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Random-field effects in antiferromagnets with classically degenerate ground states

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Abstract. We consider the influence of impurities on the ground-state selection in vector spin systems with non-trivial degeneracy of classical ground states. The main result is that, in the case of body central tetragonal antiferromagnets, random fields generated by defects can destroy long-range magnetic order.

In some random crystals the classical ground state is infinitely degenerate, that degeneracy being not the consequence of the trivial rotation invariance but the result of an additional symmetry caused by competition between exchange interactions. Degenerate states form a continuous manifold in the phase space of the system.

The simplest example of systems of that kind are magnets which can be decoupled into interpenetrating antiferromagnetic subsystems in such a way that the effective fields produced by spins of one subsystem at any spin of the other subsystem cancel. Then the subsystems, when treated classically, turned out to be decoupled in the ground-state configuration; a rigid rotation of the spins of a single subsystem does not change the energy. For example, the type 2 antiferromagnetic structure in the BCC lattice as well as the BCT lattice (figure 1) can be decomposed into two subsystems of that kind.

The structure shown in figure 1 is stable if $|J_2/J_1| < 1; J_1, J_3 < 0$, where J_1 and J_3 are intrasubsystem coupling constants and J_2 stands for the interaction between spins of

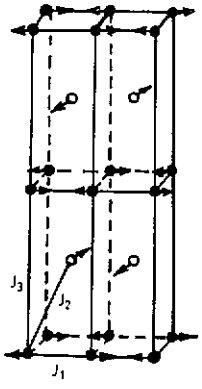

Figure 1. The ground-state spin arrangement in the BCT antiferromagnet when $J_3 < 0$. The spin systems in the sites denoted by full and open circles are decoupled in the classical treatment.

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different subsystems. Such a structure can be considered as a set of antiferromagnetically ordered planes, coupled in the classical treatment only by the J_3 interaction. The planes form two interpenetrating simple tetragonal antiferromagnets. This model describes the magnetism of lanthanum cuprate when a small orthorhombic distortion is neglected and is also relevant to such materials as K_2NiF_4 [1].

If J_3 is negligibly small, the staggered magnetizations of planes are independent in the mean-field approximation, i.e. we have an infinite number of decoupled subsystems (it should be noted that the degenerate helical states in the BCT lattice, studied in [2], are only a part of the whole degeneracy manifold).

The existence of a non-trivial ground-state degeneracy does not necessarily mean that a system can be divided into several decoupled parts. We shall also study below the infinitely degenerate helices which are classical ground states for the rhombohedral antiferromagnets [3, 4].

In all these systems, quantum spin fluctuations remove the classical degeneracy and select a ground state [4, 5] (the contribution to this effect due to thermal fluctuations was considered in [6]). The mechanism was convincingly demonstrated by an inelastic neutron scattering experiment [7] where a gap in the 'phason' mode spectrum induced by quantum breaking of the degeneracy [5, 8] was observed.

It is clear that quenched disorder which locally violates a balance between competing interactions can lift the degeneracy as well. We would like to emphasize that even small amounts of defects could be responsible for ground-state selection because the spin-fluctuation corrections to the ground-state energy prove to be small at any values of the parameters inside the classical zero-temperature stability region of degenerate phases [4, 5].

The main problem considered in this paper is the following: does a random effective interaction induced by impurities destroy the long-range magnetic order, or do defects select a ground state, without destroying order? We show that the behaviour of systems considered depends on the type of disorder, the properties of the degeneracy manifold, and the space dimension. Bond disorder destroys the long-range ordering provided that the degree of ground-state degeneracy is sufficiently high or the system is two dimensional. As for site disorder, in a linear approximation of the concentration of impurities the magnetic order is preserved and one of the states from the degeneracy manifold is selected as the ground state. For the specific case of the site-diluted two-dimensional lattice, our result agrees with that obtained earlier by Henley [6].

