Quantum origins of moment fragmentation in $Nd_2Zr_2O_7$

Owen Benton
Okinawa Institute of Science and Technology Graduate University, Onna-son, Okinawa 904-0495, Japan
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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, E. Lhotel, B. Canals, M. Ciomaga Hatnean, J. Ollivier, H. Muttka, E. Ressouche, A. R. Wildes, M. R. Lees, and G. Balakrishnan, Nat. Phys. **12**, 746 (2016)] suggest that this scenario may be realized in the pyrochlore magnet $Nd_2Zr_2O_7$. 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-neighbor exchange, appropriate to $Nd_2Zr_2O_7$. 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 wave functions and the structure of equations of motion of the magnetic degrees of freedom. The results of our analysis suggest that $Nd_2Zr_2O_7$ 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.

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I. INTRODUCTION

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 [1–6]. 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 behavior of a strongly interacting system [8–15].

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 magnetization 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 [22–24], which hosts an emergent "magnetic flux" obeying Gauss's 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 $Nd_2Zr_2O_7$ [25]. $Nd_2Zr_2O_7$ is known to undergo magnetic ordering at a temperature $T_N \approx 0.3$ K [26–28], with the formation of magnetic Bragg peaks consistent with an antiferromagnetic, "all-in-all-out," ordered state shown in Fig. 1(a). Alongside these Bragg peaks, the authors of Ref. [25] observed pinch-point singularities, like those shown in Fig. 1(b). 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 $Nd_2Zr_2O_7$ remains open.

Here, we explain the quantum origins of the moment fragmentation observed in $Nd_2Zr_2O_7$. 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 those doublets. Our theory goes beyond previous work by reconciling the magnetic ground state of $Nd_2Zr_2O_7$ 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 $Nd_2Zr_2O_7$ is proximate to a $U(1)$ spin-liquid phase.

II. MODEL

To model the magnetism of $Nd_2Zr_2O_7$ we must begin with the physics of Nd^{3+} ions in their local crystal field. The ground state of the crystal field (CF) 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 has 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 $\mathbf{J}$, where the z axis is defined locally as the C_3 symmetry axis pointing from a magnetic site through the centers of the two pyrochlore tetrahedra which share it [cf. Fig. 1(a)]. 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 (DO) doublets, we introduce on each site i a vector of pseudospin-1/2 operators $\vec{\tau}_i = (\tau_i^x, \tau_i^y, \tau_i^z)$. Since $\langle J_x \rangle = \langle J_y \rangle = 0$ within

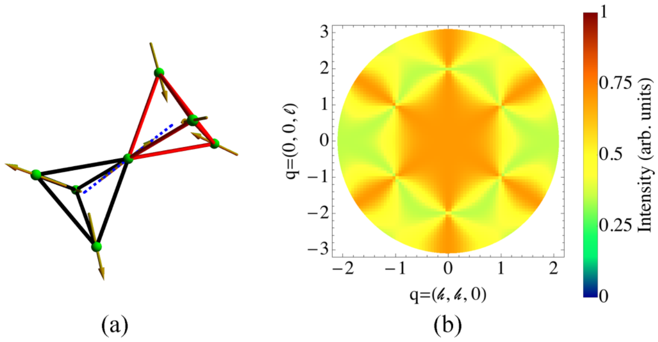


FIG. 1. (a) All-in-all-out configuration of magnetic moments on the pyrochlore lattice. The blue dashed line indicates the local z axis on the central site. (b) Pinch-point singularities in the neutron-scattering structure factor $\mathcal{S}(\mathbf{q},\omega = \Delta_{\text{flat}})$ [Eq. (14)] at finite energy above the all-in-all-out ground state in dipolar-octupolar 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 $\Delta \approx 0.07$ meV in recent experiments on the pyrochlore material $Nd_2Zr_2O_7$ [25], along with Bragg peaks signifying an all-in-all-out ordered phase. This suggests that $Nd_2Zr_2O_7$ 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 $\mathcal{H}^{\text{DO}}_{XYZ}$ [Eq. (4)] and the exchange parameters in Eq. (18).

the doublet, the magnetization on site i is given by

$$\mathbf{m}_i = g_z \mu_B \tau_i^z \hat{\mathbf{z}}_i, \tag{1}$$

where $\hat{\mathbf{z}}_i$ is a unit vector in the local z direction and g_z is the z component of the g-tensor.

