

Ordering due to disorder in dipolar magnets on two-dimensional lattices

Sona Prakash

Department of Physics, Boston University, Boston, Massachusetts 02215

Christopher L. Henley

*Department of Physics, Boston University, Boston, Massachusetts 02215
and Department of Physics, Cornell University, Ithaca, New York 14853**

(Received 26 March 1990)

We calculate the effects of thermal fluctuations, dilution, and a uniform external field on the continuously degenerate ground state of dipolar magnets on the square and honeycomb lattices. In all cases, these perturbations make terms in the free energy that favor particular states, reducing the continuous degeneracy to a discrete symmetry. Dilution and thermal selection compete; i.e., they select different states. For the cases of thermal fluctuations and dilution, the discrete symmetry is fourfold on the square lattice and sixfold on the honeycomb lattice; for a uniform field it is twofold and threefold, respectively. In addition, dilution generates effective random axes, which disorder all of the selected phases in each system, except for the dilution-selected phase of the square lattice. We present a proposed phase diagram including speculations on the finite-temperature transition.

I. INTRODUCTION

Frustrated vector spin systems are often found to have continuously degenerate ground states at zero temperature. The ground states can usually be parametrized by angles that describe the relative orientations of the staggered magnetizations of different sublattices.^{1,2}

However, fluctuations can remove this degeneracy, i.e., they can select certain states from the continuous manifold. Quite often, this selection effect results in the reduction of the continuous *degeneracy* to a discrete *symmetry*.^{1,2}

This is rather analogous to the selection effects which have been studied in numerous *Ising* systems at special points with an infinite ground-state entropy, and which were termed "ordering due to disorder."³ An example of such a system is the *Ising* antiferromagnet with nearest-neighbor interactions only on a face-centered-cubic lattice.⁴

We consider here a dipolar magnet on a square lattice and on a honeycomb lattice with the dipoles confined to rotate in the plane of the lattice.⁵ In both cases, the ground state at $T=0$ and in a small external field is continuously degenerate.⁶ The various states of the one-dimensional (1D) manifold can be obtained by rotating two different sets of dipoles [even and odd numbered, Figs. 1(a) and 2(a)] in opposite directions by the same angle.

The ground-state manifold of both systems is described in Sec. II. In Sec. III, we calculate the effect of thermal fluctuations on the ground-state manifold of both systems, in Sec. IV that of an external field and in Sec. V, that of quenched fluctuations due to dilution. Dilution is found to have, in addition, another effect, discussed in Sec. VI: that of creating an *effective random axis* which disorders either kind of discrete symmetric state (that selected by temperature or that selected by dilution) in

both the systems considered. In Sec. VII we predict a phase diagram in the temperature versus dilution plane on the basis of these selection effects and discuss the nature of the various phases and transitions between them. Finally, in Sec. VIII we summarize the results obtained in this paper, relate them to similar work done earlier,^{1,2} and discuss the experimental significance of these results.

The Hamiltonian of the system is given by

$$H = H_{\text{dip}} + H_h, \tag{1.1a}$$

where

$$H_{\text{dip}} = \frac{1}{2} \sum_{i,j} J_{ij} [\boldsymbol{\mu}_i \cdot \boldsymbol{\mu}_j - 3(\boldsymbol{\mu}_i \cdot \hat{\mathbf{r}}_{ij})(\boldsymbol{\mu}_j \cdot \hat{\mathbf{r}}_{ij})] \tag{1.1b}$$

is the dipolar energy, and

$$H_h = -\mathbf{h} \cdot \sum_i \boldsymbol{\mu}_i \tag{1.1c}$$

is the field energy. Here $\hat{\mathbf{r}}_{ij}$ is the unit lattice vector from site i to site j . In the real system, $J_{ij} = 1/r_{ij}^3$. For simplicity, we restrict the interactions to nearest neighbors only, i.e., we take $J_{ij} = J$ for nearest neighbors and 0 otherwise.

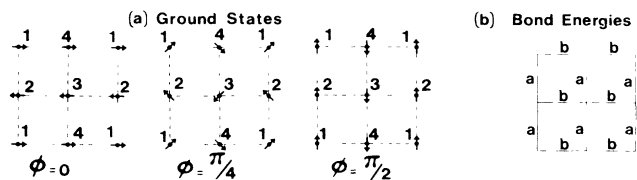


FIG. 1. (a) Parametrization of the ground states of the square lattice by ϕ : note that $\phi \rightarrow \phi \pm n\pi/2$ is a symmetry of the system. Thermal fluctuations select $\phi = 0 \pm n\pi/2$ while dilution selects $\phi = \pi/4 \pm n\pi/2$. (b) The bond energies for the various ground states of the system are given by $(E_a, E_b) = -J(1 + \sin^2\phi, 1 + \cos^2\phi)$.

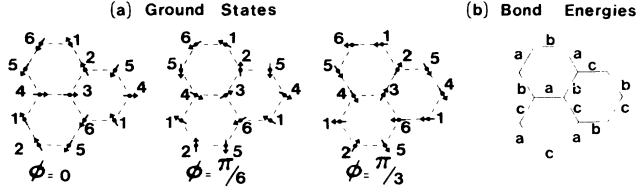


FIG. 2. (a) Parametrization of the ground states of the honeycomb lattice by ϕ : here $\phi \rightarrow \phi \pm n\pi/3$ is a symmetry of the system. Thermal fluctuations select $\phi = 0 \pm n\pi/3$ while dilution selects $\phi = \pi/6 \pm n\pi/3$. (b) The bond energies for the various ground states of the system are given by $(E_a, E_b, E_c) = -J(1 + \cos^2\phi, 1 + \cos^2(\phi + \pi/3), 1 + \cos^2(\phi - \pi/3))$.

erwise (it can be shown that the continuous degeneracy exists in either case). Then we can write (1.1b) as

$$H_{\text{dip}} = \frac{J}{2} \sum_{i,j} [\cos(\theta_i - \theta_j) - 3 \cos(\theta_i - \psi_{ij}) \cos(\theta_j - \psi_{ij})] \quad (1.2a)$$

and (1.1c) as

$$H_h = -h \sum_i \cos(\theta_i - \phi_h), \quad (1.2b)$$

where θ_i , ψ_{ij} and ϕ_h are the angles that the dipole at site i , the lattice vector \hat{r}_{ij} , and the external field h , respectively, make with the x axis. The sum is over nearest neighbors i and j . In (1.2a) and (1.2b), the magnitude of the dipole moment has been absorbed into J and h (i.e., $J\mu^2 \rightarrow J, h\mu \rightarrow h$).

II. GROUND STATES

Equation (1.2a) can also be written as

$$H_{\text{dip}} = -\frac{J}{2} \sum_{i,j} \left[\frac{3}{2} \cos(\theta_i + \theta_j - 2\psi_{ij}) + \frac{1}{2} \cos(\theta_i - \theta_j) \right]. \quad (2.1)$$

For both square and honeycomb systems, there is a one-parameter family of ground states parametrized by an angle ϕ . In any ground state the quantity $\theta_i + \theta_j - 2\psi_{ij}$ that appears in (2.1) is equal to zero. As ϕ is varied, spins on the even and odd sublattices rotate in opposite senses, keeping $\theta_i + \theta_j$ constant for nearest neighbors i and j :

A. Square lattice

The one-parameter family of ground states is parametrized by the angle ϕ as follows:

$$\theta_\alpha = (\phi, -\phi + \pi, \phi + \pi, -\phi), \quad (2.2a)$$

where $\alpha = 1, \dots, 4$ for the four sites α in the magnetic unit cell of the square lattice. Different ground states are obtained from each other by rotating the two sets of dipoles (even α and odd α) by the same angle ϕ in opposite directions [see Fig. 1(a)]. Then we can write

$$\theta_\alpha - \theta_\beta = 2\phi - m\pi, \quad (2.2b)$$

where α and β are nearest neighbors and m labels the two kinds of bonds and takes the values

$$m = \begin{cases} 0 & \text{for } (\alpha, \beta) = (1, 4) \text{ or } (3, 2) \\ 1 & \text{for } (\alpha, \beta) = (1, 2) \text{ or } (3, 4), \end{cases} \quad (2.2c)$$

i.e., each site is connected to four others through two bonds of each type. Then (2.1) becomes, for the ground states of the square lattice (with $h = 0$),

$$H = -\frac{JN}{4} \sum_{\alpha=1}^4 \sum_{m=[0,1]} [\frac{3}{2} + \frac{1}{2} \cos(2\phi - m\pi)]. \quad (2.3)$$

The second term in (2.3) cancels out between the four nearest neighbors for any ϕ , so the ground-state energy is $E_0 = -3JN$ independent of ϕ and thus the ground state is infinitely degenerate. This is due to the competition between antiferromagnetic and ferromagnetic behavior of the first and second terms, respectively, in the dipolar interaction given by (1.1b). The configurations $\phi = 0, \pi/4$, and $\pi/2$ for the square system are shown in Fig. 1(a). One can see that this system has a discrete fourfold symmetry (for $h = 0$) with respect to rotation in spin space of the even- and odd-numbered dipoles in opposite directions, by comparing the $\phi = 0$ and $\pi/2$ states in Fig. 1(a). One expects this symmetry to remain even after selection effects due to thermal fluctuations and dilution are accounted for.

The difference between the continuous degeneracy and the discrete symmetry can be seen as follows: when we are in a ground state, from (2.2c) we see that there are two kinds of bonds a, b in the square system [see Fig. 1(b)] for some generic ϕ , and the energy of the ground state is

$$E_0 = \frac{N}{2} (2E_a + 2E_b)$$

since each site is connected to four others through two bonds of each type. The bond energies are given by

$$(E_a, E_b) = -J(1 + \sin^2\phi, 1 + \cos^2\phi). \quad (2.4a)$$

The continuous degeneracy corresponds to invariance of the ground-state energy under *continuous variation* of the distribution of the ‘‘amount of frustration’’ among the two kinds of bonds, whereas the discrete symmetries correspond to invariance of the system under *permutations* of bond energies. This can be seen from the values of the bond energies for the different states obtained from (2.4a)

$$(E_a, E_b) = \begin{cases} (-1, -2)J & \text{for } \phi = 0 \\ (-\frac{3}{2}, -\frac{3}{2})J & \text{for } \phi = \pi/4 \\ (-2, -1)J & \text{for } \phi = \pi/2, \end{cases} \quad (2.4b)$$

where a bond energy of $-J$ corresponds to the most frustrated bond while one of $-2J$ corresponds to the least frustrated bond. We see from (2.4b) that $E_a + E_b = -3J$ independently of ϕ . Therefore, ϕ can be varied continuously leaving the ground-state energy E_0 invariant. But, for example, the transformation $\phi \rightarrow \phi + \pi/2$ simply permutes the values $E_a \rightarrow E_b, E_b \rightarrow E_a$ due the fourfold symmetry of the system apparent from Fig. 1(a).