Let us consider a system of classical three-component vectors S_i coupled via an isotropic exchange interaction. The ground-state spin configuration in a pure crystal is supposed to be planar (in the x–y plane). To find the ground state of a system with defects we follow [6] and expand the system energy $E = -\sum_{i,j} J_{ij} S_i \cdot S_j$ up to second order with respect to out-of-plane (angle θ_i) and in-plane (angle φ_i) deviations in the orientation of the spin at site i. We assume these deviations to be small and get

$$E = E_0 + E_\theta + E_\varphi \qquad E_0 = -S^2 \sum_{i,j} J_{ij} \cos(\Delta\Phi_{ij}) \tag{1a}$$

$$E_\theta = -S^2 \sum_{i,j} J_{i,j}[\theta_i\theta_j - \theta_i^2 \cos(\Delta\Phi_{ij})] \tag{1b}$$

$$E_\varphi = -S^2 \left(\tfrac{1}{2}\sum_{i,j} A_{ij}\varphi_i\varphi_j - \sum_i h_i\varphi_i\right) \tag{1c}$$

$$A_{ij} = J_{ij}\cos(\Delta\Phi_{ij}) - \delta_{ij}\sum_k J_{ik}\cos(\Delta\Phi_{ik})$$

$$h_i = \sum_j J_{ij}\sin(\Delta\Phi_{ij}) \qquad \Delta\Phi_{ij} = \Phi_i - \Phi_j. \tag{1d}$$

The set of azimuthal angles $\{\Phi_i\}$ of spins determines a specific state, chosen as the ground state in the ideal crystal. Then we shall calculate the impurity correction to the energy and minimize it over all sets of $\{\Phi_i\}$.

In order for the deviations θ_i and φ_i to be small, all local perturbations of exchange energies should be small as well.

We consider two different kinds of disorder. First of all, we consider a bond disorder, when all exchange energies fluctuate about their ideal crystal values $J_{ij}^{(0)}$ independently according to a probability distribution with a small variance $(V^2)_{ij} \ll (J_{ij}^{(0)})^2$. The bond disorder can easily arise in a real material as a result of local deformations or disorder in an arrangement of non-magnetic atoms mediating the superexchange interaction. Another type of randomness arises when some of the host spins are substituted by impurity spins. It can be simulated by changing all bonds connected to an impurity spin located at a site i: $J_{ij} = J_{ij}^{(0)}(1-\alpha)$. To make all local perturbations small, we assume that $\alpha \ll 1$ and that the impurity concentration x is small as well.

Equations (1*a*)–(1*c*) demonstrate that up to second-order in-plane deviations φ_i are decoupled from out-of-plane deviations θ_i. A disorder does not produce terms linear in θ_i in the expression for the energy but only slightly changes coefficients of the positive definite quadratic form E_θ. Hence $\theta_i = 0$ for any i in a ground state, i.e. the spin system remains coplanar under the weak disorder considered here.

In contrast, when we consider the change in the energy produced by in-plane deviations φ_i, we see that there is linear coupling between φ_i and the local random fields h_i. Minimizing the corresponding energy E_φ we find that the in-plane deviation in a site i is equal to $\varphi_i = \Sigma_j h_j (A^{-1})_{ij}$. Let us stress that the non-vanishing values of all random fields h_i are a purely disorder-induced effect caused by fluctuations in J_{ij}. Therefore, it is evident that, in order to calculate ground-state energy corrections $\delta E = \overline{E_\varphi}$ and the mean square $\overline{\varphi_i^2}$ (the bar denotes an averaging over spatial disorder) to the lowest order in small random perturbations of coupling constants, we can neglect small fluctuations in A_{ij} caused by defects. These fluctuations are irrelevant and could not induce any breaking of the long-range magnetic order. Going into momentum space we find that

$$\delta E = \tfrac{1}{2}S^2\frac{\overline{h_q h_{-q}}}{A_q} \qquad \overline{\varphi_i^2} = \sum_q \frac{\overline{h_q h_{-q}}}{A_q^2}. \tag{2}$$

Here h_q and A_q are the Fourier transforms of the corresponding quantities.

The correlation function of random fields h_i can be easily calculated using the expressions for coupling constants J_{ij} perturbed by impurities presented above. We get

$$|h_q|^2 = \begin{cases} \alpha^2 x|W_q|^2 \\ P_0 - P_q \end{cases} \qquad \text{for} \begin{cases} \text{site disorder} \\ \text{bond disorder} \end{cases} \tag{3}$$

where W_q and P_q are Fourier transforms of $J_{ij}\sin(\Delta\Phi_{ij})$ and $(V^2)_{ij}\sin^2(\Delta\Phi_{ij})$, respectively.

