Intrinsic quantum Ising magnet with intertwined multipolarness on a triangular lattice

Changle Liu$^{1,2,3}$ and Chun-Jiong Huang$^{4,5,6}$

$^1$State Key Laboratory of Surface Physics and Department of Physics, Fudan University, Shanghai, 200433, China
$^2$Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing, 210093, China
$^3$Department of Physics and Center of Theoretical and Computational Physics, The University of Hong Kong, Pokfulam Road, Hong Kong, China
$^4$Shanghai Branch, National Laboratory for Physical Sciences at Microscale and Department of Modern Physics, University of Science and Technology of China, Shanghai, 201315, China
$^5$CAS Center for Excellence and Synergetic Innovation Center in Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei, Anhui 230026, China
$^6$CAS-Alibaba Quantum Computing Laboratory, Shanghai, 201315, China

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Material realizations of frustrated magnets whose effective model can be solved in a controlled manner theoretically or numerically, have a high significance to the strong correlation community. The triangular lattice magnet TmMgGaO$_4$ is a realistic quantum Ising magnet with an intertwined multipolar order described by the antiferromagnetic transverse field Ising model (TFIM) on the triangular lattice. Particularly, for this material the transverse field has an intrinsic origin that comes from the weak splitting of low-energy crystal field levels. Moreover, this model can be implemented by unbiased quantum Monte Carlo methods. Motivated by recent experiments, we perform a comprehensive study on the effective model of this system and consider the effect of the external magnetic field. We show that the TFIM with three-sublattice intertwined ordered ground state agrees with the inelastic neutron scattering and thermodynamic measurements on TmMgGaO$_4$. We make specific predictions for the experiments when the external field is applied. Furthermore, we demonstrate interesting emergent $U(1)$ continuous symmetry may emerge in thermal melting of the underlying orders or at the quantum critical point. We discuss the relevance of intrinsic quantum Ising models to other systems.

I. INTRODUCTION

Frustrated magnetism is an exciting field in condensed matter physics which has been under quite active research for the past decades$^1$. Generally, it arises from competing interactions among local moments which cannot be satisfied simultaneously. The strong competitions can give rise to exotic low-energy behaviors in frustrated magnets. This feature retains in the simplest classical antiferromagnetic Ising model, where for some particular frustrated lattices (triangular$^2$, Kagome$^3$, pyrochlore$^{4,5}$, etc.) there are macroscopic degenerate ground states associated with a finite zero-point entropy.

An interesting and important question is to consider the fate of classical macroscopic degeneracy in presence of quantum fluctuations. Quantum fluctuations allow tunneling within the macroscopic degenerate manifold, therefore will lift the macroscopic degeneracy. Depending on lattice structures, the resulting quantum ground-state can be either magnetically ordered or disordered$^6$-$^{10}$, owing to the so-called “order-by-disorder” or “disorder-by-disorder” mechanism$^{6,11,12}$. In practice, the simplest way to add quantum fluctuations is to add a transverse field to the quantum spins. The resulting model is the TFIM, which has not only received a considerable theoretical attention, it is also achievable in experiments. Moreover, this model is sign-problem free in any lattice structures, therefore can be efficiently dealt with by unbiased quantum Monte Carlo simulations. These qualities render TFIM a good platform for collaborations among experimental, theoretical and numerical communities.

In realistic materials, many physical origins of the transverse field was proposed, and has been summarized in Ref. 13. In some systems transverse spin components act as ordinary magnetic dipole moments, hence the transverse field is directly achievable with physical magnetic field in transverse directions. However, in many cases the transverse spin components exhibit magnetic multipolar properties imposed by lattice symmetry therefore inaccessible by magnetic field. A lot of proposals has been made to deal with this issue, such as introducing non-magnetic disorder$^{14}$ and applying external shear strain$^9$. In these proposals the transverse field is generated extrinsically and suffer from the issue that the introduced perturbations always break some symmetries of the original systems.

