

Braided Ising spin-tube physics in a purported kagome magnetJ. Nagl^{1,*}, D. Flavián¹, B. Duncan¹, S. Hayashida², O. Zaharko³, E. Ressouche⁴, J. Ollivier⁵, Z. Yan¹, S. Gvasaliya¹, and A. Zheludev^{1,†}¹Laboratory for Solid State Physics, *ETH Zürich*, 8093 Zürich, Switzerland²Neutron Science and Technology Center, Comprehensive Research Organization for Science and Society (CROSS), Tokai, Ibaraki 319-1106, Japan³Laboratory for Neutron Scattering and Imaging, *PSI Center for Neutron and Muon Sciences*, Forschungsstrasse 111, 5232 Villigen, PSI, Switzerland⁴Université Grenoble Alpes, CEA, IRIG, MEM, MDN, 38000 Grenoble, France⁵Institut Laue-Langevin, 71 Avenue des Martyrs, CS 20156, 38042 Grenoble Cedex 9, France

(Received 12 September 2024; revised 17 March 2025; accepted 6 May 2025; published 16 May 2025)

The magnetic insulator Nd_3BWO_9 has been previously proposed to realize the highly frustrated breathing kagome lattice model. We report a combination of single-crystal neutron scattering studies and numerical simulations that debunk this interpretation. We show that it is the interplane couplings that determine the physics. To explain the exotic magnetism, we derive a simple one-dimensional Ising model composed of twisted triangular spin tubes, i.e., triple braids of Ising spin chains with almost-orthogonal anisotropy frames and competing ferro-antiferromagnetic interactions. This model can account for the ground state, excitations, the numerous field-induced fractional magnetization plateau phases, and incommensurate magnetic correlations at elevated temperatures. Nd_3BWO_9 constitutes a rare example where rich magnetic phenomena can be understood and simulated quantitatively in terms of a simple classical Hamiltonian.

DOI: [10.1103/PhysRevB.111.L180406](https://doi.org/10.1103/PhysRevB.111.L180406)

Spin systems with strong geometric frustration are the most likely to host exotic quantum magnetic phases [1]. One of the oldest known and most important models of this type is an antiferromagnet (AFM) on the so-called kagome lattice. It can be viewed as a partially depleted triangular lattice, but has a much higher degree of frustration. Precious few experimental realizations of kagome AFMs have been found to date. Unfortunately, their physics is complicated by either additional “undesirable” terms in the Hamiltonian [2,3] or intrinsic structural disorder [4]. This is one reason why the recent discovery of an entirely new family of rare-earth compounds R_3BWO_9 [5] featuring disorder-free “breathing-kagome” [6] arrangements of magnetic ions has caused a great deal of excitement and intense experimental investigation. As hoped for, the latter revealed exotic frustration phenomena and complex magnetic phase diagrams [7–12].

In this Letter, we report single-crystal neutron scattering, magnetic torque, and dilatometry studies, as well as numerical simulations for one particular species, namely Nd_3BWO_9 [7–9]. We conclude that, due to peculiarities of single-ion electronic configurations, the breathing-kagome spin arrange-

ment is largely irrelevant. The highly peculiar magnetic properties, including a plethora of magnetization-plateau phases and a commensurate-to-incommensurate transition, are due to an entirely different mechanism of frustration. The system is best described as braids of Ising spin chains with misaligned and almost-orthogonal anisotropy frames for individual spins.

Like other members of the series, Nd_3BWO_9 crystallizes in hexagonal $P6_3$ structure. From the schematic shown in Fig. 1(a), it is easy to see why it has been discussed in the context of breathing-kagome physics. The magnetic Nd^{3+} ions indeed realize this type of lattice in the ab plane of the crystal. The two in-plane nearest-neighbor Nd-Nd distances are 4.25 Å and 4.90 Å, respectively. In Fig. 1 the respective exchange constants are labeled J_Δ and J_∇ . Consistent with the resulting high degree of frustration, Nd_3BWO_9 shows a significant ratio of Weiss to Néel temperature $|\theta_{\text{CW}}|/T_N \gtrsim 10$, with AFM order observed only below $T_N \approx 0.3$ K. This picture is, however, incomplete. The shortest Nd-Nd bonds, 3.94 Å, [J_1 and J'_1 in Figs. 1(a) and 1(b) with inequivalent Nd-O-Nd superexchange pathways] actually connect magnetic ions from adjacent planes [7]. Each of these bonds forms bundles of three intertwined spin chains running along the c axis, but twisted in opposite directions [Fig. 1(b)]. Below, we shall demonstrate that it is this one-dimensional “twisted-spin-tube” arrangement rather than the breathing-kagome structure that plays a decisive role for the magnetism.

