$_7$. At the same time it is also able to account for the apparent contradiction between the Curie-Weiss temperature and the antiferromagnetic ordering and gives reasonable agreement with the strongly reduced ordered moment measured in experiments [27].

Moment fragmentation—Having established that a theory based on a linear spin wave treatment of Eq. (4) correctly reproduces the experimental phenomenology, we can now ask what this theory tells about the proposed “magnetic moment fragmentation”. In particular, is this a true example of moment fragmentation, as proposed in Ref. [17], and if so, what is its origin?

The proposal of Brooks-Bartlett et al. in Ref. [17] is based on the Helmholtz decomposition of the magnetisation density

$$ m = m_m + m_d = \nabla \psi + \nabla \times A $$

(21)

where $m_d = \nabla \times A$ is divergence-free ($\nabla \cdot m_d = 0$) and $m_m = \nabla \psi$ is “divergence-full”. The fragmentation phenomenon is observed when magnetic order occurs in $m_m$, but $m_d$ remains fluctuating quasi-independently of $m_m$. Since $m_y$ obeys $\nabla \cdot m_d = 0$ this gives rise to the pinch point correlations associated with a Coulomb phase [13, 22, 24].

We can understand the magnetic fragmentation phenomenon in Nd$_2$Zr$_2$O$_7$ by defining fields $m_i^{(g)}$ for each pseudospin component $\gamma_i^g$, according to Eq. (7) and then applying the Helmholtz decomposition to each one individually

$$ m_i^{(g)} = \nabla \psi^{(g)} + \nabla \times A^{(g)}. $$

(22)

(Note that $m_i^{(g)}$ does not contribute to the physical magnetisation field $m_i$ [Eq. (6)]).

In the all-in-all-out ground state, $m_i^{(g)}$ is completely “divergence-full”, and we may write $A^{(g)} = 0$. The fluctuations of $m_i^{(g)}$ and $m_i^{(g)}$, on the other hand, have both divergence-free and “divergence-full” components. The moment fragmentation phenomenon is observed because the equations of motion decouple the dynamics of divergence-free and “divergence-full” components of $m_i^{(g)}, m_i^{(g)}$.

Writing down the Heisenberg equations of motion for $m_i^{(g)}$ and $m_i^{(g)}$ and linearising around the all-in-all-out ground state we find

$$ \partial_t m_i^{(g)} \approx \varepsilon_{\gamma_i^g \hat{g} ^{x}} S (J_{\alpha'} \nabla_i (\nabla \cdot m^{(\gamma_i')} + (6|J_x| - 2J_{\alpha'}) \psi^{(\gamma_i')}) $$

(23)

where $\alpha' = \hat{y}$ when $\alpha = \hat{x}$ and vice versa and $\varepsilon_{\hat{x} \hat{y} \hat{z}} = -\varepsilon_{\hat{y} \hat{z} \hat{x}} = 1$. In Eq. (23), $\nabla_i$ and $\nabla \cdot$ should be interpreted as the lattice gradient and divergence. This suggestive form for the equations of motion in terms of the lattice gradient and divergence arises because the sites of the pyrochlore lattice can be considered as the bonds of a bipartite (in this case, diamond) lattice [12].

Eq. (23) can be solved in terms of the Helmholtz decompositions [Eqs. (22)], by writing

$$ \partial_t \psi^{(\gamma_i)} = \varepsilon_{\gamma_i^g \hat{g} ^{x}} S (J_{\alpha'} \nabla^2 \psi^{(\gamma_i')} + (6|J_x| - 2J_{\alpha'}) \psi^{(\gamma_i')}) $$

(24)

$$ \partial_t A^{(g)} = \varepsilon_{\gamma_i^g \hat{g} ^{x}} S (6|J_x| - 2J_{\alpha'}) A^{(\gamma_i')} $$

(25)

The important feature of Eqs. (24)-(25) is that the divergenceless fluctuations (i.e. fluctuations of $A^{(g)}$) are completely decoupled from the “divergence full” fluctuations (i.e. fluctuations of $\psi^{(\gamma_i)}$). Fluctuations of $A^{(g)}$ form a flat band at energy $\Delta_{\text{flatt}} = \sqrt{(3|J_x| - J_y)(3|J_z| - J_y)}$, while fluctuations of $\psi^{(\gamma_i)}$ form dispersive bands.