Let us now consider the BCT antiferromagnet. Taking $J_2 < 0$ for definiteness we see from equations (1*d*) and (3) that, at small momentum q,

$$|h_q|^2 = \begin{cases} \alpha^2 J_2^2 x \sin^2 \Phi\, q_x^2 q_y^2 q_z^2 \\ V^2 \sin^2 \Phi\, q^2 \end{cases} \qquad \text{for} \begin{cases} \text{site disorder} \\ \text{bond disorder} \end{cases} \tag{4}$$

$$A_q \sim (q_\perp a)^2 + j_3 (q_z c)^2 \qquad j_3 = J_3/J_1. \tag{5}$$

Here a and c are lattice constants in the plane and in the z direction, respectively, $q_\perp$ is the momentum component in the x–y plane, Φ is an angle between staggered magnetizations of subsystems and V is a variance of the J_2 interaction.

We see from equations (2), (4) and (5) that, for site disorder, the integrals (2) turn out to be convergent irrespective of the value of j_3. The correction to the energy is $\delta E \sim -\sin^2 \Phi$, which means that the 'orthogonal' state $\Phi = \pm \pi/2$ is preferred here as it was for the two-dimensional model studied in [6]. The value of $\overline{\varphi_i^2}$ which gives us the perturbation theory parameter is $\overline{\varphi_i^2} \sim \alpha^2 x j_2^2$ where $j_2 = J_2/J_1$. Hence, when the disorder is small, the perturbation theory is valid, i.e. all local spin deviations from the Néel ground state are small.

For bond dilution the situation is quite different: since $|h_q|^2 \sim q^2$ at small q, the integral for $\overline{\varphi_i^2}$ shows a power-like divergence at small transversal momentum $q_\perp$ if $j_3 = 0$. If j_3 is finite but small we have

$$\overline{\varphi_i^2} \sim V^2/J_1^2 |j_3|. \tag{6}$$

This means that spin deviations from the Néel ground state are large even at weak disorder if j_3 is sufficiently small. The divergence at $j_3 = 0$ seems to be similar to that found by Imry and Ma [9] in the random-field problem, and it is natural to think that at $j_3 = 0$ the long-range order is unstable at any amount of disorder. The j_3 interaction tends to stabilize the long-range order in a way similar to the effect of a uniform external magnetic field in the random-field problem.

The ground-state instability can be understood qualitatively in the following way. As was discussed above, if $j_3 = 0$ we have independent Néel antiferromagnets in planes perpendicular to the z axis. If we introduce a bond disorder retaining the Néel ordering of planes, we would not change the total energy since $\overline{J_{ij} - J_{ij}^{(0)}} = 0$. However, suppose that we allow spins to deviate from their ground-state configuration in the ideal crystal, with the angle between spins in a plane being of the order of π at a distance L. Let us consider a region of a plane with a characteristic size L and its interaction with an adjacent plane. Because of the non-collinear orientation of spins within each plane we have lost an energy of the order of J_1 per region. Similarly to the Imry–Ma [9] problem, the energy gain is proportional to the square root of the number of sites located within the region considered, i.e. the gain is of the order of VL. Minimizing the energy per site with respect to L, we find that $L \sim J_1/V$ and the resulting energy gain per site due to the instability is of the order of $V^2/|J_1|$. The same estimation of L can be obtained by cutting the integration over $q_\perp$ in equation (2) off at the lower limit $q_\perp \sim L^{-1}$ and equating $\overline{\varphi_i^2}$ to unity. The interaction J_3 stabilizes the magnetic order when it exceeds the energy gain V^2/J_1. We see that the same estimation follows from equation (6).

This qualitative picture permits us to understand why site defects do not destroy the magnetic ordering in the linear approximation in x. The main difference between site and bond disorder effects stems from the fact that an isolated site defect does not change the energy if spins in each of adjacent planes remain in the Néel state, while a bond

defect does change the energy. So, for isolated site defects the gain of energy in the non-collinear state is not VL, but it is proportional to a smaller power of the correlation length L. As a result, for a site-disordered system the energy of a random non-uniform state is always higher than the energy of the ordered system, which is therefore stable. The difference between effects produced by site and bond dilution in Ising systems noted in [10] can be understood in the same way.

As it was pointed out above, quantum fluctuations of spins always remove the degeneracy of a classical ground state. In particular, for the case of a BCT antiferromagnet, quantum corrections remove the soft line in the spectrum of magnons even at $j_3 = 0$ [11]. That cuts the divergency of the corresponding intergral for $\overline{\phi_i^2}$, with a combination $C_0|j_2|/\sqrt{S}$, $C_0 \approx 0.13$ [11], playing effectively the role of $|j_3|$ when estimating $\overline{\phi_i^2}$ (see (6)). Taking into account that in the case of a bond dilution, $V^2 \sim J_2^2$, we conclude that quantum fluctuations can stabilize long-range magnetic order in diluted BCT antiferromagnets when $j_2 = J_2/J_1$ is sufficiently small: $|j_2| < C_0/\sqrt{S}$.