As discussed in Ref. [29], the symmetry properties of $\vec{\tau}$ are somewhat counterintuitive: both τ_i^z and τ_i^x transform like the z component of a magnetic dipole moment. The operator τ_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-neighbor interactions between the operators τ_i^α allowed by the symmetries of the system [29,31] (time reversal ⊗ lattice symmetries):

$$\mathcal{H}^{\text{DO}}_{\text{ex}} = \sum_{\langle ij\rangle} \left[\mathsf{J}_x \tau_i^x \tau_j^x + \mathsf{J}_y \tau_i^y \tau_j^y + \mathsf{J}_z \tau_i^z \tau_j^z + \mathsf{J}_{xz}\left(\tau_i^x \tau_j^z + \tau_i^z \tau_j^x\right)\right]. \tag{2}$$

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

$$\tilde{\tau}_i^{\tilde{x}} = \cos(\vartheta)\tau_i^x + \sin(\vartheta)\tau_i^z, \quad \tilde{\tau}_i^{\tilde{y}} = \tau_i^y,$$
$$\tilde{\tau}_i^{\tilde{z}} = \cos(\vartheta)\tau_i^z - \sin(\vartheta)\tau_i^x, \quad \tan(2\vartheta) = \frac{2\mathsf{J}_{xy}}{\mathsf{J}_x - \mathsf{J}_z}. \tag{3}$$

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

$$\mathcal{H}^{\text{DO}}_{XYZ} = \sum_{\langle ij\rangle} \left[\tilde{\mathsf{J}}_x \tilde{\tau}_i^{\tilde{x}} \tilde{\tau}_j^{\tilde{x}} + \tilde{\mathsf{J}}_y \tilde{\tau}_i^{\tilde{y}} \tilde{\tau}_j^{\tilde{y}} + \tilde{\mathsf{J}}_z \tilde{\tau}_i^{\tilde{z}} \tilde{\tau}_j^{\tilde{z}}\right]. \tag{4}$$

The phase diagram of $\mathcal{H}^{\text{DO}}_{XYZ}$ is then a function of the three parameters $\tilde{\mathsf{J}}_{\tilde{x},\tilde{y},\tilde{z}}$ and does not depend on the angle ϑ.

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

$$\mathbf{m}_i = g_z \mu_B \left(\cos(\vartheta)\tilde{\tau}_i^{\tilde{z}} + \sin(\vartheta)\tilde{\tau}_i^{\tilde{x}}\right)\hat{\mathbf{z}}_i. \tag{5}$$

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

The peculiar moment fragmentation observed in $Nd_2Zr_2O_7$ stems from Eq. (5). Equation (5) can be split up in terms of its contributions from $\tilde{\tau}_i^{\tilde{z}}$ and $\tilde{\tau}_i^{\tilde{x}}$:

$$\mathbf{m}_i = g_z \mu_B \cos(\vartheta)\mathbf{m}_i^{(\tilde{z})} + g_z \mu_B \sin(\vartheta)\mathbf{m}_i^{(\tilde{x})}, \tag{6}$$

$$\mathbf{m}_i^{(\tilde{\alpha})} = \tilde{\tau}_i^{\tilde{\alpha}} \hat{\mathbf{z}}_i. \tag{7}$$