In addition, we see that

$$E_a^2 + E_b^2 = \begin{cases} 5J^2 & \text{for } \phi=0 \\ \frac{9}{2}J^2, & \text{for } \phi=\pi/4 \\ 5J^2, & \text{for } \phi=\pi/2. \end{cases} \quad (2.4c)$$

Thus, the variance of the bond-energy distribution is largest for the states given by $\phi=0 \pm n\pi/2$.

We note that the degree of *collinearity* or *anticollinearity* of the various ground states is not as obvious to see here as it is for exchange-coupled systems.² However, we can define the most collinear states to be those in which the spins can be divided into groups, within which interactions are most satisfied but between which they are least satisfied. Hence, the variation of the distribution of bond energies, or of the quantities $\hat{\mathbf{m}}_i \cdot \mathbf{h}_{ij}$ (where h_{ij} is the local field at site i due to the spin at site j), which measure the extent of alignment between a spin and its local field, is a good measure of the collinearity. From (2.4c) we can, therefore, interpret the states $\phi = \pm n\pi/2$ as being the most collinear and the states $\phi = \pi/4 \pm n\pi/2$ as being the most anticollinear.

B. Honeycomb lattice

Again, the one-parameter family of ground states can be parametrized by the angle ϕ as follows:

$$\theta_\alpha = (\phi + 2\pi/3, -\phi + 2\pi/3, \phi, -\phi, \phi - 2\pi/3, -\phi - 2\pi/3), \quad (2.2a')$$

where $\alpha = 1, \dots, 6$ for the six sites α in the magnetic unit cell of the honeycomb lattice. As for the square lattice, different ground states are again obtained from each other by rotating the two sets of dipoles (even α and odd α) by the same angle ϕ in opposite directions [see Fig. 2(a)]. Then we can write

$$\theta_\alpha - \theta_\beta = 2\phi - 2\pi m / 3 \quad (2.2b')$$

$$(E_a, E_b, E_c) = -J(1 + \cos^2\phi, 1 + \cos^2(\phi + \pi/3), 1 + \cos^2(\phi - \pi/3)). \quad (2.4a')$$

Their values for the various states are

$$(E_a, E_b, E_c) = \begin{cases} (-2, -\frac{5}{4}, -\frac{5}{4})J & \text{for } \phi=0 \\ (-\frac{7}{4}, -1, -\frac{7}{4})J & \text{for } \phi=\pi/6 \\ (-\frac{5}{4}, -\frac{5}{4}, -2)J & \text{for } \phi=\pi/3. \end{cases} \quad (2.4b')$$

In (2.4b') a bond energy of $-J$ corresponds to the most frustrated bond and one of $-2J$ corresponds to the least frustrated one. We see that $E_a + E_b + E_c = -\frac{9}{2}J$ for any ϕ and therefore ϕ can be varied continuously leaving the ground-state energy invariant. This gives rise to the continuous degeneracy of the ground-state manifold. But, in analogy with the square system, the transformation $\phi \rightarrow \phi + \pi/3$ simply permutes the values $E_a \rightarrow E_b$, $E_b \rightarrow E_c$, $E_c \rightarrow E_a$ because of the sixfold symmetry of the system apparent from Fig. 2(a). In addition, for this sys-

and, in this case, m labels the three kinds of bonds and its values are given by

$$m = \begin{cases} 1 & \text{for } (\alpha, \beta) = (3, 2) \text{ or } (1, 6) \text{ or } (5, 4) \\ 0 & \text{for } (\alpha, \beta) = (1, 2) \text{ or } (3, 4) \text{ or } (5, 6) \\ -1 & \text{for } (\alpha, \beta) = (1, 4) \text{ or } (5, 2) \text{ or } (3, 6), \end{cases} \quad (2.2c')$$

i.e., in general, m has a different value for each of the three bonds on any one site. Then (2.1) becomes, for the ground states of the honeycomb lattice (with $h=0$),

$$H = -\frac{JN}{12} \sum_{\alpha=1}^6 \sum_{m=[-1,0,1]} [\frac{3}{2} + \frac{1}{2}\cos(2\phi - 2\pi m/3)]. \quad (2.3')$$

As for the square system, the second term in (2.3') cancels out between the three nearest neighbors for any ϕ and the ground-state energy is found to be $E_0 = -\frac{9}{4}JN$ independent of ϕ thus once again resulting in an infinitely degenerate ground state. The relevant configurations for the honeycomb lattice $\phi=0, \pi/6$, and $\pi/3$ are shown in Fig. 2(a). One sees that the system has a discrete sixfold symmetry (for $h=0$) with respect to rotation in spin space of the even- and odd-numbered dipoles in opposite directions, by comparing the $\phi=0$ and $\pi/3$ states in Fig. 2(a). Again, one would expect this symmetry to remain after effects of thermal fluctuations and dilution are taken into consideration.

The difference between the continuous degeneracy and the discrete symmetry for this system can again be understood by considerations similar to those for the square lattice. There are three kinds of bonds a, b, c with the energy of the ground state given by

$$E_0 = \frac{N}{2}(E_a + E_b + E_c),$$

since here each site is connected to three others through a bond of each type [see Fig. 2(b)]. The bond energies are given by

tem, we see that

$$E_a^2 + E_b^2 + E_c^2 = \frac{57}{8}J^2 \quad (2.4c)$$

for all ϕ . Thus, unlike the square lattice, the variance of the bond distribution is the same for all the states. Therefore, from arguments given in Sec. II A, we cannot interpret particular states as being more collinear or anticollinear.

III. SELECTION BY THERMAL FLUCTUATIONS

Some of the ground states will have a higher density of states for low-energy excitations and will therefore be selected due to entropy considerations by thermal fluctuations. We need to calculate the selection term in the free energy as a function of the degeneracy parameter ϕ .

This is done by expanding in spin waves^{1,2,7} about the ground-state manifold. Then, the selection term can be minimized with respect to ϕ to find the selected ground state. Consider the effect of thermal fluctuations on the dipole configuration. Then

$$\boldsymbol{\mu}_i \rightarrow \boldsymbol{\mu}_i + \delta\boldsymbol{\mu}_i$$

that is, expanding in spin deviations $\theta_i \rightarrow \theta_i + \delta\theta_i$, we get

$$H \rightarrow H + \delta H$$

and (to lowest order in spin waves)

$$\delta H = \frac{1}{2} \sum_{i,j} \delta\theta_i A_{ij}(\phi) \delta\theta_j = \frac{1}{2} \sum_{\mathbf{q}} \delta\theta_{\mathbf{q}}^\dagger A_{\mathbf{q}}(\phi) \delta\theta_{\mathbf{q}}, \quad (3.1)$$

where we define

$$\delta\theta_{\mathbf{q}}^\dagger A_{\mathbf{q}}(\phi) \delta\theta_{\mathbf{q}} = \sum_{\alpha,\beta} (\delta\theta_{\mathbf{q}}^\dagger)_\alpha [A_{\mathbf{q}}(\phi)]_{\alpha\beta} (\delta\theta_{\mathbf{q}})_\beta.$$

One can calculate the entropy as a function of ϕ as follows: the partition function is given by

$$\begin{aligned} Z &= \prod_{\mathbf{q}} \left[\int_{-\infty}^{\infty} \delta\theta_{\mathbf{q}} \exp[-(1/2T)\delta\theta_{\mathbf{q}}^\dagger A_{\mathbf{q}}(\phi) \delta\theta_{\mathbf{q}}] \right] \\ &= \prod_{\mathbf{q}} \left[\frac{2\pi T}{\det A_{\mathbf{q}}(\phi)} \right]^{1/2}. \end{aligned}$$

Then the free energy per spin is

$$F = -T \ln Z \{\phi\} = \text{const} + \frac{T}{2} \sum_{\mathbf{q}} \ln[\det A_{\mathbf{q}}(\phi)] \quad (3.2)$$

and the entropy is

$$S(\phi, T) = - \left[\frac{\partial F}{\partial T} \right] = \text{const} - \frac{1}{2} \sum_{\mathbf{q}} \ln[\det A_{\mathbf{q}}(\phi)]. \quad (3.3)$$

From arguments given by Henley² and the form of the selection term in (3.2), we expect states with the widest dispersion in the "density of states" of $A_{\mathbf{q}}$ values (stiffness of spin mode q) over the Brillouin zone to be selected by thermal fluctuations. In real space, these are states with maximum variance of the bond-energy distribution, i.e., as mentioned in Sec. II A, states in which the spins can be divided into groups within which all couplings are the most satisfied but between which they are the least satisfied. Thus, in \mathbf{q} space the spin-fluctuation modes along the corresponding bonds will be soft and stiff, respectively. In Sec. II A, we interpreted these states as being *collinear*. We see from (2.4b) and (2.4c) that this is true for the $\phi = 0 \pm n\pi/2$ states of the square lattice but that for the honeycomb lattice [see (2.4c')] all states have the same variance. Thus, we can intuitively expect the states $\phi = 0 \pm n\pi/2$ to be selected by thermal fluctuations for the square lattice (and that is what we will find in Sec. III A) but we cannot say anything about the states selected for the honeycomb lattice, at least from dispersion arguments.

A. Square lattice

For the square system, $\alpha, \beta = 1, \dots, 4$, $A_{\mathbf{q}}(\phi)$ is a 4×4 matrix, and the $\delta\theta_{\mathbf{q}}$ are four-vectors since there are four

sites in the magnetic unit cell. The matrix is given by

$$A_{\mathbf{q}}(\phi) = \frac{J}{2} \begin{pmatrix} 3 & a\nu & 0 & b\mu \\ a\nu & 3 & b\mu & 0 \\ 0 & b\mu & 3 & a\nu \\ b\mu & 0 & a\nu & 3 \end{pmatrix}, \quad (3.4a)$$

where

$$a = 1 + \cos^2\phi, \quad b = 1 + \sin^2\phi; \quad (3.4b)$$

also

$$\mu(\mathbf{q}) = \cos q_x, \quad \nu(\mathbf{q}) = \cos q_y. \quad (3.4c)$$

The selection term to be minimized is

$$\sum_{\mathbf{q}} \ln[\det A_{\mathbf{q}}(\phi)].$$

First, let us consider the case of zero field ($\mathbf{h} = 0$). Evaluating the determinant one gets

$$\begin{aligned} \det A_{\mathbf{q}}(\phi) &= \left[\frac{J}{2} \right]^4 (3^4 - 18a^2 \cos^2 q_y - 18b^2 \cos^2 q_x \\ &\quad + a^4 \cos^4 q_y + b^4 \cos^4 q_x \\ &\quad - 2a^2 b^2 \cos^2 q_x \cos^2 q_y). \end{aligned}$$

Numerical evaluation of the selection term in the free energy per spin in (3.2) gives

$$\begin{aligned} \delta F_T &= \frac{T}{2} \int_{\text{BZ}} \frac{d^2 q}{(2\pi)^2} \ln[\det A_{\mathbf{q}}(\phi)] \\ &\simeq \frac{T}{2\pi^2} (34.28 - 0.42 \cos 4\phi - 0.01 \cos^2 4\phi). \quad (3.5) \end{aligned}$$

Thus, the free energy is minimized at $\cos 4\phi = 1$, i.e., $\phi = 0 \pm n\pi/2$, where $n = 0, 1, 2, 3$. This agrees with intuitive arguments from dispersion given earlier. The selection effect is small, since in (3.5), the term in $\cos 4\phi$ that is responsible for selection of a particular ϕ is small compared to the constant term. Hence, the effect of thermal fluctuations in the absence of an external field is to reduce the continuous degeneracy of the ground state to a four-fold discrete symmetry.

