Anisotropy-induced ordering in the quantum $J_1 - J_2$ antiferromagnet

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We study the effect of spin anisotropies on a frustrated quantum antiferromagnet using the $J_1$-$J_2^{XXZ}$ model on the square lattice. The $T = 0$ and finite-$T$ phase diagrams of this model are obtained utilizing spin-wave theory, exact diagonalization, and quantum Monte Carlo. We find that anisotropic frustration tends to stabilize XY- and Ising-like ordered phases, while the disordered spin-liquid phase is restricted to a small region of the phase diagram. The ordered phases are separated by first-order transitions and exhibit a non-trivial reentrance phenomenon.

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Frustrated quantum magnets attract significant interest because of the spin-liquid and novel ordered phases they may exhibit.\[1\] The macroscopic degeneracy of the ground state in such magnets makes them very sensitive to additional interactions that may lead to various unconventional ordered states. Perhaps the most extensively investigated, yet highly controversial model is the $S = 1/2$ $J_1$-$J_2$ Heisenberg model on the square lattice with competing nearest-neighbor, $J_1$, and next-nearest-neighbor, $J_2$, interactions.\[2\] The recent synthesis of compounds that can be closely described by the two-dimensional (2D) $J_1$-$J_2$ Hamiltonian\[3\] has also fueled interest in the properties of this basic model of magnetic frustration. In that respect, the presence of spin anisotropies in real systems raises the question of how robust the behavior of the isotropic $J_1$-$J_2$ model is against such perturbations.

In the present paper we study the effect of spin anisotropies on the properties of frustrated quantum antiferromagnets using a generalization of the $J_1$-$J_2$ model, in which the frustrating next-nearest-neighbor interaction is anisotropic. Such a $J_1$-$J_2^{XXZ}$ model is given by:

$$\hat{H} = J_1 \sum_{\text{n.n.}} S_i \cdot S_j + \frac{1}{2} \sum_{\text{n.n.n.}} \left( J_2^{XXZ} S_i^z S_k^z + J_2^{XY} S_i^x S_k^x \right),$$

where $S_i$ is the spin operator, the sites $i, j, k$ are on the square lattice, and the summation runs over all nearest-neighbor (n.n.) or next-nearest-neighbor (n.n.n.) sites. We hereafter use the dimensionless ratios $\alpha_z = J_2^{XXZ} / J_1$ and $\alpha_\perp = J_2^{XY} / J_1$.

For the extensively studied isotropic case $\alpha_\perp = \alpha_z$, analytical and numerical works have suggested the existence of a $T = 0$, non-magnetic gapped phase for $0.4 \lesssim J_2 / J_1 \lesssim 0.6$, separating the Néel state from the collinear state.\[4\] The absence of long-range order at any finite temperature in 2D systems for the isotropic $J_1$-$J_2$ case is dictated by the Mermin-Wagner theorem. In the collinear phase, however, the 2D $J_1$-$J_2$ model has been predicted to exhibit an Ising-like Chandra-Coleman-Larkin (CCL) transition\[5\] at $T > 0$ with spontaneous breaking of the discrete lattice rotational symmetry. This unusual scenario has recently received support from numerical and analytical studies.\[6\]

In this Letter we show that even a small anisotropy in the frustrating (n.n.n.) coupling leads to strong deviations from the behavior of the isotropic $J_1$-$J_2$ model. The main effect of anisotropy is to energetically favor the sector of spin space in which the frustration is minimal. This tendency of spins to avoid frustration leads to somewhat counter-intuitive results, as, e.g., Ising-like anisotropy in $J_2$ favoring the $XY$ phase and vice versa. In the case of the unfrustrated 2D Heisenberg antiferromagnet, an arbitrarily small anisotropy is found to stabilize finite-temperature ordered phases.\[7\] In this work we show that the anisotropically frustrated $J_1$-$J_2^{XXZ}$ model is generally more ordered than its isotropic counterpart.

