Ordering due to disorder in a triangular Heisenberg antiferromagnet with exchange anisotropy

1992 J. Phys.: Condens. Matter 4 2937

(http://iopscience.iop.org/0953-8984/4/11/020)

J. Phys.: Condens. Matter 4 (1992) 2937–2959. Printed in the UK

Ordering due to disorder in a triangular Heisenberg antiferromagnet with exchange anistropy

Qing Sheng and Christopher L Henley
Department of Physics, Cornell University, Ithaca, NY 14853, USA

Abstract. We discuss the effects of thermal fluctuations, quantum fluctuations and quenched fluctuations on a triangular Ising–Heisenberg antiferromagnet. Although thermal fluctuations of the quadratic order (in spin-wave expansions) are found to preserve the continuous ground-state degeneracy, higher-order spin-wave calculations show that thermal fluctuations lift the ground-state degeneracy. Long-wavelength thermal fluctuations are found to be dominant in the thermal selection. Thermal fluctuations and quantum fluctuations select states with different spin directions within the ground-state manifold. We conjecture that this competition leads to a first-order phase transition. Due to the symmetry-breaking fields introduced by the thermal/quantum fluctuations, we expect there to be another Kosterlitz–Thouless (KT) phase transition (T_6) below which the system is locked into one of the discretely selected states, in addition to the two KT transitions (T_{N_1} and T_{N_2}) found by Miyashita and Kawamura. Random site dilutions affect the system the way the discrete random anisotropy field affects an XY ferromagnet. Two KT transitions (T_{N_1} and T_6) are destroyed as real phase transitions at the appearance of the random site dilutions, but T_{N_2} is unaffected.

1. Introduction

Frustrations in vector-spin systems often lead to non-trivial continuous degeneracy in the ground states: *non-trivial* in that the Hamiltonian of the system does not bear the same symmetries as the ground-state spin configurations. Because of this, the non-trivial degeneracy of the ground states does not usually persist when we consider perturbations that involve higher states, such as (i) non-zero temperatures, (ii) quantum zero-point fluctuations or (iii) quenched randomness.

At finite temperatures, the macroscopically stable submanifold of the ground state manifold is the one with the higher density of excited states, because its larger entropy reduces the free energy. This is the thermal selection effect. Secondly, since quantum-spin components do not commute, the classical ground-state spin configurations are subject to the selection of quantum fluctuations. Thirdly, quenched fluctuations (random site/bond dilutions) reduce the non-trivial continuous degeneracy to discrete degeneracies; in addition to a selection effect, quenched fluctuations usually introduce random exchange fields into the system which may or may not destroy the selected order depending on models [2–4]. All these effects are due to the fluctuations, and are thus called *ordering due to disorder* [5, 6]. Usually it is the case that quantum and thermal fluctuations select the same subset of the ground states†, while quenched fluctuations tend to select a different subset [6, 2, 3].

† Competing quantum and thermal selections were found in another system [7] where the selection is from a continuous manifold of wavevectors, instead of spin directions.

0953-8984/92/112937+23$04.50

It is the behaviour of the system at the low-temperature limit that we are interested in, since the fluctuations at low temperature determine which subsets of the ground states are stabilized (thus selected). Since low-temperature expansions are used in this approach, the results are, strictly speaking, valid only for $T \ll J$ (where J is the energy scale of the interactions in the system). However, they guide us to the correct symmetry for labelling the states of the low-temperature phases. This suggests the universality class of the transitions between these phases and less-ordered ones at higher temperature. Furthermore, the low-temperature expansions suggest how the coefficients of the coarse-grained degrees of freedom in the free energy depend on parameters, such as the temperature T or the dilution δp.

In this paper, we study the anisotropic Heisenberg model of AFT (antiferromagnets on the triangular lattice)

$$H = \sum_{\langle ij \rangle} J_x S_{ix} S_{jx} + J_y S_{iy} S_{jy} + J_z S_{iz} S_{jz} = J \sum_{\langle ij \rangle} A \sin\theta_i \sin\theta_j \cos\phi_i \cos\phi_j + (\sin\theta_i \sin\theta_j \sin\phi_i \sin\phi_j + \cos\theta_i \cos\theta_j) \tag{1}$$

where $\langle \cdots \rangle$ means summing over all the nearest-neighbour pairs, $\{\theta_i, \phi_i\}$ are the polar angles of spin i, and

$$\begin{aligned} J_x &= AJ/S^2 \qquad A > 1 \\ J_y &= J_z = J/S^2. \end{aligned} \tag{2}$$

The symmetry of the Hamiltonian (1) is $Z_6 \times S_1$, S_1 being the global rotational symmetry about the easy axis, the x-axis. The only continuous symmetry in the Hamiltonian is the trivial global rotation symmetry about the x-axis, but the ground states are found to have an additional continuous degeneracy [1].

Our interest in this system is that, unlike the previously studied frustrated vector-spin systems, the first-order thermal selection term is still zero. Since the ground-state degeneracy of the AFT Ising–Heisenberg model is not the real symmetry of the Hamiltonian, we still expect the degeneracy to be lifted by fluctuations in a proper higher-order calculation. The aim of this paper is to compute all three kinds of selections (previously mentioned) and to use the information on the symmetry of the selected states to suggest the phase diagram as a function of four parameters: spin quantum number S, temperature T, anisotropy A and dilution δp.

In the limit $A = 1$, this model becomes the isotropic AFT Heisenberg model which has an order parameter space of $SO(3) = P_3$ [1, 8]. In the AFT Ising–Heisenberg model, if we stretch the usual notion of order parameter space so as to label the degenerate classical ground states, the order parameter space is $S_1 \times S_1$ (see section 2.1). So the order parameter space of the AFT Ising–Heisenberg model has a different topology from that of the AFT isotropic Heisenberg model. Also, the ground-state degeneracy of the isotropic model is the true symmetry of its Hamiltonian, so the degeneracy still exists when fluctuations are included. In contrast, the real symmetry of the anisotropic model is only $Z_6 \times S_1$, so the degeneracy will be lifted by fluctuations.

Since the parameter spaces of the two models have different topologies, the ordering processes at low-temperature regions are expected to be different. In the isotropic AFT Heisenberg model, the phase transition has been characterized by

binding–unbinding of Z_2-vortex pairs similar to the Kosterlitz–Thouless (KT) type phase transition [8]; and the spin correlation function decays exponentially at any finite temperature ([8] and references therein). However in the AFT Ising–Heisenberg model, the ordering process is more sophisticated. From simulations of this model, Miyashita and Kawamura [1] inferred that there exist *two* phase transitions in the temperature region of their Monte Carlo simulations: they identify the first transition (T_{N_1}) as the ordering of components of spins along the easy axis; and the second transition (T_{N_2}) as a KT-like ordering of the components perpendicular to the easy axis. (This successive ordering process has been observed experimentally in a variety of two- and three-dimensional Ising–Heisenberg AFT, such as $CsNiX_3$(X≡Cl, Br) [9], $LiNiO_2$ [10].) As they kept lowering the temperature below the second transition point, they observed the recovery of the non-trivial symmetry. This seemed consistent with their analytical result that this system does not show any symmetry-breaking at the order of harmonic spin-wave excitations. However, since the Hamiltonian has the discrete Z_6 symmetry, we expect that the continuous degeneracy of the classical ground states will be lifted by fluctuations.

The outline of this paper is as follows: Following [1] section 2 sets up the ground-state manifold and the expansion in linearized modes which is used in the rest of the paper. Section 3 shows that thermal fluctuation selection does indeed appear at higher orders of spin-wave excitations; since the naive second-order term is divergent, this necessitates a self-consistent cutoff. In section 4, we show that the quantum fluctuations also lift the ground-state degeneracies. The quantum fluctuations select different discrete sets of the ground states from the ones selected by thermal fluctuations. In section 5, we show that the random site dilutions introduce a random uniaxial anisotropy field into the system, which does not select any particular discrete subsets in the ground-state manifold. In section 6, all the competing terms are combined in a free energy which is used to suggest the form of the phase diagram as a function of S, T, A and δp. Among other things, we find that the pure system ought to have an additional transition not mentioned by Miyashita and Kawamura (section 6.1); and that in the diluted system, one transition (of KT type) is not destroyed even though it occurs in a background of other degrees of freedom whose transition is destroyed by random-field-like effects (section 6.2). We summarize the results in section 7.

2. Ground state and harmonic excitations

2.1. Non-trivial degeneracy in ground-state manifold

In the ground state the triangular lattice breaks up into three sublattices; within each sublattice, the spins are ordered ferromagnetically and can be described by two polar angles $\{\phi_s, \theta_s\}$ ($s = 1, 2, 3$ sublattice). For the case of $A < 1$, the spins are all in the yz plane and form a '120° structure' (the spins on three sublattices form 120° angles with each other), in which case the ground state has only the trivial degeneracy associated with the global rotation and reflection of the 120° structure. For the case of $A > 1$, the x-axis becomes an *easy axis*. If we had an antiferromagnet on a bipartite lattice (no frustration), the ground state for $A > 1$ would have only discrete symmetry—spins on two sublattices (aligned in opposite directions) lie along the x-axis. However, for the triangular lattice, the inherent frustration makes it impossible for all the nearest-neighbour spins to align in opposite directions. The

partial resolution of the frustration is that the spins on the three sublattices lie in a plane including the easy axis (x-axis). Surprisingly spins can lie in any direction in the plane as long as the directions of the spins in the three sublattices satisfy certain relations, as has been noted by Miyashita and Kawamura [1].