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

III. SPIN-WAVE THEORY

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

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

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

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

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

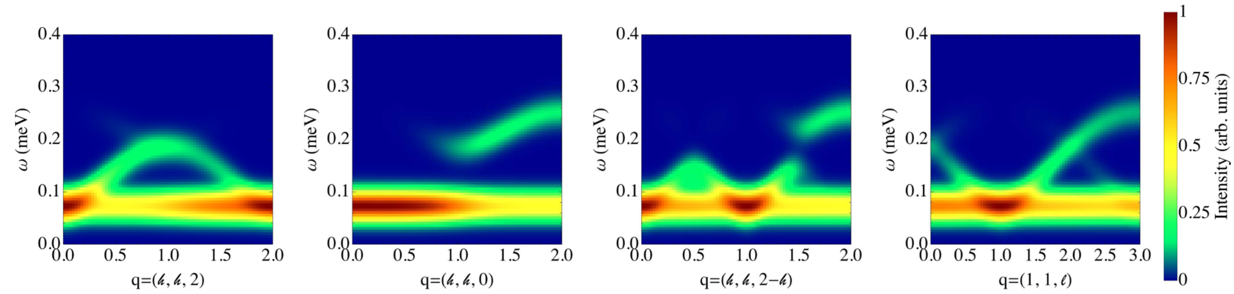
$$\tilde{\tau}_i^{-} = \tilde{\tau}_i^{\tilde{x}} - i\tilde{\tau}_i^{\tilde{y}} = a_i^\dagger \sqrt{(2S - a_i^\dagger a_i)} \approx \sqrt{2S} a_i^\dagger, \tag{11}$$

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 (LSW) Hamiltonian:

$$\mathcal{H}^{\text{DO}}_{\text{LSW}} = -3N|\tilde{J}_z|S^2 + 6|\tilde{\mathsf{J}}_z|S \sum_i a_i^\dagger a_i + \frac{S}{2}\sum_{\langle ij\rangle} (a_i^\dagger,\ a_i) \begin{pmatrix} \tilde{\mathsf{J}}_x + \tilde{\mathsf{J}}_y & \tilde{\mathsf{J}}_x - \tilde{\mathsf{J}}_y \\ \tilde{\mathsf{J}}_x - \tilde{\mathsf{J}}_y & \tilde{\mathsf{J}}_x + \tilde{\mathsf{J}}_y \end{pmatrix} \begin{pmatrix} a_j \\ a_j^\dagger \end{pmatrix}. \tag{12}$$

After Fourier transformation into momentum ($\mathbf{q}$) space and a subsequent Bogoliubov transformation [32,33] to a new set of bosonic operators $[b_{\lambda\mathbf{q}}, b^\dagger_{\lambda'\mathbf{q}'}] = \delta_{\mathbf{q}\mathbf{q}'}\delta_{\lambda\lambda'}$ we arrive at a



(a) $\mathcal{S}(\mathbf{q}=(h,h,2),\omega)$ (b) $\mathcal{S}(\mathbf{q}=(h,h,0),\omega)$ (c) $\mathcal{S}(\mathbf{q}=(h,h,2-h),\omega)$ (d) $\mathcal{S}(\mathbf{q}=(1,1,l),\omega)$

FIG. 2. Structure factor for inelastic neutron scattering [Eq. (14)] in $Nd_2Zr_2O_7$ calculated from linear spin-wave theory. Calculations are made from a linear spin-wave treatment of $\mathcal{H}^{\rm DO}_{XYZ}$ [Eq. (4)], assuming an all-in-all-out ground-state order and with exchange parameters given in Eq. (18). The flat band at $\Delta_{\rm flat} \approx 0.07$ meV contains the pinch-point structures shown in Fig. 1(b). The calculated spin-wave dispersion and intensities are in good agreement with the inelastic scattering measurements in Ref. [25]. The structure factor was calculated at $T = 0$ and convoluted with a Gaussian of full width at half maximum of 0.05 meV to mimic finite experimental resolution.

diagonalized Hamiltonian for four bands λ of bosons with dispersion $\omega_\lambda(\mathbf{q})$:

$$\mathcal{H}^{\rm DO}_{\rm LSW} = -3N|\tilde{J}_z|S(S+1) + \sum_{\mathbf{q},\lambda} \omega_\lambda(\mathbf{q})\left(b^\dagger_{\mathbf{q}\lambda} b_{\mathbf{q}\lambda} + \frac{1}{2}\right). \tag{13}$$

These four bands $\omega_\lambda(\mathbf{q})$ consist of two degenerate flat bands at energy $\Delta_{\rm flat} = \sqrt{(3|\tilde{J}_z| - \tilde{J}_x)(3|\tilde{J}_z| - \tilde{J}_y)}$ 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

$$\mathcal{S}(\mathbf{q},\omega) = \int dt\, e^{-i\omega t} \sum_{\mu\nu} \left(\delta_{\mu\nu} - \frac{q_\mu q_\nu}{q^2}\right) \times \langle m^\mu(-\mathbf{q},0) m^\nu(\mathbf{q},t)\rangle, \tag{14}$$

where $\mathbf{m}(\mathbf{q},t)$ is the lattice Fourier transform of the site magnetization [Eq. (5)] at time t.

