Ordering Due to Disorder in a Frustrated Vector Antiferromagnet

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In many continuous spin systems, competing interactions give nontrivial degeneracies of the classical ground states. Degeneracy-breaking free-energy terms arise from thermal (or quantum) fluctuations, which select for *collinear* states, and from dilution, which selects for "*anticollinear*" (yet long-range ordered) states. They are explicitly computed for an XY square-lattice antiferromagnet dominated by second-neighbor antiferromagnetic exchange. The predicted phase diagram agrees qualitatively with simulations.

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Many periodic vector spin systems with competing exchange couplings have nonunique (classical) antiferromagnetic ground states: These form a continuous manifold of degenerate states including not only the states trivially related by the global rotational symmetry, but additional sets of states related by applying different rotations to the various antiferromagnetic sublattices.[1,2]

There is a large class of such systems: many spinels;[3,4] all face-centered-cubic (fcc) antiferromagnets including type-I systems[1,5] (e.g., γ-Mn),[6] type-II systems (e.g., MnO), type-III systems (e.g., $Cd_{1-x}Mn_xTe$ for larger x),[7,8] and possibly Cu nuclear spins;[9] triangular antiferromagnets (possibly stacked);[10-12] bcc type-II (e.g., $Ca_3Fe_2Ge_3O_{12}$ garnet)[13] dipolar-coupled spins on a honeycomb lattice;[14,15] and fully frustrated cubic systems.[16] In addition, they may be realized in certain superconducting arrays at particular rational values of flux per plaquette.[2]

When diluted by substituting nonmagnetic impurities, such systems are supposed to become spin glasses:[4] e.g., $Cd_{1-p}Mn_pTe$, a diluted magnetic semiconductor[17] where the Mn ions form a diluted fcc lattice with well understood antiferromagnetic exchange constants.[18] Experimentally, at $p \approx 0.4$ this system is spin-glass-like[17] while at $p \approx 0.7$ it shows strong (but still local) antiferromagnetic order.[18(a)] Part of the motivation of this work is to distinguish the spin glass from other phases with random-field-like disorder which might be present near $p=1$.

Not surprisingly, perturbations—thermal fluctuations, quantum fluctuations, or dilution—lift these degeneracies and select specific states, reducing the continuous degeneracy to a discrete one.[12,13] I will call this "ordering due to disorder"[19] by analogy to the Ising case.

In this Letter I argue that, in exchange-coupled systems, *thermal* and *quantum* disorder favor *collinear* states, wherein spins are aligned parallel or antiparallel to a single direction (which itself remains free to rotate); but random *dilution* favors the *least* collinear states, which I will call "anticollinear." In addition, random dilution often makes effective "random exchange fields" coupling to the discrete (but not the rotational) symmetries[20] like a random field. These effects all compete, yielding two or more antiferromagnetic phases.[1,2,8]

In the rest of the Letter, I will outline the general arguments, and display the specific calculations for a 2D XY system with second-neighbor exchange,[1] the simplest possible model with both rotational symmetry and nontrivial continuous degeneracy. The rich phase diagram predicted for this case is consistent with Monte Carlo results.[21]

Model system.—Let us take XY spins on a square lattice (lattice constant $\equiv 1$) with Hamiltonian $\mathcal{H} = \frac{1}{2}\sum_{ij} J_{ij}\cos(\theta_i - \theta_j)$, where $J_{ij} = J_1$ (J_2) for nearest (second-nearest) neighbors. If $|J_1|/|J_2| < 2$, the system in its ground state breaks up into two square ($\sqrt{2}\times\sqrt{2}$) sublattices, a and b, each ordered antiferromagnetically[22] (Fig. 1). To label the ground states, choose one reference spin from each sublattice, say at [0,0] and [0,1] with angles θ_a and θ_b. Then $\phi \equiv \theta_a - \theta_b$ parametrizes a nontrivial "degeneracy," since the ground-state energy $E_0 \equiv -2N|J_2|$ is independent of ϕ.

This model might be realized in a two-layer square array of superconducting islands with one quantum of flux per cell, in two layers of MnTe in CdTe (fabricable by molecular-beam epitaxy), or in two adjacent square CuO_2 layers centered on each other (as in some high-T_c superconductors), where J_1 is a small interlayer ex-

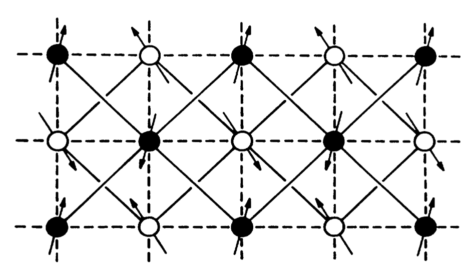

FIG. 1. Ground state on square lattice with $J_2 < -\frac{1}{2}|J_1|$.

change. The spin-$\frac{1}{2}$ version appears in a study,[23] motivated by high-T_c theories, of the loss of Néel order due to large quantum fluctuations when $|J_1|/|J_2| \approx 2$.