The TmMgGaO$_4$ material$^{15-17}$ has provided an another route to generate the transverse field in some entirely different manner. In this system the lowest crystal field levels that contribute to the local moment are not protected by symmetry therefore must be split and give rise to effective transverse field. Notice that the crystal field splitting is intrinsic and appears at atomic level, it cannot be ignored compared to exchange interactions and must be considered at the first place. Moreover, in this material the transverse and longitudinal spin components behave physically different in nature, the system exhibits antiferromagnetic dipolar order coexisting with preformed multipolar order due to the effective transverse field. The resulting state is an example the intertwined multipolar order formed due to the effective transverse field.

In this article, we present a systematic study on the effective model for the TmMgGaO$_4$ system and discuss its implications to relevant systems. The rest of the article is organized as follows: in Sec. II we review the microscopics and modeling of the TmMgGaO$_4$ material; in Sec. III we analyze the phase diagram and phase transitions; in Sec. IV we discuss the...
experimental signatures in neutron and thermodynamic measurements; in Sec. V we discuss the relevance with other systems.

II. EFFECTIVE MODEL OF TmMgGaO₄

To start with let’s first review the microscopics and effective model of this material. The Tm³⁺ non-Kramers ion has 4f² configuration with total angular momentum J = 6. The (2J + 1)-fold degenerate states are lifted by D₃d symmetric crystal field into a series of singlets and non-Kramers doublets. It has been shown that the Tm ions in these materials exhibit strong Ising character and the low-lying crystal field levels are dominated by J' = ±6 components. For the case of D₃d symmetric crystal field, there is no symmetry that protects the J' = ±6 components to form a doublet, therefore they weakly hybridize into two non-degenerate singlets

\[ |\Psi^+\rangle \sim |J' = 6\rangle + |J' = -6\rangle + \cdots, \]

\[ |\Psi^-\rangle \sim |J' = 6\rangle - |J' = -6\rangle + \cdots. \]

where the two singlets carry A₁g and A₂g representation of the D₃d point group, respectively. The local moment can be described by the following effective spin-1/2 operator \( S \) acting upon the quasi-doublet:

\[
S^z_i = \frac{i}{2} (|\Psi^+_i\rangle\langle\Psi^+_i| - |\Psi^-_i\rangle\langle\Psi^-_i|), \quad S^y_i = \frac{i}{2} (|\Psi^+_i\rangle\langle\Psi^-_i| + |\Psi^-_i\rangle\langle\Psi^+_i|), \quad S^x_i = \frac{i}{2} (|\Psi^+_i\rangle\langle\Psi^+_i| + |\Psi^-_i\rangle\langle\Psi^-_i|). 
\]

We can see from the effective spin definition that \( |\Psi^\pm\rangle \) are eigenstates of \( S^y \) with eigenvalue \( S^y = \pm 1/2 \), while the \( S^x \) and \( S^z \) components introduces hybridization between \( |\Psi^\pm\rangle \). Under crystal point group symmetry and time reversal operations, the effective spin components transform as:

\[
C_3 : S^x \rightarrow S^x, S^y \rightarrow S^y, S^z \rightarrow S^z \quad (6)
\]
\[
C'_3 : S^x \rightarrow -S^x, S^y \rightarrow S^y, S^z \rightarrow -S^z \quad (7)
\]
\[
\Theta : S^x \rightarrow S^x, S^y \rightarrow S^y, S^z \rightarrow -S^z \quad (8)
\]

This implies that the \( S^x \) and \( S^y \) operators are time reversal even and transform as multipole moments under crystal symmetries. The \( S^z \) component are time reversal odd and transform as dipole moments along z direction. While dipole moments can be probed by neutrons, the multipole moments are hidden in most conventional experimental probes.

The crystal field opens a energy gap \( h \) between \( |\Psi^\pm\rangle \) states which can be captured by the transverse field term \( -h \sum_i S^z_i \). Considering the strong Ising nature of spins and the strongly localized nature of 4f orbitals, to capture the essential physics consider only nearest neighbor Ising interactions between the Tm³⁺ local moments. The resulting effective Hamiltonian is the TFIM

\[
H = \sum_{\langle ij \rangle} J_{zz} S^z_i S^z_j - \sum_i \left( h S^y_i + B S^x_i \right), \quad (9)
\]

where \( B \equiv \mu_B g_i |B|^2 \) is the scaled longitudinal external field.