Without lose of generality, we assume spins lie in the xy plane, i.e. $\theta_1 = \theta_2 = \theta_3 = \pi/2$. We obtain ϕ_1, ϕ_2 and ϕ_3 by solving the equations

$$\frac{\partial}{\partial \phi_i} H(\phi_1, \phi_2, \phi_3) = 0 \tag{3}$$

where $i = 1, 2$ or 3,

$$\begin{cases} \cos\phi_1(\sin\phi_2 + \sin\phi_3) = A\sin\phi_1(\cos\phi_2 + \cos\phi_3) \\ \cos\phi_2(\sin\phi_3 + \sin\phi_1) = A\sin\phi_2(\cos\phi_3 + \cos\phi_1) \\ \cos\phi_3(\sin\phi_1 + \sin\phi_2) = A\sin\phi_3(\cos\phi_1 + \cos\phi_2). \end{cases} \tag{4}$$

There exists Z_6 symmetry in the solutions to these equations: if $v_1 = \{\zeta_1, \zeta_2, \zeta_3\}$ is a set of solution, then

$$\begin{cases} v_2 = \{\zeta_2, \zeta_3, \zeta_1\} \\ v_3 = \{\zeta_3, \zeta_1, \zeta_2\} \\ v_4 = \{\pi - \zeta_1, \pi - \zeta_2, \pi - \zeta_3\} \\ v_5 = \{\pi - \zeta_2, \pi - \zeta_3, \pi - \zeta_1\} \\ v_6 = \{\pi - \zeta_3, \pi - \zeta_1, \pi - \zeta_2\} \end{cases} \tag{5}$$

are equivalent sets of the solutions.

It can be shown that equations (4) lead to the following equalities:

$$\begin{cases} \cos\phi_1\cos\phi_2 + A\sin\phi_1\sin\phi_2 = -A/(A+1) \\ \cos\phi_2\cos\phi_3 + A\sin\phi_2\sin\phi_3 = -A/(A+1) \\ \cos\phi_3\cos\phi_1 + A\sin\phi_3\sin\phi_1 = -A/(A+1). \end{cases} \tag{6}$$

Combining the first and second equations in (6), we get

$$\cos\phi_2(\cos\phi_1 - \cos\phi_3)\left(1 + A\frac{\sin\phi_2}{\cos\phi_2}\frac{\sin\phi_1 - \sin\phi_3}{\cos\phi_1 - \cos\phi_3}\right) = 0 \tag{7}$$

on the other hand, the second equation in (4) gives

$$\frac{\sin\phi_2}{\cos\phi_2} = \frac{\sin\phi_1 + \sin\phi_3}{A(\cos\phi_1 + \cos\phi_3)}. \tag{8}$$

Substituting equation (8) into (7), we get the identity

$$\cos\phi_2(\cos\phi_1 - \cos\phi_3)\left(1 + \frac{\sin^2\phi_1 - \sin^2\phi_3}{\cos^2\phi_1 - \cos^2\phi_3}\right) = 0. \tag{9}$$

Therefore the three equations in (4) (or (6)) are not independent of each other. There is a continuous set of solutions to the equations parameterized by ϕ_1:

$$\begin{cases} \phi_2 = \arctan(A\tan\phi_1) + \arccos\left(-\dfrac{A}{(1+A)\sqrt{\cos^2\phi_1 + A^2\sin^2\phi_1}}\right) \\ \phi_3 = \arctan(A\tan\phi_1) - \arccos\left(-\dfrac{A}{(1+A)\sqrt{\cos^2\phi_1 + A^2\sin^2\phi_1}}\right) \end{cases} \tag{10}$$

where $\phi_1 \in [0, \phi_0]$ ($\cos\phi_0 \equiv A/(A+1)$), and the other solutions are related to (10) by the six-fold symmetry (5). So the ground state has a non-trivial continuous degeneracy S_1 (parameterized by ϕ_1). On the other hand, the ground state has a trivial continuous degeneracy associated with global rotation of the spins about the easy axis. Therefore the ground state has $S_1 \times S_1$ symmetry, and the ground-state energy is

$$E_0/J = -(A^2 + A + 1)/(A+1). \tag{11}$$

Alternatively, the non-trivial degeneracy can be parameterized in a more symmetric way by the parameter ψ defined by

$$\psi \equiv (\phi_1 + \phi_2 + \phi_3)/3. \tag{12}$$

Then $\psi \to \psi + \pi/3$ or $\psi \to -\psi$ is a symmetry operation. There are two *special* kinds of states (figure 1):

type I, the spins on one of the sublattices lie along the easy axis in which case $\psi \in \{0, \pm\pi/3, \pm 2\pi/3, \pi\}$;

type II, the spins on one of the sublattices lie perpendicular to the easy axis (we call these *loose* spins: in the limit of $A \to \infty$ the system does not care very much about which direction these spins point); in this case $\psi \in \{\pm\pi/6, \pm\pi/2, \pm 5\pi/6\}$.

2.2. *Expansion in harmonic excitations*

At finite temperatures, the spin configurations $\{\tilde\theta_i, \tilde\phi_i\}$ will deviate randomly from the ground-state configurations due to thermal fluctuations. Express the spin deviations from the ground-state configuration $\{\theta_i, \phi_i\}$ as $\{\delta\theta_i = \tilde\theta_i - \theta_i, \delta\phi_i = \tilde\phi_i - \phi_i\}$. Then the Hamiltonian (1) can be expanded in a power series of the spin deviations as

$$H = E_0 + H_2 + H_3 + H_4 + \cdots \tag{13}$$

where H_n denotes the nth order in the expansion. Since we are expanding about the ground state, the linear terms should vanish. In reciprocal space,

$$H_n = \frac{1}{n!}\prod_{i=1}^{n}\sum_{q_i\in \mathrm{BZ}}\sum_{\xi=\theta,\phi} \delta\xi_{s_1}(q_1)\cdots\delta\xi_{s_n}(q_n)\frac{\partial}{\partial\xi_{s_1}(q_1)}\cdots\frac{\partial}{\partial\xi_{s_n}(q_n)}H \tag{14}$$

where the repeated indices are summed over (we will keep this convention in the rest of the paper), BZ is the first Brillouin zone of the triangular lattice with an edge of

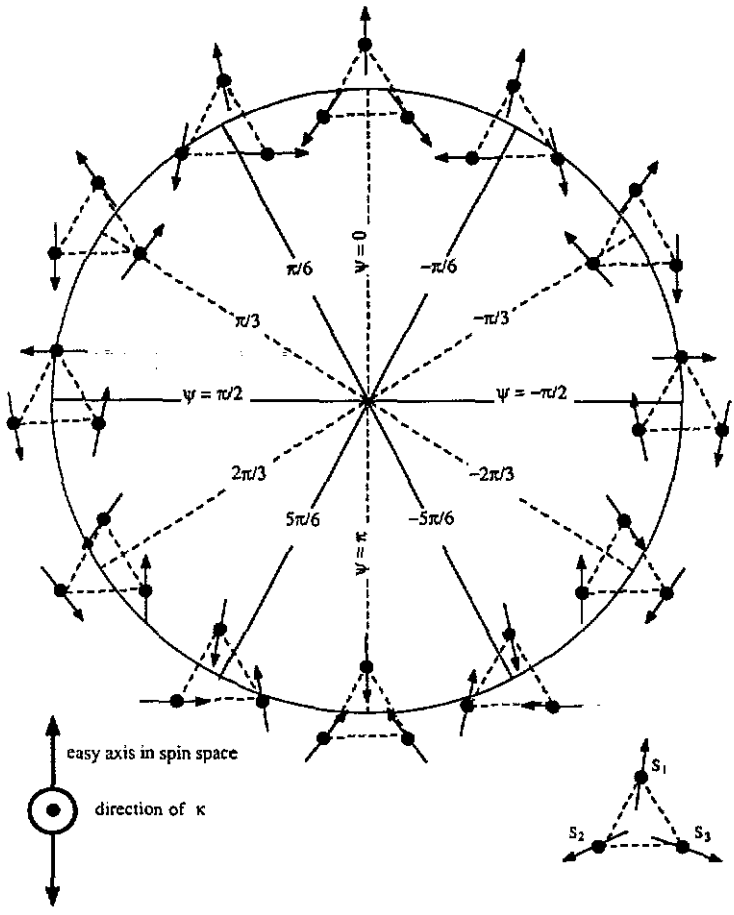

Figure 1. The ground states parametrized by ψ. The spins are in the 'distorted 120°' structure, where the spins are bent towards the easy axis compared with the isotropic 120° structure. The 'chiral' order parameter κ (defined in section 6) is pointing perpendicular out of the plane of the paper. States on broken (full) lines are related to each other by the Z_6 symmetry. The states on the broken lines are type I, and the states on the full lines are type II.

