A Scaling Theory for ac Magnetic Response in Kagomé Ice

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A theory for frequency-dependent magnetic susceptibility $\chi(\omega)$ is developed for thermally activated magnetic monopoles in kagomé ice. By mapping this system to a two-dimensional (2D) Coulomb gas and then to a sine-Gordon model, we have shown that the susceptibility has a scaling form $\chi(\omega)/\chi(0) = F(\omega/\omega_1)$, where the characteristic $\omega_1$ is related to a charge correlation length between diffusively moving monopoles, and to the sine-Gordon principal breather. The dynamical scaling is universal among superfluid and superconducting films, and 2D XY magnets above Kosterlitz-Thouless transitions.

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Frustrated spin systems have attracted considerable attention for decades, because they provide an opportunity of uncovering novel phases and excitations [1]. Even in the simplest cases of the Ising antiferromagnets on triangular [2] and kagomé [3] lattices, exact solutions revealed the absence of magnetic order and the macroscopic degeneracy in the ground states, which are viewed as the hallmark of the frustration.

Rare-earth pyrochlore oxides such as Ho$_2$Ti$_2$O$_7$ [4] and Dy$_2$Ti$_2$O$_7$ [5] proffers a new paradigm in this research area [6]. It is considered that despite large magnetic moments $\mu_{\text{eff}} \sim 10\mu_B$ the spins on the pyrochlore lattice do not order down to a quite low temperature $\sim 0.1$ K [6, 7], and exhibit a residual entropy [3] (although possibilities of a magnetic order [7] and an absence of the residual entropy [8] have been reported). The origin of these behaviors can be attributed to a strong Ising anisotropy with respect to the local $[111]$ axis and an effective ferromagnetic coupling between neighboring spins [10], which, then, force the spins at four corners of each tetrahedron to satisfy the two-in and two-out condition. Since this constraint is the same as that for proton configurations in water ice [11], those materials are named as “spin ice”.

Recently, point-defect excitations in spin ice created by breaking the ice rule [12] have been intensively investigated [13–16], since the intriguing prediction of magnetic monopoles [17, 18]. These excitations behave as quasi-particles with magnetic charges moving on the diamond lattice [12, 13, 20] like ion defects, H$_2$O$^+$ and HO$^-$, in water ice [21]. While much efforts have been paid to account for their static and dynamical properties, there still exist unclear points and subjects to explore [22]. This is partly because the monopole-like excitation is a topological defect, and is a nonlocal object emerging in the vicinity of the ground-state manifold.

A way to circumvent its intractability is to make it move in more restricted space, e.g., in two dimensional (2D) space. A 2D spin ice can be achieved by applying a magnetic field $H_{\text{dc}}$ along a $[111]$ direction, along which the pyrochlore lattice is stacking of triangular and kagomé lattices [23, 24]. When the $[111]$ field is not very high, the spins on the triangular layers are fixed parallel to the field direction at low temperatures, and consequently the spins on each kagomé layer are decoupled and remain frustrated, which endowed the name “kagomé ice” [24, 25]. The kagomé-ice state is characterized by a magnetization plateau [24, 25] and a reduced residual entropy [24, 26, 27, 30]. In the low temperature limit $T \to 0$, it is in a Coulomb phase [33] with the power-law decay of spin correlations [31]. At low $T$, kagomé ice is characterized by a long charge correlation length $\xi$ or a small monopole density $n_m \propto \xi^{-2}$ [28, 29, 31].

A minimal Hamiltonian for kagomé ice is a nearest-neighbor (NN) model [23] consisting of one kagomé layer and neighboring two triangular layers with pinned spins:

$$H_{\text{ice}} = J_{\text{eff}} \sum_{\langle ij \rangle} \sigma_i \sigma_j - \mu_{\text{eff}} \sum_i H_{\text{dc}} \cdot \hat{z}_{a(i)} \sigma_i \ (J_{\text{eff}} > 0). \quad (1)$$

$\sigma_i (= \pm 1)$ is an Ising variable for a spin $S_i = \sigma_i \hat{z}_{a(i)}$ at a site $i$ on a sublattice $a(i)$, and $\hat{z}_{a(i)}$ stands for a unit vector parallel to the local Ising axis. The NN exchange interaction is antiferromagnetic in terms of Ising variables. The ice rule, requiring $\sum_{a=1}^4 \sigma_a = 0$ for each tetrahedron, can be broken by thermal activation, creating magnetic monopoles with a charge $q = \frac{1}{2} \sum_{a=1}^4 \sigma_a = \pm 1$, which is illustrated in Fig. 1.

