Emergence of Magnetic Order in the Kagome Antiferromagnets

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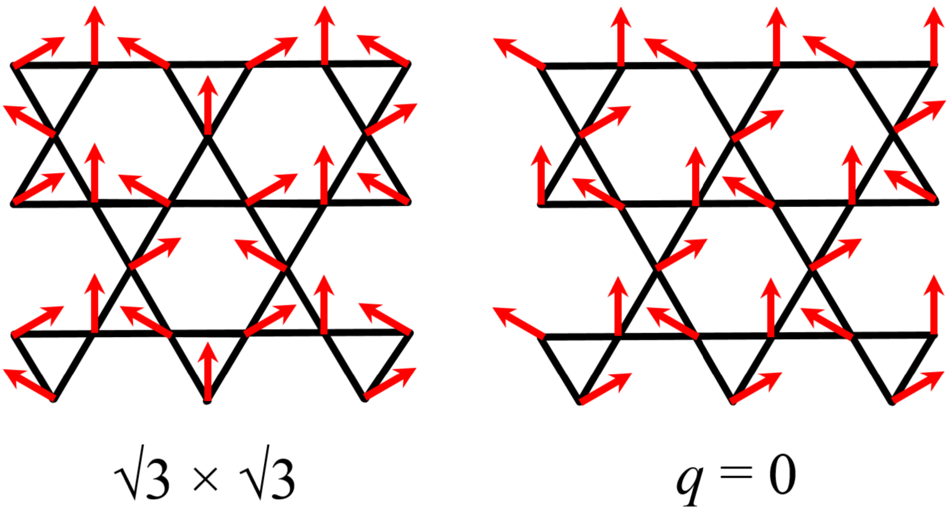

FIG. 1. Kagome lattice with the configurations of spins for the $\sqrt{3} \times \sqrt{3}$ and $q = 0$ states

Exotic quantum spin liquid (QSL) states [1] and fractionalized quasiparticles [2, 3] in frustrated magnets are of much current interest in theoretical and experimental studies of quantum magnetism. The kagome-lattice Heisenberg antiferromagnet (KAFM) provides a possible realization of just such novel topological states of matter. The kagome lattice shown in Fig. 1 is one of eleven Archimedean lattices [4, 5] in two spatial dimensions, where the word *kagomé* itself means "weave pattern" in Japanese. The Hamiltonian for the KAFM model is given by

$$H = J \sum_{\langle i,j \rangle} \mathbf{s}_i \cdot \mathbf{s}_j \quad , \tag{1}$$

where $s_i^2 = s(s+1)$ and the summation $\langle i, j \rangle$ again runs over all nearest-neighbor (NN) bonds (counting each bond once only) and where $J(> 0)$ is the bond strength. The KAFM is geometrically frustrated in the sense that not all pairs of nearest-neighbor spins can be simultaneously antiparallel, as is otherwise favored by the Heisenberg antiferromagnetic exchange interaction between pairs of spins. Even the classical behavior of this model at zero temperature is complicated because there are an infinite number of possible classical ground states to choose from potentially. Each potential classical ground state has nearest-neighboring spins on the "triangles" of these lattices that form angles of 120° to each other. Thermal fluctuations [6–10] favor the $\sqrt{3} \times \sqrt{3}$ (coplanar) state (see Fig. 1). Harmonic quantum fluctuations favor coplanar over non-coplanar states [11–13], whereas anharmonic quantum fluctuations suggest specifically that it is the $\sqrt{3} \times \sqrt{3}$ state that is favored for $s > 1/2$.

Results for the ground-state energy for the spin-half, KAFM are $Eg/(NJs^2) = -1.7544(0.002)$ (density matrix renormalization group (DMRG) [14]; see also Refs. [15, 16]), -1.72884 (entanglement renormalization [17]), -1.732 (series expansions around a dimer limit [18]), -1.754488 (coupled cluster method (CCM) [5, 19]), $-1.75008(0.00024)$ (tensor network states [20]), -1.75257 (large-scale exact diagonalizations (ED) for $N = 42$ [21, 22, 24]), and -1.75482 (ED for $N = 48$ [23]). All of the evidence from these approximation methods (see, e.g., Refs. [5, 14, 19–21]) indicates that the long-range magnetic order parameter is zero. Candidates for the ground state of the spin-half KAFM system are a gapped spin liquid [14–16, 25, 26] (gap $\sim 0.055(5)$ [16] to $0.13(1)$ [14]; see also recent experimental evidence of Ref. [27]), a gapless spin liquid [20, 22, 28, 29], and a valence-bond state [18, 30–33]. Although long-range magnetic order does not occur, the exact nature of the ground state is therefore still a topic of debate.