To understand the character of magnetic interactions, we first analyze the electronic state of individual ions. In Nd_3BWO_9 , the Nd^{3+} ions are in $4f^3$ electronic configuration

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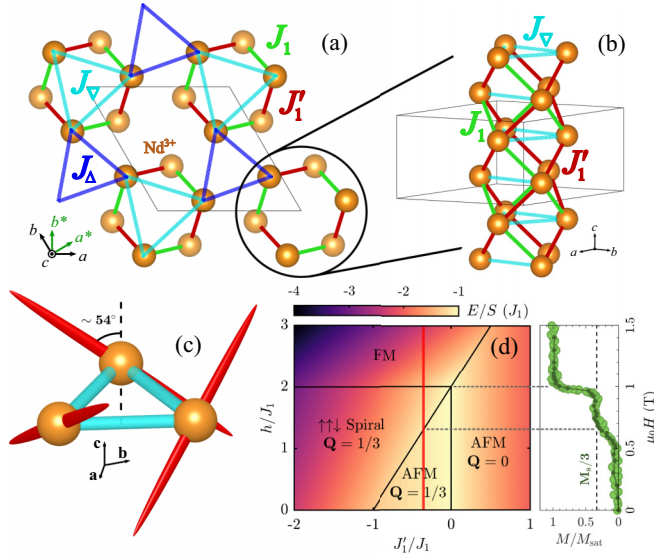


FIG. 1. (a) Exchange pathways in Nd_3BWO_9 , emphasizing the breathing kagome structure realized in the ab plane. (b) The shortest bonds connect Nd^{3+} ions in adjacent kagome planes through inequivalent superexchange pathways $(J'_1)J_1$, forming triple braids of spin-chains, i.e., frustrated triangular spin tubes, running (counter)clockwise along the c axis. (c) Schematic of the Ising-like g -tensor ellipsoids for a triangle of spins, depicting the nearly orthogonal anisotropy frames. (d) Phase diagram of the single spin-tube model Eq. (1) in an axial magnetic field. The exchange ratio $J'_1/J_1 \simeq -0.35$ (red line) can be estimated from the relative width of the $m_z = 1/3$ plateau in the magnetization data shown on the right [7].

and have trivial point symmetry (C_1), being caged by eight O^{2-} ions. Previously reported high-energy inelastic neutron scattering (INS) spectra [7] show the first crystal electric field (CEF) excitation at 15.6 meV. Thus, only the lowest-energy Kramers doublet is relevant to the low-temperature properties. The low symmetry makes a direct determination of the 27 independent crystal field parameters unfeasible. Instead, we derive an approximate single-ion Hamiltonian from a point-charge calculation [13]. We model the crystal electric field produced by the neighboring anions, approximating them as idealized point charges. For rare earth ions this approach is known to work very well [14]. Assuming a charge $-2e$ for each O^{2-} site already gives an agreement with observed excitation energies to within $\sim 20\%$. By fitting the magnitude of the charge for each of the three inequivalent sites we obtain a near perfect agreement with INS data [15]. The thus-determined CEF Hamiltonian can adequately reproduce both the temperature dependence of the excitations as observed with neutron spectroscopy, and the single-crystal magnetometry data down to a temperature of 2 K without any additional fitting [15].

