Rapid Communications

Destruction of long-range order in noncollinear two-dimensional antiferromagnets by random-bond disorder

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We consider frustrated Heisenberg antiferromagnets, whose clean-limit ground state is characterized by noncollinear long-range order with nonzero vector chirality, and study the effects of quenched bond disorder, i.e., random exchange couplings. A single bond defect is known to induce a dipolar texture in the spin background independent of microscopic details. Using general analytical arguments as well as large-scale simulations for the classical triangular-lattice Heisenberg model, we show that any finite concentration of such defects destroys long-range order for spatial dimension $d \leqslant 2$, in favor of a glassy state whose correlation length in $d = 2$ is exponentially large for small randomness. Our results are relevant for a wide range of layered frustrated magnets.

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Spatially inhomogeneous exchange couplings are ubiquitous to magnetic solids. Such disorder, usually dubbed random-bond disorder, arises from crystalline defects or intentional chemical substitution on nonmagnetic sites, causing local changes in bond lengths or bond angles which in turn influence local exchange couplings.

The effect of bond disorder in magnets has been studied extensively, both experimentally and theoretically, with particular focus on frustrated systems [1,2] where the delicate balance of partially satisfied constraints can be easily broken by disorder. In general, systems which are gapped in the clean limit are expected to be stable against weak disorder, such that the phase realized in the clean system survives up to a critical level of disorder where it typically gives way to a spin-glass state [3]. The fate of gapless systems is more subtle, and various scenarios are possible: For strongly frustrated systems, weak bond disorder may immediately induce a spin glass, as in the classical pyrochlore Heisenberg antiferromagnet [4,5], or it may stabilize a distinct disorder-driven long-range-ordered state, as in the classical XY antiferromagnet on the pyrochlore lattice [6] or the frustrated square lattice [7]. In contrast, for weakly frustrated systems it is frequently assumed that the clean-limit order survives the introduction of weak bond disorder, as is the case without frustration.

In this Rapid Communication, we argue that long-range order (LRO) is in fact *not* stable against weak bond disorder for an important class of weakly frustrated magnets, namely SU(2)-symmetric noncollinear magnets in two space dimensions, with the triangular-lattice Heisenberg antiferromagnet being a prominent example. A single bond defect induces a dipolar texture in the spin background [8,9]. A finite concentration of defects then corresponds to spatially fluctuating dipoles which we show to destroy ground-state LRO in space dimension $d \leqslant 2$ [Fig. 1(a)]. The resulting noncoplanar glassy state is characterized by exponentially decaying spin and chirality correlations. Given that $d = 2$ is the lower critical dimension, the magnetic correlation length of this spin glass is exponentially large for weak disorder, implying that both numerical simulations and experiments will detect the destruction of LRO only beyond a resolution-dependent level of bond disorder. As a by-product, we show that site dilution has a much weaker effect, leaving bulk LRO intact in the weak-disorder limit, thus invalidating the assertion that bond disorder and site dilution have similar effects. To connect to experiments, we discuss the physics of finite temperature, weak interlayer coupling, and weak breaking of SU(2) symmetry. In all these cases, the behavior of the clean system survives up to a small finite level of bond randomness.

We note that previous numerical work [10–12] for the triangular-lattice spin-1/2 Heisenberg model suggested that LRO is destroyed only beyond a critical level of bond disorder, in favor of a randomness-dominated spin-liquid-like (or random-singlet) state [13,14]. Our results instead imply

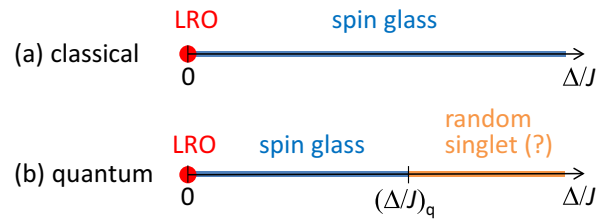

FIG. 1. Schematic ground-state phase diagram of chiral noncollinear magnets with quenched bond disorder in space dimension $1 < d \leqslant 2$, with Δ/J parametrizing the disorder strength. (a) Classical limit ($S \to \infty$): The LRO of the clean system is destroyed for infinitesimal disorder in favor of a spin glass. (b) Quantum case (conjectured): The spin glass turns into a random-singlet (or valence-bond) state at a critical level of disorder (whose value depends on microscopic details).

that true LRO is lost already for infinitesimal bond disorder [Fig. 1(b)], but this could not be detected in the numerics because of finite-size effects.

