Disorder-Induced Mimicry of a Spin Liquid in YbMgGaO_{4}

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Disorder-induced mimicry of a spin liquid in $YbMgGaO_4$

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We suggest that a randomization of the pseudo-dipolar interaction in the spin-orbit-generated low-energy Hamiltonian of $YbMgGaO_4$ due to an inhomogeneous charge environment from a natural mixing of Mg^{2+} and Ga^{3+} can give rise to orientational spin disorder and mimic a spin-liquid-like state. In the absence of such quenched disorder, $1/S$ and DMRG calculations both show robust ordered states for the physically relevant phases of the model. Our scenario is consistent with the available experimental data and further experiments are proposed to support it.

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Dating back to Wannier's pioneering study of the Ising model [1], triangular lattice models and materials with frustrating antiferromagnetic interactions have served as fertile playgrounds for new ideas [2–10]. These systems continue to draw significant experimental [11–15] and theoretical interest because they exhibit many intriguing novel ordered states [16–22] and unusual continuum-like spectral features [23–31], and especially because they provide a setting for spin-liquid states [32–42].

Among the latest experimental discoveries [14, 15], a rare-earth triangular-lattice antiferromagnet $YbMgGaO_4$ has recently emerged as a new candidate for a quantum spin liquid of the effective spin-1/2 degrees of freedom of Yb^{3+} ions [43, 44]. It has been argued that the spin-orbit origin of its magnetic properties and the pseudo-spin nature of the low-energy states with highly anisotropic effective spin interactions may potentially open a new route to realizing quantum spin liquids [44–46]. While the lack of ordering, anomalous specific heat, and especially continuum-like excitations in inelastic neutron scattering [45, 47] all provide strong support to the idea of an intrinsic spin liquid, other experimental findings are increasingly at odds with this picture.

First, in magnetization vs field measurements, there is no sharpening of the transition to the saturated phase upon lowering temperature and the lack of the upward curvature in $M(H)$ at the lowest T's [43, 44] is indicative of low quantum fluctuations in the ground state [49]. Second, in the high-field polarized phase, neutron scattering shows that continuum-like excitations persist, with significant smearing of magnon lines that are expected to be sharp [47]. In addition, an apparent absence of any detectable contribution of spin excitations to thermal conductivity down to the lowest temperatures, accompanied by a strong deviation of the phonon part from the ballistic T^3 form [50], both suggest strong scattering effects. These, combined with the anomalously broadened higher-energy Yb^{3+} doublet structure [47, 48] and a ubiquitous mixing of Mg^{2+} and Ga^{3+} ions in the non-magnetic layers [43, 47], implicate disorder as a key contributor to the observed properties [48].

In this Letter, we first argue that a hypothetical, disorder-free version of $YbMgGaO_4$ should exhibit a robust collinear/stripe magnetic order. We demonstrate this by extending the well-studied phase diagram of the triangular-lattice Heisenberg $J_1 - J_2$ model, which is known to have an extensive spin-liquid region for $S = 1/2$ [33–40], to the anisotropic version of the model that corresponds to the types of anisotropy allowed in $YbMgGaO_4$ with realistic restrictions from experiments. A significant XXZ anisotropy present in $YbMgGaO_4$ suppresses the spin-liquid region of the phase diagram and the pseudo-dipolar interactions further diminish it. Both types of anisotropy lower the symmetry and produce gaps in the excitation spectra, reducing quantum fluctuations that suppress the ordered states.

We then suggest that the stripe order is fragile to an orientational disorder that can be easily produced via a randomization of the subleading pseudo-dipolar interactions. The physical reason of such a sensitivity is a small energetic barrier, $\delta E \sim 0.03 J_1$ per site, between the stripe phases of different spatial orientations, which, in the absence of the pseudo-dipolar terms, are selected by order-by-disorder fluctuations. Thus, we propose that the spin-liquid-like state in $YbMgGaO_4$ is disorder-induced and is composed of nearly-classical, orientationally-randomized, short-range stripe-like spin domains. The quenched, spatially-fluctuating charge environment of the magnetic Yb^{3+} ions due to random site occupancies of Mg^{2+} and Ga^{3+} ions is seen as a likely culprit, affecting the low-energy effective spin Hamiltonian through the spin-orbit coupling.