Ground-state selection.— Broad intuitive arguments[1] suggest that thermal fluctuations favor the collinear states, defined by $\cos\phi = \pm 1$, whereas quenched fluctuations favor the perpendicular states with $\sin\phi = \pm 1$. In either case, there are two senses in which the second sublattice can orient collinear or perpendicular, so the continuous degeneracy is replaced by a discrete, Ising-type variable.

Take $\mathbf{h}_i \equiv \sum_j J_{ij}\mathbf{s}_j$, the local exchange field; in a ground state, $\mathbf{s}_i \parallel \mathbf{h}_i$. Also, for site i in sublattice a, let $\delta\mathbf{h}_i^{ba}$ be the component of $\mathbf{h}_i$ from sublattice b. The nontrivial degeneracy arises because of a cancellation: $\mathbf{h}_i^{ba} \equiv 0$ everywhere— classically, at $T=0$. In the presence of *thermal* (or *quantum*) fluctuations, if $\delta\mathbf{h}_i^{ba} \parallel \mathbf{s}_i$, it has no effect on $\mathbf{s}_i$ (to lowest order). To maximize the coupling between the fluctuations of the two sublattices, we need rather $\delta\mathbf{h}_i^{ba} \perp \mathbf{s}_i$. But $\delta\mathbf{h}_i^{ba}$ is itself perpendicular to the spins of sublattice b. Hence, a *collinear* alignment is preferred.[13]

In a *diluted* lattice, even in the classical ground state, $\mathbf{h}_i^{ba} \neq 0$. Say we remove just one spin from site j in sublattice b. For a neighbor i in sublattice a, $\mathbf{h}_i^{ba} = \mp J_1\mathbf{s}_j$; then $\mathbf{s}_i$ will cant towards this direction by an angle $\delta\theta_i \sim J_1/J_2$. The energy is minimized when the $\{\delta\theta_i\}$ are maximized, i.e., when the two sublattices are *perpendicular*.

In either case, the same logic applies to other exchange-coupled systems suggesting a universal rule for the respective selection effects.

Spin-mode calculations.— Next I confirm the asserted selection terms by calculating them from the Hamiltonian expanded about a ground-state configuration $\{\theta_i\}$,

$$\delta\mathcal{H}_\phi \equiv \tfrac{1}{2}\sum_{ij} A_{ij}(\phi)\delta\theta_i\delta\theta_j \,, \tag{1}$$

where $A_{ij}(\phi) \equiv 4|J_2|\delta_{ij} - J_{ij}\cos(\theta_i - \theta_j)$. By Fourier transforming, we find

$$\delta\mathcal{H}_\phi = N\int (2\pi)^{-2} d^2q \, \tfrac{1}{2} A_{\mathbf{q}}(\phi)\,|\delta\theta_{\mathbf{q}}|^2 \,, \tag{2}$$

where

$$A_{\mathbf{q}} = 4|J_2|(1-\cos q_x \cos q_y) - 2J_1(\cos q_x - \cos q_y)\cos\phi \,. \tag{3}$$

Firstly, for thermal fluctuations the small parameter is T. Following Ref. 12, we evaluate the free energy from Eq. (2):

$$F(\phi,T) - E_0 = -\tfrac{1}{2}NT\ln T - NTS_0(\phi) \,,$$

where the "ground-state entropy" is

$$S_0(\phi) = \text{const} - \int (2\pi)^{-2} d^2q \ln A_{\mathbf{q}}(\phi) = \text{const} + g_0(J_1\cos\phi/2J_2) \,, \tag{4}$$

with $g_0(x)$ an even function, increasing with $|x|$: $g_0(x) = 0.220 + 0.32x^2 + \cdots$. This confirms that the *collinear* state $\cos\phi = \pm 1$ is selected.

Secondly, in the case of *quantum* fluctuations (at $T=0$), the small parameter is $\hbar$. To define the quantum-mechanical spin waves, we must endow the system with reactive dynamics: $d^2\theta_i/dt^2 = -\Gamma\, dH/d\theta_i$— this is plausible for both realizations of the model[2]— so the spin-wave frequencies are $\omega_{\mathbf{q}}(\phi) = [\Gamma A_{\mathbf{q}}(\phi)]^{1/2}$. The ground-state energy difference is given by the zero-point term,

$$E(\phi) - E_0 = \tfrac{1}{2}\int (2\pi)^{-2} d^2q \, \hbar\omega_{\mathbf{q}}(\phi) = 2\hbar(\Gamma J_2)^{1/2} g_{1/2}(J_1\cos\phi/2J_2) \,, \tag{5}$$

where $g_{1/2}(x) = 0.9581 - 0.082x^2 + \cdots$, again favoring the *collinear* state with $\cos\phi = \pm 1$.[13,24]