B. Honeycomb lattice

For the honeycomb system, $\alpha, \beta = 1, \dots, 6$, $A_{\mathbf{q}}(\phi)$ is a 6×6 matrix, and the $\delta\theta_{\mathbf{q}}$ are six-vectors corresponding to the six sites in the magnetic unit cell. In this case the matrix is given by

$$A_{\mathbf{q}}(\phi) = \frac{J}{2} \begin{pmatrix} \frac{3}{2} & a\mu & 0 & b\nu & 0 & c\pi \\ a\mu^* & \frac{3}{2} & c\nu^* & 0 & b\pi^* & 0 \\ 0 & c\nu & \frac{3}{2} & a\pi & 0 & b\mu \\ b\nu^* & 0 & a\pi^* & \frac{3}{2} & c\mu^* & 0 \\ 0 & b\pi & 0 & c\mu & \frac{3}{2} & a\nu \\ c\pi^* & 0 & b\mu^* & 0 & a\nu^* & \frac{3}{2} \end{pmatrix}, \quad (3.4a')$$

where

$$\begin{aligned} a &= 1 + \sin^2 \phi, \\ b &= 1 + \sin^2(\phi + \pi/3), \\ c &= 1 + \sin^2(\phi - \pi/3), \end{aligned} \quad (3.4b')$$

also,

$$\begin{aligned} \mu(\mathbf{q}) &= \exp(-iq_x/2 + i\sqrt{3}q_y/2), \\ \nu(\mathbf{q}) &= \exp(-iq_x/2 - i\sqrt{3}q_y/2), \\ \pi(\mathbf{q}) &= \exp(iq_x) \end{aligned} \quad (3.4c')$$

Evaluating and simplifying the determinant one gets

$$\det A_{\mathbf{q}}(\phi) = (J/2)^6 [T_1(\mathbf{q}) + T_2(\mathbf{q})\cos 6\phi + T_3(\mathbf{q})\cos^2 6\phi] \quad (3.5')$$

with

$$\begin{aligned} T_1(\mathbf{q}) &= 2^{-10} [251\,262F(\mathbf{q}) + 9801G(\mathbf{q})], \\ T_2(\mathbf{q}) &= -2^{-10} [2241F(\mathbf{q}) + 198G(\mathbf{q})], \\ T_3(\mathbf{q}) &= 2^{-10} [-3F(\mathbf{q}) + G(\mathbf{q})], \end{aligned}$$

where

$$\begin{aligned} F(\mathbf{q}) &= 6 - 4 \cos(3q_x/2) \cos(3\sqrt{3}q_y/2) - 2 \cos(3q_x), \\ G(\mathbf{q}) &= 6 - 4 \cos(9q_x/2) \cos(3\sqrt{3}q_y/2) - 2 \cos(3\sqrt{3}q_y). \end{aligned}$$

Therefore, $|T_3| \ll |T_2| \ll |T_1|$ for all \mathbf{q} except $\mathbf{q}=0$. For small $|\mathbf{q}|$, $F(\mathbf{q}) \simeq \frac{27}{2}|\mathbf{q}|^2$, and $G(\mathbf{q}) \simeq \frac{81}{2}|\mathbf{q}|^2$. Consequently, T_1 , T_2 , and T_3 all go to zero as $|\mathbf{q}|$ goes to zero. Clearly, $T_2(\mathbf{q})$, the coefficient of the selection term in ϕ , is always negative (its maximum value is 0 at $\mathbf{q}=0$). So, in analogy with the square system we see that the free energy is minimized at $\cos 6\phi = 1$, i.e., $\phi = 0 \pm n\pi/3$, where $n = 0, 1, \dots, 5$. Numerical evaluation of the selection term in the free energy per spin (3.2) using (3.5') gives

$$\begin{aligned} \delta F_T &= \frac{T}{2} \int_{\text{BZ}} \frac{d^2q}{\Omega} \ln[\det A_{\mathbf{q}}(\phi)] \\ &\simeq \frac{\sqrt{3}T}{4\pi^2} (86.74 - 0.11 \cos 6\phi - 0.00057 \cos^2 6\phi), \end{aligned} \quad (3.6')$$

where Ω , the volume of the Brillouin zone, $= 8\pi^2/\sqrt{3}$ for the honeycomb lattice and the integration is performed over a quarter of this Brillouin zone bounded by the points $(q_x, q_y) = (0, 0)$, $(0, 2\pi)$, and $(2\pi, 2\pi/\sqrt{3})$, where $(0, 0)$ is a center of symmetry.

Hence, on the honeycomb lattice, thermal fluctuations (in the absence of an external field) reduce the continuous degeneracy of the ground state to a sixfold discrete symmetry.

Moreover, if we calculate the strength of this selection effect by dividing the coefficient of $\cos 6\phi$ in the selection term by the constant term in (3.6') and compare with the corresponding strength of the selection effect on the square lattice from (3.5), we find that this effect is about ten times weaker on the honeycomb lattice. This seems

to be in accordance with the observations made just after (3.3).

IV. SELECTION BY EXTERNAL FIELD

In the fully occupied lattice at zero temperature, there will still be a selection among ground states due to an external field \mathbf{h} . It is convenient to write $\mathbf{h} = h(\cos\phi_h, \sin\phi_h)$.

Interestingly, we will find that, both in the square lattice (4.5) and in the honeycomb lattice (4.21), the field selection is strongest (either positive or negative) for the states which are equivalent to $\phi = 0$ in zero field; these are precisely the states selected by thermal fluctuations [see (3.5) and (3.6'), respectively]. On the other hand, for the other kind of special symmetry states, which are those selected by dilution (Sec. V, below), the coupling vanishes and the system is insensitive to the direction of the external field.

A. Square lattice

The Hamiltonian of (1.1a)–(1.1c) can also be written in the form

$$\begin{aligned} H/N &= -\frac{3}{4}(\boldsymbol{\mu}_1^* + \boldsymbol{\mu}_3^*) \cdot (\boldsymbol{\mu}_2^* + \boldsymbol{\mu}_4^*) - \frac{1}{4}(\boldsymbol{\mu}_1 + \boldsymbol{\mu}_3) \cdot (\boldsymbol{\mu}_2 + \boldsymbol{\mu}_4) \\ &\quad - \frac{1}{4}\mathbf{h} \cdot (\boldsymbol{\mu}_1 + \boldsymbol{\mu}_2 + \boldsymbol{\mu}_3 + \boldsymbol{\mu}_4), \end{aligned} \quad (4.1a)$$

where the vectors $\boldsymbol{\mu}_i^*$ are defined by

$$\boldsymbol{\mu}_i^* \cdot \boldsymbol{\mu}_j^* = \cos(\theta_i + \theta_j - 2\psi_{ij}) \quad (4.1b)$$

and therefore their directions are given by the angles $\theta_1^* = \theta_1$, $\theta_2^* = -\theta_2 + \pi$, $\theta_3^* = \theta_3 + \pi$, and $\theta_4^* = -\theta_4$, respectively, which satisfy

$$\theta_i^* - \theta_j^* = \theta_i + \theta_j - 2\psi_{ij}.$$

Now, once again expanding in spin deviations $\theta_i \rightarrow \theta_i + \delta\theta_i$ we get, apart from a constant and without the field,

$$\begin{aligned} \delta H_{\text{dip}}/N &= \frac{3}{8}(\psi_1^2 + \psi_2^2 + \psi_3^2 + \psi_4^2) \\ &\quad - \frac{3}{4}\psi_1\psi_2 + \frac{1}{4}(\cos 2\phi)\psi_3\psi_4, \end{aligned} \quad (4.2a)$$

where $\psi_1 = \delta\theta_1 + \delta\theta_3$, $\psi_2 = \delta\theta_2 + \delta\theta_4$, $\psi_3 = \delta\theta_1 - \delta\theta_3$, and $\psi_4 = \delta\theta_2 - \delta\theta_4$.

The field coupling term is

$$\begin{aligned} \delta H_h/N &= \frac{h}{4} [(\sin\phi \cos\phi_h - \cos\phi \sin\phi_h)\psi_3 \\ &\quad + (\sin\phi \cos\phi_h + \cos\phi \sin\phi_h)\psi_4]. \end{aligned} \quad (4.2b)$$

Thus, we see from (4.2a) and (4.2b) that the degeneracy parameter ϕ couples only to the modes ψ_3 and ψ_4 . Equations (4.2a) and (4.2b) can be combined and written in the form:

$$\delta H/N = \frac{1}{2}\psi^\dagger \bar{\chi}^{-1} \psi - \bar{h} \psi, \quad (4.3a)$$

where we have considered only the ϕ -dependent part, therefore $\psi = (\psi_3, \psi_4)$. The susceptibility matrix in (4.3a) is given by

$$\bar{\chi} = [(\frac{3}{4})^2 - (\frac{1}{4}\cos 2\phi)^2]^{-1} \begin{bmatrix} \frac{3}{4} & -\frac{1}{4}\cos 2\phi \\ -\frac{1}{4}\cos 2\phi & \frac{3}{4} \end{bmatrix} \quad (4.3b)$$

and the field that couples to ψ is

$$\tilde{h} = \frac{h}{4} (-\sin\phi \cos\phi_h + \cos\phi \sin\phi_h, -\sin\phi \cos\phi_h - \cos\phi \sin\phi_h). \quad (4.3c)$$

Now, minimizing (4.3a) with respect to the deviations ψ we get

$$\delta H/N = -\frac{1}{2}\tilde{h}\bar{\chi}\tilde{h}. \quad (4.4)$$

Evaluating (4.4) using (4.3b) and (4.3c) we finally get

$$\delta H/N = (9 - \cos^2 2\phi)^{-1} \frac{h^2}{J} \times (\cos 2\phi \cos 2\phi_h - \frac{1}{8}\cos 4\phi - \frac{7}{8}). \quad (4.5)$$

