I. Introduction

A classical spin model studied by Thorpe and Blume \cite{1} (TB) showed interesting ground state behavior, where there was either simple collinear-spin long range order, or disorder. The spins were on a linear chain, with nearest-neighbor (nn) Heisenberg and biquadratic exchange interactions. Recently a next-nearest-neighbor (nnn) anti-ferromagnetic Heisenberg exchange term was added (making the Heisenberg terms frustrated), solved exactly for the ground state, and found to yield a rich phase diagram, \cite{2} with spirals and the “up-up-down-down” (uudd) state (isotropic version of the uudd state of the ANNNI model \cite{2}), plus the TB states.

It was speculated \cite{2} that extension of the model to lattice dimensionality \(d = 2\), with the rhombic symmetry of the Heisenberg model used for multiferroic manganites \cite{4, 5}, would yield the historically puzzling uudd (E-type) state observed in those materials.

Here we carry out this extension, and also treat a corresponding square-symmetry model. We again find the ground state exactly. As in \cite{2}, this is enabled by use of the LK cluster method \cite{9}; it is also an additional test of the applicability of that method.

A 2d version of the uudd state is indeed found in the rhombic model and is essentially the observed uudd state \cite{4, 8}. Spirals and highly degenerate phases are also found. A model along these lines appears to be realistic for the manganites, and provides strong support for the suggested mechanism \cite{2} behind the uudd state, namely frustrated Heisenberg plus biquadratic interactions.

For the square symmetry, a coplanar non-spiral state that is an ordered array of vortices, a “vortex lattice” (VL), is found, also discussed earlier by Henley \cite{10} (see also \cite{11}), both for XY and Heisenberg spins. Also found is a non-coplanar state, a “conical vortex lattice”.

A principal motivation for the addition of biquadratic terms to the frustrated Heisenberg model \cite{2} was that they can be large for ions with large spin \(S\) \cite{12, 13}. Two sources of these terms are i. Electronic: higher order terms in the hopping amplitudes or orbital overlap (leading order yields the Heisenberg interactions) \cite{14, 17} and ii. Lattice induced via spin-lattice interaction \cite{16, 17}. There are indications that these sources may be of roughly equal magnitude. \cite{12, 15}. For the present purposes, the source is not relevant.

The model Hamiltonian studied is

\[
H = \sum_{\langle n,m \rangle} \left[ J_1 S_n \cdot S_m - A(S_n \cdot S_m)^2 \right] + J_2 \sum_{\langle n,m \rangle} S_n \cdot S_m + J_2' \sum_{\langle n,m \rangle} S_n \cdot S_m, \tag{1}
\]

where \(S_u\) is a unit 3-vector, is the spin at site \(u\). The first term sums Heisenberg and biquadratic interactions over nn pairs: \(n, m\) go over the vectors of a square lattice. The 2nd and 3rd terms are, respectively, sums over the nn pairs along the \((1,1)\) and \((1,-1)\) diagonals of the square unit cell. We consider two cases: \(J_2 = J_2'\) (square symmetry) and infinitesimal \(J_2'\) (rhombic symmetry). The latter case is motivated by models \cite{4, 6} applied to manganites.

\(H\) extends that studied in \cite{2} to \(d = 2\). Motivations for its study are as in \cite{2}, e.g., biquadratic terms can be large for large-spin ions \cite{12, 13}, such terms are used to mimic the order-selecting effects of thermal, quantum, or dilution fluctuations ("order-by-disorder" effects) \cite{20, 21}. Its ground state phase diagram can be found analytically, and shows properties that should be of interest in statistical mechanics and for manganites particularly.

The Luttinger-Tisza method and its generalizations (see the review \cite{22}) appear to be not useful in connection with (1) because of the non-linearity in the equation for stationarity of \(H\) subject to the weak constraint,

\[
\sum_j (J_{ij} - 2 A_{ij} S_i \cdot S_j) S_j = \lambda S_i.
\]
Instead we turn to the rather unknown LK cluster method, which solves the problem exactly. Recall that method as applied here. Assume periodic boundary conditions, with the thermodynamic limit (TL) to be taken finally. Then (1) can be written

\[ H = \sum_n H_c(S_n, S_{n+x}, S_{n+y}, S_{n+y}), \]

where \( H_c \) is the cluster energy; \( h_c \equiv H_c/|J_1| \) is given by

\[
h_c(S_1, S_2, S_3, S_4) = -\frac{1}{2} \sum_{n=1}^{4} [S_n \cdot S_{n+1} + a(S_n \cdot S_{n+1})^2] + \gamma S_1 \cdot S_3 + \gamma' S_2 \cdot S_4,
\]

where \( S_5 \equiv S_1, \ a = A/|J_1|, \ \gamma = J_2/|J_1|, \ \gamma' = J_2'/|J_1|, \) and we’ve taken \( J_1 < 0 \). Clearly, \( h \equiv H/|J_1| \) satisfies

\[ h \geq \min_n h_c(S_n, S_{n+x}, S_{n+y}, S_{n+y}). \]

