

## Thermal canting of spin-bond order

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Magnetism arising from coupled spin and spatial degrees of freedom underlies the properties of a broad array of physical systems. We study here the interplay between correlations in spin and space for the quantum compass model in a finite external field, using quantum Monte Carlo methods. We find that finite temperatures cant the spin and space (bond) correlations, with increasing temperature, even reorienting spin correlations between orthogonal spatial directions. We develop a coupled mean-field theory to understand this effect in terms of the underlying quantum critical properties of crossed Ising chains in transverse fields and an effective field that weakens upon increasing temperature. Thermal canting offers an experimental signature of spin-bond anisotropy.

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### I. INTRODUCTION

The collective behavior of spins in highly correlated quantum magnets typically establishes order globally, over a large portion of the system by breaking a symmetry of the system. Conventional characterizations of magnetic order, such as ferromagnetism, therefore, rely on microscopic models with global symmetries. Recent theoretical studies have, in contrast, analyzed models that support quantum phases without long-range ordering, for which novel types of order, including intriguing topological quantum liquid phases, can nevertheless arise.<sup>1–3</sup> Quantum liquids can be difficult to identify (both theoretically and experimentally) because the lack of long-range order defies characterization by simple bulk order parameters such as magnetization. Models with quantum liquid ground states can harbor quasilocal symmetries which impose a symmetry on a small, nonextensive subset of spins.<sup>4</sup> Examples of such quasilocal operators include one-dimensional operator chains embedded in two-dimensional graphs. The qualitatively distinct characteristics of systems with reduced, nonextensive symmetry play an important role in identifying novel quantum liquid phases.

We study here one of the simplest of these models, the quantum compass model in a magnetic field  $h$ :

$$H = -\gamma \sum_{i,j} S_{i,j}^x S_{i+1,j}^x - \sum_{i,j} S_{i,j}^z S_{i,j+1}^z - h \sum_{i,j} S_{i,j}^z, \quad (1)$$

where  $S_{i,j}^\alpha = \sigma_{i,j}^\alpha/2$  are spin-1/2 operators at the  $(x, z)$  coordinates  $(i, j)$  on an  $L \times L$  square lattice. It is sufficient to consider  $\gamma > 0$  since with a  $\pi$  rotation of spins about the  $z$  axis on a single sublattice, our results apply to the  $\gamma < 0$  case as well. The coupling between bond and spin degrees of freedom separates this class of models from more conventional models of magnetism and motivates the intriguing question as to whether there are generic signatures of anisotropic spin-bond coupling.

The quantum compass and related models were first discussed in the context of Mott insulators<sup>5</sup> and have been studied as simple models of orbital order in the transition-metal oxide compounds with anisotropic coupling among orbitals

defining pseudospins.<sup>5–7</sup> For example, recent experiments<sup>8–10</sup> on a two-dimensional  $e_g$  orbital compound,  $\text{LaSrMnO}_4$ , observe intriguing thermal effects. Anomalies in thermal-expansion measurements<sup>8</sup> and in Raman scattering,<sup>10</sup> as well as structural changes measured by neutron and x-ray diffraction,<sup>9</sup> have all been interpreted as indicating a change in orbital occupation that is driven by temperature. The reorientation of orbital direction with increasing temperature implied by the experiments raises the question as to whether this results from thermal repopulation of a noninteracting orbital system, or from competition between orbital-orbital interactions and thermal effects. Detailed understanding of this phenomenon requires quantitative analysis of the competing roles of superexchange, phonon, crystal-field, and Jahn-Teller effects. However, the orbital-only degrees of freedom in these systems have been shown to be approximately described by a quantum compass model with anisotropic-pseudospin interaction terms given by superexchange contributions and an effective magnetic field determined by the crystal-field splitting.<sup>11</sup> This enables one to address in general the intriguing question of whether and how orbital-orbital interactions can influence the thermal redistribution of orbitals. We answer this question in the affirmative here, showing that a type of thermal reorientation, “thermal canting,” of nearly degenerate orbitals can indeed arise from a large anisotropic orbital-orbital interaction of the type exemplified in Eq. (1). We also show that this thermal canting can provide an observable signature of the anisotropic interactions.