The second term in (4.5) is a fourfold selection of $O(h^2)$. The first term represents the reduction of the fourfold symmetry of the system to a twofold one by the introduction of an external field. Thus, in the presence of an external field the states selected are those given by $\cos 2\phi \cos 2\phi_h = -1$. Therefore, the selection effect is strongest for a field along a symmetry direction $\phi_h = n\pi/2$ and the degeneracy parameter ϕ couples to the field direction in such a way that for a field along the x axis, the states $\phi = \pi/2 \pm n\pi$ with $n=0,1$ are selected whereas for a field along the y axis it is the states $\phi = 0 \pm n\pi$ that are selected. This is exactly like the case of an exchange-coupled antiferromagnet in which the staggered magnetization turns perpendicular to a uniform applied field so that both sublattices can gain energy to second order by canting towards it.

$$v \equiv \frac{1}{\sqrt{6}} (e^{i\phi}, e^{-i\phi}, e^{i(\phi+2\pi/3)}, e^{-i(\phi+2\pi/3)}, e^{i(\phi+4\pi/3)}, e^{-i(\phi+4\pi/3)}). \quad (4.8)$$

The vector v in (4.8) is the analog of \tilde{h} in (4.3c). To $O(\delta\theta)$, from (4.6a), the coupling to the field is

$$\delta H_h = -\mathbf{h} \cdot \delta \mathbf{M}^{(1)} = -\text{Re} \left[\frac{i}{\sqrt{6}} h_- (v, \delta\theta) \right]. \quad (4.9)$$

The dipole energy difference to second order is given by

$$\frac{1}{2}(\delta\theta, A_0 \delta\theta),$$

where A_0 is (3.4a') evaluated at $\mathbf{q}=0$; thus, A_0 , is a real and symmetric 6×6 matrix. Fortunately, the six-vector v (with its independent conjugate v^*) is an eigenvector of A_0 :

B. Honeycomb lattice

In this section, we will expand the selection energy E_h due to an external field out to $O(h^3)$; we will verify that there is no selection effect to $O(h^2)$, the coupling is of $O(h^3)$ and has the symmetry $\cos 3\phi \cos 3\phi_h$.

It is not surprising that the adjustments to an external field preserve the unit cell; thus the spin configurations in a field are just described by the six-vector $\delta\theta$ with components equal to the deviations of the six sublattices, thus $\delta\theta \propto \delta\theta_{\mathbf{q}}$ at $\mathbf{q}=0$.

1. Linear response to external field

It will be convenient to combine the x and y components of the field and magnetization (per spin) into complex scalars,

$$M_+ \equiv M_x + iM_y,$$

$$h_+ \equiv h_x + ih_y,$$

with $M_- \equiv (M_+)^*$ and $h_- \equiv (h_+)^*$. In terms of M_+ the field-coupling term defined in (1.1c) is

$$H_h = -\mathbf{h} \cdot \delta \mathbf{M} = -\text{Re}(h_- M_+). \quad (4.6c)$$

As in the square lattice, we are working in the restricted $q=0$ manifold where each of the six sublattices has a constant angle. Then the magnetization is

$$M_+ = \frac{1}{6} \sum_{i=1}^6 e^{i\theta_i}. \quad (4.6b)$$

Then we find that to linear order, the magnetization (per site) is

$$\delta M_+^{(1)} \equiv \frac{1}{\sqrt{6}} i(v, \delta\theta) \quad (4.7)$$

using the notation for the scalar product of six-vectors, with $v_i = (1/\sqrt{6})e^{i\theta_i}$; that is,

$$A_0 v = \frac{15}{4} J v, \quad A_0 v^* = \frac{15}{4} J v^*. \quad (4.10)$$

The linear response is thus

$$\delta\theta = A_0^{-1} \text{Re} \left[\frac{i}{6} (h_- v) \right] = (\frac{15}{4} J)^{-1} \text{Re} \left[\frac{i}{\sqrt{6}} (h_- v) \right]. \quad (4.11)$$

As a corollary, the linear susceptibility is isotropic:

$$\delta M_+ = \frac{i}{\sqrt{6}} (v, \delta\theta) = \chi h_+ \quad (4.12a)$$

with

$$\chi = \frac{2}{45} J^{-1}. \quad (4.12b)$$

Thus, in contrast to the square-lattice results (4.3b) and (4.5), on the honeycomb lattice the magnetic energy $\frac{1}{2}\chi h^2$ is independent of ϕ and has no selection effect to $O(h^2)$.

2. Terms of $O(h^3)$

We must compute two kinds of contributions⁸ from $O(h^3)$ terms.

(i) In the field coupling (4.9), contributions to magnetization $\delta\mathbf{M}$ of $O(\delta\theta^2)$.

(ii) Contributions to the dipolar energy of $O(\delta\theta^3)$.

Into the higher-order terms, we need only substitute

$$\delta\theta = \frac{1}{2}(\alpha v + \alpha^* v^*) \quad (4.13a)$$

involving just the two eigenvectors, where Eq. (4.11) says

$$\alpha = \frac{4}{15J} \frac{i}{\sqrt{6}} h_-. \quad (4.13b)$$

(i) Contribution to $\delta\mathbf{M}$ of $O(\delta\theta^2)$. We find

$$\begin{aligned} \delta M_+ &= -\frac{1}{6} \frac{1}{2} \sum_i e^{i\theta_i} (\delta\theta_i)^2 \\ &= -\frac{1}{6} \frac{1}{2} [e^{i\phi} (e^{2\pi i/3} \delta\theta_1^2 + \delta\theta_3^2 + e^{4\pi i/3} \delta\theta_5^2) \\ &\quad + e^{-i\theta} (e^{2\pi i/3} \delta\theta_2^2 + \delta\theta_4^2 + e^{4\pi i/3} \delta\theta_6^2)]. \end{aligned} \quad (4.14)$$

Substituting (4.13a) into (4.14), one gets

$$\delta M_+ = -\frac{1}{48} \alpha^2 \cos 3\phi. \quad (4.15)$$

Using (4.6a) and (4.13b), the net energy is

$$\text{Re}(h_- \delta M_+) = -\frac{1}{(18)15^2} \frac{h^3}{J^2} \cos 3\phi \cos 3\phi_h. \quad (4.16)$$

(ii) Contribution from dipolar term of $O(\delta\theta^3)$. Taylor expanding the dipole energy (2.1), the third-order term is

$$\delta H_3 \equiv \frac{1}{6} \sum_{i=1,3,5} \sum_{j=2,4,6} (-\frac{1}{2}J) \sin(\theta_i - \theta_j) \frac{1}{3!} (\delta\theta_i - \delta\theta_j)^3. \quad (4.17)$$

The first term in (2.1) does not contribute, since $\sin(\theta_i + \theta_j - 2\psi_{ij}) \equiv 0$ in any ground state.

Observe that the coefficients of terms $(\delta\theta_i)^3$ all cancel when grouped from the terms in (4.17). Then noting that we can write $\sin(\theta_i - \theta_j) = \text{Im} e^{i(\theta_i - \theta_j)}$, we get

$$\begin{aligned} \delta H_3 &= \frac{-J}{24} \text{Im} \{ 3e^{2i\phi} [(\delta\theta_1^2 + e^{-2\pi i/3} \delta\theta_3^2 + e^{-4\pi i/3} \delta\theta_5^2)(\delta\theta_2 + e^{-2\pi i/3} \delta\theta_4 + e^{-4\pi i/3} \delta\theta_6)] \\ &\quad - 3e^{-2i\phi} [(\delta\theta_1 + e^{-2\pi i/3} \delta\theta_3 + e^{-4\pi i/3} \delta\theta_5)(\delta\theta_2^2 + e^{-2\pi i/3} \delta\theta_4^2 + e^{-4\pi i/3} \delta\theta_6^2)] \}. \end{aligned} \quad (4.18)$$

For $\delta\theta$ of form (4.13a), we get

$$\delta H_3 = -\frac{J}{12} (3^3) 2 \text{Re}(e^{3i\phi}) \text{Im} \left[\left[\frac{\alpha}{2\sqrt{6}} \right]^3 \right]. \quad (4.19)$$

Substituting (4.13b) gives

$$\delta H_3 = -\frac{1}{(6^2)15^3} \left[\frac{h^3}{J^2} \right] \cos 3\phi \cos 3\phi_h. \quad (4.20)$$

3. Total field selection term

Adding (4.16) and (4.20), the full field selection term is

$$\delta H/N = -c_h \cos 3\phi \cos 3\phi_h \quad (4.21a)$$

with

$$c_h = \frac{31}{(30)(18)15^2} \frac{h^3}{J^2}. \quad (4.21b)$$

It is mysterious why the contribution of (4.20) is only $\frac{1}{30}$

times that of (4.16).

The selection term (4.21a) reduces the continuous degeneracy present in zero field (even though the symmetry of the system is sixfold) to a threefold symmetry in the external field. In an external field, the selected states [in which (4.21a) is minimized] are those with ϕ given by $\cos 3\phi \cos 3\phi_h = +1$. The field selection is thus strongest when the field is along a symmetry direction $\phi_h = n\pi/3$. If the field points along the x axis, the states $\phi = 2\pi n/3$ ($n=0,1,2$) are selected; if the field is along the y axis there is no selection effect, and (contrary to the square lattice case) if we reverse the field, we also reverse the selected state.

Numerical mean-field calculations in Ref. 4 also calculated field selection for the honeycomb lattice shown in their Fig. 3. For \mathbf{h} in the $+x$ direction they found $\phi = \pi$, which is the same kind of state but opposite in direction [i.e., corresponds to opposite sign of c_h in (4.21a)]; we have no explanation for this discrepancy. For \mathbf{h} in the $+y$ direction they found a selection of $\phi = \pi/2$; in this case this may reflect the thermal selection in their mean-field theory, which generally does not give the right behavior at low temperatures.

V. SELECTION BY DILUTION

The Hamiltonian for the system with site dilution is given by (1.1b) with the substitution²

$$J_{ij} \rightarrow \epsilon_i \epsilon_j J_{ij} . \quad (5.1a)$$

Here, $\epsilon_i = 1$ or 0 depending on whether the site i is occupied or not. The ϵ_i are random and independent of each other, with $\langle \epsilon_i \rangle_\epsilon = p$, where p is the occupied fraction. Here, $\langle \rangle_\epsilon$ denotes an average over realizations. Then we can write

$$J_{ij} \rightarrow p^2 J_{ij} + \eta_{ij} J_{ij} , \quad (5.1b)$$

where

$$\eta_{ij} = \epsilon_i \epsilon_j - p^2 \quad (5.1c)$$

and thus $\langle \eta_{ij} \rangle_\epsilon = 0$.