The physical magnetisation field in Nd$_2$Zr$_2$O$_7$ [Eq. (6)] thus comprises (i) a static, ordered, “divergence full” component, (ii) a finite energy divergenceless component exhibiting Coulomb-liquid-like correlations and finally (iii) another “divergence full” component corresponding to the finite energy dispersive bands.

The fact that all three components are observable within a magnetisation field which is strictly Ising like (in the sense that $m_i$ is always parallel to the local easy axis) is a consequence of the unusual symmetry of dipolar octupolar doublets—specifically that the $x$-component of the pseudospin
transforms like the $z$-component of a dipole moment. This understanding of the moment fragmentation is fully compatible with the observation that the pinch points remain observable above the ordering transition at $T_N$, but at lower frequency \cite{25}. Above the transition $m^{(x)}_n$ can fluctuate for little or no energy cost but its correlations will remain ice like due to the positive value of $J_z$.

Conclusions-- In conclusion we have explained the quantum origins of the moment fragmentation in Nd$_2$Zr$_2$O$_7$, observed in Ref. \cite{25}. It may be rationalized as the consequence of the symmetry properties of dipolar-octupolar doublets and a decoupling of divergence-free and divergence-full fluctuations in the equations of motion.

Much of the physics discussed here is generic to systems described by the exchange Hamiltonian $H^{DO}_{XYZ}$ which have an all-in-all-out ground state. Specifically, the flat band exhibiting pinch points at finite energy is present throughout the all-in-all-out phase of $H^{DO}_{XYZ}$, at least at the level of linear spin wave theory. It will therefore be interesting to investigate whether the moment fragmentation phenomenon is also observed in other Nd based pyrochlores showing an all-in-all-out ground state such as Nd$_2$Sn$_2$O$_7$ \cite{14,16,37–42}, Nd$_2$Hf$_2$O$_7$ \cite{55} and possibly Nd$_2$Pb$_2$O$_7$ \cite{55}.

The parameterisation of the exchange Hamiltonian $H^{DO}_{XYZ}$ [Eq. (4)], given in Eq. (18) suggests that Nd$_2$Zr$_2$O$_7$ is proximate to the $U(1)$ spin liquid phase which has been long sought amongst “quantum spin ice” pyrochlores \cite{14,16,37–42}. As shown in Fig. 3 the closing of the gap to the flat band containing the pinch point correlations occurs at $\frac{T}{T_N} = 3$ within linear spin wave theory. Classically, this would signal the formation of an extensive ground state manifold with ice-like character, but the mixing of these states by quantum fluctuations is known to stabilise a $U(1)$ spin liquid with dynamic emergent gauge fields \cite{33,41,42,52}. The placement of Nd$_2$Zr$_2$O$_7$ close to the point where this gap vanishes hints at the proximity of the $U(1)$ spin liquid phase. If there is a well formed Coulomb phase above $T_N$ in Nd$_2$Zr$_2$O$_7$ this may make the observed magnetic ordering a candidate for the observation of a Higgs transition in which the emergent gauge field of the Coulomb phase is gapped by the condensation of emergent gauge charges \cite{44,45}. We therefore have reason to hope that experiments on Nd$_2$Zr$_2$O$_7$ and related materials may yet reveal even more exotic phenomena.

Acknowledgements – This work was supported by the Theory of Quantum Matter Unit of the Okinawa Institute of Science and Technology Graduate University. The author is grateful to Ludovic Jaubert and Nic Shannon for careful readings of the manuscript.