The ground state Kramers doublet is composed almost exclusively of the maximal angular momentum states $|j^z = \pm 9/2\rangle$, where the local quantization axes z form an angle $\theta \simeq 54.4^\circ$ with the crystallographic c axis. For adjacent ions, these local anisotropy frames are rotated around $\mathbf{c}$ according to sixfold symmetry. As a result, and that is a crucial point, the anisotropy axes of nearest-neighbor ions in the breathing-

kagome plane are almost orthogonal, forming an angle $\phi \simeq 89^\circ$ relative to one another [see Fig. 1(c)]. The g tensor of the ground state doublet is extremely anisotropic. From our CEF model we estimate $g_{zz}/g_\perp \simeq 20$ in the local frame of each ion and $g_{zz} \simeq 6.19$. The near orthogonality of anisotropy axes results in an almost isotropic magnetic susceptibility for the bulk sample [7].

The single-ion ground state imposes strict constraints on the magnetic interactions. The latter can be written as a general bilinear form $\sim \hat{j}_i^\alpha K_{ij}^{\alpha\beta'} \hat{j}_j^{\beta'}$ of the interacting moments $\hat{j}$ in the local anisotropy frames of the two coupled ions. Since the transverse angular momentum components $\hat{j}^\pm$ only change j^z by unity, they have no matrix elements within the ground state doublet [15]. As a result, exchange interactions are rendered classical, taking an Ising form $\sim \hat{j}_i^z K_{ij}^{zz} \hat{j}_j^z$ where z and z' refer to the local principal anisotropy axes. Furthermore, if we disregard the possibility of asymmetric (Dzyaloshinskii-Moriya) interactions, $K^{zz'}$ must vanish by symmetry when the respective z axes are orthogonal. The far-reaching consequence is that the in-plane interactions J_Δ and J_∇ are suppressed and can be neglected.

What remains are nearly decoupled twisted Ising spin tubes. Representing every Kramers doublet with a $S_{\text{eff}} = 1/2$ pseudospin, we arrive at an effective spin Hamiltonian for each tube:

$$\mathcal{H}_{\text{eff}} = \sum_{i,j} (J_1 \hat{S}_{i,j}^z \hat{S}_{i+1,j}^z + J'_1 \hat{S}_{i,j}^z \hat{S}_{i+1,(j-1)\bmod 3}^z) - g_{zz} \mu_0 \mu_B \sum_{i,j} \mathbf{H} \cdot \hat{\mathbf{z}} \hat{S}_{i,j}^z. \quad (1)$$

Here the index i labels consecutive spins in each chain and $j = \{0, 1, 2\}$ refers to the three chains. In all cases, $\hat{\mathbf{z}} = \hat{\mathbf{z}}_{i,j}$ are unit vectors along the local anisotropy axes and g_{zz} refers to the principal component of the g tensor.

First, we consider the ground state of this tube model. We evaluate the energies of all possible spin configurations, assuming a unit cell periodicity of ≤ 4 along the c axis (up to 24 spins per tube). The resulting phase diagram for a field applied along the c axis is depicted in Fig. 1(d). In zero field, one finds three distinct phases. For $J_1, J'_1 > 0$ the model is unfrustrated, realizing an AFM state with a 1D propagation vector $q = 0$. Couplings of opposing sign $J'_1/J_1 < 0$ result in a tripling of the lattice period $q = 1/3$. A dominant FM J'_1 yields an $\uparrow\uparrow\downarrow$ spiral ground state with uniform magnetization $m_z = 1/3$ [Fig. 2(b)], whereas a dominant AFM J_1 results in a nonmagnetized phase with AFM stacking along the J_1 bonds and $\uparrow\uparrow\downarrow\downarrow\downarrow$ -type correlations along J'_1 [Fig. 2(a)]. The latter is the only state consistent with the experimentally observed $\mathbf{q} = (0, 0, 1/3)$ 3D propagation vector in Nd_3BWO_9 and compensated magnetization in zero field [7], and thus a strong contender for the magnetic ground state.

