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 nonuniquc (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.

There is a large class of such systems: many spinels, all face-centered-cubic (fcc) antiferromagnets including type-I systems (e.g., MnO), type-II systems (e.g., MnO), type-III systems (e.g., Cd$_{1-x}$Mn$_x$Te for larger $x$), and possibly Cu nuclear spins; triangular antiferromagnets (possibly stacked); Ca$_3$Fe$_2$Ge$_2$O$_{12}$ garnet; dipolar-coupled spins on a honeycomb lattice; and fully frustrated cubic systems. In addition, they may be realized in certain superconducting arrays at particular rational values of flux per plaquette.

When diluted by substituting nonmagnetic impurities, such systems are supposed to become spin glasses; e.g., Cd$_{1-p}$Mn$_p$Te, a diluted magnetic semiconductor, where the Mn ions form a diluted fcc lattice with well understood antiferromagnetic exchange constants. Experimentally, at $p \approx 0.4$ this system is spin-glass-like while at $p \approx 0.7$ it shows strong (but still local) antiferromagnetic order. 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. I will call this “ordering due to disorder” 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 like a random field. These effects all compete, yielding two or more antiferromagnetic phases.

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, 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.

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 (Fig. 1). To label the ground states, choose one reference spin from each sublattice, say at $[0,0]$ and $[0,1]$ with angles $\theta_a$ and $\theta_b$. Then $\phi \equiv \theta_a - \theta_b$ parametrizes a nontrivial “degeneracy,” since the ground-state energy $E_0 \equiv -2N |J_1|$ is independent of $\phi$.

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 exchange.

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,\textsuperscript{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\textsuperscript{1} suggest that thermal fluctuations favor the collinear states, defined by $\cos \phi = \pm 1$, whereas quenched fluctuations favor the perpendicular states with $\sin \phi \approx \pm 1$. 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 $h_i = \sum_j J_{ij} s_j$, the local exchange field; in a ground state, $s_i \parallel h_i$. Also, for site $i$ in sublattice $a$, let $\delta h_{ib}^a$ be the component of $h_i$ from sublattice $b$. The nontrivial degeneracy arises because of a cancellation: $\delta h_{ib}^a \equiv 0$ everywhere—classically, at $T = 0$. In the presence of thermal (or quantum) fluctuations, if $\delta h_{ib}^a \ll s_i$, it has no effect on $s_i$ (to lowest order). To maximize the coupling between the fluctuations of the two sublattices, we need rather $\delta h_{ib}^a \gg s_i$. But $\delta h_{ib}^a$ is itself perpendicular to the spins of sublattice $b$. Hence, a collinear alignment is preferred.\textsuperscript{13}

In a diluted lattice, even in the classical ground state, $h_i \neq 0$. Say we remove just one spin from site $j$ in sublattice $b$. For a neighbor $i$ in sublattice $a$, $h_{ib}^a \approx \pm J_{ij} s_i$; then $s_i$ will cant towards this direction by an angle $\delta \theta \approx J_{ij}/J_2$. 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 $\{\theta_i\}$,

$$\delta \mathcal{H}_s = \frac{1}{2} \sum_{ij} A_{ij}(\phi) \delta \theta_i \delta \theta_j,$$

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

$$\delta \mathcal{H}_s = N \int (2\pi)^{-2} d^2q \frac{1}{2} A_q(\phi) |\delta \theta_q|^2,$$

where

$$A_q = 4|J_2| \left(1 - \cos q_x \cos q_y\right) - 2J_1 \left(\cos q_x - \cos q_y\right) \cos \phi.$$

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

$$F(\phi, T) - E_0 = -\frac{1}{2} NT \ln T - NT S_0(\phi),$$

where the "ground-state entropy" is

$$S_0(\phi) = -\int (2\pi)^{-2} d^2q \ln A_q(\phi),$$

$$= \int g_0(J_1 \cos \phi/2J_2),$$

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: $i \hbar \partial \theta_q/\partial t = -\Gamma \Delta \mathcal{H} Q_d \theta_q$—this is plausible for both realizations of the model\textsuperscript{1}—so the spin-wave frequencies are

$$\omega_q(\phi) = \sqrt{g_0(J_1 \cos \phi/2J_2)}.$$

The ground-state energy difference is given by the zero-point term,

$$E(\phi) - E_0 = \frac{1}{2} \int (2\pi)^{-2} d^2q \hbar \omega_q(\phi),$$

$$= 2\hbar (\Gamma J_2)^{1/2} g_{1/2}(J_1 \cos \phi/2J_2),$$

where $g_{1/2}(x) = 0.958 - 0.082x^2 + \cdots$, again favoring the collinear state with $\cos \phi = \pm 1$.\textsuperscript{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.\textsuperscript{2,25} The selecting free energies (4) and (5) have the algebraic structure

$$\int d^2q A_q(\phi),$$

depending on the $\phi$ only through $A_q(\phi)$. For thermal fluctuations $f(x) \propto \ln x$ and for quantum fluctuations $f(x) \propto \sqrt{x}$. If the overall scale of $A_q$ depended on $\phi$, this would control the selection, but in fact its integral is constrained to be $2E_0$ for every $\phi$ value.\textsuperscript{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 $\phi$, we expand about the corresponding ground state up to $O(\delta \theta^2)$:

$$\delta \mathcal{H} = -\Delta \cos \phi - J_1 \sin \phi \sum \gamma_q \delta \theta_q + p^2 \delta \mathcal{H}_s.$$

