Emergent Halperin-Saslow mode and Gauge Glass in quantum Ising magnet TmMgGaO$_4$

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We propose quenched disorders could bring novel quantum excitations and models to certain quantum magnets. Motivated by the recent experiments on the quantum Ising magnet TmMgGaO$_4$, we explore the effects of the quenched disorder and the interlayer coupling in this triangular lattice Ising antiferromagnet. It is pointed out that the weak quenched (non-magnetic) disorder would convert the emergent 2D Berezinskii–Kosterlitz–Thouless (BKT) phase and the critical region into a gauge glass. There will be an emergent Halperin-Saslow mode associated with this gauge glass. Using the Imry-Ma argument, we further explain the fate of the finite-field $C_3$ symmetry breaking transition at the low temperatures. The ferromagnetic interlayer coupling would suppress the BKT phase and generate a tiny ferromagnetism. With the quenched disorders, this interlayer coupling changes the 2D gauge glass into a 3D gauge glass, and the Halperin-Saslow mode persists. This work merely focuses on addressing a phase regime in terms of emergent U(1) gauge glass behaviors and hope to inspire future works and thoughts in weakly disordered frustrated magnets in general.

Disorder is unavoidable in quantum materials. Topological phases, such as the gapped spin liquids with intrinsic topological orders and topological insulators with symmetry-protected topological properties, are robust against weak disorders, so disorder effects are not often invoked in these contexts $^1$ $^2$. For conventional ordered phases, critical phases, and phase transitions, disorder effects require a more serious consideration. This is particularly relevant for many frustrated materials. Clearly, disorder and frustration play an important role in spin glass that is unambiguously one of most challenging subjects in condensed matter physics $^3$. Quenched impurities were shown to select spiral magnetism among degenerate spiral states in classically frustrated magnets $^4$, and quenched disorders could bring a strong quantum entanglement and generate highly entangled quantum states $^5$. However, there have not yet been many solid and conclusive results for the effects of quenched/annealed disorders in frustrated quantum magnetism $^6$. An impressive example of the disorder effect is the spin-1/2 random Heisenberg chain that was studied with the real-space renormalization group analysis and the master equation by D.S. Fisher $^7$. It establishes that any weak randomness in the exchange due to disorders would drive the system into the strong disordered random fixed point. More substantially, this analysis is asymptotically exact. For disorder effects, one should distinguish the long-distance and low-energy physics from the short-distance and high-energy one. The numerical study of small systems with buried disorders is certainly difficult to provide much useful information about the former with D.S. Fisher’s remarkable result as an example, while the understanding of the latter is feasible. Thus, theoretical arguments or making connection to established results in statistical physics can sometimes be helpful.

We are partly motivated from the triangular lattice antiferromagnet TmMgGaO$_4$ $^8$ $^{15}$. TmMgGaO$_4$ is a Mott insulator where the magnetic Tm$^{3+}$ ions form a triangular lattice. This material is isostructural to the triangular lattice spin liquid candidate YbMgGaO$_4$ that caught some attention earlier $^{16}$ $^{20}$. TmMgGaO$_4$ was shown to have an anisotropic magnetic behavior with a nearly zero magnetic response to the external magnetic field in the triangular plane $^8$ and a large magnetic moment normal to the triangular planes, i.e. the $z$ direction. Despite the structural disorders due to the Tm$^{3+}$ positional disorder and the Ga-Mg site disorder $^8$, further measurements found an ordering transition $\sim$1K $^9$ $^{11}$ $^{15}$. Detailed neutron scattering revealed a dominant Bragg peak at the $K$ point of the Brillouin zone, suggesting a three-sublattice magnetic order. Our previous theoretical efforts proposed a weakly-split doublet for the Tm$^{3+}$ ion to understand the low-temperature magnetic properties $^9$ $^{31}$, instead of the conventional non-Kramers doublet $^{10}$ $^{13}$ $^{32}$. The weak splitting between the doublet was modeled as an intrinsic transverse field on the effective spin-1/2 local moment $S_i$, and the exchange is primarily Ising-like with $^9$ $^{32}$