Model.—Although the magnetism of $YbMgGaO_4$ is dominated by spin-orbit coupling, which can result in large spin anisotropies of various types [51–54], it is restricted by the high symmetry of the lattice [44, 45], yielding the familiar XXZ anisotropy accompanied by the so-called pseudo-dipolar terms. Moreover, the local character of the f-shells on Yb dictates that the dominant interactions are between the nearest-neighbor spins, further restricting possible spin models.

Thus, we are compelled to explore the phase diagram of the following $S = 1/2$ model as relevant to $YbMgGaO_4$ [43–45, 47] and also to a broader family of the rare-earth triangular-lattice materials [55]: $\mathcal{H}=\mathcal{H}_{\rm XXZ}^{J_1-J_2}+\mathcal{H}_{\rm pd}$, with

$$\mathcal{H}_{\rm XXZ}^{J_1-J_2} = \sum_{\langle ij\rangle_n} J_n \left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z\right), \qquad (1)$$

where the sums are over the (next-)nearest-neighbors with $J_1 > J_2 \geq 0$, the XXZ anisotropy $0 \leq \Delta \leq 1$, and the pseudo-dipolar terms introduced as [44, 45, 47]

$$\mathcal{H}_{\rm pd} = J_{\pm\pm} \sum_{\langle ij\rangle} \left(e^{i\tilde{\varphi}_\alpha} S_i^+ S_j^+ + e^{-i\tilde{\varphi}_\alpha} S_i^- S_j^-\right), \qquad (2)$$

where $S^\pm = S^x \pm iS^y$ and $\tilde{\varphi}_\alpha = \{0, -2\pi/3, 2\pi/3\}$ are the bond-dependent phases for the primitive vectors $\boldsymbol{\delta}_\alpha$, with $\boldsymbol{\delta}_\alpha$'s and x and y axes as in Fig. 1(a). Although this is not obvious from (2) [56], the pseudo-dipolar terms favor the direction of the spins on a bond to be either parallel or perpendicular to the bond [52]. Because of the high symmetry of the lattice, the Dzyaloshinsky-Moriya interactions are forbidden [43, 57] and we also omit the couplings of $S^{x(y)}$'s to the out-of-plane S^z's, referred to as the $J_{z\pm}$ terms, as they are negligible in $\mathrm{YbMgGaO_4}$ [44, 47] and do not affect our conclusions. An intuitive derivation of the Hamiltonian is given in [58].

XXZ only.—In $\mathrm{YbMgGaO_4}$, electron spin-resonance (ESR), magnetic susceptibility, and neutron scattering [44, 47] have suggested strong XXZ anisotropy, $\Delta \sim 0.5$, and put rather stringent bounds on the pseudo-dipolar terms, indicating their subleading role. Thus, we study the pure XXZ model (1) first, considering effects of the pseudo-dipolar terms next. The anisotropy for J_1 and J_2 bonds, Δ_1 and Δ_2, is assumed equal [47] as it originates from the magnetic state of $\mathrm{Yb^{3+}}$ ions, with no qualitative changes expected for $\Delta_1 \neq \Delta_2$.

While the Heisenberg version of (1) at $\Delta=1$ is well-explored [33–40], its anisotropic extension has been studied only rarely [59, 60]. For $J_2/J_1<1$, two ordered states compete, the 120° and the collinear state, where in the latter ferromagnetic rows ("stripes") of spins align antiferromagnetically, see Fig. 1(a). Their classical energies are $E_{\rm gs}^{120^\circ}=-3(J_1/2-J_2)$ and $E_{\rm gs}^{\rm str}=-J_1-J_2$ (per NS^2), yielding a transition at $J_2=J_1/8$ [34, 35] independent of Δ. It is important to note that XXZ anisotropy leads to an overlap of the J_2-ranges of stability for magnon spectra of the competing phases [58, 59]. This implies that the spin-wave instabilities do not yield an intermediate magnetically disordered state for $S\gg 1$, favoring instead a direct transition between the two orders.

The $J_2-\Delta$ phase diagram of $\mathcal{H}_{\rm XXZ}^{J_1-J_2}$ for $S=1/2$, obtained via spin-wave theory (SWT) and DMRG calculations, is shown in Fig. 1(b). The color map shows the ordered moment $\langle S\rangle$ and the $\langle S\rangle=0$ boundaries of a non-magnetic phase (gray) according to SWT. The solid black line marks the crossing of $\langle S\rangle$ from the 120° to the stripe phase. It outlines a region where SWT predicts a direct transition with no intermediate state. Note that the SWT groundstate energies indicate this transition to be on the left of the classical $J_2=J_1/8$ line for $\Delta<1$ [58].