Equation (1) has been studied in other contexts as well. A duality mapping<sup>12</sup> relates Eq. (1) to models of  $p$ -wave superconductors.<sup>13</sup> Recent proposals put forward to realize Eq. (1) in optical lattices, using asymmetric tunneling<sup>14</sup> or polar molecules,<sup>15</sup> and in Josephson-junction arrays<sup>16</sup> (finite-sized versions of which have recently been realized<sup>17</sup>) have been motivated by the recognition that the  $h=0$  limit is characterized by sets of low-energy twofold-degenerate states that may provide protected subspaces for encoding quantum information.<sup>16,18</sup> In addition, related<sup>4</sup> anisotropic-spin models [Eq. (4) of Ref. 3] show topological order which allows a robust form of quantum information processing.<sup>2,19</sup> A generic

experimental signature of the spin-bond asymmetric models would be a valuable tool in the search for realizations of models with anisotropy and related quasilocal symmetries.

In this paper we study the finite-temperature properties of the quantum compass model for  $h > 0$ , using both quantum Monte Carlo (QMC) and mean-field theory to identify generic signatures of the set of one-dimensional symmetries underlying Eq. (1). We find a remarkable thermal canting effect whereby finite temperatures *enhance* spin-bond correlations  $r_x$ , where  $r_\alpha = L^{-2} \sum_{i,j} \langle S_{i,j}^\alpha S_{i+1,j}^\alpha \rangle$  and  $\alpha = x, z$ . The enhancement resembles an order-by-disorder process.<sup>6,20,21</sup> The reorientation of net spin-bond correlations (from  $r_z$  to  $r_x$ ) defines a phase akin to a liquid-crystal smectic  $C$  phase where molecular moments order along a vector (“director”) tilted with respect to translationally ordered columns. We develop a mean-field theory using intersecting Ising models and their crossover phase diagrams<sup>22</sup> to show that the finite-temperature reduction in the  $z$  magnetization leads to thermal canting at the mean-field level and should therefore play a role in similar models. For  $\gamma > 1$  we find that finite temperatures can completely reorient spin-bond correlations to point in orthogonal directions in spin and real space.

## II. SYMMETRIES IN ZERO-FIELD LIMIT

We begin our study with an analysis of the unique symmetries of the quantum compass model in several well-defined limits, for  $h=0$ . Under high strain,  $\gamma \gg 1$  ( $\gamma \ll 1$ ), the system forms a series of  $L$  weakly coupled spin chains lying along the  $x$  ( $z$ ) direction in real space. At zero temperature the chains order to point along the  $x$  ( $z$ ) direction in spin space. Note that with  $h=0$ , Eq. (1) commutes with the quasilocal symmetry operators  $P_i = \prod_j 2S_{i,j}^x$  and  $Q_j = \prod_i 2S_{i,j}^z$ , which act on ordered chains to generate spin flips along  $z$  and  $x$  chains, respectively ( $P$  is not an exact symmetry for  $h \neq 0$ ). Gapped excitations along chains are then finite length domains of flipped spins, as in one-dimensional Ising models. The operators  $Q$  ( $P$ ) can flip all spins along the ordered chains in the ground state, resulting in yet another ground state. This results in a twofold degeneracy of each chain and therefore a massive ground-state degeneracy of the two-dimensional system of at least  $2^L$ .

As  $\gamma \rightarrow 1$ , the interchain coupling allows a set of interchain excitations. Reference 23 showed that high-order interchain fluctuations of the magnetization cost zero energy (for  $L \rightarrow \infty$ ), preserving a large [ $\mathcal{O}(2^L)$ ] ground-state degeneracy for *all*  $\gamma$ . The  $\gamma=1$ ,  $h=0$  limit yields discrete global rotational symmetries (see Ref. 12 for a discussion). A nematic-like order parameter has been defined,<sup>12,24</sup>  $r \equiv r_z - r_x$ , to characterize smecticlike bond ordering along the  $x$  and  $z$  directions in spin and real space, analogous to the directional ordering of molecular moments along columns in liquid crystals.  $r=0$  implies a disordered phase with no preferred direction. Finite-temperature studies<sup>7,25</sup> suggest a phase transition from an ordered,  $|r| > 0$ , phase to a disordered phase with  $T_c \approx 0.055$  (Ref. 25) in units of the interaction strength in Eq. (1). Example interaction strengths in corresponding experimental systems are  $\sim 1000K$  for orbitals<sup>8</sup> or  $\sim 1K$  for Josephson junctions.<sup>17</sup>