III. PHASE DIAGRAM

The TFIM on the triangular lattice has been well-studied in the absence of external magnetic field, while the situation with longitudinal field has not been investigated yet. To gain insight into the ground-state phase diagram, we first tackle with Weiss mean-field approximation by decoupling interactions between different sites

\[
S^z_i S^z_j \rightarrow \langle S^z_i \rangle S^z_j + \langle S^z_j \rangle (S^z_i) - \langle S^z_i \rangle \langle S^z_j \rangle. \quad (10)
\]

Here the mean-field order parameter \( \langle S^z_i \rangle \) needs to be solved self-consistently. The mean-field phase diagram is shown in Fig. 2(a).

At the Ising limit, the system lies at a classically critical state that hosts macroscopic ground-state degeneracy: any spin configurations with “2-up-1-down” or “1-up-2-down” has the minimal energies. With introducing the transverse field \( h \), quantum fluctuations will allow tunneling within the massively degenerate manifold: it lifts the macroscopic degeneracies and eventually, stabilizes a three-sublattice long-range ordered phase (dubbed as three-sublattice “1” state) as the ground-state owing to the quantum order-by-disorder mechanism. Since the three-sublattice ordering is entirely contributed by quantum fluctuations, it is relatively weak and is controlled by quantum fluctuation \( h \) in non-monotonic fashion: with \( h \) being too small the quantum order-by-disorder effect is weak, while too much \( h \) quantum fluctuations suppresses the three-sublattice ordering and drives the system into the “quantum disordered” state where spins are fully polarized along the transverse direction. Notice that although the
above results are obtained mean-field level, they are consistent with those obtained via quantum dimer mapping where quantum fluctuations are taken into account in a more serious manner.\textsuperscript{6,7}

As external longitudinal field $B$ is applied, at the Ising limit the system immediately becomes unstable against magnetic ordering due to criticality at this point. The resulting state is an another three-sublattice ordered state called “1/3-plateau” state with “2-up-1-down” structure at the three sublattices respectively. Unlike the pure quantum origin in the “I” phase, the three-sublattice ordering of plateau state arise at classical level therefore can be very large. The plateau state remains as the ground-state upon increasing magnetic field until the system becomes fully polarized at $B_c = 3$ through a first-order transition. When the quantum fluctuation $h$ is switched on, the three-sublattice “plateau” state becomes the “quasi-plateau” phase (dubbed as three-sublattice “II” state) due to quantum fluctuations. Moreover, as the three-sublattice “I” phase is protected by quantum fluctuations, it stable against small perturbations. But since that the ordering is weak, a very small external field $B$ can drive the system to the quasi-plateau state across a phase transition. The transition from “I” to “II” state is of the second-order, while the transition from “II” to the polarized state is of the first-order, consistent with what happens at $h = 0$ limit. The two phase boundaries terminate the classical critical point $h = 0$, and the quantum critical point $h_{Q}\text{MF} = 1.5$, both located along $B = 0$ axis.

To examine our mean-field results, we perform quantum Monte-Carlo simulations. Through the path integral with $\{S_i^z\}$ basis, the partition function of the original quantum model is mapped onto a worldline representation:

$$\mathcal{Z} = \text{Tr} \left[ e^{-g\mathcal{H}} \right] = \sum_{\{\alpha\}} \langle \alpha_0 | e^{-g\mathcal{H}} | \alpha_0 \rangle$$

$$= \lim_{\beta \to 0} \sum_{\alpha = \{1\}} \langle \alpha_{\beta} | e^{-\beta g \mathcal{H}} | \alpha_{\beta-1} \rangle \cdots \langle \alpha_1 | e^{-\beta g \mathcal{H}} | \alpha_0 \rangle$$

$$= \sum_{\alpha} \sum_{k = 0} \int_{0}^{\beta} \cdots \int_{0}^{\beta} \prod_{i = 1}^{2k} d\tau_{i} \exp \left[ -\beta \mathcal{H} - \frac{\beta}{\hbar} \text{U}(\tau) d\tau \right],$$  \hspace{1cm} (11)$$

where $U(\tau) = \langle \alpha(\tau) | \left( \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z - \sum_i BS_i^z \right) | \alpha(\tau) \rangle$. Fig. 3(a) is a worldline configuration of partition function. The term of $\sum_i h S_i^z$ causes spins to reverse and we call a flipping as a kink. $|\alpha(0)\rangle = |\alpha(\beta)\rangle$ demand the number of kinks $N_k$ must be even $N_k = 2k$ as in Eq. (11). Due to the presence of longitudinal field $B$, the cluster update fails and we design a metropolis algorithm which contains two update schemes, creation/deletion flat and shift kink, as shown in Fig. 3(b).