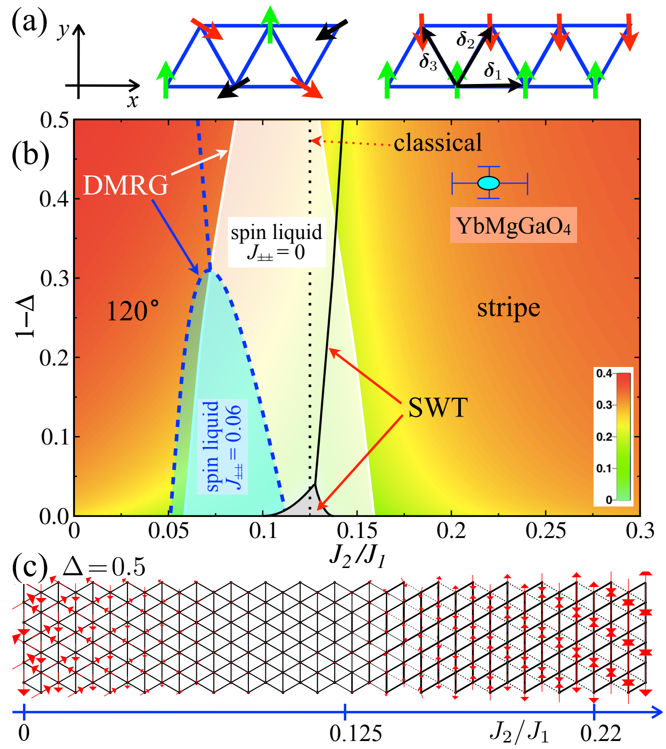

FIG. 1: (a) Axes, primitive vectors, and a sketch of the 120° and stripe states. (b) $1-\Delta$ vs J_2 phase diagram of the XXZ model (1). The $\langle S\rangle$ color map and boundaries (solid lines) are by SWT; dotted line is the classical phase boundary. The shaded white area is the spin-liquid region by DMRG, see text. The dashed line with the shaded region is the same for the model with $\mathcal{H}_{\rm pd}$ with $|J_{\pm\pm}|=0.06$, see Fig. 2. The error bars mark $\mathrm{YbMgGaO_4}$ parameters from [47]. (c) The DMRG scan of (1) vs J_2 for $\Delta=0.5$ with up to 2000 states.

Fig. 1(c) shows a DMRG calculation of the model (1) for $\Delta=0.5$ where J_2 is varied along the length of the cylinder so that different phases appear at different regions. The orders are pinned at the boundaries and the spin patterns give a faithful visual extent of their phases. Similar scans for several Δ's allow us to map out the phase diagram of the model [33, 61]. To roughly estimate the J_2-boundaries for the spin liquid (SL), we use the cut-off value of $\langle S\rangle=0.05$, below which the system is assumed to be in a SL state. This procedure matches the SL boundaries for the isotropic ($\Delta=1$) J_1–J_2 model found in [33] by a more accurate method. The resultant extent of the SL phase is shown in Fig. 1(b) by the white shaded area. We note that the $\langle S\rangle$ cut-off value that we use may overestimate the SL region at $\Delta<1$ as the anisotropy tends to stabilize ordered phases, while the SWT clearly underestimates it, as expected.

The ellipse with error bars in Fig. 1(b) marks $J_2/J_1=0.22(2)$ and $\Delta=0.58(2)$, proposed for $\mathrm{YbMgGaO_4}$ [47]. For these parameters (with $J_{\pm\pm}=0$), we find a close agreement between the DMRG and SWT on the ordered moment, 0.29 and 0.32, respectively, implying that $\mathrm{YbMgGaO_4}$ is deep in the stripe phase.