Model and general considerations. To be specific, we will consider the spin-S triangular-lattice Heisenberg model with antiferromagnetic first-neighbor and second-neighbor couplings,

$$\mathcal{H} = \sum_{\langle ij\rangle} J_{1,ij}\vec{S}_i \cdot \vec{S}_j + \sum_{\langle\langle ij\rangle\rangle} J_{2,ij}\vec{S}_i \cdot \vec{S}_j, \tag{1}$$

whose ground state in the clean limit, $J_{1,ij} \equiv J_1$ and $J_{2,ij} \equiv J_2$, displays coplanar spiral 120° LRO with propagation wave vector $\vec{Q} = \pm(4\pi/3, 0)$ for $\alpha \equiv J_2/J_1 < 1/8$. We will vary α to tune the stiffness of the 120° LRO. The 120° state is chiral: For spins in the x-y plane in spin space, the vector chirality $\vec{S}_i \times \vec{S}_j$ for any given directed pair of sites i, j can point along either $+\hat{z}$ or $-\hat{z}$, corresponding to the two possible propagation wave vectors [15]. As a result, inversion symmetry is broken as well, which plays an important role for the defect physics.

We will be interested in the random-bond case where the $J_{1,ij}$ and $J_{2,ij}$ are taken as (independent) random variables. Bond disorder can be made weak either by employing a narrow distribution of J, or by having a small concentration of defect bonds deviating from the majority coupling strength.

Our main focus will be on semiclassical spin order—this is appropriate for the clean system in the presence of LRO, and by continuity also for weakly disordered systems. In this regime, quantum effects will only yield quantitative corrections, and we will show explicit results for the classical case, formally $S \to \infty$. Strong quantum effects can be expected to be relevant at large disorder and small S: Here, the formation of singlet bonds akin to a random-singlet state has been proposed [10–12]; we will comment on this at the end of this Rapid Communication.

Most generally, our qualitative results will apply to all two-dimensional Heisenberg magnets with semiclassical noncollinear, but coplanar LRO, both with spontaneous and explicit (i.e., crystallographic) chirality.

Single bond defect. A single defect bond in an otherwise homogeneous system has been studied before [8,9], and we summarize the key results. We consider

$$J_{1,ij} = \begin{cases} J_1 + \delta J, & (i,j) = (0,1), \\ J_1, & \text{otherwise}, \end{cases} \tag{2}$$

and $J_{2,ij} \equiv J_2$. The presence of the defect locally relieves frustration, leading to a readjustment of the spin directions. Numerical results for a bond defect with $\delta J = -J_1$ are shown in Fig. 2(a). The spin configuration remains coplanar and can be analyzed in terms of angles $\delta\Theta_i$ which describe the defect-induced in-plane rotation of the spins $\vec{S}_i$ with respect to the 120° LRO. The Hamiltonian with couplings (2) has inversion symmetry with respect to the center of the defect bond, and $\delta\Theta_i$ is found to be *odd* under this inversion: $\delta\Theta_i$ has a p-wave-like shape and decays proportional to $1/r$, where r is the distance from the defect [Fig. 2(b)]. This implies that the bond defect acts as a *dipolar* perturbation; this can be contrasted to the case of a vacancy which induces an octupolar texture, with an f-wave shape and a $1/r^3$ decay of $\delta\Theta_i$ [16,17].