The calculation of acceptance rates of update schemes is standard through the detailed balance equation and we will not show them explicitly here. Thermal annealing procedure is employed to deal with the freezing issue. The result is shown in Fig. 2(b). We can see that the Monte-Carlo phase diagram well agrees with the mean field one at the qualitative level. However, the qualitative locations the phase boundaries are very different. For example, the critical field $h_{Q}\text{MC} \approx 0.82\text{9,20}$ is almost half of the mean-field result $h_{Q}\text{MF} = 1.5$. This is as expected, as mean-field approximations underestimates quantum fluctuations especially near phase boundaries.

To discover the finite-temperature properties and phase transitions, it is necessary to perform field theoretical analysis. Basically, the three-sublattice order parameter is characterized by the Fourier transformed $S^z$ dipolar component at the K point. Locally, this can be captured by the following complex field

$$\psi = (m_1 + m_2 e^{i2\pi/3} + m_3 e^{-i2\pi/3}) / \sqrt{3},$$  \hspace{1cm} (12)$$

where $m_i (i = 1, 2, 3)$ are the dipolar magnetizations of three sublattices at neighboring sites. We can see that $\psi$ characterizes the three-sublattice ordering, since $\psi = 0$ occurs only when $m_1 = m_2 = m_3$, where the three-sublattice order vanishes. The transformation of the field variable $\psi$ under the translation $T_\vec{z}$ and time-reversal $\Theta$ operation take the following form

$$T_\vec{z} : \psi \rightarrow \psi e^{i2\pi/3}$$ \hspace{1cm} (13)$$

$$\Theta : \psi \rightarrow -\psi \hspace{1cm} (14)$$

For the three-sublattice “I” state, spin alignment at three sublattices are different from one another, therefore the

FIG. 2. Phase diagram of the model (9). Here we set the unit $J_{zz} = 1$. Two three-sublattice ordered phases, I (orange) and II (green) and a polarized phase are found in the phase diagram. The red dot represents quantum phase transition with (2+1)D XY universality class.

FIG. 3. The worldline configuration under imaginary time evolution and update schemes. (a) the worldline configuration of four spins in a chain. Different colors mean different spin states. Along the imaginary time, every spin worldline may be divided into several flats by cause of $S^z$. (b) the diagrammatic sketch of update schemes.
ground-state is six-fold degenerate. At zero external magnetic field, the $\psi$ corresponding to the ground states are located at a circle in the complex plane with $\text{Arg } \psi = (2n + 1)\pi/6$ ($n = 0, 1, ..., 5$), protected by the translation and time-reversal symmetry, see Fig. 4(a). Notice that such clock anisotropy is robust against short-range interactions such as transverse exchange and next-nearest-neighbor Ising interactions which also exists in materials, therefore our analysis remains valid against these perturbations. The coarse-grained Landau-Ginzburg-Wilson Hamiltonian dictating the mains valid against these perturbations.

The high temperature transition at $T > T_c$ is dual to the high temperature phase transition at $T_c$ is dual to the high temperature one thus is called the “inverse KT” transition. Besides thermal melting, the emergence of continuous symmetry also shows up at the quantum phase transitions from three-sublattice state to the “quantum disordered” polarized state at the zero field (the red dot in Fig. 2). It has been shown that in $(2+1)d$ the $\mathbb{Z}_3$ clock anisotropy is dangerously irrelevant at the quantum critical point. Therefore, this quantum phase transition belongs to $(2+1)d$ XY universality class, which also shows emergent $U(1)$ continuous symmetry.