To check this against experiment, we performed detailed single crystal neutron diffraction studies to determine the magnetic structure in zero field. The data were collected on the lifting counter instruments D23 at ILL (France) [16] and ZEBRA at PSI (Switzerland). Experimental details and group-theory based data analysis procedures are described in the Supplemental Material [15]. The obtained magnetic structure belongs to the Γ_2 representation of the symmetry group. This

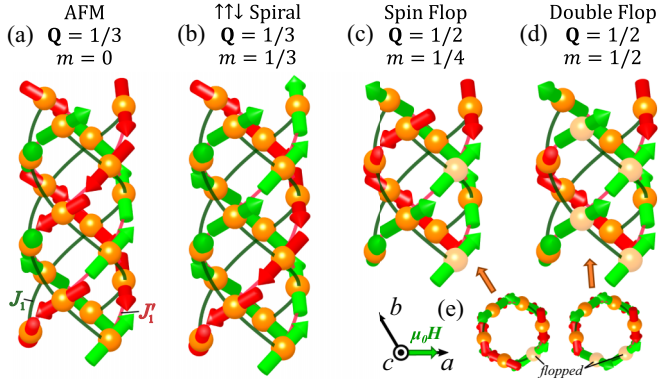


FIG. 2. Magnetic structures realized for our single spin-tube model, including (a) the AFM state (zero field), (b) the $\uparrow\uparrow\downarrow$ spiral phase ($\mathbf{H} \parallel \mathbf{c}$), and (c), (d) the spin-flop (SF) and double spin-flop (DSF) phases ($\mathbf{H} \perp \mathbf{c}$). (e) Top-down view of the latter two structures.

is consistent with the AFM $q = 1/3$ phase predicted for our Ising tube model [see Fig. 2(a)]. Moreover, the refined orientations of all magnetic moments in Nd_3BWO_9 is less than 4° away from the easy axes predicted by the point-charge model.

We conclude that Nd_3BWO_9 is in the frustrated regime $J_1 > -J'_1 > 0$. While each individual chain is clearly unfrustrated, these chains are braided together, intertwining at every third lattice position (nonbipartite) to give rise to a highly frustrated exchange topology. In an axial magnetic field $\mathbf{H} \parallel \mathbf{c}$, the $\uparrow\uparrow\downarrow$ spiral phase is stabilized before the fully polarized ferromagnetic state, resulting in a fractional $m_z = 1/3$ magnetization plateau with $q = 1/3$, as seen experimentally. From the width of this plateau phase $h_c/h_{\text{sat}} = 1 + J'_1/J_1 \simeq 0.65$ [see Fig. 1(d)], we can estimate the exchange ratio $J'_1/J_1 \approx -0.35(5)$ for the spin-tube couplings. This is very close to the value $J'_1 \approx -0.37J_1$ determined in the sister compound Pr_3BWO_9 from INS data [10]. Of course, our purely 1D model is unable to account for the stacking of adjacent tubes in the 3D crystal, which is governed by weak residual intertube exchange or dipolar interactions [15].

Additional plateau phases appear in magnetic fields applied in the plane. Figures 3(a)–3(c) summarizes magnetization curves measured using Faraday balance and neutron diffraction magnetometry (NDM) [7]. For $\mathbf{H} \parallel \mathbf{a}$, new NDM data were collected using the ZEBRA diffraction setup with a 1.8 T horizontal cryomagnet, by tracking the field dependence of the (002) structural reflection [15]. The prominent plateau seen in low transverse fields has been previously assigned fractional values of $m_z = 1/4$ or $m_z = 1/3$ [7,8]. The new measurements and a careful re-examination of data of Ref. [7] clearly favor $m_z = 1/4$. An additional weak kink can be seen around $\mu_0 H \sim 1.2$ T, pointing to yet another plateau, with $m_z = 1/2$. It is barely visible in previous Faraday-balance experiments [7], but quite prominent in the new NDM dataset. That it corresponds to a distinct thermodynamic phase is separately confirmed by magnetic torque and dilatometry measurements [15]. While the presence of a multitude of plateau states is not unheard of in highly frustrated 2D geometries such as Shubnikov-Sutherland [17,18] or kagome [19–21], there have been few 1D examples reported to date.