If states that minimize \( h_c \) ”propagate”, i.e. if there is a state of the whole system such that every cluster (every square plaquette with its 4 spins) achieves the minimum \( h_c \), it follows that the state is a ground state of \( H \) (the global minimum). To minimize \( h_c \), we find, analytically, stationary states, construct a phase diagram by comparing their \( h_c \)-values and check that there are no lower states by calculating \( h_c \) on a mesh over the whole range of the variables. This and other related matters are discussed in the Appendix.

Case 1. Infinitesimal \( \gamma' \) (rhombic symmetry)

For clarity, we first consider coplanar spins (spin dimensionality D=2, i.e. XY spins). Because of the spin isotropy of \( h_c \), it is only a function of 3 angles. FIG. 1 is the phase diagram. The state, all spins parallel, occurs in the Ferro region. In the upper-right and lower-left regions there is large degeneracy that is lifted by \( \gamma' \neq 0 \) in favor of the states given.

II. Results. See Appendix for their derivations.

Case 2. \( \gamma = \gamma' \) (square symmetry)

In the lower left region a canted ferromagnet, CF\(_2\), shown in Fig. 4, or a spiral are ground states for \( \gamma' < \gamma > 0 \). The spiral wave vector is \((q_1, -q_1), \cos q_1 = -1/(2a), q_1 \) being also the canting angle.

The phase diagram is unchanged for Heisenberg spins.

Non-coplanar states

FIG. 1: (Color online) Phase diagram, \( \gamma' = 0 \) (rhombic symmetry). In the upper-right and lower-left regions there is large degeneracy that is lifted by \( \gamma' \neq 0 \) in favor of the states given.

FIG. 2: The ground states in the uudd, \((\pi, 0)/(0, \pi)\) region of FIG. 1.

FIG. 3: (Color online) Phase diagrams, \( \gamma' = \gamma \) (square symmetry), for XY and Heisenberg (HEIS) models, respectively

In the lower left region a canted ferromagnet, CF\(_2\), shown in Fig. 4, or a spiral are ground states for \( \gamma' < \gamma > 0 \). The spiral wave vector is \((q_1, -q_1), \cos q_1 = -1/(2a), q_1 \) being also the canting angle.

The phase diagram is unchanged for Heisenberg spins.

In the lower left region a canted ferromagnet, CF\(_2\), shown in Fig. 4, or a spiral are ground states for \( \gamma' < \gamma > 0 \). The spiral wave vector is \((q_1, -q_1), \cos q_1 = -1/(2a), q_1 \) being also the canting angle.
We found the ground state to be non-coplanar in the region Conical VL (FIG. 3HEIS). FIG. 5 shows an example. There appears no obvious symmetry, although it was found that at all points in the region, \( \theta_2 = \theta_4 \) and \( \phi_3 = (1/2)\phi_4 \). After FIG. 5 was drawn, and much puzzlement, we found that a particular uniform rotation of the spins brings the state to a highly symmetric one: The spins in each plaquette lie on the surface of a cone, of half-angle \( \Omega \), and the azimuthal angles are equally spaced (i.e. the spacing is \( \pi/2 \)). Thus the name "Conical VL". \( \Omega \) varies smoothly from 0 at the Ferro boundary to \( \pi/2 \) at the VL boundary. But at the CF_2 boundary there is a first-order transition. Note that there is a net spin, i.e. this is ferro- (or ferri-) magnetic.

**Degeneracies**

In classical systems variables vary continuously. However, in the XY case, fixing just one spin in our ground states makes them countable: They derive from various propagations of the degenerate cluster ground states, which are clearly countable when one spin is fixed. This allows the definition of entropy \( S = \ln (\text{number of states}) \), which we will use for XY spins.