$$H_{TFIM} = \sum_{ij} J_{ij} S_i^x S_j^x - h \sum_i S_i^z. \tag{1}$$
This transverse field Ising model (TFIM) with an intrinsic microscopic origin provides a reasonable explanation of both the magnetic structure and the magnetic excitation from the inelastic neutron scattering measurements [9]. The intrinsic transverse field in $H_{\text{TFIM}}$ generates quantum tunneling events between different classical Ising ground states in Fig. 1, and the resulting three-sublattice order is a quantum effect, known as the “order-by-disorder” [33–35]. Due to the multipolar nature of the local moments, only the $z$ component, $S^z$, is visible in the magnetic measurement [9,10,13]. It is understood that $S^z$ flips the transverse component and generates quantum dynamics for the inelastic neutron detection [9]. More quantitative aspects of the experiments can be more sensitive to disorder effects on the exchange and Landé factors as well as the residual coupling to the high-order multipolar moments [8,11,15].

Aligned with the well-known result of the triangular lattice TFIM [34,35], a finite-temperature Berezinskii–Kosterlitz–Thouless (BKT) phase with an emergent U(1) symmetry would exist above the low-temperature three-sublattice order [12,13,33]. In an interesting Ga-based NMR experiments on TmMgGaO$_4$ [14], a hump in the $1/T_1$ dynamics was found from $\sim0.9K$ to $\sim1.9K$, indicating significant low-energy spin excitations in this temperature window, and was attributed as an evidence for the BKT phase, while there is no such hump in the Knight shift. As the magnetic susceptibility in the BKT phase exhibits a power-law divergence [26] with the field in the zero field limit, Ref. [14] then studied the magnetic susceptibility from 0.6T-0.9T and numerically fitted to power laws. This field range, equivalent to $\sim4K$-$6K$ for the Tm moment, may be a bit too large to be perturbative and would drive the system into a “up-up-down” state in Fig. 1 [13]. Ref. [15] performed an advanced neutron scattering measurement on TmMgGaO$_4$ and extracted the spin correlation length, $\xi$, as a function of temperature. They found that, $\xi$ is $\sim3$-$7$ lattice spacing in the claimed BKT phase, and saturates to $\sim30$ lattice spacing in the zero-temperature limit. This finite $\xi$ is in contrary to the divergent $\xi$ expected for the BKT phase.

Our work is partly to resolve different pieces of experiments in TmMgGaO$_4$. Considering several suggestion of non-magnetic disorders [8,11,15], we begin with the analysis of the zero field regime for the single triangular layer and incorporate the non-magnetic disorder by assuming a weak disorder. This is a continuation of the disorder analysis that was advertised in the end of Ref. [13]. We further include the interlayer coupling and consider the three dimensional limit. As a convention, we assume the disorders are uncorrelated and local. In the clean limit, the low-energy physics of the triangular lattice TFIM is well captured by a coarse-grained model [36]

$$H \simeq -J \sum_{\langle ij \rangle} \cos(\theta_i - \theta_j) - J_0 \sum_i \cos(6 \theta_i),$$

where $\langle ij \rangle$ refers to the nearest-neighbor bond on the coarse-grained triangular lattice, and $\theta$ is the phase of the complex order parameter at the $K$ point. Here, the order parameter is given as the three-sublattice order parameter $\psi = m_1 + m_2 e^{i2\theta/3} + m_3 e^{-i2\theta/3}$, where $m_i$ ($i = 1,2,3$) refers to $\langle S^z \rangle$ of three sublattices at the neighboring sites, and $\theta = \text{arg}[\psi]$, where we have set the lattice constant to unity. The coupling $J$, $J > 0$, is the coarse-grained exchange and favors a uniform phase $\theta$. The second term is a dangerously irrelevant 6-fold anisotropic term [37–40]. It is irrelevant in the BKT phase, and there is an emergent U(1) symmetry. At the low temperatures below the BKT phase, it pins the system to the three-sublattice order. We expect $J_0 < 0$ for TmMgGaO$_4$. In the BKT phase, if the spacing in the coarse-grained lattice is large enough, $J_0$ becomes very small and can be neglected in the initial analysis. As the BKT phase is compressible, it is more susceptible to the disorder. Once the non-magnetic disorder is considered, the coarse-grained model for the phase $\theta$ takes the following effective form,

$$H_{\text{gg}} \simeq -\tilde{J} \sum_{\langle ij \rangle} \cos(\theta_i - \theta_j - A_{ij}),$$