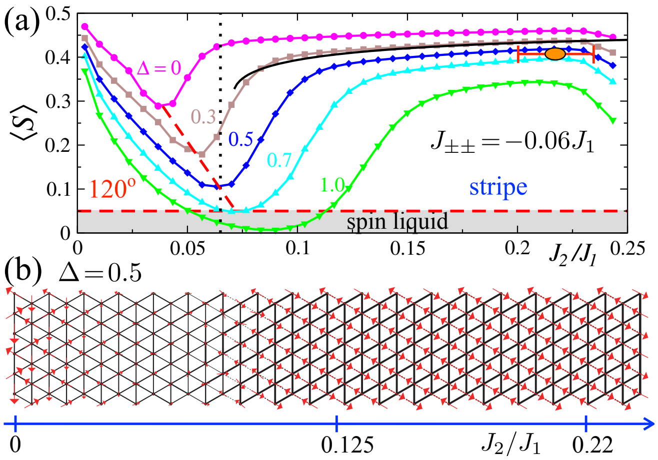

FIG. 2: (a) DMRG results for $\langle S\rangle$ vs J_2 with $|J_{\pm\pm}|=0.06J_1$. Dotted and dashed lines denote classical and DMRG phase boundaries. The error bar is the same as in Fig. 1(b). The solid black line is the SWT result for $\Delta=0.5$. (b) A long-cylinder DMRG scan for $\Delta=0.5$ and $J_{\pm\pm}=-0.06J_1$.

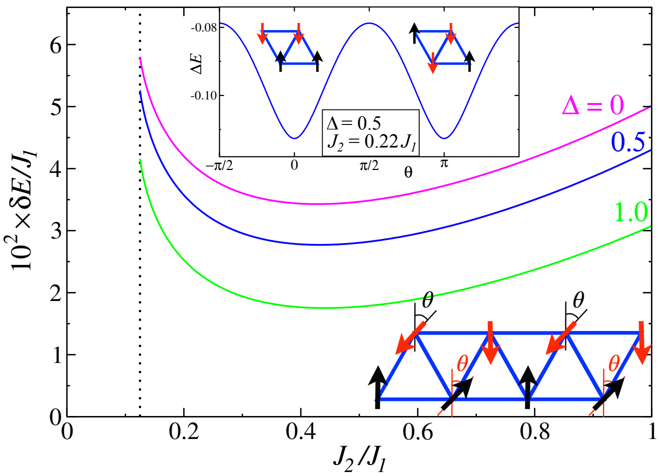

FIG. 3: Energy barrier between the stripe states of different orientations in the XXZ J_1–J_2 model vs J_2 for various Δ and $S=1/2$. Upper inset: quantum energy correction vs angle θ. Lower inset: a sketch of the degenerate classical ground states with $\theta=0(\pi)$ corresponding to two stripe orientations.

Pseudo-dipolar terms.—The anisotropic terms in (2) explicitly break the $U(1)$ symmetry of the XXZ model (1) and are expected to pin the spin directions to the lattice. This is indeed true for the stripe phase, in which the pseudo-dipolar terms make the spin orientation parallel ($J_{\pm\pm}<0$) or perpendicular ($J_{\pm\pm}>0$) to the stripe direction [58] as in Figs. 2(b) or 1(a), see also [57]. From the $1/S$ perspective, no pinning and no change of the classical energy occurs due to (2) for the 120° phase, which, however, remains stable [58]. On the other hand, the partially frustrated pseudo-dipolar terms in (2) lower the classical energy of the stripe phase by $-4|J_{\pm\pm}|S^2N$ and expand its stability range by shifting the classical phase boundary to a lower $J_2=J_1/8-|J_{\pm\pm}|$.

In Figs. 2 and 1(b), we show the effect of adding $J_{\pm\pm}$ to the model, using $|J_{\pm\pm}|=0.06J_1$, as suggested by ESR [44]. The classical transition between the 120° and stripe phases is at $J_2=0.065J_1$ for this value of $|J_{\pm\pm}|$, with the DMRG long-cylinder scans showing it tilting toward smaller J_2 at smaller Δ. Using the same generous criteria for the spin liquid as above, the DMRG results show that $J_{\pm\pm}$ shrinks the SL region [light blue in Fig. 1(b)], and moves it farther from the $YbMgGaO_4$ parameters. It also strengthens the stripe order [Fig. 2(a)], in close agreement with the SWT (solid line). The agreement for the ordered moment for $YbMgGaO_4$ parameters [47] is very close, $\langle S\rangle\approx 0.419(0.433)$ by DMRG (SWT), and the magnitude of the order parameter is large.