Another motivation of this work originates from our recent experimental studies on the dynamics of monopoles moving in the kagomé plane of Dy$_2$Ti$_2$O$_7$ [34]: As pointed out there, the 2D dynamics of monopoles excited from the kagomé-ice state can be investigated by applying an ac magnetic field $H_{\text{ac}}(t) = \text{Re}[H_0 e^{i\omega t}]$ perpendicular to $H_{\text{dc}}$, which works as driving force for monopoles and measuring induced magnetization $M_{\text{ac}}(t) = \text{Re}[\chi(\omega)H_0 e^{i\omega t}]$. The frequency-dependent ac susceptibility $\chi(\omega)$ has been indeed measured. In the low-frequency ranges of the experiments [34, 35], the monopole motion is thought to be governed by diffusion [37], characterized by a diffusion constant $D$.

In this letter, we have theoretically studied $\chi(\omega)$, and have found that it gives deep insight into the monopole dynamics in the kagomé-ice state. Using the monopole...
A link between the magnetization ofices possibly including the artificial spin ice [44–49].

Kosterlitz-Thouless transition [42, 43], and generic 2D superconducting films [40], 2D XY magnets above the Kosterlitz-Thouless transition [42, 43], and generic 2D superconducting films [40], kagomé ice can be mapped to the 2D Coulomb gas model [38], in which monopoles interact via the logarithmic Coulomb potential, which is caused by the entropic interaction in kagomé ice [31, 32]. The magnetic response accompanied by creations, annihilations and rearrangements of monopoles: the string flip along

\[ \chi = \frac{1}{2k_B T} \left( M^2 \right)_{\text{ice}} \approx \frac{1}{2k_B T} \left( P^2 \right)_{\text{ice}} [51]. \]

Since kagomé ice exhibits an isotropic charge correlation, we rewrite \( \chi \) by introducing the charge-density distribution function \( \rho(x) = \sum_{i \in \Lambda} q_i \delta(x - x_i) \), as

\[ \chi = \Omega \pi \mu_{\text{eff}}^2 \frac{k_B T a^2}{\pi \mu} \int_0^\infty dr \ r^3 C(r), \]

where \( r = |x| \) and \( \Omega \) is the area of \( \Lambda \). The charge correlation function was defined as \( C(r) = \left< \langle \rho(x) \rho(0) \rangle_{\text{ice}} \right> \).

Equation (3) exhibits the magnetic response in terms of the monopole degrees of freedom. However, it is restricted to the static case, so its extension to the dynamical case is our next task. For this purpose, second, we address Ambegaokar’s argument, which provides a link between the ac response and a static charge correlation [39]. In the analysis of the superfluid film on the oscillating substrate, Ambegaokar et al. supposed a diffusive motion of vortices, and then obtained the ac response by focusing on a role of the mean diffusion length during a period of an ac field \( L_\omega = \sqrt{D}/\omega \). One intuitive reasoning is as follows: Consider a pair of monopoles with separation \( r \), and suppose its time-dependent polarization keeping in phase with the ac field. Then, a monopole should move a reachable distance by diffusive motion within one period. Thus, in order to give the in-phase response, the pair should satisfy a condition \( r < O(L_\omega) [38] \). Here, we also assume a diffusive motion and apply this heuristic argument to express the in-phase component (real part) \( \chi'(\omega) \) of \( \chi(\omega) \). The result is given by replacing the upper bound of the integral in Eq. (3) with the length of \( O(L_\omega) \):