length $\sqrt{3}$, and $\xi_s(q)$ are $\theta_s(q)$ or $\phi_s(q)$ (s=1, 2, 3 sublattices). The derivative in the reciprocal space is related to that in the real space by

$$\frac{\partial}{\partial \xi_s(q)} = \sum_{i \in s} e^{iq \cdot r_i} \frac{\partial}{\partial \xi_i} \tag{15}$$

where the sum is over all the spins on sublattice s. Also

$$\delta \xi_s(q) = \sum_{i \in s} e^{-iq \cdot r_i} \delta \xi_i. \tag{16}$$

The quadratic term in the expansion of the Hamiltonian is

$$H_2 = \sum_q \delta \phi_s^*(q) A_{ss'}(q) \delta \phi_{s'}(q) + \sum_q \delta \theta_s^*(q) B_{ss'}(q) \delta \theta_{s'}(q) \tag{17}$$

where $\mathbf{A}(q)$ and $\mathbf{B}(q)$ are 3×3 matrices,

$$\mathbf{A}(q) = \begin{pmatrix} A_{11} & A_{12}\epsilon_q & A_{13}\epsilon_q^* \\ A_{21}\epsilon_q^* & A_{22} & A_{23}\epsilon_q \\ A_{31}\epsilon_q & A_{32}\epsilon_q^* & A_{33} \end{pmatrix} \tag{18}$$

$$\mathbf{B}(q) = \begin{pmatrix} B_{11} & B_{12}\epsilon_q & B_{13}\epsilon_q^* \\ B_{21}\epsilon_q^* & B_{22} & B_{23}\epsilon_q \\ B_{31}\epsilon_q & B_{32}\epsilon_q^* & B_{33} \end{pmatrix} \tag{19}$$

with

$$\begin{cases} A_{11} = B_{11} = -\frac{3}{2}[\sin\phi_1(\sin\phi_2 + \sin\phi_3) + A\cos\phi_1(\cos\phi_2 + \cos\phi_3)] \\ A_{22} = B_{22} = -\frac{3}{2}[\sin\phi_2(\sin\phi_3 + \sin\phi_1) + A\cos\phi_2(\cos\phi_3 + \cos\phi_1)] \\ A_{33} = B_{33} = -\frac{3}{2}[\sin\phi_3(\sin\phi_1 + \sin\phi_2) + A\cos\phi_3(\cos\phi_1 + \cos\phi_2)] \\ A_{12} = A_{21} = \frac{1}{2}(\cos\phi_1\cos\phi_2 + A\sin\phi_1\sin\phi_2) = -A/[2(A+1)] \\ A_{23} = A_{32} = \frac{1}{2}(\cos\phi_2\cos\phi_3 + A\sin\phi_2\sin\phi_3) = -A/[2(A+1)] \\ A_{31} = A_{13} = \frac{1}{2}(\cos\phi_3\cos\phi_1 + A\sin\phi_3\sin\phi_1) = -A/[2(A+1)] \\ B_{12} = B_{21} = B_{23} = B_{32} = B_{31} = B_{13} = \frac{1}{2} \end{cases} \tag{20}$$

and

$$\epsilon_q = \exp[-iq_x] + \exp[i(q_x + \sqrt{3}q_y)/2] + \exp[i(q_x - \sqrt{3}q_y)/2]. \tag{21}$$

It should be noted that due to the continuous degeneracy $S_1 \times S_1$ of the ground state, there are two *zero modes* [11]:

$$\rho(\psi) = \frac{d}{d\psi}\{\phi_1, \phi_2, \phi_3\} \tag{22}$$

corresponds to a mode in which the ground state moves along the ψ *direction* within the ground-state manifold, and $\rho(\psi)$ is the eigenvector of $\mathbf{A}(q = 0)$ at the ground state $\{\phi_1, \phi_2, \phi_3\}$ with zero eigenvalue (at anisotropy $A > 1$, ρ depends on the value of ψ, while for $A = 1$, $\rho(\psi) \equiv \{1, 1, 1\}$); and the eigenvector of $\mathbf{B}(q = 0)$ with zero eigenvalue corresponds to a mode in which the ground state rotates about the x-axis within the ground-state manifold.

The stable ground-state spin configuration at finite temperature is the one that minimizes the free energy

$$\beta F_0 = -\ln\left\{\prod_q \prod_{s,s'} \int d\delta\theta_s(q) d\delta\phi_{s'}(q) \sin\theta_{s'} e^{-\beta H}\right\}$$

$$\simeq \beta N E_0 + N\ln(\pi\beta) + \tfrac{1}{2}\sum_q [\ln\det\mathbf{A}(q) + \ln\det\mathbf{B}(q)] \tag{23}$$

if we only keep the expansion up to H_2. In the frustrated vector-spin systems that have been studied before [6, 12, 13, 3, 2], the free energy F_0 shows a dependence on the ground-state configuration, i.e. the thermal-fluctuation selection effect emerges at

the order of harmonic excitations (which is the last term in (23)). However, in this model, though each of the three eigenvalues $\lambda_\mu^{(A)}(q)$ ($\lambda_\mu^{(B)}(q)$) (where $\mu = 1,2,3$ is the index for eigenvalues) of matrix $\mathbf{A}(q)(\mathbf{B}(q))$ has dependence on ψ, there is a perfect cancellation among the logarithm of the three eigenvalues so that the determinants

$$\det \mathbf{A}(q) = \lambda_1^{(\mathbf{A})}(q)\lambda_2^{(\mathbf{A})}(q)\lambda_3^{(\mathbf{A})}(q)$$

and

$$\det \mathbf{B}(q) = \lambda_1^{(\mathbf{B})}(q)\lambda_2^{(\mathbf{B})}(q)\lambda_3^{(\mathbf{B})}(q)$$

are independent of the value of ψ [1]. As a result, the free energy in (23) is constant for all the ground-state spin configurations, i.e. the non-trivial degeneracy of ground state does not break down in harmonic excitations. In order to see the thermal selection effect, we need to go to higher orders.

3. Higher-order interactions

3.1. Diverging high-order contributions from thermal fluctuations

As we know from the introduction, the non-trivial continuous degeneracy usually does not persist into the non-zero temperatures because thermal fluctuations have the effect of reducing the degeneracy into the discrete symmetry that the Hamiltonian possesses. Usually the thermal fluctuation selection effect takes place at the lowest non-trivial perturbation expansion (quadratic order). This system, however, has an additional 'pseudo-higher symmetry' so that there is no broken symmetry in the harmonic excitations. Based on our belief that there must be a symmetry-breaking in this system, we continued the perturbation expansion of the free energy to the next non-trivial order: $O(T^2)$, i.e. terms from the third and fourth order in the Hamiltonian.

The third term is

$$H_3 = \frac{1}{3!}\sum_{q_1,q_2,q_3}[c^{(1)}(s_1,q_1;s_2,q_2;s_3,q_3)\delta\phi_{s_1}(q_1)\delta\phi_{s_2}(q_2)\delta\phi_{s_3}(q_3) + c^{(2)}(s_1,q_1;s_2,q_2;s_3,q_3)\delta\theta_{s_1}(q_1)\delta\theta_{s_2}(q_2)\delta\phi_{s_3}(q_3) \tag{24}$$

where

$$\begin{cases} c^{(1)}(s_1,q_1;s_2,q_2;s_3,q_3) = \dfrac{\partial}{\partial\phi_{s_1}(q_1)}\dfrac{\partial}{\partial\phi_{s_2}(q_2)}\dfrac{\partial}{\partial\phi_{s_3}(q_3)}H \\ c^{(2)}(s_1,q_1;s_2,q_2;s_3,q_3) = \dfrac{\partial}{\partial\theta_{s_1}(q_1)}\dfrac{\partial}{\partial\theta_{s_2}(q_2)}\dfrac{\partial}{\partial\phi_{s_3}(q_3)}H. \end{cases} \tag{25}$$

The fourth term is

$$\begin{aligned} H_4 = \frac{1}{4!}\sum_{q_1,q_2,q_3,q_4}[&d^{(1)}(s_1,q_1;s_2,q_2;s_3,q_3;s_4,q_4) \\ &\times \delta\phi_{s_1}(q_1)\delta\phi_{s_2}(q_2)\delta\phi_{s_3}(q_3)\delta\phi_{s_4}(q_4) \\ &+ d^{(2)}(s_1,q_1;s_2,q_2;s_3,q_3;s_4,q_4)\delta\phi_{s_1}(q_1)\delta\phi_{s_2}(q_2)\delta\theta_{s_3}(q_3)\delta\theta_{s_4}(q_4) \\ &+ d^{(3)}(s_1,q_1;s_2,q_2;s_3,q_3;s_4,q_4)\delta\theta_{s_1}(q_1)\delta\theta_{s_2}(q_2)\delta\theta_{s_3}(q_3)\delta\theta_{s_4}(q_4)] \end{aligned} \tag{26}$$

where

$$\begin{cases} d^{(1)}(s_1,q_1;s_2,q_2;s_3,q_3;s_4,q_4) = \dfrac{\partial}{\partial\phi_{s_1}(q_1)}\dfrac{\partial}{\partial\phi_{s_2}(q_2)}\dfrac{\partial}{\partial\phi_{s_3}(q_3)}\dfrac{\partial}{\partial\phi_{s_4}(q_4)}H \\ d^{(2)}(s_1,q_1;s_2,q_2;s_3,q_3;s_4,q_4) = \dfrac{\partial}{\partial\phi_{s_1}(q_1)}\dfrac{\partial}{\partial\phi_{s_2}(q_2)}\dfrac{\partial}{\partial\theta_{s_3}(q_3)}\dfrac{\partial}{\partial\theta_{s_4}(q_4)}H \\ d^{(3)}(s_1,q_1;s_2,q_2;s_3,q_3;s_4,q_4) = \dfrac{\partial}{\partial\theta_{s_1}(q_1)}\dfrac{\partial}{\partial\theta_{s_2}(q_2)}\dfrac{\partial}{\partial\theta_{s_3}(q_3)}\dfrac{\partial}{\partial\theta_{s_4}(q_4)}H. \end{cases} \tag{27}$$

In these equations, s_i are sublattice indices 1, 2 and 3.