Then, making the substitution (5.1b) and again expanding in spin deviations we get

$$\delta H_{\text{dil}} = p^2 \delta H + \delta H' , \quad (5.2a)$$

where δH is given by (3.1) as before, and

$$\begin{aligned} \delta H' = & \frac{J}{2} \sum_{\langle ij \rangle} \eta_{ij} [\cos(\theta_i - \theta_j) \\ & - 3 \cos(\theta_i - \psi_{ij}) \cos(\theta_j - \psi_{ij})] \\ & - J \sum_i \gamma_i \delta \theta_i \end{aligned} \quad (5.2b)$$

with

$$\gamma_i = \sum_{\langle j \rangle} \eta_{ij} [\sin(\theta_i - \theta_j) + 3 \cos(\theta_i - \psi_{ij}) \sin(\theta_j - \psi_{ij})] . \quad (5.2c)$$

In (5.2c), $\langle j \rangle$ indicates that the sum is over all j that are nearest neighbors of i .

Now, we have to minimize δH_{dil} with respect to the deviations $\delta \theta_q$, i.e., minimize the function

$$\frac{p^2}{2} \sum_q \delta \theta_q^\dagger A_q(\phi) \delta \theta_q - J \sum_q \gamma_q^\dagger(\phi) \delta \theta_q . \quad (5.3)$$

Minimization gives

$$\delta \theta_q = \frac{J}{p^2} \gamma_q^\dagger(\phi) A_q^{-1}(\phi)$$

and

$$\begin{aligned} \langle \delta H_{\text{dil}} \rangle_\epsilon &= - \frac{J^2}{2p^2} \sum_q \sum_{\alpha, \beta} \left\langle [\gamma_q^*(\phi)]_\alpha [A_q^{-1}(\phi)]_{\alpha\beta} [\gamma_q(\phi)]_\beta \right\rangle_\epsilon \\ &= - \frac{J^2}{2p^2} \int_{\text{BZ}} \frac{d^2 q}{\Omega} \sum_{\alpha, \beta} \langle [\gamma_q^*(\phi) \gamma_q(\phi)]_{\alpha\beta} \rangle_\epsilon [A_q^{-1}(\phi)]_{\alpha\beta} \end{aligned} \quad (5.4a)$$

with

$$\langle [\gamma_q^*(\phi) \gamma_q(\phi)]_{\alpha\beta} \rangle_\epsilon = \sum_{i \in \alpha} \sum_{k \in \beta} e^{-iq \cdot (\mathbf{r}_k - \mathbf{r}_i)} \sum_{\langle l \rangle} \sum_{\langle j \rangle} \langle \eta_{ij} \eta_{kl} \rangle_\epsilon X_{ij} X_{kl} . \quad (5.4b)$$

In (5.4a), Ω is the volume of the Brillouin zone and the integral is to be done over this Brillouin zone. In (5.4b), $\langle l \rangle$ and $\langle j \rangle$ indicate that the sums are over all l and j that are nearest neighbors of k and i , respectively, and

$$X_{ij} = \sin(\theta_i - \theta_j) + 3 \cos(\theta_i - \psi_{ij}) \sin(\theta_j - \psi_{ij}) \quad (5.4c)$$

and the correlation in (5.4b)² satisfy

$$\langle \eta_{in} \eta_{kl} \rangle_\epsilon = \begin{cases} p^2(1-p^2) & \text{if } i-j \text{ and } k-l \text{ are the same bond ,} \\ p^3(1-p) & \text{if bonds } i-j \text{ and } k-l \text{ have one site in common ,} \\ 0 & \text{if they have no site in common .} \end{cases} \quad (5.4d)$$

It is hard to predict from (5.4a) which states would be selected by dilution from dispersion arguments, as was done for thermal fluctuations earlier. However, earlier work^{1,2} leads us to expect dilution to select the states that are opposite to those selected by temperature, i.e., the states given by $\phi = \pi/4 \pm n\pi/2$ for the square lattice and $\phi = \pi/6 \pm n\pi/3$ for the honeycomb lattice, and that is indeed what we shall find in the following sections.

A. Square lattice

For this system, the $\delta \theta_q$ and the $\gamma_q(\phi)$ in Eqs. (5.3) and (5.4) are four-vectors. Equation (5.2b) can be written as

$$\delta H' = - \frac{J}{4} \sum_i \sum_{m=[0,1]} (-1)^m \eta_{im} \cos 2\phi - J \sum_i \gamma_i \delta \theta_i \quad (5.5a)$$

and γ_i can be written as

$$\gamma_i = -\frac{1}{2} \sum_{m=[0,1]} \eta_{im} (-1)^m \sin 2\phi . \quad (5.5b)$$

In (5.5a) and (5.5b), m labels the two kinds of bonds; each site is connected to four bonds—two of each kind. The quantity η_{im} is the same as η_{ij} defined in (5.1c) with i and j being nearest neighbors (NN) and m labeling the type of bond between them. Evaluation of expression (5.4b) using the Fourier transform of (5.5b) gives, for the square lattice (with $\delta p = 1 - p$),

$$\langle [\gamma_q^*(\phi) \gamma_q(\phi)]_{\alpha\beta} \rangle_\epsilon = \begin{cases} \frac{N}{8} p^3 \delta p \sin^2 2\phi (1 + \cos 2q_x + \cos 2q_y) + \frac{N}{4} p^2 \delta p \sin^2 2\phi & \text{if } \alpha = \beta \\ -\frac{N}{8} p^2 (\delta p)^2 \sin^2 2\phi \cos q_x & \text{if } \alpha \text{ and } \beta \text{ are NN and } m = 1 \\ -\frac{N}{8} p^2 (\delta p)^2 \sin^2 2\phi \cos q_y & \text{if } \alpha \text{ and } \beta \text{ are NN and } m = 0 \\ -\frac{N}{2} p^3 \delta p \sin^2 2\phi \cos(q_x / \sqrt{2}) \cos(q_y / \sqrt{2}) & \text{if } \alpha \text{ and } \beta \text{ are NNN,} \end{cases} \quad (5.6)$$

where NN and NNN denote first and second neighbors, respectively.

Numerical integration over the Brillouin zone and subsequent evaluation of the right-hand side of (5.4a) with $\Omega = (2\pi)^2$ for the square lattice then gives for the selection energy per spin

$$\langle \delta H_{\text{dil}} \rangle_\epsilon = -\frac{J}{\pi^2 p^2} [D_0(p) - D_1(p) \cos 4\phi - D_2(p) \cos^2 4\phi + \dots] \quad (5.7a)$$

with $D_1(p) > 0$ and thus the minimum occurs at $\cos 4\phi = -1$, i.e., $\phi = \pi/4 \pm n\pi/2$ with $n = 0, 1, \dots, 3$. Thus, dilution also results in replacing the continuously degenerate ground state by one with a fourfold discrete symmetry, but opposite to that selected by thermal fluctuations.

The coefficients $D_0(p)$, $D_1(p)$, and $D_2(p)$ in (5.7a) were found by numerically evaluating the values of expression (5.4a) for different values of the degeneracy parameter ϕ and of the occupied fraction p , and expressing the result as a polynomial in $\cos 4\phi$. The dependence on p up to $O(\delta p^2)$ was found to be

$$\begin{aligned} D_0(p) &\simeq 2.42(\delta p) - 5.34(\delta p)^2, \\ D_1(p) &\simeq 2.41(\delta p) - 5.34(\delta p)^2, \\ D_2(p) &\simeq 0.00295(\delta p) - 0.00766(\delta p)^2. \end{aligned} \quad (5.7b)$$

Note that the expansion in $\cos 4\phi$ and δp [(5.7a) and (5.7b)] must terminate at $O(\delta p^4)$ in view of (5.6) but goes to all orders in $\cos 6\phi$ due to $A_q^{-1}(\phi)$ in (5.4a). Thus, for a small amount of dilution δp , the selection free energy is given by

$$\delta F_D(\phi) = \frac{J\delta p}{\pi^2} (2.41) \cos 4\phi . \quad (5.7c)$$

Therefore, to $O(\delta p)$, the physical meaning of this is that

the selection free energy is a sum of independent contributions of isolated missing sites. If we got the term correct for one missing site, then we are correct to linear order and the energy should be accurate for moderate δp , up to the point where pairs of adjacent removed spins become significant. However, in reality there is no limit in which the deviations near the removed site go to zero, and so our coefficient would be wrong even for $\delta p \rightarrow 0$. Nevertheless, it is easy to show that *for these dipolar systems*, the deviations happen to be quantitatively small compared to 1. For example, take a spin, remove one of its neighbors, and freeze the rest, not allowing them to relax. Then the local field direction changes by 0 (for $\phi = 0$), and by, at most, $\tan^{-1}(\frac{1}{9}) = 0.111$ (for $\phi = \pi/4$). This shows that quantitatively it is a good approximation to linearize.

B. Honeycomb lattice

Here, the $\delta\theta_q$ and the $\gamma_q(\phi)$ in (5.3) and (5.4) are six-vectors. Further, for the honeycomb lattice, (5.2b) can be written as

$$\begin{aligned} \delta H' &= -\frac{J}{4} \sum_i \sum_{m=[-1,0,1]} \eta_{im} \cos(2\phi - 2\pi m/3) \\ &\quad - J \sum_i \gamma_i \delta\theta_i, \end{aligned} \quad (5.5a')$$

where

$$\gamma_i = -\frac{1}{2} \sum_{m=[-1,0,1]} \eta_{im} \sin(2\phi - 2\pi m/3) . \quad (5.5b')$$

Here, m labels the three kinds of bonds; each site is connected to three bonds—one of each kind. Evaluation of expression (5.4b) using the Fourier transform of (5.5b') gives, for the honeycomb lattice (with $\delta p = 1 - p$)

$$\langle [\gamma_q^*(\phi)\gamma_q(\phi)]_{\alpha\beta} \rangle_\epsilon = \begin{cases} \frac{N}{16}p^2\delta p & \text{if } \alpha=\beta, \\ -\frac{N}{24}p^2(\delta p)^2\sin^2(2\phi-2\pi m/3)e^{iq\cdot r_{\alpha\beta}} & \text{if } \alpha \text{ and } \beta \text{ are NN}, \\ \frac{N}{24}p^3\delta p \sum_\gamma \sin(2\phi-2\pi m/3)\sin(2\phi-2\pi n/3)e^{iq\cdot r_{\alpha\gamma}}e^{iq\cdot r_{\gamma\beta}} & \text{if } \alpha \text{ and } \beta \text{ are NNN}, \end{cases} \quad (5.6')$$

where m and n label the corresponding bonds $\alpha-\beta$, $\alpha-\gamma$, and $\gamma-\beta$ and their values are given by (2.2c'). In general, m is equal to $-1, 0$, and 1 , respectively, for the three bonds connected to any one site.