The $T = 0$ phase diagram of classical ($S = \infty$) $J_1$-$J_2^{XXZ}$ model is shown in Fig. 1.\[8\] We find four phases characterized by different order parameters, $m_\theta^\theta = \langle \sum_i e^{i\theta} S_i^\theta \rangle$, whereafter indicated as: (i) $XY$-Néel phase with $m_\theta^\theta(\pi,\pi)$, (ii) $Z$-collinear phase with $m_\theta^\theta(0,0)$ or $m_\theta(0,0)$, (iii) $XY$-collinear phase with $m_\theta^\theta(\pi,0)$ or $m_\theta(0,\pi)$, and (iv) $Z$-Néel phase with $m_\theta^\theta(\pi,\pi)$. The diagonal line $\alpha_\perp = \alpha_z$
corresponds to the isotropic $J_1$-$J_2$ model. All four phases are separated by first-order transition lines. Taking into account quantum corrections within linear spin-wave theory results in the opening of a spin-liquid region where all the order parameters vanish. Along the main diagonal this reproduces the well-known results for the isotropic case.\[1\] Our analysis suggests that in the vicinity of the $\alpha_1 = \alpha_2$ line the $T = 0$ transitions from the ordered phases to the spin-liquid phase is of second order. However, we also find that the collinear phases are separated from the spin-liquid phase mostly by the first-order transition lines, as shown in Fig. 1. This means that the XY- and Z-collinear orders are particularly robust, so that even quantum-renormalized order parameters vanish with a finite jump when crossing such boundaries.

Given the anisotropic nature of the ordered phases found in the $T = 0$ phase diagram, one can anticipate qualitatively the finite temperature behavior. In the Ising regions magnetic order is expected to survive at $T > 0$ up to a second-order transition point. Topological order is instead expected at $T > 0$ in the XY regions up to a Berezinski-Kosterlitz-Thouless (BKT) transition.

The study of the quantum $J_1$-$J_2$ model at finite temperature by quantum Monte Carlo (QMC) techniques is generally precluded by the sign problem due to the transverse frustrating coupling $\alpha_1$. However, by removing that term completely we obtain the sign-problem-free $J_1$-$J_2^Z$ model, where the frustration is Ising-like, corresponding to the $\alpha_1 = 0$ line of the phase diagram in Fig. 1. A comprehensive study of this limiting case is, therefore, very important since its thermodynamics can shed light on the main features of a large region of the phase diagram ($\alpha_1 < \alpha_2$). Moreover, in this limit of $J_2^Z$-only frustration, the Hamiltonian of Eq. (1) is also relevant for the broad class of strongly correlated bosonic systems. In fact, this $J_1$-$J_2^Z$ model can be mapped exactly onto the frustrated hard-core Bose-Hubbard model.\[8\]

We have thus investigated the thermodynamics of the $S = 1/2$ $J_1$-$J_2^Z$ model by means of the Stochastic Series Expansion QMC based on the directed-loop algorithm.\[10\] A large interval of frustration values, $0 < \alpha_2 < 1.7$, has been scanned using different lattices $L \times L$ up to $L = 96$ to perform an extensive finite-size scaling analysis. The resulting phase diagram of the $J_1$-$J_2^Z$ model is shown in Fig. 2. As main features we observe the occurrence of the topologically ordered phase (quasi-long-range XY-Néel) for small $\alpha_2$ and the Z-collinear order for large $\alpha_2$. These two phases are separated by a first-order transition line. Phases with finite-temperature topological order and Z-collinear order in the spin language correspond, in the bosonic language, to quasi-long-range superfluidity and striped solid order, respectively.

The study of the first-order transition line has been performed by making use of a quantum parallel tempering technique\[11\] to overcome critical slowing down at first-order transition points.\[12\] In the collinear phase we have also used thermal parallel tempering\[13\] to overcome the well-known loss of efficiency of standard cluster algorithms in Ising-like frustrated antiferromagnets.\[14\]