The above scenario no longer presents in presence of external magnetic field. This is because that the magnetic field breaks the time-reversal symmetry, which brings about $\mathbb{Z}_3$ clock anisotropy to the system:

$$H_{\text{L-GW}} = -K|\psi|^2 + rv^*\psi + u_4(|\psi|^2)^2 + u_6(|\psi|^4)^2$$

$$+ v_6(\psi^6 + \psi^{-6})$$

The thermal melting of the three-sublattice order takes two-step manner which can be understood from self-duality of the effective theory (15), and is also clearly identified in the order parameter histogram as is shown in Fig. 4. At low temperature phase $T < T_{c1}$, the $\mathbb{Z}_6$ clock term is relevant, therefore we have the long-range ordered state. This can be seen in the histogram of $\psi$ in Fig. 4(a). The dual phase at $T > T_{c2}$ is the high temperature disordered phase in which vortices proliferate. In our case $T_{c1}$ and $T_{c2}$ do not coincide, therefore in the intermediate temperature $T_{c1} < T < T_{c2}$ we have an extended phase in which both vortices and the clock anisotropy become irrelevant, see Fig. 4(b). Notice that the irrelevance of clock anisotropy indicates emergent continuous symmetry, therefore system behaves just like low-temperature quasi-long-range ordered phase of the XY model without anisotropy term. The high temperature transition at $T_{c2}$ belongs to the Kosterlitz-Thouless (KT) universality class, while the low temperature transition at $T_{c1}$ is dual to the high temperature one thus is called the “inverse KT” transition.

Having figured out the entire phase diagram, here we discuss about the experimental consequences. The three-sublattice order is characterized by the order parameter $\psi$ defined by dipolar magnetization, which is directly reflected as magnetic Bragg peaks at K point. Meanwhile, due to the intrinsic crystal field, there is always non-vanishing ordering in

IV. RELATION TO EXPERIMENTS

FIG. 4. Histogram of the order parameter $\psi$ in thermal phases. Left: Low temperature three-sublattice long range ordered state; Middle: intermediate temperature “KT phase”; Right: high-temperature disordered state. In all cases external magnetic field $B$ is set to zero.

FIG. 5. Finite temperature phase diagram of the three-sublattice state with external magnetic field $B$. The parameter we take is $J_{\parallel} = 1$, $h = 0.65$. The “KT phase” at $B = 0$ axis is marked by thick purple line. The solid-dot lines refer to continuous transition while the upper and lower red dots at $B = 0$ axis correspond to KT and inverse KT transitions, respectively.

$\mathbb{Z}_3$ clock term is always relevant at phase transition. Therefore, the successive KT transition scenario in thermal melting as well as emergent continuous symmetries are no longer presented. Instead, the thermal transition between the three-sublattice and the disordered state now becomes three-state Potts universality class. The finite-temperature phase diagram of three-sublattice state is shown in Fig. 5.

$$H_3 = v_3(\psi^3 + \psi^{-3}).$$

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transverse components that arises not from spontaneous symmetry breaking but from the polarization effect. Since that the transverse components are multipoles they do not directly couple to neutrons hence hidden in the elastic neutron probes. However, due to the peculiar local moment structure of this system, the elementary excitations of the multipole moment can be measured in dynamical probes such as inelastic neutron scattering, owing to the non-commutative relation between dipole and multipole moments. This idea was initially pointed out by some of the authors in the context of non-Kramers doublets and also applies here. As neutron spins only directly couple to the dipole components, in inelastic neutron scattering what is measured is the \( S^z(S^z) \) correlation

\[
S^{zz}(\mathbf{q}, \omega) = \frac{1}{2\pi N} \sum_{ij} \int_{-\infty}^{+\infty} dt \ e^{i \mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j) - i \omega t} \langle S_i^z(0) S_j^z(t) \rangle, \quad (17)
\]