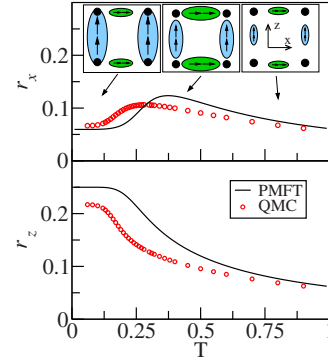


FIG. 1. (Color online) The top (bottom) panel plots the spin-bond correlations versus temperature along the  $x$  ( $z$ ) direction computed via quantum Monte Carlo (open circles) and partial mean-field theory (solid line) for the quantum compass model on a square lattice in a magnetic field. The peak in  $r_x$  indicates a reorientation of bond correlations with increasing temperature depicted schematically in the inset, where  $\uparrow$  and  $\rightarrow$  correspond to spins aligned along the  $z$  and  $x$  directions, respectively. The data are plotted here for  $\gamma=1$  and  $h=0.08$ .

## III. QUANTUM MONTE CARLO SIMULATIONS AT FINITE FIELDS

A weak finite magnetic field picks out a single ground state with a gap to excitations set by  $h$ , simplifying QMC simulations. We employ the stochastic series expansion using directed loops.<sup>26–28</sup> A modification of the code in the ALPS (Ref. 29) package allows a treatment of  $S_{i,j}^x S_{i+1,j}^x$  bond terms in the updating scheme. All QMC results presented here are converged to the thermodynamic limit in studies of several system sizes up to  $L=30$  for periodic boundary conditions.

Our results show that competition between the interaction and the magnetic field sets up an interesting interplay in the energetics along both spatial directions. The inset of Fig. 1 (top panel) shows a schematic of the generic behavior observed in the QMC calculations for  $\gamma \leq 1$  as we increase temperature. At low temperatures the weak magnetic field aligns the net magnetization along the  $z$  direction. The magnetic field and  $z$ -interaction term favor spin chains that are bond correlated along the  $z$  direction, i.e., large  $r_z$ . Using mean-field theory we will show that interchain interactions contribute to an effective magnetic field that suppress excitations along the  $x$  direction. As we increase temperature above the gap set by  $h$ , the applied magnetic field becomes less relevant while the mean-field effective magnetic field is reduced with increasing temperature. Excitations along  $x$  are then allowed to recover. Consequently, bond correlations build up along the  $x$  direction, yielding  $r_x \rightarrow r_z$  as can be seen in the QMC and mean-field results in Fig. 1. At high temperatures both types of correlations are trivially suppressed as the system becomes thermally disordered.

## IV. PARTIAL MEAN-FIELD THEORY

At first sight, the buildup of bond correlations along the  $x$  direction is puzzling since one would expect a suppression of bond correlations with increasing temperature. To understand

this buildup of correlations along the  $x$  direction, we construct a coupled partial mean-field theory (PMFT) in which spin interactions along a given bond direction are treated at the mean-field level, while the interactions along the orthogonal direction are treated exactly. In our two-step protocol we first choose the  $z$  direction (indicated by  $\langle \cdots \rangle_z$ ) and make a mean-field decoupling,  $S_{i,j}^z S_{i,j+1}^z \rightarrow S_{i,j}^z \langle S_{i,j+1}^z \rangle_z$  to generate the effective field  $B_x^{\text{eff}} = \langle S_{i,j+1}^z \rangle_z + \langle S_{i,j-1}^z \rangle_z + h$  for spins along  $x$ . We then repeat the process by exchanging  $x$  and  $z$ , resulting in a set of two coupled mean-field equations:

$$H_{\text{MF}}^z = - \sum_{i,j} S_{i,j}^z S_{i,j+1}^z - h \sum_{i,j} S_{i,j}^z - B_z^{\text{eff}} \sum_{i,j} S_{i,j}^z, \quad (2)$$

$$H_{\text{MF}}^x = - \gamma \sum_{i,j} S_{i,j}^x S_{i+1,j}^x - B_x^{\text{eff}} \sum_{i,j} S_{i,j}^x. \quad (3)$$