where $i,j$ here should be interpreted as different phase puddles due to disorder for the U(1) phase $\theta$ (see Fig. 2), and the gauge field $A_{ij}$ takes care of the disorder effects and is a random variable from 0 to $2\pi$. The parameter $\tilde{J}$, with $\tilde{J} > 0$, is an effective coupling between the puddles, and the neglecting of the randomness in the magnitude does not qualitatively change the physics. The puddle size is determined by the disorder strength. The model in Eq. (3) is known as the Hamiltonian of the U(1) gauge glass model in two dimensions [41–43]. It should be a good approximation of the physics as long as the anisotropic term becomes negligibly small at the scale of the puddle size, and we do not really need to sit right on the BKT phase in the clean limit. Numerical studies have shown that the lower critical dimension of this model is
FIG. 2. The schematic plot of phase puddles and couplings for the U(1) gauge glass.

greater than 2, but less than 3 [44, 45]. The model in Eq. (3) without the dangerously irrelevant anisotropic term would not have a thermal transition in the 2d limit. At low temperatures, the anisotropic term is expected to pin \( \theta \) and generate the three-sublattice order.

The well-known excitation of the U(1) gauge glass is the gapless Halperin-Saslow mode [46, 47]. This mode is related to the slow and smooth variation of the phase angle \( \theta \) and is of the Goldstone-type spin wave mode. It has been proposed in the context of the triangular lattice antiferromagnets NiGa\(_2\)S\(_4\) and FeAl\(_2\)Se\(_4\) with consistent thermodynamic behaviors [47–51]. Over there, the system is expected to have a continuous spin rotational symmetry and the exchange scale is fairly large so that the low-energy properties of this mode can be well-accessed [48, 50]. In our case, the U(1) symmetry is emergent and is not microscopically originated, thus the Halperin-Saslow mode is emergent and will in principle develop a tiny gap due to the irrelevant anisotropic term in Eq. (2). This tiny gap is determined by the magnitude of the anisotropic term at the puddle scale, and may not be well-resolved with the experimental resolution. Since this nearly-gapless emergent Halperin-Saslow mode is a spin-wave mode in nature, it shows up around the \( K \) point in the inelastic neutron scattering measurement and contributes to the NMR \( 1/T_1 \) dynamics.

The emergent U(1) symmetry is immediately destroyed once the magnetic field is applied along the \( z \) direction. The system quickly breaks the Ising-triangle degeneracy and enters an “up-up-down” state in the intermediate magnetic field regime [13] (see Fig. 1). The system no longer has the time reversal symmetry, and this state breaks the \( C_3 \) rotational symmetry around the centers of the triangular plaquettes. The Ginzburg-Landau analysis yields a strongly first-order transition, and this result was supported by the numerical calculation [13]. As the frustration is lifted in this finite field regime, the transition temperature is enhanced compared to the zero-field one. From the symmetry point of view, the non-magnetic disorder would directly couple to the \( C_3 \) order parameter, e.g. the nearest-neighbor bond spin correlation. Therefore, these non-magnetic disorders behave like random fields for the \( C_3 \) symmetry breaking. If we restrict ourselves to a single Tm\(^{3+}\) layer and neglect the weak interlayer coupling, the standard Imry-Ma argument [52] suggests that the long-range \( C_3 \) order should be destroyed by any arbitrarily weak disorder strength. The system would be separated into different domains of the \( C_3 \) orders, and the domain size is determined by the balance between the surface energy of the domains and the random field strength. The strong first order transition is expected to be rounded by the disorder. As two dimensions is the lower critical dimension for the stability of the ordered phase based on the Imry-Ma’s argument, the correlation length for the \( C_3 \) order is an exponential function of the disorder strength below the transition temperature. This may be further measured by advanced neutron scattering techniques in Ref. [15].