In the CF_2 and uudd regions of FIG. 1 there is a large degeneracy coming from many ways of propagating the cluster ground states: the corresponding entropy \( S \geq N^{1/2} \ln 2 \), where \( N \) is the number of spins. Non-zero \( \gamma' \) removes this degeneracy. In the Spiral-CF_4 region of FIG. 3XY there is a similarly large degeneracy.

The propagation of the Ferro and Spiral states, FIG. 1, is unique; but we cannot conclude they are non-degenerate (see Appendix). Similarly, all the regions in FIG. 3 other than Spiral-CF_4 show unique propagation.

The emphasized line segments at \( \gamma = 0 \) and 1 in FIG. 1 and at \( \gamma = 0 \) in FIG. 3 are closely related to the disorder lines in the 1d case [2]. The 2d generalization of the TB disordered states [1] occurs at \( \gamma = 0 \). In 1d, \( S = N \ln 2 \). Whether a similar conclusion holds in 2d is an interesting question that should be addressed. We find \( S \) is at least \( O(N^{1/2}) \) (see Appendix) The line at \( \gamma = 1 \), FIG. 1, is the 2d isotropic generalization of the highly degenerate states of the ANNNI model [2] at the multiphase point.

**III. Discussion**
Under the nn interaction $J_1 \to -J_1$, the net spin in the CF$_2$ and CF$_4$ remains non-zero, although at a smaller value. Interestingly, this net spin occurs despite having only antiferromagnetic interactions in a Bravais lattice. Uniform rotation by $\pm \pi/2$ of the horizontal arrows in the VL state in FIG. 4 changes it to one of the ($\pi,0$), $(0,\pi)$ states of FIG. 2. At $\alpha = 0$, such a uniform rotation through an arbitrary angle $\phi$ has energy independent of $\phi$ for any $\gamma \[10,30\]$, explaining why the boundary between VL and $(\pi,0), (0,\pi)$ is the line $\alpha = 0$.

The question of what removes the degeneracy was considered: Randomness due to dilution was found to give preference to $\phi = 0 \[10,20,29\]$ while quantum fluctuations stabilize $\phi = \pm \pi/2$, i.e. the collinear states $(\pi,0), (0,\pi) \[10,30\]$. Furthermore, as we have seen, the same effect is caused by the biquadratic terms, illustrating the use of the latter to mimic the fluctuations $\[21\]$. In view of the appreciable size of the biquadratic terms, shown by experiment $\[12,13\]$, true biquadratic interactions might be as least as important as the fluctuations.

The purely electronic mechanism for the (2-body) biquadratic terms also gives, in the same order in the hopping amplitude, 3-body, e.g. $S_1 \cdot S_2 S_2 \cdot S_3$, and 4-body terms, like $S_1 \cdot S_2 S_3 \cdot S_4$. To be complete one needs information about the coefficients of these various terms, particularly their signs. The only unambiguous experiments, in that they can contain only 2-body terms, are studies of magnetic dimers. Two examples: Mn impurities in MgO $\[12\]$, where Mn-Mn pairs were studied, and an example involving Ni$^{2+}$ dimers $\[31\]$. In the former case $a > 0$, in the latter $a < 0$. Understanding of how either sign can occur can be seen in the perturbation calculation of Bastardis et al $\[18\]$. Unfortunately, such a conclusive result is not available for the 3- and 4-body terms, as far as we’re aware. There is a calculation of the 3-body terms for a rather special case $\[18\]$, and the 4-body terms have been calculated only for $S = 1/2$ spins $\[11,14\]$. The lattice-induced mechanism is similar in that it also gives 4-body terms $\[11\]$, and sufficiently general explicit calculations of these terms are not available. Fortunately, the experiments on MnO, NiO $\[13\]$, where these extra terms will appear, show the same physics as represented by the biquadratic terms with $a > 0$, namely a preference for collinearity, thus a stiffening of the collinear antiferromagnetic state. i.e., the extra terms do not necessarily spoil the reason for the existence of the uudd or E-type state in our model. Thus we feel that the mechanism presented here for the uudd state is probably correct.

In summary, we have shown that an essentially realistic model for the insulating manganites (the rhombic case) captures the main ground-state magnetic features seen in these materials, spirals, A-type and uudd or E-type ordering. Isotropic corrections to frustrated Heisenberg interactions, in the simplified form of biquadratic terms, characterize the model, a square symmetry version having also been studied. And, despite the model’s complexity, the LK cluster method $\[9\]$ has been shown to enable simple and exact determination of the classical ground states. Finally, the square symmetry case shows a novel spin ordering, the conical vortex lattice, which might be accessible in real materials.