just as if one is performing polarized neutron measurements. As neutron measures longitudinal dipolar moments, it flips the multipole moment that is orthogonal to the dipole moments, creating coherent spin-wave excitations. Therefore, in inelastic neutron experiment what is measured is the dynamical excitations of multipole components which contain information on the underlying hidden multipole structures. We have calculated the dynamical structure factors for three representative parameters. The results are shown in Fig. 6. For the paramagnetic phase, there is only one branch of excitation, reflecting one-sublattice structure of the ferro-multipoles ordering, see Fig. 6(a). Meanwhile, for the three-sublattice phase, one can clearly identify two branches of excitations, which correspond to amplitude and phase fluctuations of the order parameter \( \psi \). Meanwhile, for the three-sublattice phase, one can calculate the dynamical structure factors for three representative parameters. The results are shown in Fig. 6. For the paramagnetic neutron experiment what is measured is the dynamical excitations of multipole components which contain information on the underlying hidden multipole structures. We have calculated the dynamical structure factors for three representative parameters. The results are shown in Fig. 6. For the paramagnetic phase, there is only one branch of excitation, reflecting one-sublattice structure of the ferro-multipoles ordering, see Fig. 6(a). Meanwhile, for the three-sublattice phase, one can clearly identify two branches of excitations, which correspond to amplitude and phase fluctuations of the order parameter \( \psi \), respectively. See Fig. 6(c). For the TmMgGaO\(_4\) material, previous neutron experiment shows a tiny spin gap of ~ 0.08 meV at K point. This suggests that this system is in proximity to the quantum critical point at \( h_c \). Indeed, our calculated excitation spectrum with parameter \( h_c/\Delta_{\text{MF}} \approx 0.87 \) [see Fig. 6(b)] shows great agreement with the experimental data. Moreover, the vicinity to the quantum critical point allows possibility of performing quantum criticality experiments for this material. One possibility to achieve the quantum critical point is by tuning external pressure.

The three-sublattice phase and the polarized phase can be also identified in thermodynamic experiments. The three-sublattice ordering is melted in two-step manner through two successive KT-like transitions. Since for KT transitions the correlation length diverges too fast near the critical point, the diverging behavior of the specific heat near the transition temperatures cannot be observed experimentally or even numerically. This is exactly what happens for TmMgGaO\(_4\): no diverging behavior is revealed in specific heat data, instead only tiny anomaly is shown at ~ K. Further, it has been discussed that this two-step melting process no longer presents with external magnetic field. Therefore, the divergence behavior will be expected with external magnetic field. This is, again consistent with the experimental observation, as is shown in the magnetic specific heat data in Ref. 16 and 17.

For the polarized state which does not break any symmetry, no phase transitions are expected as increasing temperatures with or without external field. This behavior is fundamentally different from those of the three-sublattice ordered state, which can be used to identify these two phase without performing neutron scattering experiments. The susceptibility \( \chi^{zz} \) as the function of temperature is calculated by mean-field theory and is plotted in Fig. 7(a). We can see that high temperatures the magnetic susceptibility satisfies the Curie-Weiss law

\[
\chi^{zz} \sim \frac{C}{T - \Theta_{\text{CW}}}, \quad (18)
\]

where \( \Theta_{\text{CW}} = -\frac{1}{2} J_{zz} \) from which the exchange parameter \( J_{zz} \) can be extracted from experiments. There is a crossover to the low temperature behavior where \( \chi^{zz} \) saturates to a constant value due to lack of dipolar ordering. With simple mean-field theory calculations one can obtain the low-temperature \( \chi^{zz} \) value

\[
\chi^{zz} \approx \frac{1}{6 J_{zz} + 2h}. \quad (19)
\]

Therefore, one can extract the model parameter \( J_{zz} \) and \( h \) simply from the high temperature and low temperature behavior of \( \chi^{zz} \).

Finally, we discuss about the magnetization process of the three-sublattice state which is relevant with TmMgGaO\(_4\). As has been discussed in Sec. III, in the absence of external magnetic field, the three-sublattice ordering has pure quantum origin as the quantum order-by-disorder mechanism. Therefore, in this state the three-sublattice ordering is weak and the spin gap is relatively small, see Fig. 6(b)(c). This property makes the three-sublattice state very fragile against external magnetic field: a very small field \( B_{c1} \) (for TmMgGaO\(_4\), \( B_{c1} \approx 0.04 \) T) will cause closing of spin gap and drive the system towards the intermediate quasi-plateau state. With increasing external magnetic field, the spin gap reopens and the intermediate state becomes stable. But further increasing magnetic field the spin gap drops until the system is fully polarized by magnetic field at \( B_{c2} \).