While in general one may take  $B_z^{\text{eff}} = \gamma (\langle S_{i+1,j}^x \rangle_x + \langle S_{i-1,j}^x \rangle_x)$  and make a self-consistent solution to Eqs. (3) and (2), in what follows we keep the mean-field solutions analytic by setting  $B_z^{\text{eff}} = 0$ . This approximation does not qualitatively alter our results for  $\gamma$  near unity. We also assume a uniform net magnetization  $\langle S^z \rangle_z = \langle S_{i,j}^z \rangle_z$ . The magnetization  $\langle S^z \rangle_z$  obtained from solving Eq. (2) can then be substituted directly into Eq. (3). This mean-field approach is motivated by the work in Ref. 30 but differs by: (i) having a set of two coupled mean-field equations, (ii) addressing a different parameter regime of finite temperatures and magnetic fields, and (iii) obtaining qualitatively accurate results without a self-consistency loop. The mean-field approach is expected to be qualitatively accurate because the finite magnetic field suppresses fluctuations along the  $z$  direction.

We can now solve the PMFT equations analytically. Solving Eq. (2) using the well-known solutions of the classical Ising model,<sup>31</sup> we obtain  $\langle S^z \rangle_z = \tanh(C)/(2G)$ , where  $G \equiv \{1 + [\exp(-4K) - 1] \text{sech}^2(C)\}^{1/2}$ ,  $K \equiv 1/4T$ , and  $C \equiv h/2T$ , and can estimate  $r_z$  as  $\langle S_{i,j}^z S_{i,j+1}^z \rangle_z = (1/4) \partial_K \ln\{\exp(K) \cosh(C)(1+G)\}$ . This first estimate for the magnetization is then used as input to Eq. (3), which is the Ising chain in a transverse field and can be solved exactly via the Jordan-Wigner transformation.<sup>32</sup> We obtain the total energy:  $\langle H_{\text{MF}}^x \rangle_x = \sum_k \epsilon_k (n_k - 1/2)$ , where  $\epsilon_k = [(B_x^{\text{eff}})^2 - \gamma B_x^{\text{eff}} \cos(k) + \gamma^2/4]^{1/2}$  and  $n_k$  is the Fermi distribution function. Our PMFT estimate for the magnetization of the two-dimensional system becomes  $\langle S^z \rangle_x = \sum_k [2B_x^{\text{eff}} - \gamma \cos(k)] (4\epsilon_k)^{-1} \tanh(\epsilon_k/2T)$ . We can also obtain an estimate for  $r_x$ :  $\langle S_{i,j}^x S_{i+1,j}^x \rangle_x = -(1/\gamma L) \langle H_{\text{MF}}^x \rangle_x - (1/\gamma) B_x^{\text{eff}} \langle S^z \rangle_x$ . By setting  $B_z^{\text{eff}} = 0$  we ignore the influence of the magnetization along  $x$  on  $r_z$ . The result is an overestimate for  $r_z$  at low temperatures. Figure 1 and the inset of Fig. 2 show a comparison between the PMFT and QMC results for the temperature dependence of  $r_x$ ,  $r_z$ , and  $\langle S^z \rangle_z$ . It is evident that the PMFT captures the qualitative features of the QMC results.

The success of the PMFT analysis implies that finite-temperature excitations of the two-dimensional quantum compass model can be approximated by excitations along crossed (coupled) one-dimensional Ising models. The peculiar enhancement of  $r_x$  with temperature can now be under-