Again, numerical integration over a quarter of the Brillouin zone and subsequent evaluation of the right-hand side of (5.4a) with the volume of the Brillouin zone given by $\Omega=8\pi^2/\sqrt{3}$ as before for the honeycomb lattice gives, for the selection energy per spin,

$$\langle \delta H_{\text{dil}} \rangle_\epsilon = -\frac{\sqrt{3}J}{2\pi^2 p^2} [D_0(p) - D_1(p) \cos 6\phi - D_2(p) \cos^2 6\phi + \dots] \quad (5.7a')$$

with $D_1(p) > 0$ and therefore the minimum occurs at $\cos 6\phi = -1$, i.e., $\phi = \pi/6 \pm n\pi/3$ with $n = 0, 1, \dots, 5$.

Thus, we conclude that, for the honeycomb lattice also, dilution replaces the continuously degenerate ground state by one with a discrete sixfold symmetry, but opposite to that selected by thermal fluctuations.

As for the square case, the coefficients $D_0(p)$, $D_1(p)$, and $D_2(p)$ in (5.7a') were found by numerically evaluating the values of expression (5.4a) for different values of the degeneracy parameter ϕ and of the occupied fraction p , and fitting to a polynomial in $\cos 6\phi$. The dependence on p up to $O(\delta p^2)$ was found to be

$$\begin{aligned} D_0(p) &\simeq 1.46(\delta p) - 2.19(\delta p)^2, \\ D_1(p) &\simeq 0.13(\delta p) - 0.53(\delta p)^2, \\ D_2(p) &\simeq 0.00166(\delta p) - 0.00582(\delta p)^2. \end{aligned} \quad (5.7b')$$

For the same reasons as the square case, the expansion [(5.7a'), and (5.7b')] terminates at $O(\delta p^4)$ but goes to all orders in $\cos 6\phi$. Also, for small δp we have

$$\delta F_D(\phi) = \frac{\sqrt{3}J\delta p}{2\pi^2} (0.13) \cos 6\phi. \quad (5.7c')$$

Thus, again to $O(\delta p)$ the free energy consists of a sum of independent contributions to isolated missing sites. Also, as for the square case, we can argue for the validity of our expansion that the deviations near the removed site are small. Again, removing one neighbor site and freezing the rest, we find the maximum change in the local field to be

$$\tan^{-1}(\sqrt{3}/13) = 0.132$$

for ($\phi=0$) and

$$\tan^{-1}(\sqrt{3}/11) = 0.156$$

(for $\phi=\pi/6$). Thus, for the honeycomb lattice linearizing is also a good approximation.

In addition, we infer from calculating $D_1(p)/D_0(p)$ from (5.7b') and comparing with the same quantity in (5.7b) that the selection effect due to dilution is about ten times smaller here than it is for the square lattice, as was the case with the selection effect due to thermal fluctuations.

VI. EFFECTIVE RANDOM AXIS

Now consider the first term in Eq. (5.2b). For the square lattice, we can write this term as [see (5.5a)]

$$-\frac{J}{4} \sum_i \sum_{m=[0,1]} (-1)^m \eta_{im} \cos 2\phi = \tilde{H} \cos 2\phi, \quad (6.1a)$$

where

$$\tilde{H} = -\frac{J}{4} \sum_i \sum_m (-1)^m \eta_{im}. \quad (6.1b)$$

For the honeycomb lattice, we get [see (5.5a')]

$$\begin{aligned} -\frac{J}{2} \sum_i \left[\cos 2\phi (\eta_{i,0} - \frac{1}{2}\eta_{i,1} - \frac{1}{2}\eta_{i,-1}) + \sin 2\phi \left(\frac{\sqrt{3}}{2} (\eta_{i,1} - \eta_{i,-1}) \right) \right] &= \tilde{H}_1 \cos 2\phi + \tilde{H}_2 \sin 2\phi \\ &= \tilde{H} \cos 2(\phi - \phi_0), \end{aligned} \quad (6.1a')$$

where

$$\begin{aligned}\tilde{H}_1 &= -\frac{J}{2} \sum_i (\eta_{i,0} - \frac{1}{2}\eta_{i,1} - \frac{1}{2}\eta_{i,-1}), \\ \tilde{H}_2 &= -\frac{J}{2} \sum_i \frac{\sqrt{3}}{2} (\eta_{i,1} - \eta_{i,-1}), \\ \tilde{H} &= (\tilde{H}_1^2 + \tilde{H}_2^2)^{1/2}, \\ \phi_0 &= \frac{1}{2} \tan^{-1}(\tilde{H}_2/\tilde{H}_1).\end{aligned}\tag{6.1b'}$$

The terms given by (6.1a) and (6.1a'), respectively, for the two systems considered, represent the energy contribution due to the possibility of constrained⁹ rigid rotation of the two groups of dipoles with respect to each other, assuming perfect antiferromagnetic alignment within each group.

This term is the twofold analogue of *random exchange fields*^{2,10,11} which couple to the degeneracy degrees of freedom (in this case ϕ) in the same way as an ordinary random field couples to spin directions. They are unlike ordinary random fields in that they do not break the global rotation symmetry of the system (however, in the systems considered here, there is no global rotation symmetry, so this fact is not relevant). These random exchange fields couple to the degeneracy parameter, and, as with ordinary random fields, result in random local selection of one of the degenerate ground states (i.e., one value of ϕ) resulting in the formation of domains. In the systems considered in this paper, however, the corresponding term is twofold in ϕ . Thus, its effect is to disorder a discrete symmetric state by random local selection of a particular *axis*, resulting in the formation of a domain state with local order within each domain given by $\phi, \phi + \pi$, where ϕ is one of the original four equivalent ground-state configurations of the system for the square case, or one of the six equivalent configurations for the honeycomb case. Hence, we shall refer to it as an *effective random axis*.

For the honeycomb lattice, it can disorder either of the two kinds of selected states (the kind selected by temperature or the kind selected by dilution). For the square lattice the direction of (6.1a) is nonrandom. Consequently, the effective random axis affects only the temperature-selected states since the random axis term (6.1a) is proportional to $\cos 2\phi$ which is zero in the dilution selected states. Note that, in any case, a discrete twofold symmetry is still preserved in the presence of the effective random axis.

In any ground state, every site is equivalent to every other site by some symmetry operation. Also, an isolated impurity makes one missing bond in each bond direction; it is easy to see that it makes a zero net contribution to (6.1b) and (6.1b'). Consequently, as observed by Fernandez,¹⁰ only pairs contribute towards the effective random axis, i.e., the net contribution to \tilde{H} in (6.1b) and (6.1b') is of $O(\delta p^2)$.

Since discrete random fields are known to have a lower critical dimension of 2, the system is expected to be marginally disordered. Thus, we do not expect any long-range order to be present in either kind of selected state of the honeycomb lattice or in the temperature-selected

state of the square lattice. The domain state will be invariant under the global transformation $\phi \rightarrow \phi \pm \pi$ due to the remaining twofold symmetry. However, the dilution selected state for the square lattice should have long-range order since the random axis has no effect on this state.

VII. PHASE DIAGRAM

A. General results

1. Finite-temperature behavior of pure systems

With the information we have, we can make some crude conjectures about the phase diagram at finite temperatures. Our starting point is the long-wavelength free energy appropriate at low temperatures. It is of the form

$$F = \int d^2\mathbf{x} \left[\frac{1}{2} K_{\lambda\mu}(\phi, T) (\nabla_\lambda \phi) (\nabla_\mu \phi) + \sum_m h_m(T) \cos[m\phi(\mathbf{x})] \right]. \tag{7.1}$$

Equation (7.1) is similar to the free energy of an *XY* ferromagnet with an m -fold anisotropy. In that case, ϕ was the spin angle, whereas here it represents instead the degeneracy parameter. Just as the exchange symmetry of the *XY* ferromagnet leads to a squared gradient ("Goldstone mode"), so here the degeneracy leads to a squared gradient form at $T=0$. The additional m -fold terms are typically a temperature-independent m -fold anisotropy in the *XY* ferromagnet, while in the dipolar systems they come from thermal or dilution selection terms. The first h_m terms are, respectively, h_4 for the square case and h_6 for the honeycomb case.

We can apply to (7.1) the apparatus of Kosterlitz-Thouless critical phenomena imitating José *et al.*¹² One difference is that, in the ferromagnet, the interactions are isotropic exchange apart from the weak perturbations h_m , so that the spin stiffness tensor K just couples to $|\nabla\phi|^2$; then, since the unperturbed ground states are related by a symmetry, the stiffness has the same value in each. However, when the soft degree of freedom is a degeneracy rather than a symmetry, K may be anisotropic with respect to the gradient directions in real space, and also its value may depend on ϕ , i.e., some ground states are stiffer than others. Note that by symmetry, the spin stiffness tensor must have the rotational symmetry of the spatial part of the symmetry group of the spin structure. Also, at finite temperature $K \rightarrow K_R(T)$, which includes renormalization due to vortices which decrease the stiffness. In fact, the importance of this renormalization of K_R (and hence of the value T_K) decreases with the ratio of the spin stiffness to the vortex core energy.

The result of José *et al.* is that (assuming fully isotropic, ϕ -independent K) a Kosterlitz-Thouless transition occurs to a state with algebraically decaying rotational (ϕ) correlations at the temperature T_K when

$$K_R(T_K)/T_K = 2/\pi, \tag{7.2}$$

where K_R is the renormalized value of K .

Furthermore, at a temperature T_m given by

$$K_R(T_m)/T_m = m^2/8\pi, \quad (7.3)$$

the degeneracy-breaking field h_m becomes relevant in a renormalization-group sense. If the stiffness is not greatly renormalized, (7.3) implies

$$T_m \approx (16/m^2)T_K. \quad (7.4)$$

For $T < T_m$, an arbitrarily small value of h_m will result in the system locking into a phase with a clock order parameter, i.e., m degenerate ordered phases with $\phi = 2\pi n/m$ for $n = 1, \dots, m$, and possessing long-range order. If $m < 4$, we see on comparing (7.2) and (7.3) that the locking would occur above T_K , so that the paramagnet goes directly into the locked state.

Note how the m -fold locking depends principally on $K_R(T)$, not on the value of $h_m(T)$. Thus, although $h_m(T)$ has a different T dependence than in the usual model,¹² this does not matter since the $K_R(T)$ should have a similar T dependence. Indeed, if h_m is large at the transition point, this can raise the value of T_m and affect the (continuously variable) exponents.

In the same spirit, we can use the results of Sec. IV to speculate about the universality class of the phase transition in the presence of a finite field. On both square and honeycomb lattices, this has the effect $m \rightarrow m/2$ so that h_m now becomes highly relevant, giving critical properties of the corresponding discrete system rather than the Kosterlitz-Thouless behavior.