A remarkable feature of the phase diagram is that even an infinitesimal frustration, $\alpha_2 \ll 1$, induces a finite-temperature BKT transition. The critical temperature $T_{\text{BKT}}$ has been extracted through scaling analysis of the helicity modulus $\Theta$ and of the transverse structure factor $S^zz(y)(\pi, \pi)$.\[8\] $T_{\text{BKT}}$ depends logarithmically on the frustration as $T_{\text{BKT}} \approx 4\pi \rho_s/(C + |\ln \alpha_2|)$, where $\rho_s$ is the spin stiffness of the unfrustrated Heisenberg antiferromagnet and $C$ is a constant. A fit of our data to the above law for $\alpha_2 \lesssim 0.1$ gives $\rho_s = 0.175(2)$ in excellent agreement with the best available estimates,\[16\] and $C = 5.4(1)$. For larger frustration, the BKT transition temperature reaches a maximum which falls very close to the critical temperature of the $S = 1/2$ XY model, $T_{\text{BKT}}/J = 0.3427(2)$,\[15\] indicating that the $J_1$-$J_2^Z$ model with frustration $\alpha_2 \approx 0.65$ is an almost ideal realization of the quantum XY model. At zero temperature, the XY-Néel and the Z-collinear phases are separated by a first-order transition at the critical value $\alpha_2^{(c)} = 1.252(5)$. We notice that quantum fluctuations cause a 25% shift of this critical value with respect to the classical value $\alpha_2^{(c)} = 1$, thus promoting the Néel phase against the collinear phase. When increasing the temperature the first order transition line shows a reentrant behavior, as seen in the inset of Fig. 2. For $1.21 \lesssim \alpha_2 \lesssim 1.25$, when temperature is increased, the topological order becomes unstable to the onset of the collinear magnetic order, which carries a higher entropy content. One can argue that the bending of the first-order transition line reflects the reduced role of quantum fluc-

![FIG. 2: Phase diagram of the $S = 1/2$ $J_1$-$J_2^Z$ model. Magnetic (and corresponding bosonic) phases are indicated. The horizontal dashed line marks the BKT critical temperature of the $S = 1/2$ XY model. The dotted line is a logarithmic fit for small $\alpha_2$ (see text). Inset: zoom on the tricritical point.](image-url)
we should expect a 2D Ising transition at a critical lattice. In this limit, according to Onsager's solution, the first-order transition line meets the BKT line at the point $(\alpha_z, T/J_1) = (1.210(5), 0.28(1))$, above which it maintains its first-order nature up to a temperature $T/J_1 \approx 0.36$. This is shown in Fig. 3, where the phase coexistence manifests itself in the double-peak structure of the distribution for the collinear magnetization $m_{\text{coll}} = m_{\alpha=(\pi,0)} + m_{\alpha=(0,\pi)}$ around the transition point. We have checked that the double-peak feature persists when increasing the lattice size up to $L = 48$. For $T \gtrsim 0.36 J_1$, determining the order of the transition to collinear order becomes more complicated. In fact the distribution of the order parameter $m_{\text{coll}}$ loses the strong two-peak feature, suggesting the transition to be weakly first-order. For $\alpha_z \gg 1$ the model reduces to two disconnected Ising models, living on the two sublattices of the original square lattice. In this limit, according to Onsager’s solution, we should expect a 2D Ising transition at a critical temperature $T_{\text{coll}} = 2 J_2^* \beta^2 / \ln(1 + \sqrt{2}) = 0.567 J_2^*$. The crossover from a weakly first-order to a second-order 2D Ising transition might happen with a continuous change of the critical exponents for increasing $\alpha_z$, as argued to occur in the frustrated 2D Ising antiferromagnet.\[15\]

We have determined the transition points $T_{\text{coll}}$ for $T \gtrsim 0.36$ by the crossing of Binder’s fourth cumulant $U_4^{(m)}(L) = 1 - (m_{\text{coll}}^4)/3(m_{\text{coll}}^2)^2$. In Fig. 3 we observe that, for $\alpha_z = 1.3$ as well as for the other cases studied, the values of $U_4^{(m)}(L)$ for different lattice sizes do not cross at the 2D Ising critical value $U_4 = 0.6107$\[16\], which suggests the universality class of the transition to be distinct from 2D Ising.