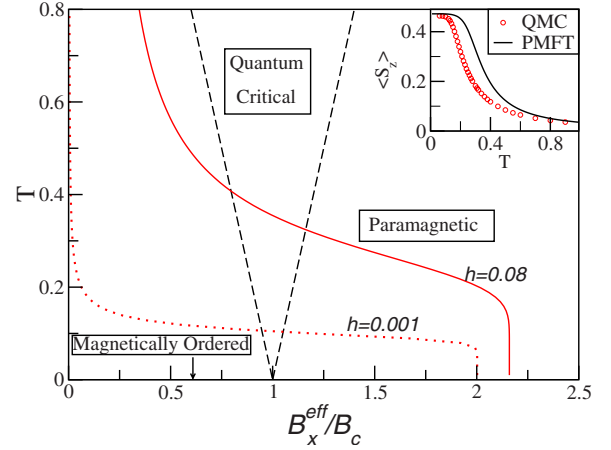


FIG. 2. (Color online) Crossover diagram for a one-dimensional transverse field Ising model. The  $x$  axis indicates the effective magnetic field which can be lowered to tune from the paramagnetic side (weak nearest-neighbor correlations), through a quantum critical point,  $B_c \equiv \gamma/2$ , to a regime with relatively strong nearest-neighbor correlations. The dashed lines schematically define the quantum critical regime. For the PMFT defined by Eq. (3),  $B_x^{\text{eff}}$  is temperature dependent and plotted for  $h=0.08$  (solid line) and  $h=0.001$  (dotted line) with  $\gamma=1$ . Inset: The same as Fig. 1 but for magnetization density. The PMFT curve plots  $\langle S^z \rangle_x$ .

stood from the crossover phase diagram of the transverse field Ising model [Eq. (3)]. Figure 2 plots a schematic of this crossover phase diagram. In our mean-field analysis,  $B_x^{\text{eff}}$  carries a temperature dependence because the  $z$  spins demagnetize with increasing temperature. In Fig. 2 we superpose  $B_x^{\text{eff}}(T)$  from a solution of the mean-field equations, which shows that an increase in temperature lowers the effective magnetic field seen by spins along the  $x$  direction. The bond correlations along the  $x$  direction are therefore enhanced because of a consequent drastic reduction in  $\langle S^z \rangle_z$ . We conclude that the nonlinear temperature dependence in the effective magnetic field experienced by the spin chains lying along the  $x$  direction is therefore responsible for the thermal canting of spin-bond correlations observed in QMC.

## V. MAGNETIC FIELD DEPENDENCE OF THERMAL CANTING

We now examine thermal canting in finite fields  $h$  in greater detail. Figure 3 indicates that the net spin-bond correlations completely reorient with increasing temperature. To understand this behavior, first note that the  $h \sim \gamma \gg 1$  limit of Eq. (1) can be thought of as a set of nearly independent transverse field Ising models with a quantum critical point ( $B_c = \gamma/2$ ). Lowering  $h$  takes the system from the paramagnetic phase through the quantum critical point (at  $T=0$ ) to an ordered chain phase along the  $x$  direction. For a specific choice of  $\gamma > 1$  we can therefore completely reorient bond correlations by tuning  $h$ . Thermal canting also tunes  $B_x^{\text{eff}}$  but with a directionally independent parameter, temperature. At low temperatures the finite magnetic field favors bond correlations along the  $z$  direction,  $r > 0$ , while at higher temperatures thermal canting of spin-bond correlations now favors a

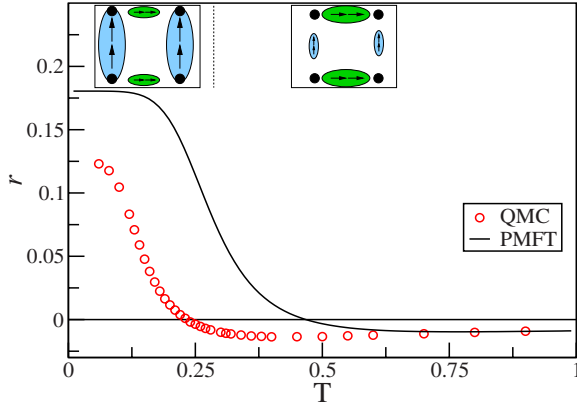


FIG. 3. (Color online) Net spin-bond correlations as a function of temperature for  $h=0.1$  and  $\gamma=1/0.85$ . Thermal canting reorients the spin-bond correlations to lie predominantly along the  $x$  direction in both spin and space for  $T>0.25$ .

net orientation along the  $x$  direction,  $r<0$ . This implies a canting transition at some temperature  $T_{\text{cant}}$ , which is illustrated in Fig. 3 for  $\gamma=1/0.85$ , where  $T_{\text{cant}}\sim 0.25$ . These results predict a striking effect, namely, a complete reorientation of net bond correlations in both spin and real space with increasing temperature. This thermal reorientation arises from a distinct mechanism, thermal canting, that can again be understood in the PMFT calculations as being due to a weakening of the effective magnetic field with increasing temperature: thus it is a consequence of the two-dimensional vector nature of the spin-bond correlations in Eq. (1).