The presence of intermediate quasi-plateau state renders the magnetization process highly non-trivial, see Fig. 7(b)(c). For small \( h \), the magnetization curve shows clear 1/3 quasi-plateau feature in the intermediate regime. Meanwhile, deep in the quasi-plateau state, the system has approximate “2-up-1-down” structure, which contributes significant three-sublattice ordering compared with the case without external field. Therefore, in elastic neutron experiments the intensity of magnetic Bragg peak at K point (proportional to \( |\langle \psi \rangle|^2 \)) is expected to show non-monotonic behaviors: deep in the quasi-plateau state the intensity is large, while approaching the three-sublattice I state the intensity is expected to decrease; the intensity is also expected to decrease when the field is large enough where the system becomes nearly polarized, see Fig. 7(b).

For the case relevant with TmMgGaO\(_4\) where the transverse field \( h \) is large, in the quasi-plateau regime the “2-up-1-down” structure is heavily distorted by strong quantum fluctuations, therefore the plateau feature of the intermediate regime is not
FIG. 6. Dynamical correlation function $S_{\mathbf{q}}(\mathbf{q}, \omega)$ of (a): the polarized state and (b),(c): the three-sublattice state calculated within linear spin-wave theory. The parameter we take for the three representative points are (a): $h = 2$, (b): $h = 1.3$ (c): $h = 0.8$. In all cases we take $J_\parallel = 1$ and $B = 0$.

FIG. 7. (a): Calculated magnetic susceptibility $\chi^{zz}$ vs. temperature $T$ for the polarized state. Here we take parameter $J_\parallel = 1$, $h = 3$. The magnetic susceptibility is defined as $\chi^{zz} \equiv \frac{m}{V}$ where $m = \frac{1}{N} \sum_i S_i^z$ is the average dipolar magnetization per site. (b),(c): Magnetization $m$ (red) and scaled K point magnetic Bragg peak intensity $|\langle \psi \rangle|^2$ (blue) for the three-sublattice state at zero temperature. The parameter we take is $J_\parallel = 1$ and for (b): $h = 0.2$, for (c): $h = 1.3$. All results are calculated within mean-field theory.

observed in the magnetization curve. Instead, the line shape curves slightly downwards at $B_{c2}$, see Fig. 7(c). This feature is also found in the magnetization data of TmMgGaO$_4$ at about 3.5 T, which marks the transition field $B_{c2}$. Above $B_{c2}$ the system becomes polarized, but not fully aligned along the $z$ direction due to the presence of transverse field. In order to let the magnetization approach the saturation value, larger external field has to be applied. Notice that this feature is in stark contrast to ordinary systems where the internal transverse field is absent. For the magnetic Bragg peak intensity, the non-monotonic behavior still persists with large quantum fluctuations, see Fig. 7(c).

V. DISCUSSION

In this paper, we have performed a comprehensive study on the triangular lattice transverse field Ising model relevant with the TmMgGaO$_4$ material. We clarify the intrinsic origin of transverse field of this material as crystal field splitting. We figure out the phase diagram using combined mean-field and quantum Monte-Carlo techniques. We point out that an emergent continuous symmetry emerges in the thermal melting and at the quantum critical point. We also discuss the properties of phases in neutron and thermodynamic experiments. The available experimental data shows that this material is well consistent with the transverse field Ising model with three-sublattice intertwined multipolar ordered ground-state.

Besides TmMgGaO$_4$, the intrinsic transverse field can also appear in other systems. A series of rare-earth triangular lattice magnets has been summarized in Ref. 18. We expect that other materials, especially some Tm based compounds, can be also described by TFIM, and share similar physics with TmMgGaO$_4$.