An interesting question is whether the CCL transition\[17\] of the isotropic $J_1-J_2$ antiferromagnet, with spontaneous breaking of the discrete lattice rotational symmetry, survives in the collinear phase of the anisotropic $J_1-J_2^X$ model. The order parameter for the transition reads:\[18\] $\sigma = \langle(S_{i,j}^z - S_{i+1,j+1}^z)(S_{i,j+1}^z - S_{i+1,j}^z)\rangle/4$. We have studied the scaling of Binder’s fourth cumulant related to the order parameter $\sigma$, $U_4^{(\sigma)} = 1 - \langle(\sigma_{\text{coll}}^4)/3(\sigma_{\text{coll}}^2)^2\rangle$. As shown by the crossing of the Binder’s cumulant in Fig. 3 the CCL transition seems to occur at the same (or slightly higher) temperature with respect to the magnetic transition. Let us now return to the general $J_1-J_2^X$ model to investigate how the ground state properties evolve in presence of the transverse component of frustration, $\alpha_\perp \leq \alpha_z$. Making use of exact diagonalization on lattices of $N = 16, 20,$ and 32 spins we have determined the dependence of the order parameters, $m_{\alpha=(\pi,\pi)}$ and $m_{\text{coll}}$, on $\alpha_z$ at fixed $\alpha_\perp$. Using a dense mesh of points, we obtain a qualitative finite-size estimate of the phase boundaries from the vanishing of the second derivatives $\partial^2 m_{\alpha}^4(\alpha_z)/\partial \alpha_z^2$, marking an inflection point in the order parameters. In this way we have obtained the $T = 0$ phase diagram shown in Fig. 4. For sufficiently low transverse frustration ($\alpha_\perp \leq 0.4$) both order parameters display an inflection at the same point, suggesting that the transverse frustration does not introduce any intermediate phase. The sharpness of the peaks in the derivatives of the order parameters suggests that the transition maintains the first-order nature found in the $J_1-J_2^X$ model. This first-order transition line runs roughly parallel to the classical line, being shifted to higher $\alpha_z$ by about 20% due to quantum fluctuations. As the transverse frustration is increased, the inflection points of the order parameters separate from each other, leaving space for an intermediate region with no magnetic order, continuously connected with the spin-liquid phase in the isotropic limit. Nonetheless, we observe the intermediate non-magnetic phase to shrink dramatically as the cluster size is increased. The small size of the clusters does not allow us to conclude on the real extension of the intermediate non-magnetic phase and on the nature of the phase transition line(s) for $\alpha_\perp \gtrsim 0.4$. Further investigations of this issue are currently in progress. A suggestive scenario is that the transition line between the collinear and the non-magnetic phase remains of first order all the way to
the isotropic limit, in agreement with the predictions of previous works [22].

We finally propose a global phase diagram for the $J_1$-$J_2^{XXZ}$ model, shown in Fig. 5 by merging the $T = 0$ data with the finite-temperature data. Here the finite-temperature $Z$-collinear and topological phases are separated by a first-order transition surface, and upper-bounded by a BKT transition surface and a weakly-first-order/second-order surface, respectively. For $\alpha_z = \alpha_\perp$ the above critical surfaces vanish. Moreover, in the high-frustration region of $\alpha_\perp \approx \alpha_z \approx 0.5$ we adopt the picture of an extended spin-liquid region where all transition temperatures are also vanishing. For $\alpha_\perp \to \alpha_z$ outside the spin-liquid region, the critical temperatures $T_c$ vanish with a very steep slope, following a logarithmic dependence on the anisotropy $\Delta = |\alpha_z - \alpha_\perp| \ll 1$, $T_c \sim |\ln(\Delta)|^{-1}$. This dependence is obtained through the mean-field condition $k_B T_c \sim J_1 \Delta [\xi(J_1 - J_2)]^2$, assuming an exponential divergence of the correlation length of the isotropic $J_1$-$J_2$ model, $\xi(J_1 - J_2) \approx T^{-1} \exp(2\pi |J_1 - J_2|/T)$ (as found, e.g., by modified spin-wave theory [22]). At the spin-liquid region boundaries, instead, the spin stiffness $\rho_s$ vanishes; to account for that, along the boundary between the spin-liquid and the Neél region we introduce a $\Delta$-dependent stiffness in the mean-field equation, $\rho_s \sim (\Delta - \Delta_\perp)^3$, where $\Delta = \Delta_\perp$, marks the boundaries of the spin-liquid region. In drawing the phase diagram we have taken $\beta = 1$, as suggested by the data in Ref. [22]. In this way we get a much smoother vanishing of $T_c$, $T_c \sim (\Delta - \Delta_\perp)/|\ln(\Delta - \Delta_\perp)|$. For the isotropic case $\alpha_z = \alpha_\perp$ and in the collinear phase, the only finite-$T$ transition line is the CCL one, recently estimated for $S = 1/2$ in Ref. [3]. Having observed the CCL transition to occur very close to the magnetic transition for $\alpha_\perp = 0$, we expect the CCL transition surface to quickly merge with the steep magnetic transition surface as $\alpha_\perp \lesssim \alpha_z$. Thus, we suggest that the CCL transition exists as a distinct thermodynamic feature only in a very close vicinity of the isotropic limit.