Our two-dimensional spin-bond analysis at small finite  $h$  also provides a microscopic rationale for the  $|r|\rightarrow 0$  transition observed in the  $h=0$  QMC simulations<sup>7,25</sup> in terms of an immediate thermal canting of spin-bond correlations. Our QMC and PMFT results both show that the  $r_x$  peak in Fig. 1 moves to lower temperatures with decreasing magnetic field. The mean-field analysis indicates that this enhancement of  $r_x$  at low temperatures derives from the rapid depolarization of  $\langle S^z \rangle_z$  with increasing temperature when  $h$  is small. Mean-field results for extremely small  $h$  values and  $\gamma=1$  are shown in Fig. 4. We see that the increase in  $r_x$  with temperature at low  $h$  concomitantly tends to decrease  $r$  (see inset). Extrapolating the PMFT results to  $h\rightarrow 0$  yields, for  $\gamma=1$ ,  $r_x=(1/4)\tanh(1/4T)+\mathcal{O}(h^2)$  and  $r=0+\mathcal{O}(h^2)$ . Thus  $r$  disappears altogether for  $h\rightarrow 0$ . This contrasts with the  $h=0$  QMC results of Refs. 7 and 25 where a low-temperature plateau in  $|r|$  was followed by a rapid decrease at larger  $T$  and assigned to a disordering transition. The difference may be due to a lack of fluctuations in the PMFT which could play a large role in the  $h\rightarrow 0$  limit.

## VI. SUMMARY

We have found a distinct thermal canting effect in the  $x$ - $z$  plane of the two-dimensional quantum compass model. Ex-

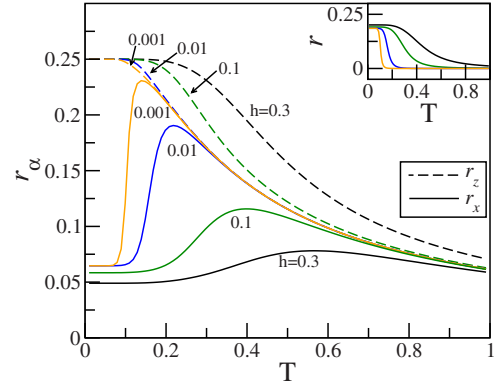


FIG. 4. (Color online) PMFT results for the bond correlations along the  $x$  (solid line) and  $z$  (dashed line) directions versus temperature for  $\gamma=1$  and several values of  $h$ . The inset shows the behavior of the scalar difference  $r=r_z-r_x$ .

citations governed by the rather unique set of chain symmetries implicit in the model provide a microscopic mechanism responsible for this effect. Thermal excitations above the paramagnetic gap can be approximated by those of intersecting Ising chains that are generated by quasilocal operators stemming from the exact symmetry  $Q$  and the approximate symmetry  $P$ . Such excitations lead to an enhancement of spin-bond correlations along the  $x$  direction at higher temperatures. This study shows that temperature can be used as a parameter to experimentally tune the balance between the excitations generated by these operators and to thereby reveal the underlying chain symmetries and anisotropic interactions. Thermal canting arises in our mean-field theory (as well as QMC). Our partial mean-field argument applies to a number of other lattice geometries, in addition to the rectangular lattice of Eq. (1).

In summary, our combined QMC and partial mean-field analysis shows that anisotropic interactions can lead to a thermal redistribution of spin-bond correlations. The results suggest that observation of thermal canting can provide an indicator of spin-bond anisotropy, with significant implications for the transition-metal oxide compounds that are described by pseudospin models of orbital degrees of freedom. Our demonstration that thermal canting can be driven by anisotropic interactions suggests that the recent observations of a thermal reorientation of nearly degenerate orbitals in  $\text{LaSrMnO}_4$  (Refs. 8–10) warrant further analysis in terms of anisotropic-pseudospin models of orbitals.

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