Treat H_3 and H_4 terms perturbatively,

$$F = F_0 - T\ln[\langle e^{-\beta H_3}\rangle_o \langle e^{-\beta H_4}\rangle_o] \tag{28}$$

where F_0 is the free energy to the order of harmonic excitations, $\langle\rangle_o$ is the average over the harmonic excitations:

$$\langle\Theta\rangle_o \equiv \int \prod_q \prod_{s,s'} d\delta\theta_{s'}(q) d\delta\phi_s(q) \sin\theta_s \Theta e^{-\beta H_2} \left(\int \prod_q \prod_{s,s'} d\delta\theta_{s'}(q) d\delta\phi_s(q) \sin\theta_s e^{-\beta H_2} \right)^{-1}. \tag{29}$$

Using the cumulant expansion the free energy is

$$F = F_0 + \langle H_4\rangle_o - \frac{1}{2T}\langle H_3^2\rangle_o + \cdots. \tag{30}$$

The averages over the harmonic excitations are calculated by *pair contractions* [14]:

$$\begin{aligned} &\langle\delta\theta_s(q)\delta\theta_{s'}(q')\rangle_o \equiv (T/2)G^{(\theta)}_{0ss'}(q) \equiv (T/2)[B^{-1}(q)]_{ss'}\delta_{q+q',0} \\ &\langle\delta\theta_s(q)\delta\phi_{s'}(q')\rangle_o = 0 \\ &\langle\sin\theta_s\delta\phi_s(q)\sin\theta_{s'}\delta\phi_{s'}(q')\rangle_o \equiv (T/2)G^{(\phi)}_{0ss'}(q) \equiv (T/2)[A^{-1}(q)]_{ss'}\delta_{q+q',0} \end{aligned} \tag{31}$$

where $G_0^{(\theta,\phi)}(q)$ are the Green functions in the harmonic approximation. The result is

$$\begin{aligned} \langle H_4\rangle_o - \frac{1}{2T}\langle H_3^2\rangle_o = \left(\frac{1}{2}\right)^5 T^2 \int_{BZ} \frac{d^2q_1}{(2\pi)^2}\frac{d^2q_2}{(2\pi)^2} \\ \times \{[A^{-1}_{s_1s_2}(q_1)A^{-1}_{s_3s_4}(q_2)d^{(1)}(s_1,q_1;s_2,-q_1,s_3,q_2;s_4,-q_2) \\ + A^{-1}_{s_1s_2}(q_1)B^{-1}_{s_3s_4}(q_2)d^{(2)}(s_1,q_1;s_2,-q_1,s_3,q_2;s_4,-q_2) \\ + B^{-1}_{s_1s_2}(q_1)B^{-1}_{s_3s_4}(q_2)d^{(3)}(s_1,q_1;s_2,-q_1,s_3,q_2;s_4,-q_2)] \\ - \tfrac{1}{3}[A^{-1}_{s_1s_1'}(q_1)A^{-1}_{s_2s_2'}(q_2)A^{(-1)}_{s_3s_3'}(-q_1-q_2) \\ \times c^{(1)}(s_1,q_1;s_2,q_2;s_3,-q_1-q_2)c^{(1)}(s_1,-q_1;s_2,-q_2;s_3,q_1+q_2) \\ + B^{-1}_{s_1s_1'}(q_1)B^{-1}_{s_2s_2'}(q_2)A^{-1}_{s_3s_3'}(-q_1-q_2) \\ \times c^{(2)}(s_1,q_1;s_2,q_2;s_3,-q_1-q_2)c^{(2)}(s_1,-q_1;s_2,-q_2;s_3,q_1+q_2)]\}. \end{aligned} \tag{32}$$

After the integration over BZ in equation (32), we find numerically that the integrands coming from H_3^2 and H_4 both have singularities at $q \to 0$ that lead to logarithmic divergence and the singular parts do not cancel. The divergence disappears in the limit of $A \to 1$. More specifically, the integrations of ϕ fluctuations (the fluctuations within the plane of the ground-state configuration) diverge logarithmically with the size of the system (infrared divergence); while the integrations of θ fluctuations (the fluctuations out of the plane) do not have singular behaviour. The leading divergent behaviour of (32) is

$$\langle H_4\rangle_o - \frac{1}{2T}\langle H_3^2\rangle_o \sim g(\psi, A)T^2 \int_0^{\Lambda} \frac{dq^2}{Dq^2} \tag{33}$$

where $\det \mathbf{A}(q) = Dq^2 + O(q^4)$, Λ is of order unity and $g(\psi, A)$ has a dependence on ψ as $g_1 + g_2\cos(6\psi)$ with $g_2 > 0$. In the limit of $A \to 1$, $g(\psi, A) \to 0$. So the resultant free energy is divergent but varies with the ground-state configurations.

3.2. Self-energy and self-consistent equation

Though the free energy is divergent, the perturbations around different ground-state spin configurations generate different prefactors ($g(\psi, A)$ in equation (33)) which multiply to a common logarithmically divergent quantity. We therefore interpret this as a sign of this system having thermal selection effect. This degeneracy-breaking leads to 'mass generation' (or *self-energy*) in our theory, i.e. the two-point Green function

$$G(q) = [G_0^{-1}(q) + \Sigma(q)]^{-1} \tag{34}$$

where $G(q), G_0(q)$ and $\Sigma(q)$ are 6×6 matrices. Here $\Sigma(q)$ is the self-energy matrix, the sum of all one-particle irreducible graphs of $G(q)$ [15]. To the order of one-loop, the graphs in figure 2 appear in $\Sigma(q)$.

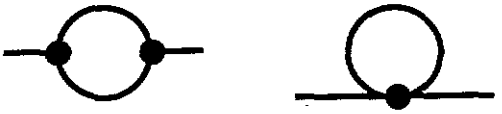

Figure 2. Graphs used to calculate $\Sigma(q)$ to the order of one-loop. Contributions from (*a*) H_3^2 and (*b*) H_4.

Since the θ fluctuations do not have singular behaviour, the major contribution to the thermal selection is from the ϕ fluctuations, i.e. in-plane fluctuations. So this system behaves the same way as an XY system would in the thermal selections. We therefore only concentrate on the in-plane fluctuations.

We can write out the expression for $\Sigma(q)$ from the graphs in figure 2 . The result is

$$\Sigma_{ss'}(q) = \frac{T}{8}\sum_{q'}\left\{G_{s_1s_2}(-q')\frac{\partial}{\partial\phi_s(q)}\frac{\partial}{\partial\phi_{s'}(-q)}\frac{\partial}{\partial\phi_{s_1}(q')}\frac{\partial}{\partial\phi_{s_2}(-q')}H\right.$$

$$-\frac{1}{2}\sum_{q''}\delta_{q+q'+q'',0}G_{s_1s_2}(-q')G_{s_1's_2'}(-q'')$$
$$\times\left[\frac{\partial}{\partial\phi_s(q)}\frac{\partial}{\partial\phi_{s_1}(q')}\frac{\partial}{\partial\phi_{s_1'}(q'')}H\right]$$
$$\times\left[\frac{\partial}{\partial\phi_{s'}(q)}\frac{\partial}{\partial\phi_{s_2}(-q')}\frac{\partial}{\partial\phi_{s_2'}(-q'')}H\right]\Bigg\}. \tag{35}$$

In particular , at $q = 0$,

$$\Sigma_{ss'}(0) = \frac{T}{4}\sum_{q'}\mathrm{Tr}\left\{G(q')\frac{\partial}{\partial\phi_s}\frac{\partial}{\partial\phi_{s'}}A(q')\right.$$
$$\left.-\left[G(q')\frac{\partial}{\partial\phi_s}A(q')\right]\left[G(q')\frac{\partial}{\partial\phi_{s'}}A(q')\right]\right\} \tag{36}$$

where we have dropped superscript ϕ for the two-point Green function. The self-energy is of order $O(T)$, which is quite small in the temperature range in which we are interested. For most of the Brillouin zone, the self-energy matrix is very small compared to $\mathbf{A}(q)$. Only for sufficiently small q such that $\det\mathbf{A}(q) \sim O(T)$ does $\Sigma(q)$ have a dominant role. Therefore, we made an assumption that the self-energy is independent of q, i.e.

$$\Sigma(q) \simeq \Sigma(q=0) \equiv \Sigma. \tag{37}$$

Furthermore, we assume that Σ is non-zero only in the subspace of *zero-mode* (the ground-state spin configuration subspace).

$$\Sigma = \sigma\hat{\varrho}(\psi)\hat{\varrho}(\psi) \tag{38}$$

where $\hat{\varrho}(\psi)$ is the normalized eigenvector of $A(\psi, q=0)$ with zero eigenvalue (see equation (22)).

Keeping terms in (36) consistently at the order of one-loop, we have

$$\sigma = \frac{T}{4}\sum_{q'}\mathrm{Tr}\left\{G(q')\frac{\partial}{\partial\phi_s}\frac{\partial}{\partial\phi_{s'}}A(q')\right.$$
$$\left.-\left[G(q')\frac{\partial}{\partial\phi_s}A(q')\right]\left[G(q')\frac{\partial}{\partial\phi_{s'}}A(q')\right]\right\}\varrho_s\varrho_{s'} \tag{39}$$

with

$$G(q) = [A(q) + \sigma\hat{\varrho}\hat{\varrho}]^{-1}. \tag{40}$$

As we see from the previous section, the long-wavelength fluctuations make the major contributions to the thermal selection in this system. This is different from the other systems with *normal* thermal selection [6, 12, 3, 2]. In the *normal* selection case (where the selection takes place at the order of harmonic excitations), the fluctuations on all different wavelength scales have equally important contributions to

the selection, in which case the short-wavelength fluctuations are important in these systems. So it is important to sum over the contributions from the fluctuations of *all* the wavevectors in the Brillouin zone. In our system, however, the long-wavelength fluctuations are the dominant contribution to the selection. So we can neglect the contributions from the short wavelength fluctuations, and only sum over a small neighbourhood of $q = 0$.