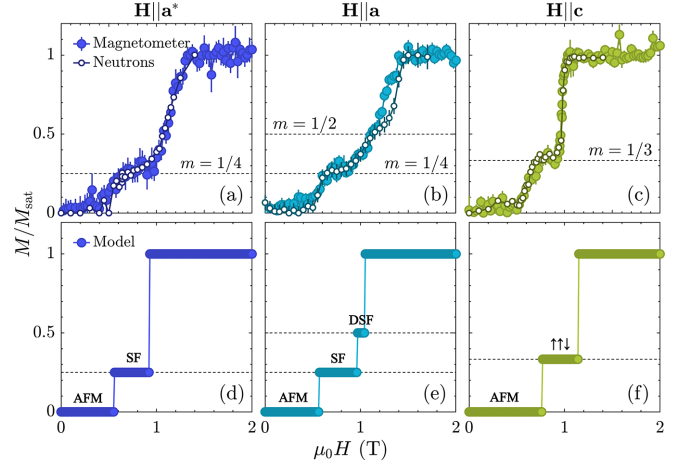


FIG. 3. (a)–(c) Magnetization curves in Nd_3BWO_9 taken at $T \lesssim 120$ mK for various field directions using Faraday balance magnetometry (large symbols) and neutron diffraction (small symbols). The neutron data in (b) are from this work, the rest is from Ref. [7]. (d)–(f) Model calculations of the same magnetization curves using $J_1 = 0.24$ meV and $J'_1 = -0.35J_1$, showing excellent qualitative agreement for all phases. In (e) we assume a 4° misalignment of the field in the ac plane.

All these plateau phases naturally occur in our Ising spin-tube model. Due to the canted anisotropy frames, all six Nd^{3+} ions in the unit cell have different g -factors $g_{\text{eff}} \approx g_{zz} \hat{\mathbf{H}} \cdot \hat{\mathbf{z}}_i$. As a result, a magnetic field applied in a general direction may influence some sites more than others, favoring different spin arrangements. For example, a $m_z = 1/4$ state with $q = 1/2$ is stabilized for all planar orientations $\mathbf{H} \parallel \mathbf{ab}$, shown in Fig. 2(c). It corresponds to a spin-flop (SF) phase where one spin with the largest g -factor is oriented along the field, while all others are stacked antiferromagnetically. A $m_z = 1/2$ state with $q = 1/2$ may occur at higher fields but is more fragile. For a strictly in-plane field it is only stable for a narrow range of orientations centered around $\phi \simeq 6.5^\circ$ from $\mathbf{H} \parallel \mathbf{a}$ (modulo 60°) [15]. This special angle corresponds to a high symmetry configuration where four ions share the same effective g -factor and the remaining two are fully decoupled from the field. Two spins become aligned with the field in this “double spin-flop” (DSF) phase, while the others remain staggered, as shown in Fig. 2(d). Although this phase is not realized for $\mathbf{H} \parallel \mathbf{a}$ exactly, it is stabilized by a small out-of-plane field component, appearing already for a tiny $\sim 2^\circ$ misalignment in the c plane. This is well within the precision of aligning the sample in a cryomagnet and explains why this phase is repeatedly seen experimentally.

Not only the presence of all these phases, but also the corresponding transition fields are quantitatively reproduced by our model. The computed magnetization curves are plotted in Figs. 3(d)–3(f) for a direct comparison with the experiment. Here we keep $J'_1/J_1 = -0.35$ fixed and use $J_1 = 0.24$ meV as precisely determined through neutron spectroscopy (see below). In order to reproduce the observed $m = 1/2$ plateau, we assumed a 4° misalignment of the field with the a axis in the ac plane.