\[ \chi'(\omega) = \Omega \pi \mu_{\text{eff}}^2 \frac{k_B T a^2}{\pi \mu} \int_0^{L_\omega} dr \ r^3 C(r). \]
We have introduced a constant $b$ less than order of unity. In order to infer the value of $b$, we assume a uni-
dimensional motion of monopoles parallel to $H_{dc}$. Then, the round-trip path length of monopoles is about $4r$ so that the condition $4r < L_ω$ is satisfied. We thus assume $b = \frac{1}{4}$ \cite{52}. The imaginary part $\chi''(Ω)$ is obtained by applying the Kramers-Kronig (KK) relation to $\chi'(Ω)$. The above formula is doubly important: It gives the magnostic response in terms of the charge correlation, which is possible only for magnets to afford their defect representations like the monopole system for kagomé ice. Also, since the frequency dependence is introduced as a finite-size effect, it may be governed by a ratio of $L_ω$ to the characteristic length $ξ$ in kagomé ice. Below, one can see that this is indeed an origin of a scaling nature of $\chi(Ω)$.

Now, we can obtain $\chi(Ω) = χ'(Ω) - iχ''(Ω)$ via $C(ξ)$, which is given as an average with respect to $H_{ice}$. This is rather convenient for numerical calculations; in fact, we perform Monte Carlo simulations to evaluate Eq. (4). (However, to analytically evaluate $\chi(ξ)$, a monopole representation of $H_{ice}$ is necessary. It has been argued that a gaseous model gives its effective description

$$H_{CG} = k_B T \sum_{l,m ∈ Λ; l > m} - gq_m g(ξ_l - ξ_m) κ,$$

(5)

where a neutrality condition $\sum_{i ∈ Λ} g_i = 0$ is imposed \cite{32}. $g(ξ)$ is a lattice propagator to give the correlation between two monopoles with a separation $ξ$ in the ground-state manifold, and represents an entropic interaction. Since we are focusing on the system with a long correlation length $ξ ∝ 1/\sqrt{m}$, the interaction can be approximated by its asymptotic behavior $ln(ξ/\alpha_0)$, where $\alpha_0$ is a monopole core radius. In this dilute-gas regime, we can obtain a simple universal system by coarse-graining a lattice structure and neglecting short-range fluctuations \cite{43}. Equation (5) defines the 2D coulomb-gas (CG) model \cite{53}, where $κ$ is the inverse CG temperature, and is independent of $T$ ($κ ∝ \frac{1}{T}$ for an ice-rule system \cite{54}).

A large reduction of the problem has been attained, but as its drawbacks, we should explicitly control a number of monopoles. Let us write a $N$-monopole partition function as $Z_N$, then the grand-partition function is given as $Z_{CG} = \sum_{N, \text{even}} y^N Z_N$, where the fugacity $y = e^{-Δ/k_B T}$. $Δ$ is a monopole creation energy, which is, to some extent, controlled by $H_{dc}$. Then, we can estimate the asymptotic behavior of the charge correlation function as $C(ξ) ∝ -<ρ(ξ)ρ(0)>_{CG}$, where $<⋯>_{CG}$ means an average with respect to $Z_{CG}$.

While CG possesses a low-temperature phase ($κ ≥ 4$) where monopoles with opposite charges are bounded into pairs \cite{43}, kagomé ice corresponds to CG in the high-temperature phase ($κ ≤ \frac{1}{2}$), and thus, exhibits a screened charge correlation due to free monopoles. To evaluate $C(ξ)$, we utilize the well-known equivalence between CG and the sine-Gordon model \cite{38,41} defined by the partition function $Z_{CG} = ∫ [dθ] e^{-S_{cg}}$ with the action \cite{55}:

$$S_{cg} = ∫ d^2 x \left[\frac{1}{2πκ} (∇θ)^2 - 2z cos √2θ\right],$$

(6)

where $z ≃ (y/ξ) a_0^{3/2}$ ($ξ$ is an area of unit cell of $Λ$). Then, in the sine-Gordon language, $C(ξ) = 4z^2 c(ξ)$ ($ξ ≠ 0$) with

$$c(ξ) = <sin √2θ(x) sin √2θ(0)>_{CG},$$

(7)