We thank C. Henley, C. Piermarocchi, A. Chubukov, J. B. Goodenough, M. Mochizuki for helpful discussions, and M. Dykman and A. Kamenev for encouragement.
APPENDIX

This contains some comments explaining further the basic cluster method, the derivations of the ground spin states, and of statements about the degeneracy of various states.

Additional explanatory remarks concerning the cluster method

At the urging of referees, we add some hopefully clarifying remarks about the LK cluster method (despite these having been made rather extensively in the original paper (1964)). We first note that there is an infinite number of ways of writing $H$ of (1) in terms of cluster energies. (1) itself is one way, where each cluster is either a nearest-neighbor pair of spins, a pair of (1,1)-diagonal nnn’s, or a (1,-1)-diagonal nnn pair; and one sums over all these “clusters” precisely as written in (1). One will readily see that the ground state of the individual clusters will not propagate when there are competing interactions, i.e. frustration. E.g., if $J_1 < 0$ and $A > 0$, then the minimum for every nn-pair cluster will force the spins in each such pair to be parallel; but given $J_2 > 0$, (1,1)-pair clusters will be minimized with antiparallel spins (assuming $A \geq 0$). Thus the essential idea of the method is to see if there is a more judicious choice. Experience has suggested that highly-symmetric clusters have a much better chance of producing cluster ground states that propagate. Thus the spins on a square plaquette were chosen in this case. The next step is to choose the cluster energy such that the sum over every such cluster (i.e. every plaquette) returns the original Hamiltonian, i.e. we need to prove (2) for our choice (3). Substituting the nn pairs along $\hat{x}$ in (3) (from $n=1$ and $n=3$) into the right side of (2) we have the all horizontal bonds

$$H_x = -(1/2) \sum_{n} [S_n \cdot S_{n+\hat{x}} + a(S_n \cdot S_{n+\hat{x}})^2] + \sum_{n} [S_{n+\hat{y}} \cdot S_{n+\hat{y}+\hat{x}} + a(S_{n+\hat{y}} \cdot S_{n+\hat{y}+\hat{x}})^2].$$

Changing the summation variable $n$ to $n-\hat{y}$ in the second sum leads precisely to the first sum, together they cancel the 1/2, giving precisely the horizontal-bond terms in (1). The nnn Heisenberg bonds along (1,1) come from the terms in $H_c$ in (2) of the form $S_n \cdot S_{n+\hat{x}+\hat{y}}$, and summing these over all $n$ gives directly all the corresponding terms in (1), etc.

Derivation of the macroscopic ground states via the cluster method

As seen from equations (2) and (3), the 4 spins in a cluster are labelled 1,2,3,4 going counterclockwise around the square (the $x$ and $y$ directions are to the right and up, respectively). For coplanar states, $h_c$ depends only on the angles $\theta_2, \theta_3, \theta_4$, of spins 2,3, and 4 relative to spin 1:

$$h_c \equiv h(\theta_2, \theta_3, \theta_4) = -(1/2)(\cos \theta_2 + \cos \theta_3 + \cos \theta_4 + \cos \theta_4)
- (a/2)(\cos^2 \theta_2 + \cos^2 \theta_3 + \cos^2 \theta_4 + \cos^2 \theta_4)
+ \gamma \cos \theta_4 + \gamma' cos \theta_24,$$

where $\theta_{nm} = \theta_n - \theta_m$. These states are denoted ($\theta_2, \theta_3, \theta_4$), and are discussed first (in Cases 1 and 2 below). The procedure is to determine stationary states analytically, solutions of $\partial h / \partial \theta_n = 0$, see that they propagate, compare their energies, and create a tentative ground state phase diagram. We then check that no lower cluster states were missed by various numerical and other methods. For clarity, we first discuss the initial cluster states and their propagation into crystal states, assuming our tentative phase diagram is correct. See the last section of the Supplement for discussion of the checks made.

We will often refer to states related by symmetry (giving rise to “trivial degeneracy” in Henley’s terms (ref. [26] main text)) as “a state” and to states not related by symmetry as “distinct states”.