The simplest and most direct route that order may "emerge from magnetic disorder" for the KAFM is to increase the spin quantum number, s. Results for the ground-state energy for the spin-one KAFM are $Eg/(NJs^2) = -1.3950$ (series expansions) [34], -1.40315 (CCM) [19, 35], $-1.4109(2)$ (tensor network) [36], $-1.410(2)$ (DMRG) [37], and -1.41095 (also DMRG) [38]. Results for the $s = 3/2$ KAFM are $Eg/(NJs^2) = -1.253022$ (series expansions) [34], -1.26798 (CCM) [19, 35], and $-1.265(2)$ (tensor network) [39]. CCM calculations [19, 35] also indicate that the ground-state energy scales with s (for $s \geq 3/2$) via the following equation,

$$\frac{E_g}{NJs^2} = -1 - \frac{0.4140}{s} + \frac{0.0180}{s^2} \quad . \tag{2}$$

The order parameter is given by

$$M = \frac{1}{N} \sum_{i}^{N} \langle \Psi | s_i^z | \Psi \rangle \quad , \tag{3}$$

where s_i^z is defined with respect to local axes of one of the classical ground states at site i. We expect that the order parameter M/s will tend to $M/s \to 1$ in the limit $s \to \infty$. The effect of quantum fluctuations is to reduce to the amount of magnetic order and so we expect $M/s < 1$ for finite values of s. Results of most approximate methods for the spin-one KAFM suggest that there is no magnetic long-range order, although Ref. [40] indicated $\sqrt{3} \times \sqrt{3}$ ground-state long-range order for integer spin quantum numbers, including $s = 1$. Series expansion calculations [34] indicated that $M/s = 0.14 \pm 0.03$ for the $s = 3/2$ KAFM and tensor network calculations [39] also indicate that the $s = 3/2$ system is $\sqrt{3} \times \sqrt{3}$ long-range ordered. CCM calculations [19, 35] indicate that the system is $\sqrt{3} \times \sqrt{3}$ ordered for $s = 3/2$ and that M/s is in the range 0.074 to 0.417. The KAFM demonstrates $\sqrt{3} \times \sqrt{3}$ ground-state long-range order for $s \geq 3/2$ [19, 35, 40, 41]. CCM calculations [19, 35] indicated that the order parameter (with respect to the $\sqrt{3} \times \sqrt{3}$ state) scales with s (for $s \geq 3/2$) via the following equation,

$$\frac{M}{s} = 1 - \frac{1.0676}{s^{0.5}} + \frac{0.0810}{s} \quad , \tag{4}$$

whereas self-consistent spin-wave theory [11] suggested that M/s scales with $s^{-1/3}$ to first order.

The next route to the emergence of magnetic order in the KAFM model is to introduce easy-plane anisotropy into the Hamiltonian [34, 41–45], which we shall refer to as the *XXZ* model, where

$$H = \sum_{\langle i,j \rangle} \left\{ \Delta s_i^z s_j^z + s_i^y s_j^y + s_i^x s_j^x \right\} \quad . \tag{5}$$

The summation $\langle i, j \rangle$ again runs over all NN bonds (counting each bond once only). The *XXZ* model on the kagome lattice is predicted to be magnetically ordered for all values of $\Delta \geq 0$ for $s \geq 3/2$. In the limit $s \to \infty$, CCM calculations [42] indicate that a phase boundary between $\sqrt{3} \times \sqrt{3}$ order at $\Delta = 1$ (KAFM) and $q = 0$ order (see Fig. 1) at $\Delta = 0$ (kagome *XY* model) occurs at the point $\Delta_c(s \to \infty) = 0.727$ [42], whereas results of non-linear spin-wave theory (NLSWT) [41] place this boundary at $\Delta_c(s \to \infty) = 0.72235$. CCM results also suggest that the boundary $\Delta_c(s)$ between these two phases at finite s *increases* with increasing s, whereas NLSWT suggest (tentatively) that the boundary $\Delta_c(s)$ between these two phases *decreases* with increasing s. (Note that CCM results for this boundary are based on the direct comparison of numerical evidence for the two states, whereas statements via NLSWT for this boundary are more speculative.) However, both NLSWT and the CCM predict also that the spin-one, NN KAFM is disordered, which agrees with the conclusions presented above for this model (see also [43]). However, CCM results also predict that a reduction in Δ from $\Delta = 1$ leads to the onset of $q = 0$ order, whereas NLSWT predicts that a reduction in Δ from $\Delta = 1$ leads to the onset of $\sqrt{3} \times \sqrt{3}$ order. Finally, a range of approximate methods (namely, CCM [42], NLSWT [41], DMRG [44], and variational Monte Carlo [45]) predict that the spin-half XXZ model on the kagome lattice is disordered for all values of $\Delta \geq 0$ (i.e., including the XY model).

