Numerical evidence of quantum melting of spin ice: quantum-classical crossover

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Unbiased quantum Monte-Carlo simulations are performed on the simplest case of the quantum spin ice model, namely, the nearest-neighbor spin-$\frac{3}{2}$ XXZ model on the pyrochlore lattice with an antiferromagnetic longitudinal and a weak ferromagnetic transverse exchange couplings, $J$ and $J_\perp$. On cooling across $T_{C\text{SI}} \sim 0.2J$, the specific heat shows a broad peak associated with a crossover to a classical Coulomb liquid regime characterized by a remnant of the pinch-point singularity in longitudinal spin correlations as well as the Pauling ice entropy for $|J_\perp| < J$, as in classical spin ice. On further cooling, the entropy restarts gradually decaying to zero for $J_\perp > J_{\text{lc}} \sim -0.103J$, as expected for bosonic quantum Coulomb liquids. With negatively increasing $J_\perp$ across $J_{\text{lc}}$, a first-order transition occurs at a nonzero temperature from the quantum Coulomb liquid to an XY ferromagnet. Relevance to magnetic rare-earth pyrochlore oxides is discussed.

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A compact U(1) gauge theory hosts dual electric and magnetic monopoles as well as photons, as emerge in non-Abelian gauge theories for grand unified theories \cite{1,2}. In condensed matter, it is expected to appear in a nearest-neighbor classical spin ice (CSI) model. It includes, for instance, Pr$_2$Ir$_2$O$_7$ \cite{9}, Pr$_2$Zr$_2$O$_7$ \cite{6,7,10}, Yb$_2$Ti$_2$O$_7$ \cite{11,12}, and Tb$_2$I$_2$O$_7$ \cite{13,14}, all of which show vital roles of quantum effects due to nonzero $J_\perp$.

The particular limit $J_\perp = 0$ of the model is reduced to the nearest-neighbor classical spin ice (CSI) model. It involves a macroscopic degeneracy of the ground states satisfying the 2-in, 2-out spin ice rule \cite{15,19} and the Pauling residual entropy $S_P = \frac{1}{2} \ln 3$ \cite{15}, as observed in Ho$_2$Ti$_2$O$_7$ \cite{19,21} and Dy$_2$Ti$_2$O$_7$ \cite{22}. This CSI has been well understood in terms of a classical Coulomb phase physics in a gauge theory on the dual diamond lattice \cite{1,23,24}: the Hamiltonian is given by $H = \sum_{(r,r')} [J s_r^+ s_{r'}^- + J_\perp (s_r^+ s_{r'}^\perp + s_r^\perp s_{r'}^+)]$, with an spin-$\frac{1}{2}$ operator $s_r = (s_r^x, s_r^y, s_r^z)$ at a pyrochlore lattice site $r$, and the nearest-neighbor longitudinal ($z$) and transverse ($xy$) exchange couplings $J(>0)$ and $J_\perp$. This model gives the most simplified case of low-energy effective spin models for prototypical magnetic rare-earth pyrochlore oxides, defined in the $C_2$-invariant local spin frames with their $z$ axes pointing inwards or outwards from the center of the tetrahedron \cite{4,8}. These oxides include, for instance, Pr$_2$Ir$_2$O$_7$ \cite{9}, Pr$_2$Zr$_2$O$_7$ \cite{6,7,10}, Yb$_2$Ti$_2$O$_7$ \cite{11,12}, and Tb$_2$I$_2$O$_7$ \cite{13,14}, all of which show vital roles of quantum effects due to nonzero $J_\perp$.

In this Letter, we reveal a finite-temperature phase diagram of the simplest QSI model \cite{1}, uncovering two form \cite{23}, indicating deconfined monopoles.
successive crossovers and a single first-order phase transition shown with dashed and a solid lines in Fig. 1. From a high-temperature side, the system first crosses over to a classical Coulomb liquid or CSI regime with the entropy approximating to the Pauling entropy, and then for small enough $|J_\perp/J|$, to a quantum Coulomb liquid or QSI regime where the Pauling entropy is gradually released to zero. For $J_\perp < J_{\perp c}$ with $J_{\perp c}/J = -0.103$ [3], a first-order phase transition occurs to an XY-ferromagnet (XY-FM) [5].

All the numerical results presented in this Letter are obtained with unbiased worldline QMC simulations based on the path integral formulation in the continuous imaginary time [33]. To update worldline configurations, we adopt a directed-loop algorithm [34] in the $\{s^z_i\}$ basis, with the modification previously introduced for soft-core bosonic systems to reduce the computational cost [35]. To moderate the freezing problem often arising in frustrated systems, we employed the thermal annealing, i.e., the temperature is gradually decreased in the simulations. We performed typically $\sim 10000$ Monte-Carlo sweeps for each temperature.