Case 1. Infinitesimal $\gamma'$ (rhombic symmetry)

For $\gamma' = 0$, in the region of FIG. 1 labelled Ferro, the minimum $h_c$ occurs for the state (0, 0, 0). In the region uudd,$(\pi,0)/0, \pi)$ the cluster ground states are $(\pi, \pi, \pi)$, $(\pi, \pi, 0)$, and $(0, \pi, \pi)$. Taking $S_1$ up, these can be written uudd, uduu, and uudd. The first, uudd, and its symmetry equivalents duuu, udud, and ddud (since $\gamma' \neq \gamma$, uudd and udud are not equivalent to uudd), can be seen to propagate in the crystal state labelled uudd in FIG.2, establishing this state as a ground state (in the TL). The symmetry equivalent cluster states uudd and udud are seen to propagate in the $(\pi, 0)$ state (on the left in FIG.2), the one on the right $(0, \pi)$ comes from the udd and uudd cluster states. The fact that all three cluster states are degenerate can be seen by inspection of FIG. 2 (the nn Heisenberg contribution is zero, the nnn contribution is the same for every plaquette). The degeneracy between the uudd and $(0, \pi)$ is removed by $\gamma' \neq 0$, seen by inspection of FIG.2

In the spiral region of FIG. 1, the lowest cluster state for $\gamma > 0$ is $(q_0, 2q_0, q_0)$, $\cos q_0 = (2\gamma - 2a)^{-1}$. From the uniform spin rotation invariance of $h_c$, this is seen to propagate as a simple spiral, with wave vector $q = (q_0, q_0)$.

In the “CF2-Spiral(1,-1)” region, which occurs at $\gamma < 0$, $a < -1/2$, there are two degenerate cluster ground states. One is $(q_1, 0, q_1)$ (cos $q_1 = -1/(2a)$), pictured in FIG.8a. It propagates uniquely into the canted state CF2 (FIG. 4). The other is $(q_1, 0, -q_1)$, (Fig. Bc), which is seen to propagate as a spiral with wave vector $q = (q_1, -q_1)$. But propagation can involve both these cluster states, leading to large degeneracy, as discussed further below. The $\gamma$-independence of $q_1$ is
an obvious consequence of spins 1 and 3 always being parallel for any \( q \) in the (1,-1) direction. This parallelism explains the \( \gamma \)-independence of the Spiral/CF\(_2\)-Ferro boundary.

**Case 2.** \( \gamma = \gamma' \) (square symmetry)

In the regions of FIG.3 (XY and HEIS) labelled \((\pi,0)/(0,\pi), \) the cluster ground state is \((\pi,\pi,0) = udu\) (plus its symmetry equivalents), which, as we just saw, leads to \((\pi,0),(0,\pi)\) shown in FIG. 2.

In the VL (vortex lattice) regions, minimum \( h_c \) occurs for \((-\pi/2,\pi,\pi/2)\) and its symmetry equivalents. It is convenient to consider the particular equivalent states obtained by reflection \( \sigma_h \) of the spin positions in the horizontal line or \( \sigma_v \) in the vertical line (symmetry operations of \( h_c \) in Case 2):

\[
\sigma_h \begin{pmatrix} 4 & 3 \\ 1 & 2 \end{pmatrix} = \begin{pmatrix} 1 & 2 \\ 4 & 3 \end{pmatrix},
\]
\[
\sigma_v \begin{pmatrix} 4 & 3 \\ 1 & 2 \end{pmatrix} = \begin{pmatrix} 3 & 4 \\ 2 & 1 \end{pmatrix}.
\]

Also define \( T_x, T_y \) as translations through a lattice constant in the \( x,y \) directions respectively. Applying \( T_x \sigma_v \) successively to the plaquette in the lower left of FIG. 4 VL, then applying \( T_y \sigma_h \) successively to that result, and so on, one sees that the whole figure is reproduced. (This is a series of checker moves, moving a column (or row) over the other column (or row), but not removing the “jumped” spins.) Hence every plaquette has minimum \( h_c \) so that VL is a crystal ground state in this region. Essential to this propagation is the commutation, \( T_y \sigma_h T_x \sigma_v = T_x \sigma_v T_y \sigma_h \), giving the 4th plaquette (the central one in the figure) the same for each possible path to it.

These considerations lead directly to the following: Any set of 4 cluster spins propagates in this way for square symmetry. Thus the cluster method rigorously reduces the 8-spin problem to a 4-spin problem for any square-symmetric interactions which can be described in terms of the square plaquette clusters.