Thus, in this model for $YbMgGaO_4$, the easy-plane and pseudo-dipolar anisotropies both lead to a stronger stripe order. Yet, the experiments show no sign of it.

Alternative sets of parameters with much larger values of $|J_{\pm\pm}|=0.26J_1$ [62] and $0.69J_1$ [47] were obtained by fitting the high-field magnon dispersion in $YbMgGaO_4$ [47] without the J_2-term in (1). Both values strongly deviate from the ESR data [44] and imply an almost classical stripe state with nearly saturated ordered moments and large magnon gaps [58], inconsistent with the observed substantial spectral weight at low energies [47]. For $|J_{\pm\pm}|\gtrsim 0.2$'s, there is no 120° state left in the phase diagram to compete with, leaving no SL state in sight.

Barrier.—Before we attempt to reconcile our finding of strong stripe order in the model with the lack of order in $YbMgGaO_4$, we give the J_1-J_2 XXZ model (1) a second look. Classically, in the absence of the pseudo-dipolar terms, the stripe phases of Fig. 1(a) are degenerate with a manifold of spiral phases in Fig. 3, in which four spins in the two side-sharing triangles add up to zero [34, 35]. Their degeneracy is lifted via order-by-disorder mechanism [63, 64], selecting the three stripe states that break rotational lattice symmetry. The tunneling barrier between them, $\delta E(J_2,\Delta)/N$, shown in Fig. 3, is obtained from the quantum energy correction $\Delta E(\theta)=c+\frac{1}{2}\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}(\theta)$, where $c=-(J_1+J_2)S$ and $\varepsilon_{\mathbf{k}}(\theta)$ is the magnon energy, which depends on the angle θ of the spiral state from the degenerate classical manifold. As one can see from Fig. 3, the tunneling barrier is small, $\delta E\sim 0.03J$ per site, similar to the J_1–J_2 model on the square lattice [65]. Thus, in the XXZ model, despite being strongly-ordered, the stripe phases of different orientations are separated by a low energetic barrier.

Disorder.—As discussed above, a number of experiments indicate a substantial disorder in the low-energy effective spin Hamiltonian of $YbMgGaO_4$ [43, 47, 48, 50]. Most direct are the neutron studies, suggesting strong variations in the effective g-factors and, possibly, magnetic couplings [48] due to a random charge environment from mixing of the non-magnetic Mg^{2+} and Ga^{3+}.

We do not attempt to analyze all forms of disorder that can naturally occur in the Hamiltonian (1) and (2). Instead, we propose that a disorder in the $J_{\pm\pm}$ terms should be potentially very destructive. Because of their pseudo-

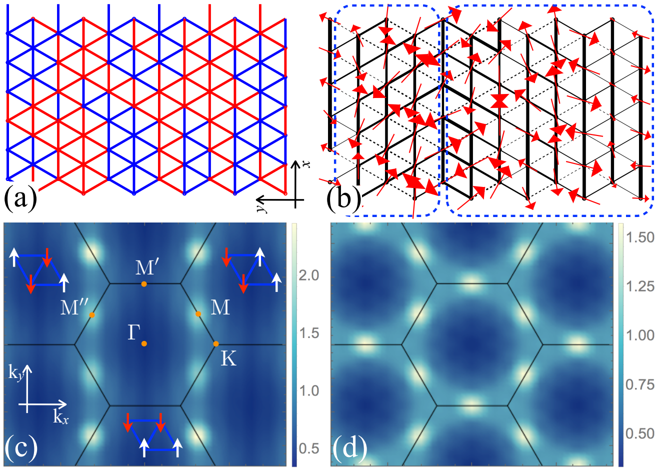

FIG. 4: (a) Positive/negative $J_{\pm\pm}$ bonds in a typical disorder realization. (b) Two stripe domains (dashed boxes) for random $|J_{\pm\pm}|=0.2J_1$, $\langle S\rangle$ up to 0.33. (c) $\mathcal{S}(\mathbf{q})$ [66] for random $|J_{\pm\pm}|=0.1(0.05)J_1$. (d) Averaged $\mathcal{S}(\mathbf{q})$ from (c), see text.

dipolar nature, random $J_{\pm\pm}$'s are not unlike fluctuating pinning fields that can locally stabilize stripes with different orientations by overcoming the low tunneling barrier between them. In addition, for the relevant values of $|J_{\pm\pm}|\sim 0.1J_1 \gtrsim \delta E$, fluctuations of the diagonal elements of the exchange tensor at the level of $0.1-0.2J_1$, that are consistent with the variations suggested in [48], translate into completely random $J_{\pm\pm}$ [58].