[1] J. E. Greedan, Geometrically frustrated magnetic materials, J. Mater. Chem. 11, 37-53, (2001)
[2] J. S. Gardner, M. J. P. Gingras and J. E. Greedan, Magnetic Pyrochlore Oxides, Rev. Mod. Phys. 82, 53-107, (2010)
[3] Y. Shimizu, K. Miyagawa, K. Kanoda, M. Maesato, and G. Saito, Spin Liquid State in an Organic Mott Insulator with a Triangular Lattice, Phys. Rev. Lett. 91, 107001 (2003)
[4] T. H. Han, J. S. Helton, S. Y. Chu, D. G. Nocera, J. A. Rodriguez-Rivera, C. Broholm and Y. S. Lee, Fractionalized excitations in the spin-liquid state of a kagome-lattice antiferromagnet Nature 492, 406-410 (2012)
[5] K. Kimura, S. Nakatsuji, J. J. Wen, C. Broholm, M. B. Stone, E. Nishihori and H. Sawa, Quantum fluctuations in spin ice-like $Pr_2Zr_2O_7$, Nature Commun. 4, 2914 (2013)
[6] R. Sibille, E. Lhotel, V. Pomjakushin, C. Baines, T. Fennell and M. Kenzelmann, Candidate Quantum Spin Liquid in the Ce$^{3+}$ Pyrochlore Stannate $Ce_2Sn_2O_7$, Phys. Rev. Lett. 115, 097202 (2015)
[7] L. Balents, Spin liquids in frustrated magnets, Nature 464, 199-208 (2010)
[8] X.-G. Wen, Quantum orders and symmetric spin liquids, Phys. Rev. B 65, 165113 (2002)
[9] A. Kitaev, Anyons in an exactly solved model and beyond, Annals of Physics 321, 2, (2006)
[10] C. Castelnovo, R. Moessner and S. L. Sondhi, Magnetic monopoles in spin ice, Nature 451, 42 (2008)
[11] R. Moessner and S. L. Sondhi, Irrational charge from topological order, Annu. Rev. Condens. Matter Phys. 1, 1 (2010)
[12] C. L. Henley, The “Coulomb phase” in frustrated systems, Annu. Rev. Condens. Matter Phys. 1, 1 (2010)
[13] S. M. Yang, D. A. Huse and S. R. White, Spin-Liquid Ground State of the $S=1/2$ Kagome Heisenberg Antiferromagnet, Science 332, 1173-1176, (2011)
[14] M. J. P. Gingras and P. A. McClarty, Quantum spin ice: a search for gapless quantum spin liquids in pyrochlore magnets, Rep. Prog. Phys. 77, 056501 (2014).
[15] O. Benton, L. D. C. Jaubert, H. Yan and N. Shannon, A spin-liquid with pinch-line singularities on the pyrochlore lattice, Nature Commun. 7, 11572 (2016)
[16] L. Savary and L. Balents, Coulombic quantum liquids in spin-1/2 pyrochlores, Phys. Rev. Lett. 108, 037202 (2012).
[17] M. E. Brooks-Bartlett, S. T. Banks, D. C. Jaubert, A. Harman-Clarke and P. C. W. Holdsworth, Magnetic Moment Fragmentation and Monopole Crystallization, Phys. Rev. X 4, 011007, (2014).
[18] S. Powell, Ferromagnetic Coulomb phase in classical spin ice, Phys. Rev. B 91, 094431 (2015).
[19] L. D. C. Jaubert, Monopole Holes in a Partially Ordered Spin Liquid, Spin 5, 1540005 (2015).
[20] B. Canals, I.-A. Chioar, V.-D. Nguyen, M. Hahn, D. Lacour, F. Montaigne, A. Locatelli, T. Onur Mentes, B. Santos Burgos and N. Rougemaire, Fragmentation of magnetism in artificial kagome dipolar spin ice, Nature Commun. 7, 11446 (2016)
[21] J. A. M. Padden, H. S. Ong, J. O. Hamp, P. Mukherjee, X. Bai, M. G. Tucker, N. P. Butch, C. Castelnovo, M. Mourigal and S. E. Dutton, Emergent Order in the Kagome Ising Magnet $Dy_3Mg_2Sb_3O_{14}$, arXiv:1605.01423
[22] S. V. Isakov, K. Gregor, R. Moessner, and S. L. Sondhi, Dipolar Spin Correlations in Classical Pyrochlore Magnets, Phys. Rev. Lett. 93, 167204 (2004).
[23] C. L. Henley, Power-law spin correlations in pyrochlore antiferromagnets, Phys. Rev. B 71, 014424 (2005).
[24] 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, Magnetic Coulomb phase in the spin ice $Ho_2Ti_2O_7$, Science 326, 415-417 (2009).
[25] S. Petit, E. Lhotel, B. Canals, M. Ciomaga Hatnean, J. Olivier, H. Muttka, E. Ressouche, A. R. Wildes, M. R. Lees
and G. Balakrishnan, Observation of magnetic fragmentation in spin ice, Nature Phys., Advance online publication, (2016). doi:10.1038/nphys3710

[26] H. W. J. Blöte, R. F. Wielinga and W. J. Huiskamp, Heat-capacity measurements on rare-earth double oxides R2M2O7, Physica 43, 549 (1969)

[27] J. Xu, V. K. Anand, A. K. Bera, M. Frontzek, D. L. Barnett, N. Casati, K. Siemensmeyer and B. Lake Magnetic structure and crystal-field states of the pyrochlore antiferromagnet Nd2Zr2O7, Phys. Rev. B 92, 224430 (2015)