In the region labelled “Spiral, CF\(_4\)”, the cluster ground state is the same, \((\theta,2\theta,\theta)\) with \( \theta = q_0 \), as in the spiral region of FIG. 1. This can propagate as a spiral with wave vector \( q = (q_0, q_0) \), or its symmetry-caused degenerate counterpart, the spiral with wave vector \( (q_0, -q_0) \) (from cluster state \((\theta,0,-\theta)\), as well as the spirals with \( q \to -q \). However, surprisingly, there is more than one way that this cluster state can propagate, one of which is the 4-sublattice canted ferromagnet CF\(_4\) shown in FIG. 4, which comes from propagating by repeated application of \( T_x \sigma_y \) and \( T_y \sigma_z \) to the basic cluster \((\theta,2\theta,\theta)\). In fact there is a large number of degenerate states, discussed below.

One can view these different propagations generally as applying a lattice translation \( T_n \) times a symmetry operation of the cluster. For the spiral the cluster symmetry operation is a uniform spin rotation \( R_\theta \); in the CF\(_4\) case the cluster symmetry operation is either \( \sigma_v \) or \( \sigma_h \). Since in this case these operations are seen to yield no contradiction, (again, essentially because \([\sigma_v, \sigma_h] = 0\)), the CF\(_4\) state is established as a ground state. The other degenerate states come from applications of \( T_n \) times one or the other of \( R, \sigma_v, \sigma_h \).

Note that large degeneracy of crystal states has originated from degeneracy of two distinct cluster states (in the rhombic symmetry case), whereas for square symmetry, it came from different propagations of a single (symmetry-induced) cluster state.

**Non-coplanar states** To examine the possibility that

\[
\text{FIG. 6: Variation of angles and energy with } \gamma \text{ at } a = -0.4.
\]

The non-coplanar boundaries are at \( \gamma = 0.1 \) and 0.5, with Ferro for \( \gamma < 0.1 \) and VL for \( \gamma > 0.5 \). Energy vs. \( \gamma \) is linear in the latter regions.

the Heisenberg ground state is not coplanar, we calculated \( h_c \) over a mesh with 5 angles varying independently (polar angles \( \theta_n \) for \( n=2,3,4 \), azimuthal angles \( \phi_n \) for \( n=3,4; S_1 \equiv \hat{z} \)). This is completely general due to the spin-rotational symmetry. Doing this at sample points in each of the regions of FIG.‘s 1 and 3, we found instability with respect to deviation from coplanarity only in the region of FIG.3HEIS labelled “Conical VL”. We then did a closer examination as follows. We analytically found instability on the boundary between Spiral,CF\(_4\) and VL in FIG. 3XY. We then used the \( \theta \)-values of the spiral (or CF\(_4\)) at a point on this boundary as an estimate in the FindMinimum program of Mathematica to determine a nearby minimum of \( h_c \). We then repeated the calculation of FindMinimum at neighboring points thus generating the ground state over the phase diagram, yielding FIG. 3HEIS. The results revealed the general property \( \theta_2 = \theta_4, \phi_3 = \phi_4/2 \). We found that the states approached...
the VL state on the vertical boundary $\gamma = 1/2$, and the ferromagnetic state on the same line $a = \gamma - 1/2$ as the Ferro-Spiral, CF boundary in FIG.3. A sample behavior of the angles and energy as $\gamma$ varies with fixed $a$ is shown in FIG. A.

Looking at the example non-coplanar state in FIG. 5, we saw no symmetry at all. This seemed strange in view of the very simple boundary structure found (FIG.3HEIS). After much puzzling over this aesthetically unsatisfying situation, we realized that there is a very simple picture of the non-coplanar state! From the numerically-determined cluster state, we found the scalar products of all 4 nn spins to be equal. This implies that the spins in a single plaquette lie on the surface of a cone, 1/2-angle $\Omega$, with equally spaced azimuthal angles $\phi$, i.e. the nn $\phi$ spacing is $\pi/2$. This is described by

$$S_n = \sin \Omega(\hat{x} \cos n \pi/2 + \hat{y} \sin n \pi/2) + \cos \Omega \hat{z},$$

$n = 1, \ldots, 4$.