Let us start with the disordered side $J_\perp > J_{\perp c}$ of the phase diagram. Figure 2 shows for $J_\perp/J = -1/11$ the temperature dependence of (a) the energy density $\varepsilon \equiv \langle H \rangle/N_s$ with $N_s (= 4 \times L^3)$ being the total number of spins, (b) the specific heat $C \equiv \partial \varepsilon / \partial T$, and (c) the entropy $S$ for $J_\perp/J = -1/11 > (J_\perp/J)_c$. Dashed and solid lines in (a) are obtained through the cubic and the basis spline interpolation of the QMC data, respectively. The wider lines in (b) correspond to the numerical derivative of the basis spline functions of $\varepsilon$. The thinner solid lines in the inset of (b) are the fits to $C_{\text{photon}} = \frac{1}{2} T^2 e^{-\frac{1}{2} \varepsilon^2 / T^3}$.

From a high-temperature local-moment regime with the entropy of the order of $\log 2$ to a classical Coulomb liquid or CSI regime where the entropy gradually decays to the spin ice plateau $S_P$ as shown in Fig. 2 (c). On further cooling across $T_{\text{QSI}}$, the specific heat $C$ shows an upturn, gradually releasing the Pauling entropy, indicating that the spin ice is melt by quantum fluctuations [3] and the system crosses over to a quantum Coulomb liquid or QSI regime. The lowest-energy excitations of the quantum Coulomb liquid are linearly dispersive “photons”, which describe a gauge-charge-0 harmonic oscillator mediated coupled transverse and longitudinal spin fluctuations [3]. Assuming the dispersion

![FIG. 1: (Color online) Finite-temperature phase diagram for $J_\perp < 0$, obtained with QMC simulations. Below the phase boundary (solid line) the transverse ($xy$) component of spins are ferromagnetically ordered, i.e., the order parameter $\langle s^x \rangle$ and the stiffness $\rho_s$ are both finite. The blue dots are extracted from Ref. [3]. The dashed lines indicate the crossover temperatures estimated from the position of the broad peaks in specific heat curves.](image)

![FIG. 2: (Color online) Temperature dependence of (a) the energy density $\varepsilon$, (b) the specific heat $C \equiv \partial \varepsilon / \partial T$, and (c) the entropy $S$ for $J_\perp/J = -1/11 > (J_\perp/J)_c$. Dashed and solid lines in (a) are obtained through the cubic and the basis spline interpolation of the QMC data, respectively. The wider lines in (b) correspond to the numerical derivative of the basis spline functions of $\varepsilon$. The thinner solid lines in the inset of (b) are the fits to $C_{\text{photon}} = \frac{1}{2} T^2 e^{-\frac{1}{2} \varepsilon^2 / T^3}$.](image)
$$S_{\parallel} = \frac{1}{2} \log \frac{3}{2}$$

FIG. 3: (Color online) Temperature dependence of (a) $C$ and (b) $S$ for $J_{\perp}/J = -1/5 < (J_{\perp}/J)_c$. Dashed lines are the cubic spline interpolation of the QMC data in (a). The entropy is computed from the cubic spline interpolation of the specific heat data with $T_{\text{max}}/J = 4$.

$\varepsilon_{\text{photon}}(k) \sim c|k|$, the density of states is computed as $D(\varepsilon) = \frac{2}{\pi} \left( \frac{a}{2\pi} \right)^3 \int d^3k \delta(\varepsilon - c|k|) = \frac{\pi^2 a^3}{3 \varepsilon^2}$, where $a$ is the cubic lattice constant, and the two polarizations and the four FCC primitive unit cells inside the cubic unit cell have been taken into account. Thus for $T \ll |J_{\parallel}^3/J^2|$, the energy density is derived from this density of states as $\varepsilon \simeq \frac{\pi^2 a^3}{3} T^4 + \text{const.}$, leading to the specific heat of “photons”, $C_{\text{photon}} = \frac{\pi^2 a^3}{3} T^3$. The speed of “light” $c$ is estimated by fitting the QMC data of the energy density at $T/J < 0.001$ as $c \simeq (1.3 \pm 0.2) a g h^{-1}$ for $J_{\perp}/J = -1/11$ where $g = |3J_{\parallel}^3/(2J^2)|$. This value lies between $c = (1.8 \pm 0.1) a g h^{-1}$ for $J_{\perp}/J = -1/9.7$ [25] and $c = (0.6 \pm 0.1) a g h^{-1}$ in the limit of $|J_{\perp}/J| \to 0$ [26].

The higher temperature crossover to the CSI regime at $T_{\text{CSI}} \sim 0.2 J$ is also observed in the case of $J_{\perp} < J_{\perp c}$, as marked with (red) dashed line in Fig. 4. To be explicit, it is demonstrated for $J_{\perp}/J = -1/5 < (J_{\perp}/J)_c$ in Fig. 3 (a). The entropy $S$ computed from the specific heat $C$ (Fig. 3 (b)) resembles the case of $J_{\perp}/J = -1/11 > (J_{\perp}/J)_c$, except that the plateau at the Pauling entropy in the CSI regime is masked by the spiky peak in $C$ due to a ferromagnetic transition at $T_{\text{c}}/J = 0.124(3)$.