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

$$\mathcal{S}(\mathbf{q},\omega) = \mathcal{S}^{(\tilde{z})}(\mathbf{q},\omega) + \mathcal{S}^{(\tilde{x})}(\mathbf{q},\omega). \tag{15}$$

The first contribution $\mathcal{S}^{(\tilde{z})}(\mathbf{q},\omega)$ comes from the ordered and static correlations of $\mathbf{m}_i^{(\tilde{z})}$ and gives rise to magnetic Bragg peaks at $\omega = 0$. The second contribution to $\mathcal{S}(\mathbf{q},\omega)$ in Eq. (15) comes from the dynamic correlations of $\mathbf{m}_i^{(\tilde{x})}$, which have the form

$$\mathcal{S}^{(\tilde{x})}(\mathbf{q},\omega) \approx \sin^2(\vartheta) g_z^2 \mu_B^2 \sum_\lambda s_\lambda(\mathbf{q}) \times [(1+n_B(\omega))\delta(\omega - \omega_\lambda(\mathbf{q})) + n_B(\omega)\delta(\omega + \omega_\lambda(\mathbf{q}))], \tag{16}$$

where $s_\lambda(\mathbf{q})$ are coefficients calculated from the Bogoliubov transformation.

Due to the flat bands, $\mathcal{S}^{(\tilde{x})}(\mathbf{q},\omega)$ exhibits a peak at $\omega = \Delta_{\rm flat}$ for all $\mathbf{q}$. The intensity of this flat-band peak is plotted in Fig. 1(b). For all choices of exchange parameters $\tilde{J}_\alpha$ within the all-in-all-out phase it exhibits a pattern of pinch-point singularities, as observed at finite energy in $Nd_2Zr_2O_7$ [25]. Our calculations thus simultaneously reproduce the observation of pinch points and the correct ground-state order for $Nd_2Zr_2O_7$, 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

$$\tilde{J}_x = 0, \quad \tilde{J}_y = -0.047 \text{ meV}, \quad \tilde{J}_z = 0.103 \text{ meV}. \tag{17}$$

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

$$\tilde{J}_x = 0.103 \text{ meV}, \quad \tilde{J}_y = 0, \quad \tilde{J}_z = -0.047 \text{ meV}. \tag{18}$$

This transformed set of parameters also gives equally good agreement with the inelastic spectrum while correctly reproducing the experimental ground state [34]. 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 (CW) temperature of $Nd_2Zr_2O_7$ [26–28,30], in spite of the antiferromagnetic ground state. Specifically, the Curie-Weiss temperature for the model in Eq. (4) is

$$T_{\rm CW} = \frac{1}{2k_B}(\tilde{J}_z \cos^2(\vartheta) + \tilde{J}_x \sin^2(\vartheta)), \tag{19}$$

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

$$\frac{m^{\rm ord}}{m^{\rm sat}} = \cos(\vartheta)\left(\frac{S - \langle a_i^\dagger a_i\rangle}{S}\right). \tag{20}$$

The spin-wave calculation gives us $(\frac{S-\langle a_i^\dagger a_i\rangle}{S}) \approx 0.87$. The fact that this number is close to unity is a good indicator that the linear spin-wave approach is valid. Combining this with the estimated value of ϑ gives an ordered moment fraction $\frac{m^{\text{ord}}}{m^{\text{sat}}} \approx 0.59$. This is close to the value $\frac{m^{\text{ord}}}{m^{\text{sat}}} \approx 0.5$ obtained in Ref. [27], but a bit higher than the value $\frac{m^{\text{ord}}}{m^{\text{sat}}} \approx 0.33$ obtained in Ref. [28]. It is interesting to note that most of this moment reduction comes from the pseudospin rotation ϑ, not from the zero-point fluctuations, as one might typically expect.