[28] E. Lhotel, S. Petit, S. Guitteny, O. Florea, M. Ciomaga Hatnean, C. Colin, E. Ressouche, M. R. Lees, and G. Balakrishnan, Fluctuations and All-In-All-Out Ordering in Dipole-Octupole Nd2Zr2O7, Phys. Rev. Lett. 115, 197202 (2015)

[29] Y.-P. Huang, G. Chen and M. Hermele, Quantum Spin Ices and Topological Phases from Dipolar-Octupolar Doublets on the Pyrochlore Lattice, Phys. Rev. Lett. 112, 167203 (2014).

[30] M. Ciomaga Hatnean, M. R. Lees, O. A. Petrenko, D. S. Keeble, M. J. Gutmannn, V. V. Klekovina and B. Z. Malkin, Structural and magnetic investigations of single-crystal neodymium zirconate pyrochlore Nd2Zr2O7, Phys. Rev. B 91, 174416 (2015).

[31] J. Carrasquilla, Z. Hao and R. G. Melko, A two-dimensional spin liquid in quantum kagome ice, Nature Commun. 6, 7421 (2015)

[32] P. Fazekas, Lecture Notes on Electron Correlation and Magnetism, (World Scientific, Singapore, 1999).

[33] M. Roger, J. H. Hetherington, and M. Delrieu, Magnetism in Solid He-3, Rev. Mod. Phys. 55, 1 (1983).

[34] A. Bertin, P. Dalmas de Réotier, B. Fak, C. Marin, A. Yaouanc, A. Forget, D. Sheptyakov, B. Frick, C. Ritter, A. Amato, C. Baines and P. J. C. King, Nd2Sn2O7: An all-in-all-out pyrochlore magnet with no divergence-free field and anomalously slow paramagnetic spin dynamics, Phys. Rev. B 92, 144423 (2015)

[35] V. K. Anand, A. K. Bera, J. Xu, T. Herrmannsdorfer, C. Ritter and B. Lake, Observation of long-range magnetic ordering in pyrochafnate Nd2Hf2O7: A neutron diffraction study, Phys. Rev. B 92, 184418 (2015).

[36] A. M. Hallas, A. M. Arevalo-Lopez, A. Z. Sharma, T. Munsch, J. P. Attfield, C. R. Wiebe, and G. M. Luke, Magnetic frustration in lead pyrochlores, Phys. Rev. B 91, 104417 (2015).

[37] M. Hermele, M. P. A. Fisher, and L. Balents, Pyrochlore photons: The U(1) spin liquid in a S=1/2 three-dimensional frustrated magnet, Phys. Rev. B 69, 064404 (2004).

[38] A. Banerjee, S. V. Isakov, K. Damle and Y.-B. Kim, Unusual liquid state of hard-core bosons on the pyrochlore lattice, Phys. Rev. Lett. 100, 047208 (2008).

[39] O. Benton, O. Sikora, and N. Shannon, Seeing the light: Experimental signatures of emergent electromagnetism in a quantum spin ice, Phys. Rev. B 86, 075154 (2012).

[40] N. Shannon, O. Sikora, F. Pollmann, K. Penc, and P. Fulde, Quantum ice: A quantum Monte Carlo study, Phys. Rev. Lett. 108, 067204 (2012).

[41] Z. Hao, A. G. R. Day and M. J. P. Gingras, Bosonic many-body theory of quantum spin ice, Phys. Rev. B 88, 144402 (2013).

[42] Y. Kato and S. Onoda, Numerical evidence of quantum melting of spin ice: quantum-to-classical crossover, Phys. Rev. Lett. 115, 077202 (2015).

[43] P. A. McClarty, O. Sikora, R. Moessner, K. Penc, F. Pollmann and N. Shannon, Chain-based order and quantum spin liquids in dipolar spin ice, Phys. Rev. B 92, 094418 (2015).

[44] S. Powell, Higgs transitions of spin ice, Phys. Rev. B 84, 094437, (2011).

[45] L.-J. Chang, S. Onoda, Y. Su, Y.-J. Kao, K.-D. Tsuei, Y. Yasui, K. Kakurai and M. R. Lees, Higgs transition from a magnetic Coulomb liquid to a ferromagnet in Yb2Ti2O7, Nature Commun. 3, 992, (2012).