Next, we clarify the spin correlations on the disordered side. Figures 4 present the energy-integrated polarized magnetic neutron-scattering cross-sections $\sigma_{\text{SF}}(q)$ in the spin flip channel, except the non-vanishing clear form factor, where

$$\sigma_T(q) = \sum_{\mu,\mu'} \langle b^*_\mu q e^{i(q \cdot R_+ + b_\mu/2)} \rangle, \nonumber$$

$$\sigma_{\text{NSF}}(q) = \sum_{\mu,\mu'} \langle b^*_\mu Z - (b^*_\mu \cdot \hat{q}) (Z \cdot \hat{q}) \rangle \nonumber$$

with $Z \equiv (1, -1, 0)/\sqrt{2}$, $\hat{b}_\mu = b_\mu/|b_\mu|$, and $\hat{q} \equiv q/q$. A broad scattering intensity along the [100] and [111] rods appears at high temperatures $T > T_{\text{CSI}}$, e.g., for $J_{\perp}/J = -1/11$ at $T/J = 4$ (Fig. 4 (a)), as experimentally observed in Pr$_2$Zr$_2$O$_7$ [11] and well above $T_c$ in Yb$_2$Ti$_2$O$_7$ [11, 12]. In the CSI regime, we clearly see remnants of the pinch-point singularity [23] at every reciprocal lattice vectors except at $q = (0, 0, 0)$, both for $J_{\perp}/J = -1/11 > (J_{\perp}/J)_c$ at $T/J = 0.1$ (Fig. 4 (b)) and for $J_{\perp}/J = -1/5 < (J_{\perp}/J)_c$ at $T/J = 0.2$ (Fig. 4 (c)), as observed in Dy$_2$Ti$_2$O$_7$ [24] and Ho$_2$Ti$_2$O$_7$ [25]. In fact, the pinch point cannot evolve into a real singularity because $T_{\text{c}}$, the 2-in, 2-out ice rule is dynamically violated by the spin-flip processes of Eq. [11, 12]. Unfortunately, the current algorithm did not provide a sufficiently good statistics for $\sigma_{\text{SF}}(q)$ at lower temperatures,
i.e., in the QSI regime. We note that the non-spin-flip channel $\sigma_{\text{NSF}}(q)$ always gives just a featureless constant.

![Graphs](image)

**FIG. 5**: (Color online) Determination of transition temperature. The energy density $\varepsilon$ for $J_{\perp}/J = -1/8$ (a) and for $J_{\perp}/J = -1/5$ (b). The spin stiffness $\rho_S$ (c) and the uniform transverse spin susceptibility $\chi_{\perp}$ scaled by $L^{-1-\eta}$ (d) for $J_{\perp}/J = -1/5$, with the critical exponent $\eta = 0.038$ for the 3D XY universality class [39].

Now we focus on the case of $J_{\perp}/J < J_{\text{c}}/J = -0.103$, where a phase transition occurs to the XY-FM [2]. The transition temperature increases with negatively increasing $J_{\perp}$, as shown with the solid curve in the phase diagram (Fig. 1). When $J_{\perp}$ is close to $J_{\text{c}}$, the energy density $\varepsilon \equiv \langle H \rangle / N_s$ exhibits a clear discontinuous jump, as shown for $J_{\perp}/J = -1/8$ in Fig. 3(a), which gives the first-order transition temperature $T/J = 0.020(2)$. On the other hand, with negatively increasing $J_{\perp}/J$, the jump becomes less clear within our linear system sizes $L = 4, 6, 8$, as shown for $J_{\perp}/J = -1/5$ in Fig. 3(b). In this case, we just observe a sharp specific heat peak gradually growing with $L$ (Fig. 3(c)) and crossings of the transverse spin susceptibility $\chi_{\perp}$ (Fig. 3(c)) and the spin stiffness $\rho_S$ (Fig. 3(d)) at different $L’s$, where

$$\chi_{\perp} \equiv \frac{1}{3N_s} \int_0^\beta d\tau \sum_r \langle s^z_r(\tau) s^z_r(0) \rangle,$$

$$\rho_S \equiv \frac{8\sqrt{2}}{3\beta J_{\perp}|N_s|} \langle W^2 \rangle,$$

with the total winding number of worldlines $W$ [40]. In fact, the finite-size scaling does not hold in this case within our linear system sizes $L = 4, 6, 8$, as is seen from slightly different crossing temperatures of $\rho_S$ and scaled $\chi_{\perp}$, as shown in Figs. 3(c) and (d). Hence, we cannot conclude whether the transition is of either the weak first order or the second order in the case of $J_{\perp}/J \leq -1/6$. Then, the transition temperature in Fig. 1 is estimated from the average of the two crossing temperatures, while the error is from the difference. Note that this transition temperature also reasonably coincides with the specific heat peak temperature.

In principle, no classical spin models can describe the physics down to zero temperatures. Even so-called dipo-