In summary, we have shown that the Heisenberg antiferromagnet on the square lattice with anisotropic nearest-neighbor frustration shows a very rich phase diagram characterized by several finite-temperature transitions, which we studied using QMC, exact diagonalization, and spin-wave theory. The competition between ordered phases leads to non-trivial reentrance phenomena, which is also common to related bosonic systems. Since anisotropic spin-spin interactions are a general feature of real magnets, the ordering effect due to anisotropic frustration is a realistic mechanism to explain magnetic transitions in recently synthesized frustrated quantum antiferromagnets.

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[1] G. Misguich and C. Lhuillier, in Frustrated spin systems, edited by H. T. Diep, World-Scientific, Singapore (2003).
[2] H. Kawamura, J. Phys.: Condens. Matter 10, 4707 (1998).
[3] P. Chandra et al., Phys. Rev. Lett. 64, 88 (1990).
[4] P. Chandra and B. Doucet, Phys. Rev. B 38, 9335 (1988); H. J. Schulz et al., J. Phys. I (France) 6, 675 (1996); J. Oitmaa and Z. WeiHông, Phys. Rev. B 54, 3022 (1996).
[5] R. Melzi et al., Phys. Rev. B 64, 024409 (2001); P. Carretta et al., Phys. Rev. B 66, 094420 (2002).
[6] L. Capriotti et al., cond-mat/0312050; C. Weber et al., Phys. Rev. Lett. 91, 177202 (2003).
[7] A. Cuccoli et al., Phys. Rev. B 67, 104414 (2003), and references therein.
[8] P. Henelius et al., Phys. Rev. B 62, 1102 (2000).
[9] F. Hébert et al., Phys. Rev. B 65, 014513 (2001).
[10] O. F. Syljuåsen et al., Phys. Rev. E 66, 046701 (2002).
[11] P. Sengupta et al., Phys. Rev. B 65, 155113 (2002).
[12] W. Janke, Physica A 254, 164 (1998).
[13] E. Marinari, in Advances in Computer Simulation, J. Kertész and I. Kondor eds., Springer-Verlag (1998).
[14] D. Kandel et al., Phys. Rev. Lett. 65, 941 (1990).
[15] K. Harada et al., J. Phys. Soc. Jpn. 67, 2765 (1998).
[16] A. W. Sandvik, Phys. Rev. B 56, 11675 (1997).
[17] G. Schmid et al., Phys. Rev. Lett. 88, 167208 (2002); G. Schmid and M. Troyer, cond-mat/0304657.
[18] K. Binder et al., Phys. Rev. B 21, 1941 (1980).
[19] G. Kamié and S. T. J. Arovas, Phys. Rev. A 26, 291 (1993).
[20] A. Chubukov, Phys. Rev. B 49, 392 (1991).
[21] S. Chakravarty et al., Phys. Rev. Lett. 60, 1057 (1988).
[22] N. B. Ivanov et al., Phys. Rev. B 46, 8206 (1992).
[23] T. Emarsson et al., Phys. Rev. B 51, 6151 (1995).