These spin-wave results for thermal and quantum fluctuations can be connected to the intuitive arguments so as to show that the collinear selection should apply to general exchange-coupled systems.[2,25] The selecting free energies (4) and (5) have the algebraic structure

$$\int d^2q \, f(A_{\mathbf{q}}(\phi)) \,,$$

depending on the ϕ only through $A_{\mathbf{q}}(\phi)$. For thermal fluctuations $f(x) \propto \ln x$ and for quantum fluctuations $f(x) \propto \sqrt{x}$. If the overall scale of $A_{\mathbf{q}}$ depended on ϕ, this would control the selection, but in fact its integral is constrained to be $2E_0$ for every ϕ value.[2] Now, $f(x)$ is convex upwards in both cases. Then to minimize the average of $f(x)$ with the average of x fixed, we need the x values to be as strongly dispersed as possible. But the collinear state has the maximum coupling between sublattices, and hence [see Eq. (3)] the strongest dispersion of spin waves; therefore it is selected.

Thirdly, consider the effect of *dilution*, with occupied fraction p; here $T \equiv 0$ and J_1/J_2 is the small parameter, which ensures that the $\{\delta\theta_i\}$ are small. For every possible ϕ, we expand about the corresponding ground state up to $O(\delta\theta_i^2)$:

$$\delta\mathcal{H} = -\tilde{H}\cos\phi - J_1\sin\phi\sum \gamma_{\mathbf{q}}\delta\theta_{-\mathbf{q}} + p^2\delta\mathcal{H}_\phi \,. \tag{6}$$

A constant has been dropped. Here $\gamma_{\mathbf{q}}$ is the Fourier transform of $\gamma_i \equiv \sum_j' (-1)^{x_i+y_j}$ (sum over *occupied* near neighbors of i) and $\tilde{H} \equiv J_1\sum_{ij}' (-1)^{y_i - y_j}$ (sum over *occupied* neighbor pairs). These ϕ-dependent constant and linear terms are zero on average, with

$$\langle \tilde{H}^2 \rangle \cong (\delta p)^2 N \,, \tag{7}$$

and

$$\langle |\gamma_{\mathbf{q}}|^2 \rangle \cong 4\,\delta p(\cos aq_x - \cos aq_y)^2 \tag{8}$$

for $\delta p \equiv 1 - p \ll 1$. The quadratic term has been replaced in (6) by its average over realizations, with $\delta\mathcal{H}_\phi$ given by (1). Minimizing the sum of the linear and quadratic terms gives $\delta\theta_{\mathbf{q}} = -(J_1\sin\phi)^2\gamma_{-\mathbf{q}}/p^2A_{\mathbf{q}}$; inserting this into (6) and using (8), the average energy (for $p \rightarrow 1$, $J_1/J_2 \rightarrow 0$) is

$$\langle \delta \mathcal{H}(\phi) \rangle \cong -(J_1^2/2J_2)\, \delta p\, G_{-1} \sin^2\phi \,, \tag{9}$$

where

$$G_{-1} \equiv \int (2\pi)^{-2} d^2q \frac{(\cos q_x - \cos q_y)^2}{1-\cos q_x \cos q_y} \approx 0.727 \,,$$

this time favoring $\phi = \pm\pi/2$.

It is clear that "anticollinear" selection holds generally in systems with degeneracies, for the energy analogous to (9) always takes its maximum value (zero) for collinear states.

Random exchange fields.—As Fernandez has noted,[20,22] the $\tilde{H}$ term in (6) can couple to the degeneracy degree of freedom ϕ exactly as a random field couples to the spin direction. However, unlike a true random field, it does not break the O(2) rotational symmetry,[26] so I propose instead calling such a term (which appears in all diluted systems with "degeneracies") a "random exchange field" (REF). A strong motivation for investigating REF's is that the "spin-glass" phase in $(Cd_{1-p}Mn_p)Te$ behaves experimentally somewhat like a random-field system: The antiferromagnetic correlation length grows with decreasing T and saturates roughly around the susceptibility cusp,[18(a)] while the ac susceptibility exhibits "activated" (logarithmic) dynamic scaling.[27]

The correct approach to the random exchange fields is based on symmetry: Assume that *locally* the system is essentially in one of the ground states selected by the bulk term (9); determine whether the REF couples to the order parameter, and if so whether the gain from following the REF locally outweighs the cost of the associated gradients in sublattice orientations.