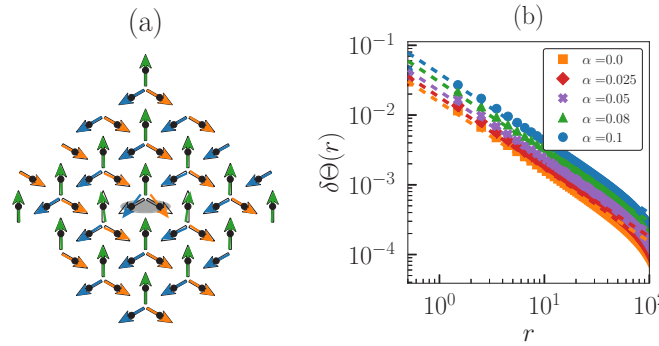

FIG. 2. Single bond defect in a triangular-lattice Heisenberg antiferromagnet. (a) Spin configuration near the defect bond (shaded) for $\delta J = -J_1$ (2) and $J_2 = 0$; the unperturbed 120° order is shown in gray. (b) Defect-induced spin rotation angles $\delta\Theta$ as a function of distance r from the defect, for $\delta J = -J_1/10$ and different $\alpha \equiv J_2/J_1$. The dashed lines are fits to $1/r$ behavior for $2 < r < 20$; deviations at large r arise from finite sample size and periodic boundary conditions, $L = 300$.

The connection between the local release of frustration and the long-range nature and shape of the distortion can be understood at a linear-response level: The frustration is released such that the two spins 0,1 on the defect bond align more (less) antiparallel compared to the 120° state for $\delta J > 0$ ($\delta J < 0$), respectively. This rotation can be induced by a locally transverse field, $h^\perp \propto \delta J$, acting on $\vec{S}_0$ and $\vec{S}_1$ with opposite signs. Working in a local frame of spin coordinates such that the order is uniform along the $\hat{x}$ axis, the locally transverse field acts as $h^\perp \sum_{j=0}^{1} \beta_j S_j^y$ with $\beta_j = (-1)^j$. The long-distance pattern follows as the response of the ordered state to this *local dipolar* field. The relevant in-plane susceptibility $\chi^\parallel(\vec{q})$ is dominated by the in-plane spin-wave modes whose dispersion is $\omega_q \propto |\vec{q}|$ such that $\chi^\parallel(\vec{q}) = N_0^2/(\rho_s q^2)$, where ρ_s is the spin stiffness against in-plane twists, and N_0 is the magnitude of the order parameter [18], both taken for the clean system. This yields, in the continuum limit, $\delta\Theta(\vec{r}) \propto \int d^d q e^{i\vec{q}\cdot\vec{r}} \beta_{\vec{q}} \chi^\parallel(\vec{q})$ in d space dimensions where $\beta_{\vec{q}} = \hat{e} \cdot \vec{q}$ is the p-wave form factor of the local perturbation, with $\hat{e}$ being the lattice vector of the directed defect bond. Together, we obtain

$$\delta\Theta(\vec{r}) = \kappa\, \delta J \frac{N_0^2}{\tilde{\rho}_s} \frac{\hat{e} \cdot \vec{r}}{r^d}, \tag{3}$$

where $\tilde{\rho}_s = \rho_s/\mathcal{A}$ with $\mathcal{A}$ the unit-cell area [19], and κ a numerical prefactor (see Ref. [20] for details). Being inversely proportional to the stiffness, the defect response thus depends significantly on $\alpha = J_2/J_1$, consistent with Fig. 2(b).

The fact that a bond defect produces an antisymmetric (or inversion-odd) texture is intimately connected to the chirality of the ground state: The Hamiltonian itself is inversion-even, and a single defect cannot spontaneously break this symmetry. However, the ground state is chiral, with broken inversion symmetry, enabling the defect to produce an odd perturbation. In other words, the ground-state chirality endows the bond

defect with a direction, as required for a dipole, and reversing the chirality will reverse the sign of the dipole, $\hat{e} \leftrightarrow -\hat{e}$.

As an aside, we note that the state with a single bond defect has a finite uniform magnetization $m_{\rm imp}$ which takes a nonuniversal fractional value, similar to the vacancy case [16]. For the bond defect with $\delta J = -J_1$ and $\alpha = 0$ we have found $m_{\rm imp}/S = 0.396 + \mathcal{O}(S^{-1})$. We also note that a single (weak) defect on a second-neighbor bond has no effect on the classical spin order, as second-neighbor spins are parallel in the 120° state.