where $<⋯>_{CG}$ means an average with respect to $Z_{CG}$. In this formulation, $c(ξ)$ is short-ranged due to the relevant nonlinear term in Eq. (6). In such cases, it can be calculated reliably by using the formfactor perturbation (FFP) method. While the method tells us elementary processes to be considered and provides a simple expression for $c(ξ)$, its explanation is devoted to a rather technical aspect of the massive sine-Gordon theory. Thus, here we only provide the result—for readers interested in details, please see the Supplementary Material (SM):

$$c(ξ) = \left(\frac{λn_m}{2z}\right)^2 \frac{K_0(m_1 r)}{π},$$

(8)

where $K_α$ denotes the $α$th-order modified Bessel function of the second kind. Thus, $C(ξ)$ is short-ranged with $ξ = 1/m_1$, where $m_1$ is the mass of the principal breather $B_1$.

Now, we are in a position to obtain the ac susceptibility. Performing the integral transform of Eq. (10) \cite{40,50}, we obtain the real part as

$$\chi'(Ω)/\chi(0) ≃ 1 - \frac{γ^3}{8} [K_1(γ) + K_3(γ)] \right).$$

(9)

We defined a characteristic frequency of the $B_1$ excitation (see SM) as $ω_1 = D/ξ^2$ and a ratio as $γ = bL_ω/ξ = b√ω_1/ω$. The static susceptibility $χ = χ(0)$ which gives a magnitude of $χ(Ω)$ is found to obey Curie’s law:

$$χ(0) ≃ Ω \frac{4μ_{eff}^2}{k_B T a^2} λ^2 n_m^2 ∈^4 = \frac{C}{k_B T}.$$  

\right. 

(10)

Intriguingly, Curie’s constant depends on $κ$ characterizing the ground-state manifold ($C ≃ 0.55 × μ_{eff}^2 N_λ$ for $κ = \frac{1}{2}$ and $N_λ$ spins), which may give one aspect of the 2D cooperative paramagnets. The imaginary part is obtained by the KK relation: the result is written, in terms of Meijer’s $G$ functions \cite{57}, as

$$\chi''(Ω)/\chi(0) ≃ \frac{2}{8π} \left[ G_{1,1}^{4,1} \left( \frac{4}{28} \right| 0 \right| a] - G_{1,1}^{4,1} \left( \frac{4}{28} \right| 0 \right| b \right],$$

(11)

where vectors $a = (0, 0, 1, \frac{1}{2}, -\frac{1}{2})$ and $b = (0, 0, 1, \frac{1}{2}, 1, \frac{1}{2})$.

Before exploring ingredients of Eqs. (9)-(11), two comments are in order: (i) Because we have focused in the vicinal region from the ground-state manifold of kagomé ice, the temperature should not be so high to bring the system out of the region. Moreover, $H_{dc}$ should be at least in the interval to give the magnetization plateau.
We employ a system of kagomé ice by using the NN model of $H_{\text{ice}}$.

In conclusion, we have investigated the ac susceptibility $\chi(\omega)$ of kagomé ice: We clarified that $\chi(\omega)$ takes an universal scaling form in terms of the ratio of $\omega$ to $\omega_1$, the characteristic frequency for the principal breather—it is a localized excitation composed of a soliton and antisoliton, but possibly looks more similar to an ordinary wave. Furthermore, we performed MC simulations, and provided data that represent the scaling form as expected in our theory. The present results suggest that breather’s dynamics characterizes the low-$T$ behavior of the magnetic monopoles in kagomé ice. In this letter, it has been explained that the universal dynamics of the magnetic monopole-like defects in the 2D spin ice can be captured by the theory of the ac magnetic response. Now, in view of the universality concept, it is natural to expect that our theory can also serve for analysis of the dynamics observed in other systems such as the vortices in superfluid and superconducting films, as well as in 2D XY magnets above the Kosterlitz-Thouless transition, and also the charged particles in 2D electrolytes. These remain as interesting future applications.