The energy $h_c$ is now easily written down:

$$h_{CVL}(\Omega) = -2 \cos^2 \Omega - 2 a \cos \Omega + 2 \gamma (2 \cos^2 \Omega - 1).$$

The projection of the spins on the x-y plane propagates to exactly the vortex lattice with reduced spin lengths; thus the name “Conical VL” (CVL). For $a < 0$, $h_{CVL}$ is minimum at

$$\cos^2 \Omega = (2 \gamma - 1)/(2a) \equiv \cos^2 \Omega_0$$

for $0 \leq (2 \gamma - 1)/(2a) \leq 1$, with corresponding energy

$$h_{CVL} = (1 - 2 \gamma)^2/(2a) - 2 \gamma.$$

The other cluster energies relevant to FIG. 3HEIS are

$$h_{Ferr} = -2 - 2a + 2\gamma$$

$$h_{(\pi, 0)} = -2\gamma - 2a$$

$$h_{VL} = -2\gamma$$

$$h_{CF_2} = 1/(2a) + 2\gamma.$$

It is readily verified that these equations yield the boundaries in FIG.3HEIS, those bounding the CVL region having previously been determined numerically. It is also seen that the cluster state $S_n, n = 1 \ldots 4 \rightarrow$ the Ferro state as $\Omega \rightarrow 0$ and the VL state as $\Omega \rightarrow \pi/2$. This implies continuous transitions at the respective boundaries (see also FIG.A). At the CF$_2$-CVL boundary, $\gamma = 0, a < -1/2$, one checks that the energies are the same, but the spin states differ, implying a 1st order phase transition.

**On the degeneracy in various regions.**

When $\gamma = 0$, for either the rhombic or square case, there is a transition from ferromagnetism to a highly degenerate ground state as $a$ decreases past -1/2. This occurs because at $a < -1/2$, the combination of nn ferromagnetic Heisenberg and perpendicular-orientation-favoring biquadratic interactions requires an angle between nn spins given by $\theta = \theta_0 = \cos^{-1} \frac{1}{2a}$. Thus for some direction of a given spin, its nn’s each are only restricted to lie on a cone of 1/2-angle $\theta$ measured from that spin. For simplicity we consider XY spins, so the restriction is just to two relative directions $\pm \theta$.

For $d=1$ (TB), the degeneracy is asymptotically $2^N$: given one spin, and moving in one direction, say to the right, along the chain, its nn to the right has two possible directions, and for each of these, its nn to the right has 2 possible directions, etc. But for $d=2$, there are restrictions on the degeneracy of a pair of nn spins depending on what the other nn’s are, because of the loops that occur.

![FIG. 7: Degenerate plaquette states ($\gamma = 0$).](image)
occur only through the reflections and leads uniquely to CF2. For the case \( \gamma' = 0, \gamma > 0 \), only one cluster state, FIG.Bb, is lowest, so the bound is unity, and the only state is the spiral (the rhombic symmetry removes the reflections \( \sigma_v, \sigma_h \) as symmetry operations); the latter case is discussed in more detail in the next section.

**Uniqueness of the cluster propagation in some regions.**

The existence of a huge number of different ways to propagate the given cluster ground states in some regions forces investigation of a similar possibility in other regions. We find unique propagation in the regions Ferro, Spiral(1,1) (FIG. 1), and \((\pi,0)/(0\pi)\), VL, and Conical VL (FIG. 3). We give a proof in the case of Spiral (1,1), illustrating the procedure used for the other cases.

To propagate a cluster state one must consider translations \( T_2 \) and \( T_y \). But, as we’ve seen, there are symmetries of the cluster states that can also be involved. One can see that \( O \) is such a symmetry operation, a necessary condition for propagation is

\[
\hat{O}S_1 = S_2 \text{ and } \hat{O}S_4 = S_3.
\]

This comes from the anticipated application of \( T_2 \). A similar condition occurs for \( T_y \).

We have \( (\theta, 2\theta, \theta) \equiv \psi \) as the cluster state associated with the \((1,1)\) spiral, wave vector \( Q = (q_0, q_0), q_0 > 0 \), and we are considering the case of rhombic symmetry. We confine the proof to XY spins. The symmetry operation that yields the propagation into this spiral is \( \hat{O} = R_{\theta} \), rotation of the four spins by \( \theta \), as already discussed. The question here is, “Are there any other \( O \)’s that will allow a different propagation?” Fortunately, there is only a small number of possibilities, namely the spatial operations of the rhombus, and those times some spin rotation or reflection applied to all four spins. The rhombus operations are \( \sigma_{1,1}, \sigma_{1,-1}, \rho_x \), respectively, reflection in the two diagonals, and rotation through \( \pi \).

Clearly \( \sigma_{1,\psi} = \psi \); no new information. Assume \( \theta = \pi/4 \) for simplicity.