Destruction of LRO by dipolar fluctuations. We now turn to the case of a finite defect concentration and argue that LRO is generically destroyed for $d \leqslant 2$, adopting an argument originally due to Aharony [21]. To this end, we assume LRO and employ local rotated frames as above, with order along the $\hat{x}$ axis. We distribute random bonds $J_{1,ij}$ on the lattice and define $\delta J_{ij} = J_{1,ij} - J_1$ where $J_1 = \overline{J_{1,ij}}$ is the disorder-averaged J_1, such that the disorder strength is parametrized by $\overline{\delta J_{ij}^2} \equiv \Delta^2$. For each directed bond $\hat{e}_{ij}$, $\vec{d}_{ij} \equiv \hat{e}_{ij}\delta J_{ij}$ takes the role of a local dipole strength. For weak randomness, the resulting rotation of an individual spin is proportional to its transverse magnetization in the rotated frame, $\delta\Theta_l = \langle S_l^{\perp}\rangle/N_0$. It can be estimated using the linear response as above,

$$\langle S_l^{\perp}\rangle = \kappa \frac{N_0^3}{\tilde{\rho}_s} \sum_{\langle ij\rangle} \frac{\vec{d}_{ij}\cdot\vec{r}_{l,ij}}{r_{l,ij}^d}, \tag{4}$$

where $\vec{r}_{l,ij}$ is the vector connecting site l and the center of the ij bond. The disorder-averaged transverse magnetization $\overline{\langle S_j^{\perp}\rangle}$ vanishes because the averaged dipole strength has zero mean. In contrast, the averaged magnetization correlation function is nonzero [20],

$$\overline{\langle S_l^{\perp 2}\rangle} = \tilde{\kappa}\,\Delta^2 \frac{N_0^6}{\tilde{\rho}_s^2} \int dr\, r^{1-d}, \tag{5}$$

where we have passed to the continuum limit, the angular average has been performed, and $\tilde{\kappa} \propto \kappa^2$ is a prefactor.

The above integral is infrared divergent for $d \leqslant 2$, such that the local transverse magnetization fluctuations diverge for $d \leqslant 2$: These fluctuations, arising from dipolar bond disorder and transmitted by long-wavelength modes, then destroy the assumed ordered state. In contrast, for $d > 2$ a finite defect concentration is required to destroy order. The same conclusion can be reached by a more elaborate renormalization-group (RG) treatment of a relevant nonlinear sigma model (for details see Ref. [20]). The destruction of LRO by dipolar fluctuations has been studied in different contexts before [22,23].

Emergent spin glass. We now discuss the nature of the emerging zero-temperature state without magnetic LRO. In the semiclassical limit, this must be a state with spontaneously broken SU(2) symmetry and, given the random-field character of the problem, is a spin glass with frozen short-ranged spin order [24,25].

We can estimate its magnetic correlation length ξ simply by assuming that ξ provides an infrared cutoff to the integral Eq. (5), i.e., by identifying ξ with a domain size. The stability condition $\overline{\langle S_l^{\perp 2}\rangle} \lesssim N_0^2$ translates into $\xi^{2-d} \propto \tilde{\rho}_s^2/(\Delta^2 N_0^4)$ for $d < 2$, whereas in $d = 2$ the correlation length is exponentially large for small Δ,

$$\ln \frac{\xi}{\xi_\infty} \propto \frac{\tilde{\rho}_s^2}{\Delta^2 N_0^4}, \tag{6}$$

where the constant ξ_∞ formally represents the correlation length as $\Delta \to \infty$. While the spin configurations remain coplanar in the limit of small Δ, this is no longer true at finite disorder [20].