The theory presented in this article thus presents a consistent treatment of the ground state and the finite energy spectrum in $Nd_2Zr_2O_7$. 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].

IV. 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 magnetization density,

$$\mathbf{m} = \mathbf{m}_m + \mathbf{m}_d = \nabla\psi + \nabla\times\mathbf{A}, \tag{21}$$

where $\mathbf{m}_d = \nabla\times\mathbf{A}$ is divergence free $(\nabla\cdot\mathbf{m}_d = 0)$ and $\mathbf{m}_m = \nabla\psi$ is "divergence full." The fragmentation phenomenon is observed when magnetic order occurs in $\mathbf{m}_m$, but $\mathbf{m}_d$ remains fluctuating quasi-independently of $\mathbf{m}_m$. Since $\mathbf{m}_d$ obeys $\nabla\cdot\mathbf{m}_d = 0$ this gives rise to the pinch-point correlations associated with a Coulomb phase [12,22–24].

We can understand the magnetic fragmentation phenomenon in $Nd_2Zr_2O_7$ by defining fields $\mathbf{m}_i^{(\tilde{\alpha})}$ for each pseudospin component $\tilde{\tau}_i^{\tilde{\alpha}}$ according to Eq. (7) and then applying the Helmholtz decomposition to each one individually:

$$\mathbf{m}_i^{(\tilde{\alpha})} = \nabla\psi^{(\tilde{\alpha})} + \nabla\times\mathbf{A}^{(\tilde{\alpha})}. \tag{22}$$

(Note that $\mathbf{m}_i^{(\tilde{y})}$ does not contribute to the physical magnetization field $\mathbf{m}_i$ [Eq. (6)].)

In the all-in-all-out ground state, $\mathbf{m}_i^{(\tilde{z})}$ is completely divergence full, and we may write $\mathbf{A}^{(\tilde{z})} = 0$. The fluctuations of $\mathbf{m}_i^{(\tilde{x})}$ and $\mathbf{m}_i^{(\tilde{y})}$, 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 $\mathbf{m}_i^{(\tilde{x})},\mathbf{m}_i^{(\tilde{y})}$.

Writing down the Heisenberg equations of motion for $\mathbf{m}_i^{(\tilde{x})}$ and $\mathbf{m}_i^{(\tilde{y})}$ and linearizing around the all-in-all-out ground state we find

$$\partial_t\mathbf{m}_i^{(\tilde{\alpha})} \approx \varepsilon_{\tilde{\alpha}'\tilde{\alpha}\tilde{z}} S(\tilde{\mathsf{J}}_{\alpha'}\nabla_i(\nabla\cdot\mathbf{m}^{(\tilde{\alpha}')}) + (6|\tilde{\mathsf{J}}_z| - 2\tilde{\mathsf{J}}_{\alpha'})\mathbf{m}_i^{(\tilde{\alpha}')}), \tag{23}$$

where $\tilde{\alpha}' = \tilde{y}$ when $\tilde{\alpha} = \tilde{x}$ and vice versa and $\varepsilon_{\tilde{x}\tilde{y}\tilde{z}} = -\varepsilon_{\tilde{y}\tilde{x}\tilde{z}} = 1$. In Eq. (23), ∇ 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].

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

$$\partial_t\psi^{(\tilde{\alpha})} = \varepsilon_{\tilde{\alpha}'\tilde{\alpha}\tilde{z}} S(\tilde{\mathsf{J}}_{\alpha'}\nabla^2\psi^{(\tilde{\alpha}')} + (6|\tilde{\mathsf{J}}_z| - 2\tilde{\mathsf{J}}_{\alpha'})\psi^{(\tilde{\alpha}')}), \tag{24}$$

$$\partial_t\mathbf{A}^{(\tilde{\alpha})} = \varepsilon_{\tilde{\alpha}'\tilde{\alpha}\tilde{z}} S(6|\tilde{\mathsf{J}}_z| - 2\tilde{\mathsf{J}}_{\alpha'})\mathbf{A}^{(\tilde{\alpha}')}. \tag{25}$$