At $T=0$, the dilution selection term (9) reduces the degeneracy freedom to an Ising-type discrete degree of freedom. Since $\cos\phi \equiv 0$ in the anticollinear state, the REF's do not couple at all.[28] On the other hand, $\tilde{H}$ *does* couple to the discrete degree of freedom of the *collinear* state. That is, statistical fluctuations favor alignment in one sense ($\cos\phi = 1$) in some domains and in the other sense ($\cos\phi = -1$) elsewhere. The REF's affect the collinear state exactly as random fields affect an Ising model.[20] In $d=2$ long-range order is lost and the collinear state ($AF_\parallel$ in Fig. 2) is actually part of the paramagnetic phase. However, analogous systems in $d>2$ retain collinear order.

Phase diagram.—The analytic results and the considerations discussed above predict the phases and transitions[1,2] shown in Fig. 2. Collinear and anticollinear order are characterized respectively by order parameters

$$\tilde{M}_\parallel \equiv \sum_{ij} (-1)^{y_i - y_j} \cos(\theta_i - \theta_j)$$

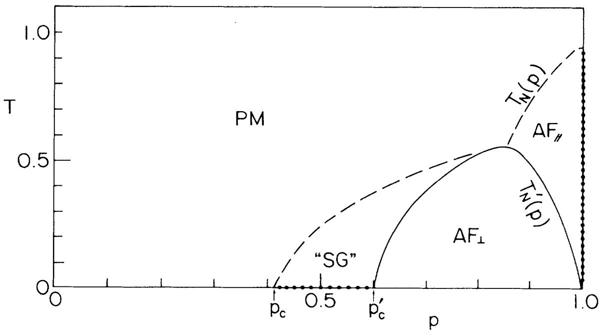

FIG. 2. Proposed phase diagram when p is the occupied fraction, and $J_2 = -1$, $|J_1| = 1$. Here "SG" denotes the "spin glass," $AF_\parallel$ and $AF_\perp$ indicate the collinear and anticollinear antiferromagnetic states, and PM is the paramagnetic state. The dashed lines are only crossover lines in $d=2$; true $AF_\parallel$ and SG phases exist only along the lines of heavy dots.

and

$$\tilde{M}_\perp \equiv \sum_{ij} (-1)^{y_i - y_j} \sin(\theta_i - \theta_j).$$

The *pure* system has the same order-parameter symmetry $[Z_2 \otimes SO(2)]$ as the triangular XY antiferromagnet[5] or the fully frustrated square XY model, and it is plausible to conjecture it has the same critical properties: a simultaneous ordering of the Ising and XY degrees of freedom at $T_N(1)$. The collinear susceptibility $\tilde{\chi}_\parallel$ corresponding to $\tilde{M}_\parallel$ and the specific heat $C(T)$ should show Ising-type critical divergences while the staggered magnetization correlations should show Kosterlitz-Thouless essential singularities.[10(b)]

The transition $T_N'(p)$ between the collinear and anticollinear states has the form $T_N'(p) \cong \text{const} \times J_2 \delta p$, since the competing selection terms (4) and (9) are, respectively, linear in T and δp. Long-range anticollinear order at $T=0$ persists for $p<1$, down to p_c' which should be slightly above the site percolation threshold (≈ 0.59) of the J_2-coupled square sublattices. The sites still percolate (by J_1 or J_2 bonds) down to $p_c \approx 0.41$. For $p_c < p < p_c'$, Villain's picture[4] suggests the ground state is a spin glass.

Monte Carlo simulations[21] (for $J_1 = 1$, $J_2 = -1$, with $N \le 50^2$ spins) confirm the expected features: Collinear ordering of the pure system occurs, at $T_N(1)/J_2 \cong 0.97 \pm 0.02$, with $T_N'(p) \cong 5.5 J_2 \delta p$, and $p_c' \approx 0.6$. This goes beyond the analytic results, which were valid only for small δp and J_1/J_2.

In conclusion, a two-dimensional XY model, chosen to be the simplest antiferromagnet exhibiting "ordering due to disorder," has a very rich phase diagram determined by competition between thermal selection favoring collinearity, dilution selection favoring anticollinearity, and random exchange fields which tend to disorder the collinear selected states. Qualitatively similar behavior is expected in most such systems; analogous calculations will be published elsewhere.[2,8,15] However, since the discrete symmetry differs from case to case, the critical behaviors and random-field responses may be quite different.[2,28]

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[22]J. F. Fernandez has independently conceived the same model.

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[24]Indeed, the spin-$\frac{1}{2}$ fcc antiferromagnet (Ref. 5) and the fully frustrated cubic lattice (Ref. 16) show collinear selection.

[25]Generally, ϕ becomes an abstract label for a ground-state manifold which often has more than one dimension of degeneracy degrees of freedom.

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[28]In other systems, REF's will couple to the anticollinear order parameter if the discrete part of the broken symmetry is associated with a direction in real space.