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Supplementary Material:
“A Scaling Theory for ac Magnetic Response in Kagomé Ice”

This Supplementary Material contains an explanation on the formfactor perturbation (FFP) calculation of the charge correlation function defined by Eq. (1) and its lowest-order result given by Eq. (3) [1]. Also, a relationship between the charge correlation length and the defect number density is obtained as a by-product.

The model Eq. (3) possesses low-energy excitations of the soliton s, the antisoliton ¯s, and breathers Bj with 0 ≤ κ < 2 (j = 1, · · ·, [1/p]) with 1/p = 4/κ − 1) [2, 3]. The mass spectrum m consists of a doublet of s and ¯s, m± = M, and singlets of Bj,

\[ m_j = 2M \sin \left( \frac{\pi p}{2} j \right). \]  

(S1)

The soliton mass varies as a power of the scaling field \( z \) [4, 5]:

\[ M = \frac{2\Gamma(p/2)}{\sqrt{\pi \Gamma((1 + p)/2)}} \left[ \frac{\Gamma(1/(1 + p))}{\Gamma(p/(1 + p))} \right]^{1/2}, \]  

(S2)

and represents an inverse length scale \( (M \approx 6.22 \times z^\frac{p}{2} \text{ for } \kappa = \frac{1}{2}) \). The FFP method expands the correlation function as \( c(r) = \sum_{N=1}^\infty c_N(r)/N! \), where the contribution from the \( N \)-excitation sector is given by

\[ c_N(r) = \left[ \sum_{\epsilon_k} \frac{d \theta_k}{2\pi} e^{-E_k(\theta_k)r} \right] \left| F_{\text{sin}}(\{\theta\};\epsilon) \right|^2. \]  

(S3)

The two sets \( \{\epsilon\} = \{\epsilon_1, \cdot \cdot \cdot, \epsilon_N\} \) and \( \{\theta\} = \{\theta_1, \cdot \cdot \cdot, \theta_N\} \) specify aforementioned species of excitations and their rapidities, respectively. An \( \epsilon_k \) excitation with a rapidity \( \theta_k \) has the energy \( E_k(\theta_k) = m_{\epsilon_k} \cos \theta_k \). The formfactor, \( F_{\text{sin}}(\{\theta\};\epsilon) \), represents a matrix element of \( \sin \sqrt{2} \theta \) between the ground state and excited states, and selects relevant excitations to the correlation function. In this respect, the invariance of Eq. (4), under the charge conjugation \( C : \theta \rightarrow -\theta \) has the central importance [2]. Since the charge-density operator transforms as \( C : \sin \sqrt{2} \theta \rightarrow -\sin \sqrt{2} \theta \), nonvanishing contributions stem from excitations with odd parity. Consequently, the leading contribution comes from the principal breather \( B_j \) which is taken into account in the expansion, i.e., \( N = 1 \) and \( \epsilon_1 = 1 \) in Eq. (S3). The formfactor is then independent of the rapidity, and is given by \( F_{\text{sin}}(\theta_1) = -\lambda e^{\sqrt{2} \theta}/s_{G} = -\lambda n_{m}/2z \) [5], where the constant

\[ \lambda = 2\cos \left( \frac{\pi p}{2} \right) \sqrt{2\sin \left( \frac{\pi p}{2} \right) \exp \left( -\int_{0}^{\pi p/2} \frac{tdt}{2\pi \sin t} \right)}, \]  

(S4)

and the defect number density [8, 9]

\[ n_{m} = \frac{(1 + p)}{4} \tan \left( \frac{\pi p}{2} \right) M^2. \]  

(S5)

As a result, the charge correlation function is simply given by Eq. (3). Since its asymptotic behavior in \( r \rightarrow \infty \) can be written as \( c(r) \sim \exp(-m_{1}r)/\sqrt{r} \), the charge correlation length \( \xi = 1/m_{1} \). Therefore, from Eqs. (S1) and (S5), we obtain the relationship between \( \xi \) and \( n_{m} \) as

\[ \xi^{-1} = \sqrt{\frac{8n_{m}}{1 + p} \sin(\pi p)}. \]  

(S6)

For instance, \( \xi^{-1} = \sqrt{7n_{m}\sin(\pi/7)} \) for \( \kappa = \frac{1}{2} \).

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