The Tm\(^{3+}\) local moment has a rather large magnetic moment and is approximately 7.3\( \mu_B \) per Tm\(^{3+}\) ion [8], suggesting the long-range dipole-dipole interaction can be particularly relevant for the low-temperature magnetic properties. Even for the clean limit in two dimensions, since the BKT phase has an infinite correlation length that suppresses the irrelevant coupling, how the (infinitely) long-range dipole-dipole interaction impacts on the BKT phase is an interesting question. It was estimated that the second (third) neighbor dipole-dipole interaction in the triangular plane is \( \sim 0.48 \)K \( (\sim 0.31 \)K) [13]. It turns out the interlayer dipole-dipole interaction can be equally relevant as the orientation of the nearest-neighbor Tm bond between the neighbor layers enhances the interaction and generates a ferromagnetic coupling. It is estimated that the nearest-neighbor interlayer coupling is \( \sim -0.30 \)K, and there are six nearest-neighbor interlayer sites (see Fig. 3). This coupling is expressed as

\[
H_{\text{int}} = \sum_{\langle ij \rangle_{\text{int}}} J_{\text{int}} S_i^z S_j^z, \tag{4}
\]

where \( J_{\text{int}} < 0 \) and “int” refers to the interlayer coupling. Although this coupling is relatively small compared to the dominant coupling for the intralayer TFIM, it is still natural to consider the role of this coupling, especially for the very low temperature properties around 1K. Once this interlayer coupling is considered, the system immediately becomes three dimensions. The BKT phase and transitions are defined in two dimensions and will disappear in three dimensions (except the vortices form strong line objects), and then the three-dimensional ordering transition is often obtained from \( T \sim \xi(T)^2 |J_{\text{int}}| \) where \( \xi(T) \) is the intralayer correlation length. On the other hand, the ferromagnetic interlayer coupling for a three-dimensional system would favor ferromagnetism. For this purpose, we start from the BKT phase at the finite temperature for each layer and couple the layers with the interlayer coupling. We can obtain the coarse-grained
interactions. The last term in \( F \) from the intralayer Ising coupling and the dipole-dipole order term in the expansion of the free energy in the three-sublattice antiferromagnetism once the weak ferromagnetism would be favored in addition to the quantum order by disorder, the spin-wave gap of this system remains with disorders. Due to the diversity of frustrated periodic lattice models and continuum models and neither have been very fruitful as we are more familiar with periodic lattice models and continuum models and neither remain with disorders. Due to the diversity of frustrated materials and the physical properties, the standard methods like Harris criteria, Imry-Ma argument, replica theory, real-space renormalization group should be properly adjusted to different contexts and disorder realization. Thus, experiments are necessary to give the guidance for the theoretical development here. This is especially so for the disorder effect in spin liquids which is an important subject along the line. While weak disorders do not cause much effect for topological spin liquids, it can be favored a weak ferromagnetism if \( \Delta > 1 / 6|J_{\text{int}}| \). Thus, even if we relax the divergent \( \chi_{2d} \) condition, a weak ferromagnetism should be expected. This above argument holds even with weak disorders. We attribute this as the origin of the weak \( \Gamma \) point Bragg peak in the neutron scattering measurement [9].

Once the interlayer coupling is included, the coarse-grained model in Eq. (3) picks up an interlayer Josephson-type coupling, and the model becomes a \( U(1) \) gauge glass model in three dimensions. Again, the interlayer Josephson-type coupling is also disordered by the Ga-Mg disorder and the Tm positional disorder. The emergent Halperin-Saslow mode persists to three dimensions. It was numerically shown that this \( U(1) \) gauge glass model has a finite-temperature phase transition [57]. The ordering would be further solidified by the anisotropic term in Eq. (2).

Discussion.—Here we give a discussion about the specific properties of TmMgGaO\(_4\), and then sketch a discussion about the disorder treatments in frustrated magnets and scenarios in spin liquids. As TmMgGaO\(_4\) is cooled from the high-temperature paramagnetic regime, it gradually builds up spin correlations. According to the careful and detailed measurements in Ref. 15, the spin correlation length grows up to the Tm-Tm lattice spacing around 6K, and this can be regarded as the entering of the cooperative paramagnetic regime. When the correlation length grows up to the puddle size set by the disorder strength in Eq. (3), an approximate \( U(1) \) gauge glass description applies. In addition to the emergent Halperin-Saslow mode, there exists more subtle low-energy excitations, i.e. “droplets” of the droplet theory for the gauge glass [11, 45, 58]. These are non-smooth deformation of the phase, and related to the vortex configurations that re-arrange, and when they do on length scale \( L \) it costs an energy that depends on \( L \) algebraically and on the dimensions. These low-energy excitations all contribute to the specific heat, while the Halperin-Saslow mode should be detectable in the inelastic neutron scattering and NMR 1/\( T_1 \) measurements. Thus, although the Ga-Mg disorder may cause a further complication in deciphering the experiment, we propose the hump in the NMR 1/\( T_1 \) dynamics [14] is a signature of the emergent Halperin-Saslow mode for the gauge glass. Compared to the BKT scenario in the clean two-dimensional case [14], the Halperin-Saslow mode of gauge glass is out of the weakly-disordered perspective and holds even for three dimensions. As the Halperin-Saslow mode would scatter the phonons strongly, we expect there will be a dip/valley-like suppression in the thermal transport. Since the gap in the three-sublattice order is quite small [9], this suppression may not be quite visible. As the system is further cooled, the dangerously irrelevant anisotropic term in Eq. (2) becomes important and drives the system into the three-sublattice order at low temperatures. The low-temperature three-sublattice order in TmMgGaO\(_4\) is a fully gapped incompressible state and thus a bit more stable to weak disorders than compressible ones. Due to the quantum order by disorder, the spin-wave gap of this order is small but is still determined by the combination of the microscopic couplings. This spin-wave gap shows up directly in the inelastic neutron scattering [9, 15], and as an Arrhenius form in the specific heat and the NMR 1/\( T_1 \) dynamics at the corresponding temperatures [14].