Keeping the leading-order terms on the right-hand side of (39), we obtain the equation to be satisfied by σ

$$\sigma = 2\pi C_1 T \int_{BZ} \frac{d^2q}{(2\pi)^2} \frac{1}{\det(\mathbf{A}(q) + \sigma\hat{\varrho}\hat{\varrho})} \tag{41}$$

and

$$\det(\mathbf{A}(q) + \sigma\hat{\varrho}\hat{\varrho}) = C_2\sigma + Dq^2 + O(q^4) \tag{42}$$

where C_1 and C_2 depend on both ψ and A, i.e. $C_1 = C_1(\psi, A)$ and $C_2 = C_2(\psi, A)$, and D only depends on A. With the approximation (42), we have

$$\sigma = C_1 T \int_0^{\Lambda} q\,dq \frac{1}{(Dq^2 + C_2\sigma)} = T\frac{C_1}{2D} \ln\left(\frac{D\Lambda^2 + C_2\sigma}{C_2\sigma}\right) \approx T\frac{C_1}{2D} \ln\left(\frac{D\Lambda^2}{C_2\sigma}\right) \tag{43}$$

where Λ is of order unity. Equation (43) gives a self-consistent equation for σ. The solution to equation (43) has singular dependence on temperature. It has leading behaviour as

$$\sigma \sim \frac{C_1}{2D} T|\ln T|. \tag{44}$$

Now the *selection part* of the free energy is

$$F_{sel} = \frac{T}{2}\sum_q \ln\det(\mathbf{A}(q) + \sigma\hat{\varrho}\hat{\varrho}) \approx \frac{T}{4\pi}\int_0^{\Lambda} q\,dq \ln(Dq^2 + C_2\sigma)$$

$$\approx \frac{T}{4\pi D} C_2\sigma \ln\left(\frac{D\Lambda^2 + C_2\sigma}{C_2\sigma}\right) \approx \frac{C_2}{2\pi C_1}\sigma^2 \approx \frac{C_1 C_2}{8\pi D^2}(T\ln T)^2. \tag{45}$$

3.3. Numerical results

For fixed A, we numerically evaluate $C_1(\psi, A)$, $C_2(\psi, A)$ (see equations (41) and (42)) and D for selected values of ψ. We find that the quantity C_1C_2/D^2 can be fitted very well by the function

$$a_0 + a_6 \cos 6\psi \tag{46}$$

with a_0 and a_6 both being positive and A-dependent. Therefore the selection free energy

$$F_{sel} \approx \frac{(T\ln T)^2}{8\pi}(a_0 + a_6\cos 6\psi). \tag{47}$$

Therefore, thermal fluctuations select the ground-state configuration in which $\cos 6\psi = -1$, i.e. the spins on one of the sublattices lie perpendicularly to the easy axis. For the case of $A = 2$, C_1C_2/D^2 was calculated for 10 values of ψ and fit to the form (46). The result is

$$F \approx \frac{(T\ln T)^2}{8\pi}(0.07 + 0.02\cos 6\psi). \tag{48}$$

4. Quantum selection

To calculate the quantum selection term, we must first find the spin-wave spectrum $\omega_\nu(q)$ for every possible state of the ground-state manifold. Then the free energy is calculated from the following formula (to harmonic order only):

$$F = \sum_q \sum_\nu \frac{\hbar\omega_\nu(q)}{2} + k_B T \sum_q \sum_\nu \ln(1 - e^{-\hbar\omega_\nu(q)/k_B T}) \tag{49}$$

the first term is the zero-point fluctuation energy, the second term is the thermally dependent part. In contrast to the thermal case (section 3), we already find a selection effect at harmonic order. Quantum selection has previously been considered only in exchange-coupled systems by this method [6, 16, 17] or by a cruder one [18].

4.1. Spin-waves

Quantum fluctuations are governed by the equation of motion:

$$\frac{dS}{dt} = \frac{1}{i\hbar}[S, H] \tag{50}$$

where $[S, H]$ is the commutator, and H is the Hamiltonian (1). For large quantum number S, each spin can be described *classically* by its two spherical angles $\{\phi_i, \theta_i\}$. In spherical coordinates, the semi-classical equations of motion are

$$\begin{cases} \hbar S \sin\theta_i \dot{\theta}_i = +\partial H/\partial\phi_i \\ \hbar S \sin\theta_i \dot{\phi}_i = -\partial H/\partial\theta_i \end{cases} \tag{51}$$

where the dot means derivative with respect to time, $i = 1, \ldots, N$ (N is the number of the spins in the system). Expanding the equations of motion about a ground-state configuration $\{\theta_i = \pi/2, \phi_i\}$, keeping only the linear order in the spin deviations $\{\delta\phi_i, \delta\theta_i\}$, we obtain

$$\begin{aligned} \hbar S \delta\dot{\theta}_i &= -J \sum_{(j)} [(A \cos\phi_i \cos\phi_j + \sin\phi_i \sin\phi_j)\delta\phi_i \\ &\quad - (A \sin\phi_i \sin\phi_j + \cos\phi_i \cos\phi_j)\delta\phi_j] \\ \hbar S \delta\dot{\phi}_i &= -J \sum_{(j)} [-(A \cos\phi_i \cos\phi_j + \sin\phi_i \sin\phi_j)\delta\theta_i + \delta\theta_j]. \end{aligned} \tag{52}$$

Fourier transforming the equations of motion (52) and using equations (18) and (19), we get

$$\frac{i\omega\hbar S}{2J} \begin{pmatrix} \delta\theta(q) \\ \delta\phi(q) \end{pmatrix} = \Omega(q) \begin{pmatrix} \delta\theta(q) \\ \delta\phi(q) \end{pmatrix} \tag{53}$$

where

$$\begin{cases} \delta\theta(q) = \{\delta\theta_1(q), \delta\theta_2(q), \delta\theta_3(q)\} \\ \delta\phi(q) = \{\delta\phi_1(q), \delta\phi_2(q), \delta\phi_3(q)\} \end{cases} \tag{54}$$

and

$$\Omega(q) = \begin{pmatrix} 0 & \mathbf{A}(q) \\ -\mathbf{B}(q) & 0 \end{pmatrix} \tag{55}$$

with $\mathbf{A}(q)$ and $\mathbf{B}(q)$ the 3×3 matrices defined in (18) and (20). The eigenvalues of the non-Hermitian matrix $\Omega(q)$ occur in imaginary pairs

$$\pm \lambda_\nu(q) = \pm i\hbar\omega_\nu(q)S/2J$$

with $\omega_\nu(q) \in$ Real ($\nu = 1, 2, 3$ is branch index). It can be verified that [19] all the frequencies $\omega_\nu(q)$ are real, if the Hessian matrix

$$\mathbf{E}(q) = \begin{pmatrix} \mathbf{A}(q) & 0 \\ 0 & \mathbf{B}(q) \end{pmatrix}$$

is positive semi-definite, i.e. the state that Hamiltonian is expanded about is a local minimum.

4.2. Quantum selection results

The set of ground states selected by quantum fluctuations is determined by the first term in (49),

$$E_{\mathrm{qm}} = \frac{1}{2}\sum_\nu \int_{q\in \mathrm{BZ}} \frac{\mathrm{d}^2 q}{(2\pi)^2}\hbar\omega_\nu(q). \tag{56}$$

For each selected ψ (with fixed A), this quantity is evaluated numerically and the result is found to be fitted very well by

$$E_{\mathrm{qm}} \approx (b_0 - b_6 \cos 6\psi)(J/S) \tag{57}$$

where b_0 and b_6 are both positive quantities which depend on the anisotropy A. Therefore, the quantum fluctuations select the set of ground-state configurations in which $\cos 6\psi = 1$, which is opposite to what the thermal fluctuations select†. For the case of $A = 2$, equation (56) was calculated for 10 values of ψ and were fit to the form (57). The numerical result is

$$E_{\mathrm{qm}} \approx (2.38 - 0.02 \cos 6\psi)(J/S). \tag{58}$$

We can conveniently model the ground-state selection by including a phenomenological biquadratic interaction in the free energy [6, 2, 17]

$$\delta H_{\mathrm{biq}} = -K\sum_{\langle ij\rangle}(S_i \cdot S_j)^2. \tag{59}$$

It was argued in previous work [6, 2, 17] that quantum selection favours $K > 0$, i.e. collinearity. It can be checked by a straightforward calculation that the biquadratic energy for $\cos 6\psi = 1$ states is lower than $\cos 6\psi = -1$ states by

$$\Delta E_{\mathrm{biq}} = 2K\left(\frac{A-1}{A+1}\right)^3 > 0.$$

This agrees with our current result $b_6 > 0$ and furthermore indicates $b_6 \sim (A-1)^3$ near $A = 1$.