The transverse systems can be extended to other lattice structures. While the case of Kagomé lattice has been discussed in a parallel paper by one of the authors, here we briefly discuss the consequences to pyrochlore systems. At the Ising limit the ground-state is the so-called spin-ice state where each tetrahedron fulfills the two-in-two-out ice rule. Like the case of triangular lattice, this spin-ice ground-state is also extensively degenerate. A $U(1)$ quantum spin ice (QSI) phase can be obtained by a transverse field term that creates quantum fluctuations within the extensively degenerate spin-ice manifold. If we treat the transverse field terms as perturbation, the leading non-trivial contribution of the transverse field comes at sixth-order:

$$H_{\text{eff}}^{(6)} = -K \sum_\mathbf{O} \left( S^+_1 S^+_2 S^+_3 S^-_4 S^+_5 S^-_6 + h.c. \right).$$

where $K = 63 \hbar^2 / 16 J_\parallel^2$. This low energy theory is identical to the XXZ model near the Ising limit, which can be mapped to compact $U(1)$ gauge theory that supports the exotic QSI phase. We point out that such scenario may be realized in the rare-earth spinel MgTm$_2$Se$_4$. In this material, the Tm$^{3+}$...
magnetic ion lives at the B site of the spinel which forms a pyrochlore lattice. In the recent neutron scattering study, it was found that the lowest crystal field levels of Tm$^{3+}$ form a quasi-doublet with dominated $J^z = \pm 6$ components separated by a small gap of $\sim 0.87$ meV. This crystal field scheme is very much similar with TmMgGaO$_4$, hence its magnetism is also described by TFIM. Therefore, this system potentially supports the exotic $U(1)$ QSI phase, and more experimental efforts are required to figure out the true ground-state of this material.

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1. L. Balents, Nature 464, 199 (2010).
2. G. Wannier, Phys. Rev. 79, 357 (1950).
3. K. Kanô and S. Naya, Prog. Theor. Exp. Phys. 10, 158 (1953).
4. M. J. Harris, S. Bramwell, D. McMorrow, T. Zeiske, and K. Godfrey, Phys. Rev. Lett. 79, 2554 (1997).
5. A. P. Ramirez, A. Hayashi, R. J. Cava, R. Siddharthan, and B. Shastry, Nature 399, 333 (1999).
6. R. Moessner, S. L. Sondhi, and P. Chandra, Phys. Rev. Lett. 84, 4457 (2000).
7. R. Moessner and S. L. Sondhi, Phys. Rev. B 63, 224401 (2001).
8. M. Hermele, M. P. Fisher, and L. Balents, Phys. Rev. B 69, 064404 (2004).
9. J. Röchner, L. Balents, and K. P. Schmidt, Phys. Rev. B 94, 201111 (2016).
10. P. Nikolić and T. Senthil, Phys. Rev. B 71, 024401 (2005).
11. J. Villain, R. Bidaux, J.-P. Carton, and R. Conte, J. Phys. France 41, 1263 (1980).
12. E. Shender, Sov. Phys. JETP 56, 178 (1982).
13. G. Chen, Unpublished.
14. L. Savary and L. Balents, Phys. Rev. Lett. 118, 087203 (2017).
15. F. A. Cevallos, K. Stolze, T. Kong, and R. J. Cava, Mater. Res. Bull. 105, 154 (2018).
16. Y. Li, S. Bachus, Y. Tokiwa, A. A. Tsirlin, and P. Gegenwart, arXiv:1804.00696 (2018).
17. Y. Shen, C. Liu, Y. Qin, S. Shen, Y.-D. Li, R. Bewley, A. Schneidewind, G. Chen, and J. Zhao, arXiv:1810.05054 (2018).
18. C. Liu, Y.-D. Li, and G. Chen, Phys. Rev. B 98, 045119 (2018).
19. S. Isakov and R. Moessner, Phys. Rev. B 68, 104409 (2003).
20. Y.-C. Wang, Y. Qi, S. Chen, and Z. Y. Meng, Phys. Rev. B 96, 115160 (2017).
21. D. Blankschtein, M. Ma, A. N. Berker, G. S. Grest, and C. Soukoulis, Phys. Rev. B 29, 5250 (1984).
22. J. V. José, L. P. Kadanoff, S. Kirkpatrick, and D. R. Nelson, Phys. Rev. B 16, 1217 (1977).
23. K. Damle, Phys. Rev. Lett. 115, 127204 (2015).
24. P. Emonts and S. Wessel, Phys. Rev. B 98, 174433 (2018).
25. D. Reig-i Plessis, A. Cote, S. van Gelderm, R. Mayrhofer, A. Aczel, and G. MacDougall, arXiv:1906.10767 (2019).