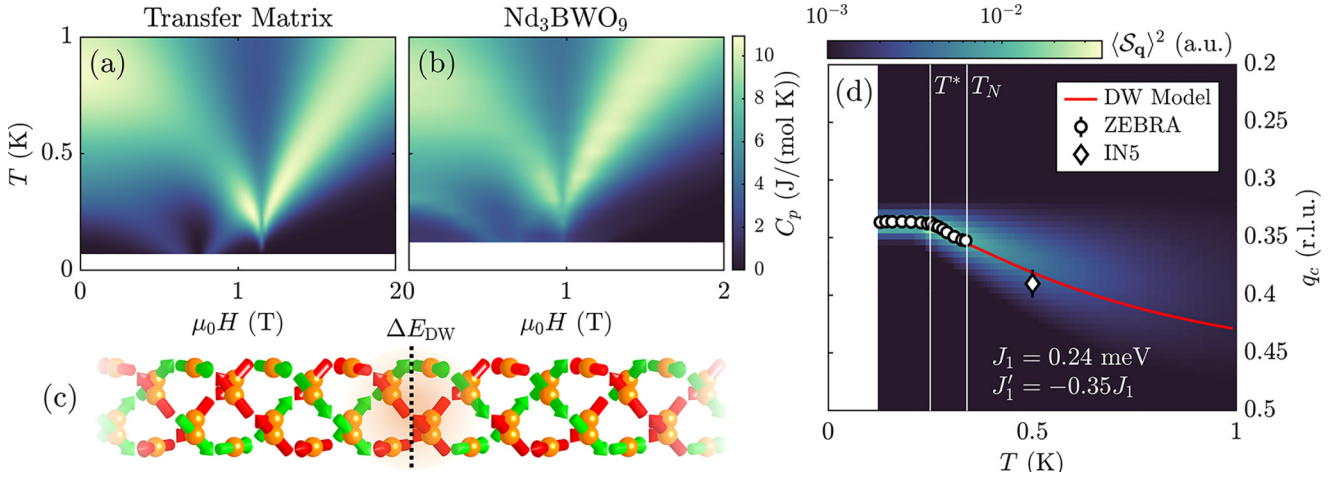


FIG. 4. (a) Numerical transfer matrix calculation of the magnetic specific heat C_m in an axial field $\mathbf{H} \parallel \mathbf{c}$, visualized as a false color plot. (b) Direct comparison to experimental heat capacity in Nd_3BWO_9 for the same configuration (data taken from [7]). (c) Schematic of a domain wall defect. Notice how the $\uparrow\uparrow\uparrow/\downarrow\downarrow\downarrow$ pattern on every chain gives way to $\uparrow\uparrow\uparrow\uparrow$ and $\downarrow\downarrow\downarrow\downarrow$ clusters across the domain wall. (d) The c axis component of the magnetic propagation vector against temperature. Symbols: data from neutron scattering (circles: [7]; diamond: this work). Red line: the Boltzmann domain wall model, as described in the text. False color plot: spin correlation function from a classical Monte Carlo simulation.

A purely one-dimensional model with short range interactions is unable to support long-range order at $T > 0$. And yet, the finite-temperature thermodynamics of Nd_3BWO_9 is completely dominated by spin-tube physics. Specific heat computed in transfer-matrix simulations for a single spin tube is shown in Fig. 4(a) as false-color plot against axial field and temperature [15]. For comparison, experimental results from Ref. [7] are shown in Fig. 4(b). All the intricate features of the experiment are reproduced. The only differences are the sharper λ anomaly at T_N in the measurement and the small shift in the transition fields. Both discrepancies are due to interchain interactions and three-dimensional long range order in the real material. A similar match is achieved for the magnetic entropy [15]. The strength of interchain interactions J_{eff} can be estimated by treating them at the mean-field (MF) level [15]. Monte Carlo simulations of Eq. (1) are used to compute the single-tube susceptibility at the ordering wave vector at a temperature equal to T_N of the real material. Solving a Weiss-style self-consistent MF equation we get $J_{\text{eff}} \sim 0.2$ K, about 7% of the leading spin-tube exchange, in support of the weakly-coupled spin-tube model for Nd_3BWO_9 .

Next we turn to the low-energy spin excitations in Nd_3BWO_9 , measured on the IN5 cold-neutron disk-chopper spectrometer at ILL [22,23]. Probing these with INS is extremely challenging, because transitions between the $|j^z = \pm 9/2\rangle$ states expected to dominate this regime are forbidden by selection rules. Only the small $\sim 4\%$ admixing of lower angular momentum states into the single-ion ground state wavefunctions contributes to a finite scattering intensity. The spectrum is composed of two flat modes at $E_1 = 0.240(2)$ meV and $E_2 = 0.306(2)$ meV, respectively (see [15]). This is consistent with expectations for an Ising magnet, where the dynamics is entirely local and due to flips of single spins. In our spin-tube model, these modes correspond to flipping a spin at the edge (in the center) of an $\uparrow\uparrow\uparrow$ or $\downarrow\downarrow\downarrow$ spin cluster, costing an energy $E_1 = J_1$ ($E_2 =$

$J_1 - J'_1$), respectively. This provides a direct measure of the dominant exchange parameters $J_1 \approx 0.240(2)$ meV and $J'_1 \approx -0.28(2)J_1$, in good agreement with our previous estimate.