† In the limit where $k_B TS/J \to 0$, we evaluated the contribution from the second term in equation (49). It is found to favour the type II states (opposite to what the first term selects, which is type I states). The selection strength from the second term is approximately $c_6 T(TS)^2$. In the case of $A = 2$, $c_6 \approx 4$. However, in the limit where $k_B TS/J$ is large (see section 4.3.), the selection effect goes away.

4.3. Relationship between quantum and thermal calculations

In the limit

$$k_B T S/J \gg |\lambda_M| \tag{60}$$

where $|\lambda_M|$ is the largest eigenvalue of the frequency matrix in (53), the second term in (49) becomes

$$\begin{aligned} F_{\text{thermal}} &\approx k_B T \sum_q \sum_\nu \ln \frac{\hbar\omega_\nu(q)}{k_B T} = -k_B T N \ln(k_B T) + k_B T \sum_q \ln\left(\prod_\nu \hbar\omega_\nu(q)\right) \\ &= -k_B T N \ln(k_B T) + k_B T \sum_q \ln\left(\prod_\nu \left\{\frac{2J}{S}[(-i\lambda_\nu(q))(i\lambda_\nu(q))]^{1/2}\right\}\right) \\ &= k_B T N \ln \frac{2J}{S k_B T} + k_B T \sum_q [\tfrac{1}{2} \ln \det \Omega(q)] \\ &= k_B T N \ln \frac{2J}{S k_B T} + \tfrac{1}{2} k_B T \sum_q [\ln \det \mathbf{A}(q) + \ln \det \mathbf{B}(q)]. \end{aligned} \tag{61}$$

Comparing this result with (23), we see that in the limit of large spin quantum number S (or large temperatures)†, the thermal fluctuation selection at the order of harmonic excitation is recovered by equally populating all the quantum spin excitation levels. This is true for any system in the harmonic limit. It can be shown *mathematically* [19] that the matrix $\mathbf{E}$ of the classical quadratic energy has the same determinant as that of the matrix Ω from which the spin excitation frequencies are obtained. Therefore equally populating all the spin excitation levels leads to the *classical* free energy as is obtained from the classical quadratic energy.

5. Effect of random site dilution

The Hamiltonian for the system with site dilution is

$$H = J \sum_{\langle ij \rangle} \epsilon_i \epsilon_j (A S_{ix} S_{jx} + S_{iy} S_{jy} + S_{iz} S_{jz}) \tag{62}$$

where $\epsilon_i = 1$ or 0 depending on whether the site i is occupied or not. ϵ_i are random and uncorrelated with each other, with $\langle \epsilon_i \rangle = 1 - \delta p$, where δp is the diluted fraction. The interactions are only between the spins that are nearest neighbours. The dilution does not change the global rotation symmetry around the easy axis (x-axis), but lifts the non-trivial degeneracy in which the $\{\phi_i\}$ vary continuously.

In the systems studied before [4, 2, 3], only pairs of removed spins can create random fields, because in any ground states of those systems, every site is *equivalent* to every other site by some symmetry operation, i.e. the local fields are site-independent. Removing an isolated site eliminates an equal number of bonds in every bond direction, and therefore has zero net contribution to the random field.

† Also in this limit, $E_{qm} \ll F_{\text{thermal}}$, so we neglect E_{qm}.

In this anisotropic AFT model, on the other hand, the local fields are site-dependent in any ground-state configuration: no symmetry operation exists that can transform the local fields on different sublattices to each other. Also, there are three kinds of bonds that connect nearest neighbours in the lattice: $b\{1,2\}$, $b\{2,3\}$ and $b\{3,1\}$. Each site is connected to its nearest neighbours through two kind of bonds; for example, a site in sublattice 1 is connected to its nearest neighbours by bonds $b\{1,2\}$ and $b\{3,1\}$. Therefore removal of an isolated site will leave *unbalanced bonds*. It favours the states in which the excessive bonds have the *minimum energy*: the spins that are connected by the excessive bonds point along the easy axis with nearest neighbours being antiparallel, while the the spins on the sublattice in which the diluted spin sits lie perpendicular to the easy axis. Thus dilution on a single site has the effect of selecting the same subset of ground states as the thermal fluctuations. Notice that the reflection about the yz plane is still a symmetry. Thus the random site dilutions create discrete random uniaxial anisotropy fields with the favoured axis being one of the three symmetry-related directions in ψ space, depending on which sublattice the diluted spin was on. Microscopically, the free energy due to the random site dilution is

$$-\sum_i v_0 \cos 2(\psi - \psi_i^{(0)}) = -\sum_i \tfrac{1}{2} v_0 [e^{i2(\psi-\psi_i^{(0)})} + e^{-i2(\psi-\psi_i^{(0)})}] \tag{63}$$

where $\psi_i^{(0)} \in \{\pi/6, \pi/2, 5\pi/6\}$, and

$$v_0 \sim (A-1)J. \tag{64}$$

Summing over a macroscopically small but microscopically large volume ΔV around $\mathbf{r}$, we obtain the random anisotropy in the continuum limit:

$$v(\boldsymbol{r}) e^{i2\psi_0(\boldsymbol{r})} = \sum_{\boldsymbol{r}_i \in \Delta V} v_0 e^{i2\psi_i^{(0)}} \tag{65}$$

where $\psi_0(\boldsymbol{r})$ labels the macroscopic easy axis. By the law of large numbers, the right-hand side of equation (65) becomes a Gaussian random variable. So $\psi_0(\boldsymbol{r})$ is a uniformly distributed random variable [3], and

$$\langle v(\boldsymbol{r})^2 \rangle \sim v_0^2 N \tag{66}$$

where $N \approx \delta p \Delta V$ is the number of impurities.

Therefore, the free energy density due to the random site dilution in the continuum limit is

$$F_{\text{random}} = -\int d^2 r\, v(\mathbf{r}) \cos 2(\psi - \psi_0(\mathbf{r})). \tag{67}$$

The correlation length should scale as the mean separation between the impurities

$$\xi_v \sim (\delta p)^{-1/2}. \tag{68}$$

6. Phase diagrams

It is complicated to describe the phase diagram since there are four non-trivial parameters: the anisotropy ratio A; the reduced temperature T/J; the occupied fraction p; the spin number S for quantum spins. (The external field would be another parameter, but we will not go into it here.) Based on the results of the previous sections, we can now give some qualitative speculations about the phase diagrams of this system.

To be more conventional, we now let the easy axis be the z-axis. To describe the rotational symmetry-breaking around the easy axis, we introduce the 'chiral' order parameter [1]

$$\kappa \equiv (S_i \times S_j + S_j \times S_k + S_k \times S_i) \tag{69}$$

where i, j, k are arranged counter clockwise for upward elementary triangles. In any classical ground state, κ lies in the xy plane, we let Φ be its angle within the xy plane.

Combining equations (45), (57) and (67) from previous sections, we get the effective continuum free energy density

$$f = \rho_s^{\Phi}(\nabla\Phi)^2 + \rho_s^{\psi}(\nabla\psi)^2 - h_6 \cos 6\psi - v(x)\cos 2(\psi - \psi_0(x)) \tag{70}$$

where $\rho_s \propto J$ are the spin stiffnesses; h_6 is the selection free energy ($h_6 > 0$ in the case of quantum selection and $h_6 < 0$ in thermal selection).

6.1. Pure system

To the extent that (70) is valid, then, the ψ and Φ degrees of freedom decouple. The continuum theory for the ψ degree of freedom in (70) is identical to that of a ferromagnetic XY model, with ψ as the spin angle. In the language of the ferromagnetic XY model, the h_6 term is a six-fold anisotropy, and the last term is a random uniaxial anisotropy. By means of this mapping, we can apply previous results on XY models to the phase transitions in our case†.

6.1.1. Thermal transitions. We first consider what transitions are associated with the ψ variable on the basis of (70), viewed as a mapping to a ferromagnetic XY model with six-fold anisotropy. At high temperatures the system is disordered by ψ vortices; at low temperatures the anisotropy is relevant and the system becomes locked into long-range order with six discrete states. José *et al* [20] showed that, for p-fold anisotropy for $p > 4$, there is a 'floating' phase between these with power-law correlations. The upper transition T_{N_1}, identified by Miyashita and Kawamura [1] is a Kosterlitz–Thouless (KT) transition associated with ordinary vortices in the ψ degree of freedom, above which the system is in a fully disordered phase; in addition there must be a lower transition T_6 above which the discrete six-fold long-range order gives way to floating quasi-long-range order (in other words, between T_{N_1} and T_6 the six-fold selection is irrelevant in the renormalization group sense and the continuous

† We have done some of the calculations in the paper on the triangular XY antiferromagnet with exchange anisotropy ($J_{xx} \neq J_{yy}$). This also shows the same degeneracy not only of the ground states but of the first $O(T)$ selection term. The principal difference is that the continuous symmetry of the Φ variable is replaced by a discrete Ising-like symmetry, corresponding to positive or negative values of the chirality κ just as in the isotropic triangular XY antiferromagnet.

ψ degeneracy behaves as a continuous S_1 symmetry). On the other hand, there is another KT transition T_{N_2} [1] associated with binding–unbinding of ordinary Φ vortices below which κ have quasi-long-range order.