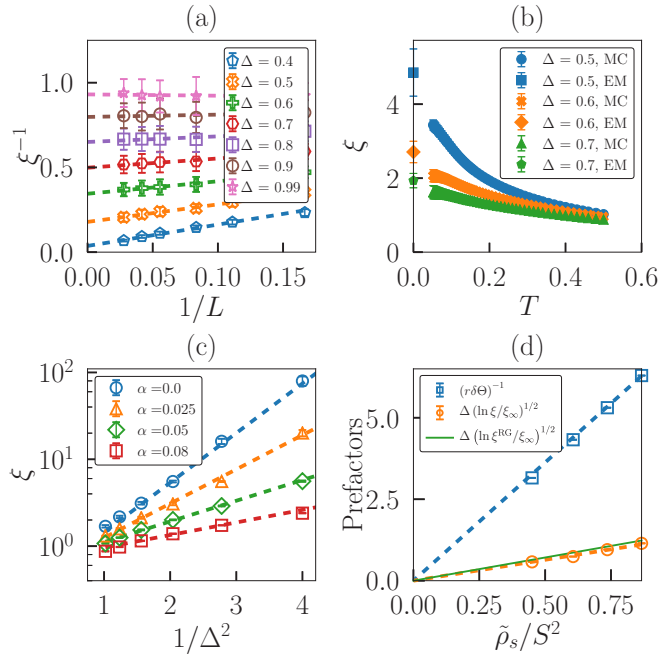

FIG. 3. Magnetic correlation length ξ for the triangular-lattice Heisenberg model with Gaussian bond disorder. (a) Finite-size scaling of $1/\xi$ as function of linear system size L for $\alpha = 0.05$, $T = 0$, and different disorder strength Δ. (b) Comparison of ξ obtained from finite-T MC simulation and $T = 0$ energy minimization (EM), for $\alpha = 0.05$ and $L = 36$. (c) $\ln\xi$ plotted as a function of $1/\Delta^2$, illustrating the scaling from Eq. (6). (d) Test of stiffness dependence: $[\ln(\xi/\xi_\infty)]^{1/2}\Delta$ and single-defect $1/[r\delta\Theta(r)]$ for different α, plotted as functions of spin stiffness. The solid line corresponds to the RG result for ξ. Dashed lines are linear fits.

Numerical results for finite disorder. Our analytical results are well borne out by large-scale simulations for the bond-disordered triangular-lattice J_1-J_2 Heisenberg model, using both ground-state energy minimization and finite-temperature Monte Carlo (MC) techniques [20]. We have employed different disorder distributions; below we show results where the $J_{1,ij}$ and $J_{2,ij}$ have been drawn independently from Gaussian distributions with mean J_1 (which we use as unit of energy) and $J_2 = \alpha J_1$ and widths Δ and $\alpha\Delta$, respectively.

We have determined the magnetic correlation length ξ from the disorder-averaged static structure factor. An example for the finite-size scaling of ξ at $T = 0$ is shown in Fig. 3(a). For large disorder, we clearly detect the absence of LRO, while for small disorder the correlation length exceeds the available system sizes, such that the finite-size scaling is inconclusive. The temperature dependence of ξ obtained via MC simulations is consistent with the data at $T = 0$ [Fig. 3(b)]. The finite-T spin-glass correlation length is found much larger than its magnetic counterpart, consistent with a spin-glass

ground state. An analysis of individual spin configurations confirms that the glassy state is noncoplanar [20].

Plotting the extrapolated data for ξ at $T = 0$ as a function of disorder strength Δ [Fig. 3(c)], we find the Δ dependence perfectly consistent with the exponential behavior predicted by Eq. (6). This strongly suggests that LRO is indeed destroyed for *any* nonzero Δ. Equation (6) also predicts that $[\ln(\xi/\xi_\infty)]^{1/2}\Delta$ is proportional to the clean-limit stiffness which reads $\tilde{\rho}_s = S^2(J_1 - 6J_2)\sqrt{3}/2$ in the classical limit [20]. This proportionality is tested in Fig. 3(d) and found to be perfectly obeyed. In fact, the RG treatment [20] also yields an approximate expression of the proportionality factor which agrees with the data. In addition, Fig. 3(d) also shows $1/[r\delta\Theta(r)]$ for the single-defect texture which is proportional to the stiffness, in quantitative agreement with Eq. (3). Finally, we do not detect any crossing points in the Δ dependence of both the magnetic correlation length and the Binder parameter for different L [20]. This confirms the absence of a critical Δ and thus the phase diagram in Fig. 1(a).