The study of disorder effects in frustrated magnets has not been very fruitful as we are more familiar with periodic lattice models and continuum models and neither remain with disorders. Due to the diversity of frustrated materials and the physical properties, the standard methods like Harris criteria, Imry-Ma argument, replica theory, real-space renormalization group should be properly adjusted to different contexts and disorder realization. Thus, experiments are necessary to give the guidance for the theoretical development here. This is especially so for the disorder effect in spin liquids which is an important subject along the line. While weak disorders do not cause much effect for topological spin liquids, it can be read.

\[
F_{3d} = \sum_{\ell} \frac{M_{\ell}^2}{2\chi_{2d}} - 3|J_{\text{int}}| \sum_{\langle \ell\ell' \rangle} M_{\ell} M_{\ell'} + c_4 \sum_{\ell} M_{\ell}^4 + \cdots,
\]

where \( M_{\ell} \) is the uniform magnetization per site on the \( \ell \)th layer, and “\( \cdots \)” refers to other terms such as the ones from the intralayer Ising coupling and the dipole-dipole interactions. The last term in \( F_{3d} \) is simply the leading order term in the expansion of the free energy in \( J_{\text{int}} \). This can also be interpreted as a mean-field treatment of the ferromagnetic interlayer coupling. This is similar to the “chain mean-field theory” [53], that has been successfully and widely applied to understand the experiments in low-dimensional magnetic systems [54-56], and is shown to be rather reliable. In the BKT regime of each layer, the uniform susceptibility \( \chi_{2d} \) was argued to diverge [30], and the first term that describes the energy cost for a finite magnetization simply vanishes. Thus, a tiny ferromagnetism would be favored in addition to the three-sublattice antiferromagnetism since the weak ferromagnetic interlayer coupling is turned on in this BKT regime. In fact, a simple calculation of Eq. (5) would favor a weak ferromagnetism if \( \chi_{2d} > 1 / (6|J_{\text{int}}|) \). Thus, even if we relax the divergent \( \chi_{2d} \) condition, a weak ferromagnetism should be expected. This above argument holds even with weak disorders. We attribute this as the origin of the weak \( \Gamma \) point Bragg peak in the neutron scattering measurement [9].

\[ F_{3d} = \sum_{\ell} \frac{M_{\ell}^2}{2\chi_{2d}} - 3|J_{\text{int}}| \sum_{\langle \ell\ell' \rangle} M_{\ell} M_{\ell'} + c_4 \sum_{\ell} M_{\ell}^4 + \cdots, \]
important for the physical properties of critical spin liquids. The perturbative analysis of weak disorders for the spinon Fermi surface $U(1)$ spin liquid indicates that the system changes from the spinon Fermi surface metal to the diffusive spinon metal and then to the spinon Anderson insulator as the disorder strength increases, while the fractionalization and emergent non-locality that define the spin liquids are actually preserved [23-59]. In contrast, the strong disorder argument would directly favor a high dimensional version of the random singlet phase [30, 60]. Depending on the sample quality and disorder strengths, both regimes may be realized in relevant materials such as the doped semiconductors [59] and the triangular lattice antiferromagnet YbMgGaO$_4$ [10, 29].

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