In the case of large A, the fluctuations of κ out of the xy plane are very small [1]. In this case, the ψ and Φ degrees of freedom are well defined, and $\rho_s^\psi \sim A$ and $\rho_s^\Phi \sim 1/A$ (the spin stiffnesses can be obtained from the eigenvalues of the matrices (18) and (19) in the long-wavelength limit $q \to 0$). Thus the energy associated with the Φ ordering is much smaller than that associated with the ψ ordering. The appearance of Φ vortices imposes a 'finite size effect' on the selection free energy when $T > T_{N_2}$, i.e. the longest wavelength contributing to the selection term is the order of the mean separation between the vortices. Thus the strength of the symmetry-breaking field h_6 is renormalized smaller by the presence of the Φ vortices. Nevertheless, h_6 *is always relevant once below* T_6 *no matter how weak* h_6 *is* [20, 21]. According to the theory of KT transitions, $T_{N_1} \propto \rho_s^\psi$, $T_{N_2} \propto \rho_s^\Phi$; furthermore it is well known [20, 21] that $T_6 \approx (4/9)T_{N_1}$ in the limit where the six-fold anisotropy is weak and the vortex core energy is large. Therefore we expect the sequence

$$T_{N_1} > T_6 > T_{N_2} \tag{71}$$

at large anisotropy $A \gg 1$.

In the limit of $A \to 1$, the fluctuation of κ is nearly isotropic. Now ρ_s^ψ and ρ_s^Φ are greatly renormalized by the fluctuation of κ out of the xy plane. It is expected that T_{N_1} and T_{N_2} merge into T_{Z_2} at $A = 1$ where the system undergoes the KT transition mediated by Z_2 vortices [1, 8]. But we have not investigated the behaviour of T_6 in this limit. It is not obvious, based on the result we have so far, whether T_6 approaches to T_{Z_2} along with T_{N_1} and T_{N_2} or whether $T_6 \to 4/9T_{Z_2}$.

6.1.2. Competition between thermal and quantum selection. Since the set of states selected by quantum fluctuations is the opposite of what the thermal fluctuations select, we expect there is a phase transition at the transition temperature T_N', roughly where the thermal selection amplitude equals that of the quantum selection. The phase transition across T_N' ought to be first order (by the usual Landau symmetry criterion), where $\langle \cos 6\psi \rangle$ changes from 1 to -1 as T increases (i.e. transition from type I to type II). Since the quantum selection is weaker for larger S, we expect that T_N' decreases with increasing spin quantum number S.

For the case where the anisotropy A is large, a rough phase diagram for the pure system is sketched in figure 3. In the limit of $A \to \infty$, the spins are all close to the z-axis. In this Ising limit, the type I states approach those of an Ising model in which one sublattice is up and two are down; the type II states approach those of an Ising model in which one sublattice is up, one is down, and the other is disordered.

6.1.3. Ferromagnetic properties. In this subsection, we will consider the possible development of a ferromagnetic moment for the cases of quantum and thermal selections.

For $T \in (T_6, T_{N_1})$, the system is in a *floating phase*, where both ψ vortices and ψ symmetry-breaking field h_6 are irrelevant. Since each magnetic unit cell has net moment, the ferromagnetic susceptibility in the z-direction diverges exponentially fast on approaching T_6 from above or T_{N_1} from below, and remains infinite throughout (T_6, T_{N_1}) [20, 22].

For $T < T_6$, the system is locked into one of the discrete states selected by the symmetry-breaking field. In the case of type I phase, there is finite ferromagnetic

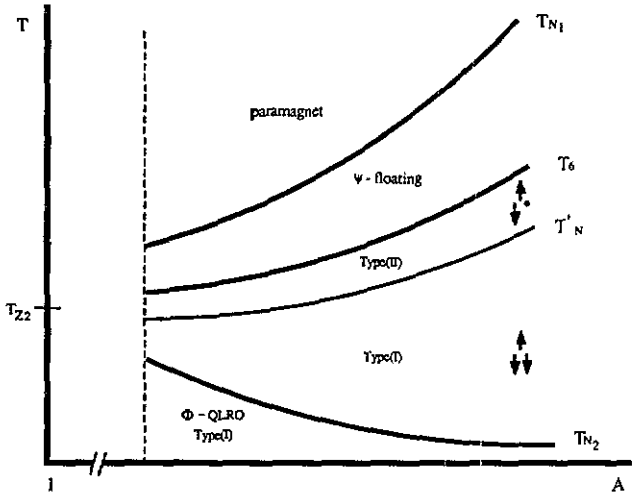

Figure 3. Phase diagram for pure ($\delta p = 0$) system in the case large anisotropy A (see the text). The phase boundary T'_N shifts lower for larger S.

magnetization per magnetic unit cell in the z-direction: for $T > T_{N_2}$, there is no order in xy plane; and still for $T < T_{N_2}$, the magnetic moment in the xy plane is purely antiferromagnetic, so there is no divergent ferromagnetic susceptibility in the xy plane. In the case of the type II phase, there is finite ferromagnetic magnetization in the xy plane per magnetic unit cell. Thus for all the temperatures $T < T_{N_2}$ the ferromagnetic susceptibility in the xy plane is divergent (this is only true for fairly large spin S when it is possible that $T'_N < T_{N_2}$ so that there exists a range over which thermal selection dominates). Because there is no net magnetic moment along the z-axis in the type II states, the ferromagnetic susceptibility is finite. The contrast in behaviours ought to make it easy to distinguish types I and II behaviour experimentally.

6.1.4. Finite-size effect on the locking transition. The recursion relation for the symmetry-breaking field is [20]

$$\left[\frac{h_6}{k_B T}\right]' \simeq b^{\lambda_6(T)} \left[\frac{h_6}{k_B T}\right] \tag{72}$$

with

$$\lambda_6(T) = 2 - 9\frac{k_B T}{\pi J_{\text{eff}}} \tag{73}$$

where b is the scale factor, J_{eff} is the effective coupling. For a system of finite size L, in order to see the locking transition T_6, there should be

$$\left[\frac{h_6}{k_B T}\right]' \simeq L^{\lambda_6(T)} \left[\frac{h_6}{k_B T}\right] \sim 1. \tag{74}$$

In the limit of $S \to \infty$, i.e. the classical spin system, the symmetry-breaking field $h_6 \sim (T \ln T)^2$. For the case $A = 2$ and system size $L = 24$, combining equations (48) and (74), we find that to see the locking transition, the temperature should be at least $T > 2J$, which is well above the KT transition temperature $T_{N_2} \approx 0.66J$. This explains why Miyashita *et al* did not see T_6 in their simulations [1].

6.2. *Random field effects*

We now turn to the diluted system ($\delta p > 0$), which has effective random anisotropies (section 5). Here we suggest their consequences for the phase diagram.

6.2.1. Behaviour at $T = 0$. We consider only the case $S < \infty$ so that the quantum selection always dominates and approaches a finite value $h_6 > 0$ in the limit $T \to 0$.

The effective random anisotropies will favour different different values of ψ in different regions. Consequently the system breaks up into domains separated by domain walls with the wall thickness

$$l_w \sim \sqrt{\rho_s^\psi / h_6} \tag{75}$$

and the energy per unit length of a domain wall

$$E_w = \sqrt{\rho_s^\psi h_6} \tag{76}$$

Note that as $A \to 1$, we have $h_6 \to 0$ and $l_w \to \infty$.

We now apply the Imry–Ma [23] argument for this system. Consider now if we construct a state in which ψ varies over a length scale (domain size) l. If $l \ll l_w$ it is not meaningful to consider discrete domains, so ψ just wanders continuously with $\nabla\psi \sim 1/l$. Hence the free energy per area is

$$F_l \sim \rho_s^\psi / l^2. \tag{77}$$

If $l \gg l_w$, then we have well defined domains of discrete selected ψ†, and

$$F_l \sim E_w / l \sim (\rho_s^\psi h_6)^{1/2} / l. \tag{78}$$

This is to be compared with the random energy, which can be easily worked out to be

$$F_{\text{random}}(l) = v_0(\delta p)^{1/2} / l = v_0 / (l\xi_v) \tag{79}$$

per site. The correlation length of the discrete order ξ_ψ is the value of l which minimizes

$$F_l + F_{\text{random}}(l).$$

For small δp, the random energy is small and this must correspond to the case where the domains are large, when it is appropriate to use the discrete approach (78). Then both competing terms scale as $1/l$, indicating marginal behaviour, and we must quote the more sophisticated reasoning which shows the randomness disorders the discrete model at sufficient distances; by Binder's argument [24] (we replaced the lattice constant by l_w since the smallest possible length scale in the discrete regime is the wall thickness),

$$\xi_\psi \sim l_w \exp[(\rho_s^\psi h_6)^{1/2} / (v_0\sqrt{\delta p})]^2. \tag{80}$$

† At $T = 0$, the symmetry-breaking field h_6 is always relevant. Therefore the system is locked into one of the discrete states selected by h_6. Due to the random anisotropy field, the system breaks up into domains to minimize the random energy F_{random}.

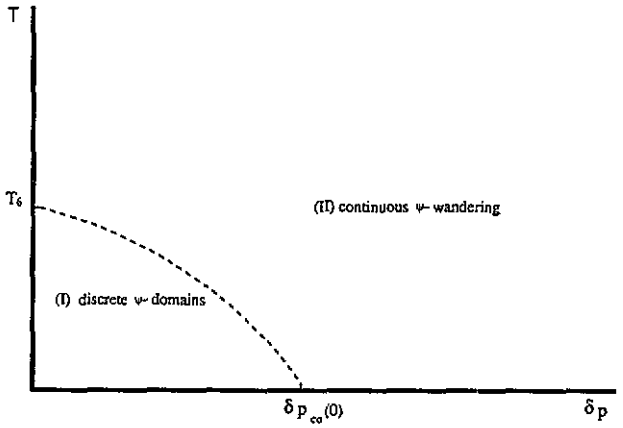

Figure 4. Phase diagram on the $(T, \delta p)$ plane. The broken curve indicates crossover not a real phase transition: I, discrete random field behaviour; II, continuous random field behaviour.