A constant has been dropped. Here $\gamma_q$ is the Fourier transform of $\gamma_i = \sum \gamma_i(-1)^{j_i+j_y}$ (sum over occupied neighbors of $i$) and $\Delta \equiv J_1 \sum \gamma_i(-1)^{j_i+j_y}$ (sum over occupied neighbor pairs). These $\phi$-dependent constant and linear terms are zero on average, with

$$\langle \Delta \phi^2 \rangle \approx (\delta p)^2 N,$$

and

$$\langle |\gamma_q|^2 \rangle \approx 4 \delta p (\cos q_x - \cos q_y)^2$$

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

\[
\langle \delta H(\phi) \rangle \approx - (J_1^2/2J_2) \delta p G_1 \sin^2 \phi,
\]

where

\[
G_1 \equiv \int (2\pi)^{-2} d^2 q \frac{(\cos q_x - \cos q_y)^2}{1 - \cos q_x \cos q_y} \approx 0.727,
\]

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 \(H\) term in (6) can couple
to the degeneracy degree of freedom \(\phi\) exactly as a random field
couples to the spin direction. However, unlike a true random field,
its exchange fields, 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
\((\text{Cd}_1-\text{Mn}_p)\text{Te}\) 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 anticonical state, the
REF’s do not couple at all. \(^{28}\) On the other hand, \(\hat{H}\) 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\(_1\) 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 anticonical order are characterized respectively by order parameters

\[
\tilde{M}_i \equiv \sum_{ij} (-1)^{\nu_i - \nu_j} \cos (\theta_i - \theta_j)
\]

and

\[
\tilde{M}_\perp \equiv \sum_{i} (-1)^{\nu_i} \sin (\theta_i - \theta_j).
\]

The pure system has the order parameter symmetry \([Z_2 \otimes \text{SO}(2)]\) 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 \(\tilde{\chi}_1\) corre-
sponding to \(M_1\) and the specific heat \(C(T)\) should show
Ising-type critical divergences while the staggered magn-
etization correlations should show Kosterlitz-Thouless
essential singularities. \(^{10(b)}\)

The transition \(T_N(p)\) between the collinear and
anticonical states has the form \(T_N(p) \equiv \text{const} \times J_1 \delta p\),
since the competing selection terms (4) and (9) are, respectively, linear in \(T\) and \(\delta p\). Long-range anticonical order at \(T=0\) persists for \(p < 1\),
down to \(p_c\) which should be slightly above the site percolation
threshold \((\approx 0.59)\) 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 \(p_c < p < p_c^t\), Villain’s picture\(^4\) suggests the ground state is a spin glass.

Monte Carlo simulations\(^{21}\) (for \(J_1=1, J_2=-1, 1\)) with
\(N \approx 50^2\) spins) confirm the expected features: Collinear
ordering of the pure system occurs, at \(T_N(1)/J_2 \approx 0.97 ± 0.02\), with \(T_N(p) \approx 5.5J_2 \delta p\,\), and \(p_c^t \approx 0.6\). This goes
beyond the analytic results, which were valid only for small \(\delta p\) 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 col-
linearity, dilution selection favoring anticollinearity, and
random exchange fields which tend to disorder the col-
linear selected states. Qualitatively similar behavior is
expected in most such systems; analogous calculations will be published elsewhere. However, since the discrete symmetry differs from case to case, the critical behaviors and random-field responses may be quite different. I would like to thank D. S. Fisher, H. Kawamura, B. E. Larson, A. T. Ogielski, and especially J. F. Fernandez and S. Prakash for useful discussion. This work was supported by DOE Grant No. DE-FG02-87ER-45323.

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21C. L. Henley and S. Prakash (to be published); S. Prakash, C. L. Henley, and J. F. Fernandez (unpublished).
22J. F. Fernandez has independently conceived the same model.
23P. Chandra and P. Coleman (unpublished).
24Indeed, the spin-1/2 fcc antiferromagnet (Ref. 5) and the fully frustrated cubic lattice (Ref. 16) show collinear selection.
25Generally, φ becomes an abstract label for a ground-state manifold which often has more than one dimension of degeneracy degrees of freedom.
28In other systems, REF's will couple to the anticoUinear order parameter if the discrete part of the broken symmetry is associated with a direction in real space.