Results for the type 2 antiferromagnetic structure can be obtained from equations (5) and (6) by putting $J_3 = J_1$. We see that both types of disorder select the same 'orthogonal' state with $\Phi = \pi/2$, which has been found in [6] for a site-diluted square lattice with sufficiently strong next-nearest-neighbour exchange interaction. It is easy to see that, if bond disorder is introduced in the lattice considered in [6], it would cause instability in the magnetic order.

We now turn our attention to the rhombohedral antiferromagnets. As was shown in [3], if $j = |J_2/J_1| < 3$ ($J_1 < 0$ and J_2 are the nearest-neighbour in-plane and out-of-plane exchange interaction energies), the classical ground states are degenerate helices with wavevectors $\boldsymbol{Q}$ forming a line $L_{\boldsymbol{Q}}$ in the momentum space given at $j \ll 1$ by the equations

$$\begin{aligned} Q_x a &= 2\pi/3 + (1/\sqrt{3})j\cos(Q_z c) + (1/6\sqrt{3})j^2\cos(2Q_z c) \\ (\sqrt{3}/2)Q_y a &= j\sin(Q_z c) - \tfrac{1}{8}j^2\sin(2Q_z c). \end{aligned} \tag{7}$$

Here a and c are the lattice distances in the x–y plane and in the z direction. The axis OX is directed along an elementary translation vector of the in-plane triangular lattice.

For a helix ground state, $\Delta\Phi_{ij} = \boldsymbol{Q}\cdot(\boldsymbol{r}_i - \boldsymbol{r}_j)$ and we get

$$A_q = \tfrac{1}{2}(J_{Q+q} + J_{Q-q} - 2J_Q) \tag{8}$$

$$\overline{|h_q|^2} = \begin{cases} \tfrac{1}{4}\alpha^2 x(J_{Q+q} - J_{Q-q})^2 \\ (V^2)_0 - (V^2)_q + \tfrac{1}{2}[(V^2)_{2Q+q} + (V^2)_{2Q-q} - 2(V^2)_{2Q}] \end{cases} \quad \text{for} \begin{cases} \text{site disorder} \\ \text{bond disorder} \end{cases} \tag{9}$$

$$J_q = 2J_1[\cos(q_x a) + 2\cos(\tfrac{1}{2}q_x a)\cos[(\sqrt{3}/2)q_y a] + j\{\cos\{\tfrac{1}{3}q_z c + (\sqrt{3}/2)q_y a] + 2\cos(\tfrac{1}{2}q_x a)\cos[\tfrac{1}{3}q_z c - (\sqrt{3}/6)q_y a]\}] \tag{10}$$

Using equations (8)–(10) we see that the integral for $\overline{\varphi_i^2}$ is convergent in the case of site disorder. The disorder-induced correction to the ground-state energy is found to be given by the following expression:

$$\begin{aligned} \delta E &= S^2|J_1|\alpha^2 x[a_0 + j^2(a_1 + \cos(2Q_z c)] \\ a_2 &= \frac{24}{\pi^2}\int_0^\pi\int_0^\pi \mathrm{d}x\,\mathrm{d}y\left(\frac{\sin x(\cos x - \cos y)}{3 - \cos x - 2\cos x\cos y}\right)^2 > 0. \end{aligned} \tag{11}$$

It shows that a helix with $Q_z c = \pi/2$ is chosen as the true ground state.

To study the effect of bond disorder we take an axis OZ in momentum space to be collinear to the tangential direction to the line L_Q at a point $Q = Q_0$ determining the wavevector of that helix structure, the stability of which we would like to investigate. As J_q has minima at $q \in L_q$, the quantities J_{Q_0+q} and A_{Q_0+q} are effectively two dimensional at small q:

$$A_{Q_0+q} = \lambda_x q_x^2 + \lambda_y q_y^2 + O(q^3). \tag{12}$$

As to the bond disorder random-field correlation function, it is proportional to a sum of the squares of all components of q. So it is easy to check that the integral (2) for $\overline{\varphi_i^2}$ is divergent at the lower limit of integration, demonstrating the instability of the structure.

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