For large δp, the random energy is large and the domains are small. In this case by comparison with equation (77) we get

$$\xi_\psi \sim (\rho_s^\psi / v_0)\xi_v. \tag{81}$$

Comparing (80) and (81), we see that the crossover into the impurity dominated regime occurs when

$$E_w \sim (\rho_s^\psi h_6)^{1/2} = v_0(\delta p)^{1/2} \sim (A-1)J(\delta p)^{1/2}. \tag{82}$$

In other words, there is a crossover value

$$\delta p_{co} \sim \frac{\rho_s^\psi h_6}{(A-1)J} \tag{83}$$

such that the decrease of ξ_ψ with δp is given by equation (80) for $\delta p \ll \delta p_{co}$ and equation (81) for $\delta p \gg \delta p_{co}$.

6.2.2. Effect on phase transitions. Since random fields make the lower critical dimension two for discrete degrees of freedom and four for continuous ones [23, 25], there is no real phase transition in $d = 2$. Therefore the random site dilution, which couples to the ψ degree of freedom, will definitely *destroy both* T_{N_1} *and* T_6 *as real phase transitions.* On the other hand, the continuous rotation symmetry around the z-axis possessed by the Φ degree of freedom is unaffected by dilution. In particular, if we travel a loop around a vortex of the ψ order, there is no frustration in the Φ coupling. Thus the effective Hamiltonian for the transverse order remains unfrustrated, both for quantum selection and thermal selection. Consequently, *the transition at* T_{N_2} *is not qualitatively changed by the disorder.*

6.2.3. Crossover behaviour for $T > 0$. At finite temperatures, the crossover behaviour will be different coming from the discrete (six-fold locked) phase in $T < T_6$ than from the floating phase in $T > T_6$. Above T_6 the selection term is zero at large scales, so for arbitrarily small dilution we have the same continuous kind of disordering response which, at $T = 0$, we found only as $\delta p > \delta p_{co}$. Thus, we can extend a crossover line $\delta p_{co}(T)$ which terminates at the point $(\delta p = 0, T = T_6)$.

6.2.4. Nature of ordering in random states. Note that the effective random axis is uniaxial and never distinguishes states with $\psi \to \psi + \pi$. In fact, this is not just coarse-grained, it is present in the original diluted microscopic model since we have an exact spin inversion symmetry. Consequently, we always have two degenerate ground states related by a discrete symmetry. According to the picture of Fisher and Huse [26], this is the essential feature of an Ising spin-glass, so we can speculate that the ground state is like that of a two-dimensional Ising spin-glass. The spin-glass does not appear as a separate phase on the phase diagram since in two dimensions the transition is at zero temperature; however, as $T \to 0$ the nonlinear susceptibility should diverge with the same power law $1/T^{\gamma}$ as in Ising systems [27], and the spatial decay of the corresponding (Edwards–Anderson) correlation function should have the same power-law decay as in Ising spin glass models†.

7. Summary

We have shown (section 3) that thermal fluctuations do have an ordering effect by selecting among the classical ground states in a triangular Ising–Heisenberg antiferromagnet. In our system, the thermal selection free energy is dominated by long-wavelength fluctuations, in contrast to all previously considered systems [6, 12, 13, 2, 3], in which short-wavelength fluctuations dominate. Since the symmetry-breaking happens at higher orders than the harmonic excitations, the selection effect is quite weak (its temperature dependence has leading behaviour $(T \ln T)^2$). While thermal selection chooses type II states, quantum selection favours type I states (section 4.2); in principle this competition may lead to a phase transition (section 6.1.2). Thus, as is typical for selection out of a degenerate manifold, in either case the continuous degeneracy is reduced to a discrete (six-fold) symmetry, in addition to the continuous symmetry which remains from the Hamiltonian. (In the process of the quantum calculation, we also show the relationship between the formalisms used for the classical and quantum selections (section 4.3), valid for quite general continuous spin systems.)

Then in section 5, we found that random dilution introduces an effective random anisotropy acting in the degenerate manifold. As in the dipolar honeycomb model [3] a true two-fold symmetry is still preserved. However, the disordering effect for site dilution is stronger in the AFT Ising–Heisenberg model than in [3], because the bond energies at different sites are not equivalent (it is analogous to the enhanced effect of *bond* dilution compared with site dilution in usual models where the sites *are* equivalent) [4].

Since the thermal (quantum) fluctuations introduce the six-fold symmetry-breaking field h_6, it is expected that there is a KT phase transition T_6 below which the system is locked into one of the six discrete states selected by h_6. Therefore we propose (section 6.1.1) that in a system with easy-axis anisotropy, there is *another phase transition T_6 in addition to the two successive phase transitions T_{N_1} and T_{N_2}* identified in [1]. For systems with large easy-axis anisotropy $A \gg 1$ and large enough spin quantum number S, we expect the sequential phase transitions $T_{N_1} > T_6 > T'_N > T_{N_2}$ upon lowering the temperature.

On the other hand, when the Ising–Heisenberg system is doped with non-magnetic impurities, the site dilutions produce a random anisotropy field that couples to the

† Even if $\delta p > \delta p_{co}$, so that we must consider ψ a continuous rather than a discrete degree of freedom, we still expect the universality class to be that of an Ising spin-glass.

degeneracy degree of freedom. Consequently there are no real phase transitions associated with this degree of freedom. Nevertheless there is a crossover between weak and strong random field behaviour characterized by whether or not there are domains within which there exist one of the six discretely selected states (sections 6.2.1 and 6.2.3). Furthermore, it turns out that the disordering of the other degrees of freedom does not frustrate the ordering of the in-plane components; since the rotation symmetry about the easy axis is preserved upon site dilution, the KT transition of these components is apparently not destroyed (section 6.2.2). Finally, the inversion symmetry is also preserved, so that the discrete degrees of freedom in such a system are expected to behave as a two-dimensional Ising spin-glass as $T \rightarrow 0$ (section 6.2.4).

Acknowledgments

We thank E F Shender, Brad Marston, H Kawamura and Ganpathy Murthy for very useful discussions. This research is supported by NSF grant DMR-9045787.

References

[1] Miyashita S and Kawamura H 1985 *J. Phys. Soc. Japan* **54** 3385
[2] Larson B and Henley C L unpublished
[3] Prakash S and Henley C L 1990 *Phys. Rev.* B **42** 6574
[4] Fernandez J F 1988 *Phys. Rev.* B **38** 6901
[5] Villain J, Bidaux R, Carton J P and Conte R 1980 *J. Physique* **41** 1263
[6] Henley C L 1987 *J. Appl. Phys.* **61** 3962
[7] Rastelli E and Tassi A 1988 *J. Phys. C: Solid State Phys.* **21** L35
[8] Kawamura H and Miyashita S 1984 *J. Phys. Soc. Japan* **53** 4138
[9] Sano M, Iio K and Nagata K 1989 *Phys. Rev.* B **39** 9753
Iio K, Sano M, Tanaka H and Nagata K 1988 *J. Appl. Phys.* **64** 5959
Maegawa S, Goto T and Ajiro Y 1988 *J. Phys. Soc. Japan* **57** 1402
[10] Itoh M, Yamada I, Ubukoshi K, Hirakawa K and Yasuoka H 1986 *J. Phys. Soc. Japan* **55** 2125
Hirakawa K *et al* 1986 *Physica* B **136** 335
[11] Negele J W and Orland H 1988 *Quantum Many-Particle Systems* (New York: Addison-Wesley)
[12] Henley C L 1989 *Phys. Rev. Lett.* **62** 2056
[13] Henley C L and Prakash S 1988 *J. Physique Coll.* C **8** 1197
[14] Wilson K G and Kogut J 1974 *Phys. Rep.* C **12** 76
[15] Amit D J *Field Theory, the Renormalization Group, and Critical Phenomena* 2nd edn (Singapore: World Scientific)
[16] Oguchi *et al* 1985 *J. Phys. Soc. Japan* **12** 4494
[17] Chubukov A V and Golosov D I 1991 *J. Phys.: Condens. Matter* **3** 69
[18] Long M W 1989 *J. Phys.: Condens. Matter* **1** 2857
[19] Walker L R and Walstedt R E 1980 *Phys. Rev.* B **22** 3816
[20] José J V, Kadanoff L P, Kirkpatrick S and Nelson D R 1977 *Phys. Rev.* B **16** 1217
[21] Nelson D R 1983 *Phase Transitions* vol 7 (London: Academic)
[22] Kosterlitz J M 1974 *J. Phys. C: Solid State Phys.* **7** 1046
[23] Imry Y and Ma S 1975 *Phys. Rev. Lett.* **24** 1399
[24] Binder K 1983 *Z. Phys.* B **50** 343
[25] Fisher D S, Frohlich J and Spencer T 1984 *J. Stat. Phys.* **34** 863
[26] Fisher D S and Huse D A 1986 *Phys. Rev. Lett.* **56** 1602; 1988 *Phys. Rev.* B **38** 386
[27] Swendsen R H and Wang J -S 1986 *Phys. Rev. Lett.* **57** 2607
Batrouni G G and Dagotto E 1988 *Phys. Rev.* B **37** 9875 and references therein