The important feature of Eqs. (24) and (25) is that the divergenceless fluctuations (i.e., fluctuations of $\mathbf{A}^{(\tilde{\alpha})}$) are completely decoupled from the divergence-full fluctuations (i.e., fluctuations of $\psi^{(\tilde{\alpha})}$). Fluctuations of $\mathbf{A}^{(\tilde{\alpha})}$ form a flat band at energy $\Delta_{\text{flat}} = \sqrt{(3|\tilde{\mathsf{J}}_z| - \tilde{\mathsf{J}}_x)(3|\tilde{\mathsf{J}}_z| - \tilde{\mathsf{J}}_y)}$, while fluctuations of $\psi^{(\tilde{\alpha})}$ form dispersive bands.

The physical magnetization field in $Nd_2Zr_2O_7$ [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 magnetization field which is strictly Ising like (in the sense that $\mathbf{m}_i$ 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 [25]. Above the transition, $\mathbf{m}_i^{(\tilde{x})}$ can fluctuate for little or no energy cost but its correlations will remain icelike due to the positive value of $\tilde{\mathsf{J}}_x$.

V. CONCLUSIONS

In conclusion we have explained the quantum origins of the moment fragmentation in $Nd_2Zr_2O_7$, observed in Ref. [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 $\mathcal{H}_{XYZ}^{\text{DO}}$ 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 $\mathcal{H}_{XYZ}^{\text{DO}}$, 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 $Nd_2Sn_2O_7$ [35], $Nd_2Hf_2O_7$ [36], and possibly $Nd_2Pb_2O_7$ [37].

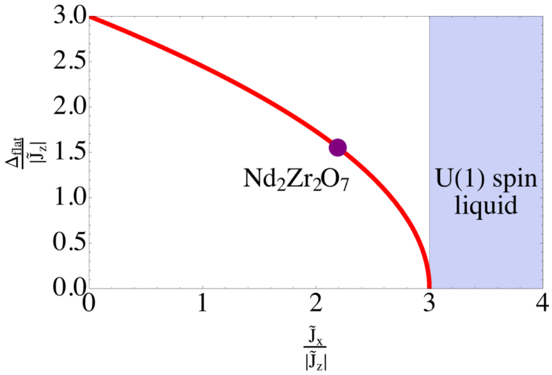


FIG. 3. Closing of the flat band gap $\Delta_{\rm flat}$ with increasing $\tilde{J}_x$ and the proximity of a $U(1)$ quantum spin-liquid phase, shown for $\tilde{J}_y = 0$. The gap to the flat band which contains the Coulomb-phase-like correlations of Fig. 1(b) closes when the ratio $\frac{\tilde{J}_x}{|\tilde{J}_z|} = 3$. This is a likely indicator of the onset of a $U(1)$ spin-liquid phase. Based on the parametrization of Eq. (18) this ratio is $\frac{\tilde{J}_x}{|\tilde{J}_z|} \approx 2.19$ in $Nd_2Zr_2O_7$, suggesting the possibility of observing a $U(1)$ spin-liquid ground state $Nd_2Zr_2O_7$ or related Nd-based pyrochlores, induced by application of chemical or physical pressure.

The parametrization of the exchange Hamiltonian $\mathcal{H}_{XYZ}^{\rm DO}$ [Eq. (4)] given in Eq. (18) suggests that $Nd_2Zr_2O_7$ is proximate to the $U(1)$ spin-liquid phase which has been long sought among "quantum spin ice" pyrochlores [14,16,38–44]. As shown in Fig. 3, the closing of the gap to the flat band containing the pinch-point correlations occurs at $\frac{\tilde{J}_x}{|\tilde{J}_z|} = 3$ within linear spin-wave theory. Classically, this would signal the formation of an extensive ground-state manifold with icelike character, but the mixing of these states by quantum fluctuations is known to stabilize a $U(1)$ spin liquid with dynamic emergent gauge fields [38,41]. The placement of $Nd_2Zr_2O_7$ 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 $Nd_2Zr_2O_7$ 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 [45,46]. We therefore have reason to hope that experiments on $Nd_2Zr_2O_7$ and related materials may yet reveal even more exotic phenomena.

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