Perturbations: Finite T, interlayer coupling, and anisotropies. Beyond the two-dimensional Heisenberg model discussed so far, a number of effects are important. First, in a strictly two-dimensional SU(2)-symmetric system, the clean-limit LRO is restricted to $T = 0$ due to the Mermin-Wagner theorem. Hence, the clean system is short-range ordered at any finite T, with the thermal correlation length ξ_T scaling as $\ln \xi_T \propto \rho_s/T$ [26]. Now, bond disorder limits the correlation length according to Eq. (6), which defines a disorder-dependent crossover temperature which scales quadratically with the disorder level, $T^* \propto \Delta^2 N_0^4/\rho_s$, below which the system settles into its $T = 0$ glassy state. We note that $d = 2$ is below the lower critical dimension for spin-glass order [27], hence, there will be no thermodynamic glass transition in a strictly two-dimensional system. Second, a small but finite interlayer coupling will render the system three-dimensional at sufficiently low temperature, leading to finite-T LRO which is also stable against weak bond disorder. As a result, LRO is destroyed in favor of a glassy state only beyond a critical level of disorder which scales with the interlayer coupling according to $J_\perp/J \propto \Delta^4 N_0^8/\rho_s^4$. Third, if SU(2) is broken down at the Hamiltonian level such that there is no spin rotation symmetry in the ordering plane, the in-plane mode of the clean system acquires a gap. As a result, the texture induced by a single defect decays exponentially. The system displays again LRO at low T which is stable against weak bond disorder. Here, the critical level of disorder scales logarithmically with the gap (for details see Ref. [20]).

Quantum effects. So far our analysis was based on semiclassical spin order. It is strictly valid for $S \to \infty$, but qualitatively also applies to finite S: $1/S$ corrections to observables can be calculated and are generically nonsingular at $T = 0$ [16,20]. However, for small S the physics can change; in particular, local spin order can be destroyed by quantum fluctuations.

For the noncollinear magnet at hand, it is conceivable that a finite amount of bond disorder can lead to the suppression of local order via the emergence of a disorder-dominated valence-bond state [28,29], similar to the celebrated random-singlet state in one dimension [13,14]. In fact, the emergence of such a state has been proposed on the basis of numerical simulations for the bond-disordered Heisenberg model both on the triangular and honeycomb lattices [10–12]. Together with our insight that infinitesimal bond disorder destroys noncollinear LRO in favor of a spin glass, we conjecture the phase diagram in Fig. 1(b), where the glass gives way to a random-singlet state at large disorder.

Conclusions. Combining analytical arguments and large-scale simulations, we have shown that random-bond defects destroy long-range magnetic order in the ground state of two-dimensional noncollinear antiferromagnets with SU(2) spin symmetry. The key insight, demonstrated explicitly for the triangular-lattice Heisenberg model, is that a finite concentration of effectively dipolar defects destabilize LRO even for weak disorder in favor of a spin-glass state.

Remarkably, the effect of random site dilution in the same system is much weaker: Vacancies induce an octupolar texture [16,17], and LRO is stable against a small vacancy concentration because the integral corresponding to Eq. (5) is *not* divergent in this case [30].

Our analysis shows that none of the two cases can be described as random-mass disorder in the relevant low-energy field theory for the ordered state, because this would miss the defect-induced random transverse fields. This underlines a fundamental difference between the present noncollinear magnets and their unfrustrated collinear counterparts, where both random-bond disorder and site dilution correspond to random-mass terms in the field theory. We note that bond disorder also tends to destroy LRO in frustrated magnets with strong spin-orbit coupling where defect-induced random transverse fields are even stronger [31,32].

We expect our ideas to motivate further studies into different types of quenched disorder in weakly frustrated magnets. Investigations for noncoplanar order are underway. On the experimental front, our results are pertinent to many materials such as $Ba_3CoSb_2O_9$ [33], $Ba_3NiSb_2O_9$ [34], $Ba_8CoNb_6O_{24}$ [35], Cs_2CuCl_4 [36], α-$CaCr_2O_4$ [37], and $NaCrO_2$ [38]. They also apply to certain iron pnictides where a noncollinear vortex crystal state has been detected [39].

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