We have performed DMRG calculations of the J_1-J_2 XXZ model (1) with $YbMgGaO_4$ parameters, $\Delta=0.58$ and $J_2=0.22J_1$ [47], and random $J_{\pm\pm}$ (2). We have used different random disorder realizations, such as in Fig. 4(a), with binary distribution of $J_{\pm\pm}$ of alternating sign and a global constraint of the same number of positive and negative $J_{\pm\pm}$ bonds to reduce the finite-size bias. We used the values of $|J_{\pm\pm}|/J_1=0.05$, 0.1, and 0.2 on the 6×12 cluster. The results are as follows.

For large values of random $|J_{\pm\pm}|=0.2J_1$, the groundstates tend to contain static, visibly disordered spin domains with mixed stripe orientations and large ordered moments, see Fig. 4(b). For smaller $|J_{\pm\pm}|$, more interesting states appear. First, there is no clear real-space order without pinning fields, as in a disorder-free $U(1)$-symmetric XXZ model, yet the structure factor [66], obtained from $\mathcal{S}^{\alpha\beta}_{\mathbf{q}}=\sum_{i,j}\langle S^\alpha_i S^\beta_j\rangle e^{i\mathbf{q}(\mathbf{R}_i-\mathbf{R}_j)}$, shows broadened peaks at *two* M-points, which are associated with *two different* stripe orderings, see Fig. 4(c). We note that the 6×12 DMRG cluster strongly disfavors the state with stripes along the shorter direction of the cylinder, parallel to the open boundaries, that would show itself as a peak at the M′ points in Fig. 4(c).

Upon a careful investigation with pinning fields, we conclude that the observed state is a *stripe-superposition state*, in which spins continue to fluctuate collectively between the two stripe states allowed by the cluster. A hint of such a state can also be seen at the right edge of Fig. 4(b). As opposed to a spin liquid, the degeneracy of such a superposed state is not extensive. This finding implies that the randomization of $J_{\pm\pm}$ leads to an effective restoration of the Z_3 lattice symmetry, broken in each individual stripe state. Whether such stripe-superposition states will be pinned to form single-stripe domains on a larger length scale, or they will survive as localized fluctuating states, remains an open question.

Note that both $|J_{\pm\pm}|/J_1=0.05$ and 0.1 yield nearly identical structure factors, with the smaller value already sufficient to destroy the long-range stripe order, supporting our hypothesis on its fragility to an orientational disorder. To overcome the lack of the third stripe direction in the DMRG cluster and provide a faithful view of a response of a spatially isotropic system, we have performed an averaging of the structure factor, see Fig. 4(d), with the results very similar to the $\mathcal{S}(\mathbf{q})$ in the neutron-scattering data for $YbMgGaO_4$ [47].

Altogether, the randomization of the small pseudo-dipolar term in the model description of $YbMgGaO_4$ results in the disordered stripe groundstates that can successfully mimic a spin liquid. Further experimental verifications of the proposed picture include possible freezing at lower temperatures, as the current lowest-temperature measurements [47] are at $T\sim 0.05J_1\sim|J_{\pm\pm}|$, and the spin pseudo-gap in the dynamical response at low energies at the M-points as a remnant of the anisotropy-induced gaps in the magnon spectra [58]. The proposed scenario implies that the anomalously low T-power in the specific heat should emerge as a result of disorder.

Summary.—We have investigated a generalization of the isotropic J_1-J_2 triangular-lattice model, known to support a spin-liquid state, and have found that the anisotropic interactions significantly diminish the spin-liquid region of the phase diagram. Our analysis finds no additional transitions near the experimentally relevant range of parameters, putting $YbMgGaO_4$ firmly in the stripe-ordered state. At the same time, the stripe states are shown to be fragile toward orientational disorder. The randomization of the pseudo-dipolar interactions due to spatially-fluctuating charge environment of the magnetic ions generates a mimicry of a spin-liquid state in the form of short-range stripe or stripe-superposition domains. This scenario is likely to be relevant to other rare-earth-based quantum magnets.

Note added. After submission of this work, we became aware of the preprint supporting our findings [67].

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