2. Competition between thermal and dilution selection

We have seen in Secs. III and IV that temperature and dilution select different ground states, i.e., different values of ϕ ; in both cases (square lattice and honeycomb lattice) preserving the discrete symmetry (for zero external field) of the system. In general, at nonzero temperature and dilution, the two kinds of selection effects will compete with each other and one can determine the kind of ground state selected at small $(T, \delta p)$ by considering the sum of the selection terms in the free energy due to thermal fluctuations and that due to dilution (there will be cross terms as well, but we expect them to be of higher order) and finding the value of ϕ for which it is a minimum. The form of the phase diagram is given in Fig. 3. The general form for the line of transition between thermal and dilution selected states is then

$$\delta F_T(\phi_1) + \delta F_D(\phi_1) = \delta F_T(\phi_2) + \delta F_D(\phi_2), \quad (7.5)$$

where ϕ_1 and ϕ_2 are the states selected by temperature and dilution, respectively. For small $(T, \delta p)$ this reduces to a linear relationship between T and δp as we shall see in Secs. VII B 2 and VII C 2 below.

3. Effects of strong dilution

With dilution, we generate an effective-random-axis (ERA) anisotropy (Sec. VI). If the axis is fully random in the sense of being isotropically distributed in space, then in $d < 4$ it will disorder systems with a continuous degree of freedom, even if it is arbitrarily small.¹³

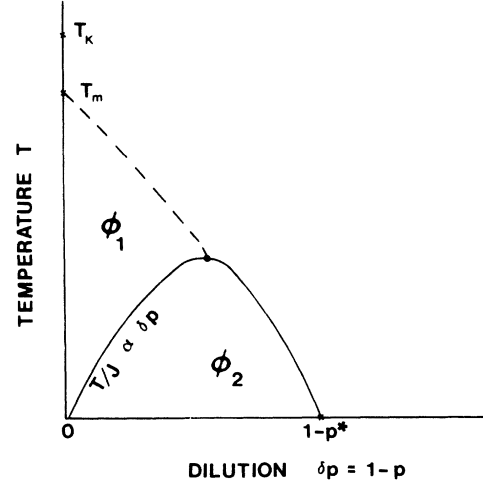


FIG. 3. Phase diagram of the system: ϕ_1 and ϕ_2 denote the temperature-selected and dilution-selected phases, respectively. The temperature T_K is the Kosterlitz-Thouless ordering temperature for the pure system $\delta p = 0$. The dotted line denotes the transition from the Kosterlitz-Thouless phase to the discrete locked phase, which takes place at T_m for the pure system. Note that $T_m = T_K$ for the square lattice. The ordering temperature of the phase ϕ_2 goes to zero at a value of the occupied fraction $p = p^*$ which is, in general, slightly above the percolation threshold for a frustrated system. The proportionality constants in the equation of the line of transition from phase ϕ_1 to ϕ_2 are 11.5 and 2.4 for the square and honeycomb lattices, respectively.

The effective random axes do not render (7.5) meaningless; as we will show next, near $(T, \delta p) = (0, 0)$ we can have arbitrarily long correlation lengths, despite the random axes. As noted in Sec. VI, the random axes come only from *pairs* of diluted sites and consequently the ERA's scale as

$$\tilde{H} \sim (\delta p)^2. \quad (7.6)$$

On the other hand, following the Imry-Ma arguments, (7.6) is to be compared with the surface energy σ per unit length of a domain wall between two of the discrete selected states. In this case σ is analogous to the wall energy of a Bloch wall, with the anisotropy being replaced by the selection energy δF :

$$\sigma \sim (\delta FK)^{1/2} \sim \delta p^{1/2} \quad (7.7)$$

since the stiffness K is independent of δp , to lowest order. Finally, it is believed^{13(b)} that the correlation length for a discrete random-field system diverges exponentially for small fields. Translating to the effective random anisotropy, and inserting (7.6) and (7.7), we have

$$\xi \sim \exp[\text{const}(\sigma/\tilde{H})^2] \sim \exp[\text{const}/(\delta p)^3]. \quad (7.8)$$

Thus, along any line radiating from $(\delta p, T) = (0, 0)$ as we approach $(\delta p, T) = (0, 0)$, we must approach arbitrarily closely to genuine long-range order.

We note here that the effect of the random axes is to add *random* anisotropies to the uniform ones in (7.1). We have described our system by treating the random aniso-

otropies as a perturbation on the uniform ones. This treatment is valid for high T and small δp since the random axes are of $O(\delta p^2)$ and the uniform selection effects are $O(T)$ and $O(\delta p)$ for thermal and dilution selection, respectively. The case of the XY model with random p -fold anisotropy has been treated by Goldschmidt and Schaub.¹⁴ This would be relevant to our model only in the portion of the phase diagram where a floating phase existed (see Sec. VII C 3). Elsewhere, the system could be described by adding the random axes to a discrete model rather than an XY model, as we have done. If we then go to higher dilution, the random axes will dominate the uniform selection and the system will be affected by vortices. Nevertheless, even with dilution, we still have the twofold symmetry under inversion of all spins as in the starting Hamiltonian (which is why we call it a “random axis,” not a “random field”). Random anisotropy ferromagnets develop order like an Ising spin-glass¹⁵ and we expect the same for these systems at $T=0$. Since spin glasses do not have long-range order in $d=2$, the main physical content of this observation is that the critical exponents of this zero-temperature spin-glass transition should be the same as those of the 2D Ising spin glass. Thus, unlike the case of Ref. 14, we still expect to have a spin-glass-like ground state at high dilution due to this twofold symmetry.

B. Square lattice

1. Pure system at $T > 0$

In the square lattice case, all the bonds in the x direction are equivalent (e.g., via a $[1,0]$ shift coupled with a reflection in spin space about the x axis), similarly all bonds in the y direction are equivalent, so $K_{xy}=0$ but possibly $K_{xx} \neq K_{yy}$. In fact,

$$K_{xx}a^2 = \frac{3}{2} - \frac{1}{2}\cos 2\phi, \quad (7.9a)$$

$$K_{yy}a = \frac{3}{2} + \frac{1}{2}\cos 2\phi. \quad (7.9b)$$

Note that K_{xx} and K_{yy} are the same as b and a , respectively, of (3.4b), which label the corresponding bonds in Fig. 1(b).

If we ignore the anisotropic stiffness for a moment, we expect a transition directly from the paramagnet into a state with a discrete order, since the selection terms have $m=4$ which is the marginal case where $T_m = T_K$. The universality class is that of the four-state clock model in $d=2$.

Now, the anisotropy in the gradient term is reminiscent of that appearing in models of 2D melting.^{16,17} In that case, the Kosterlitz-Thouless-type renormalization group must follow two stiffness constants, but they renormalize together and the gross nature of the transition is unchanged; we expect the same occurs here. In the presence of a finite field, we have a twofold selection, and thus we expect a crossover to Ising exponents at the Néel temperature.

2. Competition between thermal and dilution selection

The equation of the transition line between the two kinds of selected ground states (given by $\phi=0$ and $\pi/4$, respectively) is thus, in the notation of Secs. III and V,

$$\delta F_T(0) + \delta F_D(0) = \delta F_T(\pi/4) + \delta F_D(\pi/4). \quad (7.10)$$

From (3.5) and (5.7c) we get, to $O(\delta p)$

$$\delta F_T(\phi) = -\frac{T}{2\pi^2}(0.42)\cos 4\phi,$$

$$\delta F_D(\phi) = \frac{J\delta p}{\pi^2}(2.41)\cos 4\phi.$$

Thus, the Eq. (7.10) reduces to

$$\frac{T}{J} \simeq 11.5\delta p. \quad (7.11)$$

The form of the phase diagram, including this transition line, is given in Fig. 3.

3. Strong-dilution effects

As noted in Sec. VI, dilution creates effective random axes. This will certainly disorder the thermally selected state. Thus, the phase ϕ_1 in Fig. 3 does not possess long-range order for the square lattice and the dotted line indicates that this is not a true transition. However, in the dilution-selected state, one sees that the selected angles $\phi = n\pi/2 + \pi/4$ are precisely those where the random axis is zero. One can see more rigorously that precisely four states are degenerate, since reflection about either the x or y axes in spin space is an exact symmetry in any realization of the dilution. Thus, the situation is precisely analogous to that in the $J_1 - J_2$ antiferromagnet in the anticollinear phase.² Furthermore, if we represent this as a four-state clock model, there is *no frustration*. That is, say we restrict it to a discrete four-state clock model by allowing only the directions $(\pm 1, \pm 1)/\sqrt{2}$. Then it is easy to check that there is no frustration, and the system looks like a diluted four-state clock model, rather than a random-field four-state clock model. This situation could be compared to an XY ferromagnet with random fields only in the $\pm y$ direction. Then the spins would order in the $\pm x$ direction.¹⁸ One could then either say that the order parameter is orthogonal to the random fields or that there are two states which are exactly degenerate under reflection about the y axis (in spin space).

Thus, this phase (denoted as ϕ_2 in Fig. 3) is a true phase, locked into discrete long-range order. With dilution, its Néel temperature first increases linearly [as follows from (7.11) above] and later decreases (see Fig. 3). It goes to zero at an occupation p^* which must be above the site percolation threshold $p_c \approx 0.59$.

One expects that p^* is close to or equal to p_c . Each pairwise dipolar interaction is independently satisfied in this ground state, so in a sense there is no frustration.

C. Honeycomb lattice

1. Pure system at $T > 0$

In the honeycomb case, we have a threefold rotational symmetry of the ground states; that is, a rotation of the lattice by $2\pi/3$ coupled with the same rotation in spin space and a certain translation, gives the same ground state, for any state in the ground-state manifold. The only two-tensors with a three-fold rotation symmetry are isotropic, hence we have the form $K_{\lambda\mu} = K(\phi)\delta_{\lambda\mu}$. However, now we must extract $K(\phi)$, for example, by solving the eigenvalue equation for the matrix (3.4a) since the lowest root is

$$\sum_{\lambda\mu} K(\phi)q^2$$

to $O(q^2)$. The result is

$$K(\phi) = K_0 + K_6 \cos 6\phi$$

at $T=0$, where $K_0 = \sqrt{3}J/2$. The K_6 term is less relevant than the h_6 term so we will ignore it from here on.

Taking the approximation of bare stiffness values, $K_R = K_0$, we would have a Kosterlitz-Thouless transition at $T_K = \sqrt{3}\pi J/4$, from (7.2). For the thermal selection in the honeycomb case we found $h_6 \sim 10^{-2}$ from (3.6') so it has a negligible effect in renormalizing T_K . On the other hand, the vortex core energy should be especially low on this lattice since the vortex core can sit in the middle of a hexagon; this will renormalize T_K well below our estimate from the bare stiffness.

In the same approximation using $K_R = K_0$, we expect $T_6 \approx \frac{4}{9}T_K$ from (7.4). Between T_6 and T_K (for $\delta p = 0$), we expect a ‘‘floating’’ phase with algebraically decaying correlations, and below T_6 a locked phase with six discrete clock states and true long-range order.