Another extension of the spin-half, NN KAFM that leads to magnetic order is via the introduction of inter-layer coupling [46–48], where

$$H = \underbrace{J \sum_{n} \sum_{\langle i,j \rangle} \mathbf{s}_{i,n} \cdot \mathbf{s}_{j,n}}_{\text{Within Layer}} + \underbrace{J_{\perp} \sum_{i,n} \mathbf{s}_{i,n} \cdot \mathbf{s}_{i,n+1}}_{\text{Between Layers}} \quad . \tag{6}$$

The summation $\langle i,j \rangle$ indicates NN bonds of strength $J(>0)$ within a given layer (indicated by n). $J_{\perp}$ therefore indicates the bond strength between layers n and $n+1$. The underlying spin model is of two spatial dimensions for $J_{\perp} = 0$ and is of three spatial dimensions model for $J_{\perp} \neq 0$. Quantum magnetic systems of three spatial dimensions might well have a stronger propensity towards magnetic order than systems of lower spatial dimension and so it is not unnatural to suppose that magnetic order (of some) sort might occur. Indeed, it was observed in Refs. [47, 48] that $\sqrt{3} \times \sqrt{3}$ magnetic order is observed for both $J_{\perp}/J = 1$ (antiferromagnetic bonds) and $J_{\perp}/J = -1$ (ferromagnetic bonds). CCM results [48] then suggest that $q = 0$ magnetic order occurs between $-0.435 < J_{\perp}/J < -0.154$ and $0.151 < J_{\perp}/J < 0.310$. A region of magnetic disorder [48] is then observed in the region $-0.154 < J_{\perp}/J < 0.151$, which agrees yet again with the predicted behavior of the spin-half, NN KAFM at $J_{\perp}/J = 0$.

The final route by which magnetic order may emerge from magnetic disorder is to include interactions between spins over greater distances than nearest-neighboring spins via the J_1–J_2 model [2, 20, 49–52] and / or the J_1–J_2–J_3 model [53–57], which is given by

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{s}_i \cdot \mathbf{s}_j + J_2 \sum_{\langle\langle i,k \rangle\rangle} \mathbf{s}_i \cdot \mathbf{s}_k + J_3 \sum_{\langle\langle\langle i,l \rangle\rangle\rangle} \mathbf{s}_i \cdot \mathbf{s}_l \quad . \tag{7}$$

The summation $\langle i,j \rangle$ runs over all NN bonds, $\langle\langle i,k \rangle\rangle$ runs over all next-nearest-neighbor (NNN) bonds, and $\langle\langle\langle i,l \rangle\rangle\rangle$ runs over all next-next-nearest-neighbor (NNNN) bonds. (In each case, bonds are counted once and once only.) For the spin-half model with $J_3 = 0$, $\sqrt{3} \times \sqrt{3}$ order is stabilized by ferromagnetic NNN interactions ($J_2 < 0$) and $q = 0$ order is stabilized by antiferromagnetic NNN interactions ($J_2 > 0$). Initial CCM results [52] suggest however that the magnetically disordered regime survives for a finite range of J_2 centered around the NN KAFM, namely, $-0.070 \lesssim J_2/J_1 \lesssim 0.127$; a result that is also supported by other approximate methods [49, 51]. The addition of antiferromagnetic NNNN bonds ($J_3 > 0$) (with ferromagnetic NN and NNN bonds $J_2, J_3 < 0$) was explored in [55]. The existence of ferromagnetic ordering, $\sqrt{3} \times \sqrt{3}$ ordering, as well as non-coplanar states of magnetic order (CUBOC-1 and CUBOC-2), and finally a large magnetically disordered regime, were posited in Refs. [55, 56]. A complete picture of the ground-state phase diagram of the spin-half J_1–J_2–J_3 model is presented in Ref. [55].

Generalizations of the spin-half NN KAFM are realized physically by magnetic materials such as herbertsmithite $ZnCu_3(OH)_6Cl_2$ [3, 58], haydeeite $Cu_3Mg(OH)_6Cl_2$ [59], vesignieite $BaCu_3V_2O_8(OH)_2$ [60], kapellasite $Cu_3Zn(OH)_6Cl_2$ [61], volborthite $Cu_3V_2O_7(OH)_2{\cdot}2H_2O$ [58, 62], and francisite $Cu_3Bi(SeO_3)_2$ [63]. Hence, the study of such models where magnetic order may "emerge" from the disorder of the pure spin-half NN KAFM are crucial in understanding these materials. Furthermore, we have seen that these generalizations of the spin-half NN model lead to fascinating behavior from a theoretical point-of-view also. These generalized models also provide a useful tool in examining the disordered regimes in the spin-half, NN KAFM because one may observe those limiting cases where magnetic order is seen to vanish as a function of model parameters within the Hamiltonian. The behavior of the ground and excited states can be examined up to and beyond any order-to-disorder phase transitions. Finally, the NN KAFM model and its generalizations provide a very important, interesting, and challenging set of systems by which approximate methods of quantum many-body theory may be compared and contrasted.

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