Writing \( \psi = \left( \begin{array}{c} \uparrow \rightarrow \cr \uparrow \end{array} \right) \), we have \( \sigma_{1,-1} \psi = \left( \begin{array}{c} \rightarrow \uparrow \cr \end{array} \right) \).

There are two possibilities to operate now with spin operation \( \hat{O} \) to satisfy \( \hat{O}S_1 = S_2 \). \( \hat{O} = R_{\theta} \) or \( \sigma_s \), respectively, rotation through \( \theta \) or reflection through the line \( y = (\tan \pi/8)x \). \( R_{\theta} \sigma_{1,-1} \psi \equiv P(\psi = \left( \begin{array}{c} \uparrow \rightarrow \cr \uparrow \end{array} \right) \), showing that \( P \) takes \( S_1 \) to \( S_2 \) (by design), but takes \( S_4 \) to \( S_3 \). So this path does not lead to propagation.

The other possibility, replacing \( R_{\theta} \) by \( \sigma_s \). We have \( \sigma_{s} \sigma_{1,-1} \psi = \left( \begin{array}{c} \rightarrow \uparrow \cr \uparrow \rightarrow \end{array} \right) \), which is just \( R_{\theta} \), so nothing new.

The only remaining possibility (excluding \( \rho_x \)) is \( \sigma_y \), reflection in the line \( y = (\tan 3\pi/8)x \), applied directly to \( \psi : \sigma_y \psi = \left( \begin{array}{c} \rightarrow \uparrow \cr \uparrow \rightarrow \end{array} \right) \). But this has violated \( \hat{O}S_4 = S_3 \).

Interestingly, the last spin state would propagate as a spiral with wave vector -\( Q \). Finally we note that \( \rho_x \psi = \sigma_{1,-1} \psi \), already considered. We can conclude that the propagation in the spiral region of FIG. 1 is unique.

The reason we cannot conclude that a state is non-degenerate even if there is a unique propagation of the cluster ground state is that we know only that the state so-obtained is a ground state. This is similar to the case of the Heisenberg Hamiltonian on a Bravais lattice: we know that the ground state energy is necessarily obtained by the minimum-energy spiral or spirals (ref. 21). And while the spirals are usually the only ground states, there are quite special cases where there are additional degeneracies. See e.g. Z. Nussinov, cond-mat/0105253v12.

**Checks on the tentative ground states.**

The most straightforward check is to consider a region where we suspect \( h_s \) is the minimum and simply calculate \( h_{c} - h_s \) over a mesh that covers the full range of the (3 or 5) angle variables in \( h_c \). Usually we took the mesh step \( \delta \) as \( \pi/10 \), reasonable in view of the fact that the most rapidly changing function is \( \cos 2\alpha \) where \( \alpha \) is one of the angles (giving a “length scale” of \( \pi/2 \)). Some places we used \( \pi/20 \) instead. This procedure checked all the regions. A slight problem occurred very near first-order boundaries—quite understandable: even if the function is very well represented by the values on the mesh, if two local minima are very close in energy, depending on how the mesh points fall, the true minimum might not be found. This problem was completely overcome by using Mathematica’s FindMinimum program, which searches for a local minimum given a starting point P. We ran this with P on a mesh running over the full many-angle space. Then for any point on the P-mesh that falls within the basin of a particular local minimum immediately goes to that minimum value, with arbitrary precision. The required interval for this mesh \( \delta_P \) is not as tight as \( \delta \). As an example, using this more powerful method, the vertical (1st-order) boundary at \( \gamma = 0 \) was preserved to within one part in \( 10^4 \) or better, using \( \delta_p = \pi/4 \).

Another check was with Mathematica’s program Reduce, which analytically is supposed to return all the solutions to the stationarity equations. This worked for some regions in the sense that it ran in short time (few minutes), but in other regions it ran for at least hours, and we didn’t wait. Where it did work, it confirmed our initial results, giving a rigorous proof for those regions.

We also note that the ground state energy is rigorously known on the lines \( a = 0 \) and \( \gamma = 0 \), the former by the Luttinger-Tisza method, the latter by the cluster method, where the clusters are just the 2-body terms in the original form of the Hamiltonian. Also the limit \( a \rightarrow \infty \) is clearly correct, as well as the limit \( \gamma \rightarrow -\infty, a \rightarrow -\infty \).

These considerations have convinced us that our analytically-described phase diagrams are exact, although we can’t claim a rigorous proof due to the use of these numerical methods as checks.