In the presence of a finite field, Sec. IV B gave a threefold selection and so we expect the critical behavior at the Néel temperature crosses over to the three-state Potts model.

2. Competition between thermal and dilution selection

The equation of the transition line between the two kinds of selected ground states $\phi=0$ and $\pi/6$ is given by

$$\delta F_T(0) + \delta F_D(0) = \delta F_T(\pi/6) + \delta F_D(\pi/6). \quad (7.10')$$

Again, from (3.6') and (5.7c') we get, for small δp ,

$$\delta F_T(\phi) = -\frac{\sqrt{3}T}{4\pi^2}(0.11)\cos 6\phi,$$

$$\delta F_D(\phi) = \frac{\sqrt{3}J\delta p}{2\pi^2}(0.13)\cos 6\phi,$$

and thus Eq. (7.10') reduces to

$$\frac{T}{J} \simeq 2.4\delta p. \quad (7.11')$$

This transition line is shown in the phase diagram (Fig. 3).

3. Strong-dilution effects

In this case both kinds of selected states are affected by effective random axes. Thus, neither of the phases ϕ_1 and ϕ_2 in Fig. 3 possess long-range order. Consequently, there are no true phase transitions off the $T=0$ and $\delta p=0$ axes; we still expect a crossover in the nature of the local order along the line that is given by (7.11') above.

As we observed in Sec. VII C 1 above, along the $\delta p=0$ axis for the honeycomb lattice, there will be a ‘‘floating’’ phase for $T_K > T > T_m$. Thus, here a small amount of disorder would properly be described by adding random fields to an XY model rather than to a discrete model. For this part of the phase diagram, from arguments of Goldschmidt and Schaub,¹⁴ we expect that any small dilution would disorder the system.

VIII. DISCUSSION

We conclude from Secs. III A and V A that for the square system, both thermal fluctuations and dilution are found to reduce the continuous degeneracy of the ground state to a discrete fourfold symmetry while for the honeycomb system, from Secs. III B and V B, both reduce it to a discrete sixfold symmetry. For both systems, temperature and dilution are found to select *opposite* kinds of ground states, as was also observed for exchange-coupled XY and Heisenberg systems.^{1,2,19} Moreover, we conclude that the selection effects due to temperature and dilution are both ten times smaller on the honeycomb lattice than on the square lattice, which seems to corroborate intuitive arguments from dispersion (see Sec. III), at least for the case of thermal fluctuations. In Sec. IV we find that there is an additional selection effect due to the introduction of an external field even at $T=0$. This selection is found to be twofold for the square lattice and threefold for the honeycomb lattice, a result that agrees with expectations from intuitive symmetry arguments. From Sec. VI we see that the effective random axis induced by dilution disorders either kind of ground state by the formation of domains of the different allowed configurations, but the system retains a twofold symmetry in its presence. In Sec. VII, we deduced the form of the phase diagram for both systems by estimating the ordering temperature T_K ($\delta p=0$) for the pure system and by calculating the equation of the transition line between the two kinds of selected ground states in the temperature versus dilution plane.

We need to emphasize here that this system differs qualitatively from systems with exchange interactions which have been found to exhibit similar selection effects.^{1,2,7,19,20,21} The continuous degeneracy in this case does not correspond to an *independent* rotation of different sublattices as occurs in some systems with degeneracies.^{2,19} More fundamentally, there is no additional global rotation symmetry here.

Intuitive arguments,^{1,2} for exchange-coupled systems predict that thermal fluctuations would select the collinear states and dilution the anticollinear ones. It is not obvious what the analogs should be here. For an isolated dipolar coupled pair of spins, it can be deduced by expanding around (2.1) that fluctuations do have zero cou-

pling when one spin is parallel and the other is perpendicular to the lattice vector between them so collinearity is a somewhat useful concept. However, in the honeycomb lattice, the selection is frustrated in that we cannot achieve this condition for all pairs at the same time. The proper generalization of the concept of collinear state is that it maximizes couplings between deviations of neighboring spins; the generalization of “antcollinear” states² is that it maximizes torques on spins due to dilution. This motivated our generalized measure of “collinearity” (see Sec. II A) and this successfully identified the selected states for the square lattice. But for the honeycomb lattice all states were found to be equivalent with respect to this measure. Thus, it is an interesting mystery why dilution and temperature compete at all on the honeycomb lattice, since naive arguments from dispersion or collinearity (see Secs. II B, III, and IV) fail to predict any selection effect. [There is another anisotropic system where the selection is equally mysterious, the triangular Heisenberg²² or XY (Ref. 23) magnet with exchange anisotropy. In that case, the analog of our (3.2) gives no selection at all, but instead a selection at $O(\exp(-C/T))$ seems to be found.²³]

We expect similar effects for a dipolar magnet on any two-dimensional *bipartite* lattice in which all sites are at the same distance from their nearest neighbors and all have the same environment. Therefore, we would not expect to see this on, for example, the triangular lattice (since it is not bipartite). It would be interesting to see an eightfold selection, which we would expect to see on an octagonal lattice, but the only example that comes to mind is the eightfold Penrose tiling, on which we do not expect to see it because all sites do not have the same environment.

The study of the dipolar magnet on the honeycomb lattice is relevant to recent experiments on FeCl_3 -graphite intercalated compounds.^{6,24} The intercalate consists of finite-sized layers of FeCl_3 (with the Fe^{3+} ion arranged on

a honeycomb lattice and interacting with each other via the dipolar interaction) forming “islands” between the graphite layers. The finite size and random shape of these islands could have additional random-field-like effects: (a) Different islands of a layer, being disjoint from each other, would, in general, independently select a different ground-state configuration out of the various allowed ones and would be affected differently by an external magnetic field. (b) The degeneracy of the ground state (in the bulk) is related to cancellation of ϕ -dependent terms between the various neighbors and would not hold if we took boundary effects into account. Moreover, since the boundary is expected, in general, to be irregular, one can expect different configurations to be selected at different parts of the boundary. Thus, for sites near the boundary, one can expect random-field-like local selection effects.

Real systems would be expected to have an exchange interaction in addition to the dipolar interaction considered in this paper and a relevant question to ask would be how this would affect our results. We expect our results to be unchanged with the inclusion of a small nearest-neighbor exchange interaction. The reason for this is that, in any of the ground states, the local fields of the exchange interaction due to the nearest neighbors of a spin cancel each other out, thus leaving intact the continuous degeneracy of the ground state and, we would expect, most of the other properties.

ACKNOWLEDGMENTS

We thank G. O. Zimmermann and S. Redner for (respectively) inspiring us to study the honeycomb and the square lattice systems. S. P. would like to thank H. E. Stanley for support and encouragement. This work has been supported by the U. S. Department of Energy through Grant No. DE-FG02-ER89-45405.

*Present address.

¹C. L. Henley, *J. Appl. Phys.* **61**, 3962 (1987).

²C. L. Henley, *Phys. Rev. Lett.* **62**, 2056 (1989).

³J. Villain, R. Bidaux, J. P. Carton, and R. Conte, *J. Phys. (Paris)* **41**, 1263 (1980).

⁴R. Bidaux, J. P. Carton, and J. Villain, in *Disordered Systems and Localization*, Vol. 149 of *Lectures Notes in Physics*, edited by C. Castellani *et al.* (Springer, Berlin, 1981); A. Danielian, *Phys. Rev.* **13**, A1344 (1964).

⁵Note that we have a three-dimensional dipole interaction even though the system is two dimensional. It can easily be verified that the ground states have spins lying in the plane: states where they tilt out of the plane are energetically unfavorable. Furthermore, when we consider fluctuations about these ground states, the $\delta\mu_z$ fluctuations are not coupled to the $(\delta\mu_x, \delta\mu_y)$ fluctuations. The $\delta\mu_z$ spin waves also have an energy cost that is independent of the parameter ϕ , so they would not contribute to the selection terms computed in Secs. III and IV. Therefore, we do not lose any of the physics by confining the dipoles to the XY plane and shall do so henceforth.

⁶G. O. Zimmerman, A. K. Ibrahim, and F. Y. Wu, *Phys. Rev. B* **37**, 2059 (1988).

⁷H. Kawamura, *J. Phys. Soc. Jpn.* **53**, 2452 (1984).

⁸Of course, there will be higher-order responses of the spin deviations $\delta\theta$ of $O(h^2)$. One might worry that this produces two additional contributions when these are included in the $O(\delta\theta^2)$ dipolar energy and the $O(\delta\theta)$ magnetization, but, in fact, these two terms cancel each other quite generally in any problem with this algebraic structure.

⁹The degree of freedom ϕ in this case corresponds to a *constrained* rotation in the sense that the two groups of dipoles have to be rotated in opposite directions by the same angle.

¹⁰J. F. Fernandez, *Phys. Rev. B* **38**, 690 (1988).

¹¹J. F. Fernandez, *Europhys. Lett.* **5**, 129 (1988).

¹²J. V. José, L. P. Kadanoff, S. Kirkpatrick, and D. R. Nelson, *Phys. Rev. B* **16**, 1217 (1977).

¹³(a) Y. Imry and S. K. Ma, *Phys. Rev. Lett.* **35**, 1399 (1975); for a review, see T. Nattermann and J. Villain, *Phase Transitions* **11**, 5 (1988). (b) K. Binder, *Z. Phys. B* **50**, 343 (1983).

¹⁴Y. Y. Goldschmidt and B. Schaub, *Nucl. Phys. B* **251**, 77 (1985).

- ¹⁵R. A. Pelcovits, E. Pytte, and J. Rudnick, *Phys. Rev. Lett.* **40**, 476 (1978); R. Fisch, *Phys. Rev. B* **39**, 873 (1989), and references therein.
- ¹⁶A. P. Young, in *Ordering in Strongly Fluctuating Condensed Matter Systems*, edited by T. Riste (Plenum, New York, 1980).
- ¹⁷D. R. Nelson, in *Critical Phenomena and Phase Transitions*, edited by C. Domb and J. L. Lebowitz (Academic, New York, 1983), Vol. 7.
- ¹⁸B. J. Minchau and R. A. Pelcovits, *Phys. Rev. B* **32**, 3081 (1985); M. Ma and P. A. Lee, *ibid.* **32**, 5658 (1985).
- ¹⁹E. F. Shender, *Zh. Eksp. Teor. Fiz.* **83**, 326 (1982) [*Sov. Phys.—JETP* **56**, 178 (1982)].
- ²⁰M. W. Long, *J. Phys. Condens. Matter* **1**, 2857 (1989).
- ²¹B. E. Larson and C. L. Henley, (unpublished).
- ²²S. Miyashita and H. Kawamura, *J. Phys. Soc. Jpn.* **54**, 3385 (1985).
- ²³Q. Sheng and C. L. Henley (unpublished).
- ²⁴A. K. Ibrahim and G. O. Zimmermann, *Phys. Rev.* **B35**, 1860 (1987).