One of the lingering puzzles about Nd_3BWO_9 was the incommensurate magnetic structure observed in zero field at elevated temperatures [7]. The previously measured c -axis component of the magnetic propagation vector is plotted vs temperature in circles in Fig. 4(d), showing a commensurate-lock-in transition at $T^* \simeq 0.25$ K. As revealed by new neutron diffraction scans at even higher temperatures [15], in the paramagnetic state, the incommensurate Bragg reflections give way to a broad diffuse scattering peak. It too is incommensurate, the position being indicated by a diamond symbol in Fig. 4(d).

For a quasi-1D system it is tempting to interpret incommensurate structures as signs of a quantum longitudinal spin density wave state [24–26]. Our classical Ising spin-tube model provides a much simpler explanation. In a 1D Ising magnet, the low-energy dynamics is dominated by domain walls—kinks in the magnetic structure that act as phase slips in the local order parameter. These kinks are largely suppressed as $T \rightarrow 0$, but start to proliferate at finite temperatures, effectively changing the average periodicity of the magnetic structure and leading to incommensurate correlations. This is analogous to the modulated behavior seen in the axial next-nearest-neighbor Ising model, where entropically promoted soliton defects can form an extended incommensurate regime at elevated temperatures [27,28]. In $d \geq 3$ dimensions, these solitons may even crystallize to form a “Devil’s staircase”—a cascade of infinitely many commensurate phases.

For our model, the lowest-energy kink is depicted in Fig. 4(c) and has an energy cost of $\Delta E = (J_1 + J'_1)/2 \approx 0.9$ K. It corresponds to a phase slip of $\pi/3$ in the magnetic structure. This allows for six different types of kink-bounded domains. Toward low temperatures (i.e., the dilute limit),

we can ignore higher-lying excitations and attempt to calculate the change in the averaged lattice period by assuming Boltzmann statistics for the number of kinks $\langle n \rangle$. The resulting periodicity of magnetic correlations $\langle q \rangle = \frac{1+\langle n \rangle/N}{3}$ [15] is plotted as a red line in Fig. 4(d). There is a clear crossover from commensurate to incommensurate behavior at T^* , in excellent agreement with experiment. As a crosscheck, we can reproduce this effect by performing classical Monte Carlo (MC) simulations. We model a single spin tube of $L = 192$ unit cells (i.e., $\sim 10^4$ spins) with a simulated annealing procedure, keeping $J_1 = 0.24$ meV and $J'_1 = -0.35J_1$. In Fig. 4(d) we show the calculated magnetic structure factor $\langle S_q \rangle^2$ as a false colorplot. Just as for the Boltzmann-domain wall model, the crossover at T^* to incommensurate behavior is reproduced with no free parameters and a quasilinear increase in q that closely matches the experiment is observed at high temperatures.

In conclusion, despite previous speculations [5,8,9], Nd_3BWO_9 is hardly a model compound for kagome-lattice magnetism. It is an essentially classical quasi-1D Ising magnet with a peculiar frustration geometry of bundles of braided spin chains and an accidental cancellation of kagome-plane interactions. This unique geometry makes for a rare case where extremely rich magnetic behavior can be understood and simulated quantitatively to astounding accuracy.

This work is supported by a MINT grant of the Swiss National Science Foundation. A.Z. thanks Dr. M. Zhitomirsky (CEA Grenoble) and Dr. I. Zaliznyak (Brookhaven National Lab) for insightful discussions. We also thank Prof. Titus Neupert and Dr. Nikita Astrakhantsev (University of Zürich) for illuminating discussions. We acknowledge the beam time allocation at PSI (ZEBRA ID 20220898 and 20240065) and ILL (D23 ID CRG-3011, IN5 ID 4-05-861).

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