Frustrated Classical Heisenberg and *XY* Models in Two Dimensions with Nearest-Neighbor Biquadratic Exchange: Exact Solution for the Ground-State Phase Diagram

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The ground-state phase diagram is determined exactly for the frustrated classical Heisenberg model plus nearest-neighbor biquadratic exchange interactions on a two-dimensional lattice. A square- and a rhombic-symmetry version are considered. There appear ferromagnetic, incommensurate-spiral, ''up-up-down-down'' (UUDD), and canted ferromagnetic states, a nonspiral coplanar state that is an ordered vortex lattice, plus a noncoplanar ordered state (a ''conical vortex lattice''). In the rhombic case, which adds biquadratic terms to the Heisenberg model used widely for insulating manganites, the UUDD state found is the *E*-type state observed; this, along with accounting essentially for the variety of ground states observed in these materials, shows that this model probably contains the long-sought mechanism behind the UUDD state.

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I. Introduction.—A classical spin model studied by Thorpe and Blume [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) antiferromagnetic Heisenberg exchange term was added (making the Heisenberg terms frustrated), solved exactly for the ground state, and found to yield a rich phase diagram, [2] with spirals and the ''up-up-down-down'' (UUDD) state (isotropic version of the UUDD state of the ANNNI model [3]), plus the TB states.

It was speculated [2] that extension of the model to lattice dimensionality $d = 2$, with the rhombic symmetry of the Heisenberg model used for multiferroic manganites [4–6], 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 [2], this is enabled by use of the LK (Lyons-Kaplan) cluster method [7]; it is also an additional test of the applicability of that method.

A $d = 2$ version of the UUDD state is indeed found in the rhombic model and is essentially the observed UUDD state [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 [2] behind the UUDD state, namely, frustrated Heisenberg plus biquadratic interactions.

For the square symmetry, a coplanar nonspiral state that is an ordered array of vortices, a ''vortex lattice'' (VL), is found, also discussed earlier by Henley [9] (see also [10]), both for *XY* and Heisenberg spins. Also found is a noncoplanar state, a ''conical vortex lattice.''

A principal motivation for the addition of biquadratic terms to the frustrated Heisenberg model [2] was that they can be large for ions with large spin *S* [11,12]. Two sources of these terms are (i) Electronic: higher order terms in the hopping amplitudes or orbital overlap (leading order yields the Heisenberg interactions) [13,14] and (ii) Lattice-induced via spin-lattice interaction [15,16]. There are indications that these sources may be of roughly equal magnitude [11–14]. For the present purposes, the source is not relevant.

The model Hamiltonian studied is

$$H = \sum_{\langle \mathbf{n},\mathbf{m}\rangle} [J_1 \mathbf{S}_\mathbf{n} \cdot \mathbf{S}_\mathbf{m} - A(\mathbf{S}_\mathbf{n} \cdot \mathbf{S}_\mathbf{m})^2] + J_2 \sum_{\langle \mathbf{n},\mathbf{m}\rangle}^{1} \mathbf{S}_\mathbf{n} \cdot \mathbf{S}_\mathbf{m} + J_2' \sum_{\langle \mathbf{n},\mathbf{m}\rangle}^{2} \mathbf{S}_\mathbf{n} \cdot \mathbf{S}_\mathbf{m}, \quad (1)$$

where $\mathbf{S}_\mathbf{u}$, a unit 3 vector, is the spin at site $\mathbf{u}$. The first term sums Heisenberg and biquadratic interactions over NN pairs: $\mathbf{n}$, $\mathbf{m}$ go over the vectors of a square lattice. The second and third terms are, respectively, sums over the NN pairs along the (1,1) and (1, −1) diagonals of the square unit cell. We consider (a) $J_2 = J_2'$ (square symmetry) and (b) infinitesimal J_2' (rhombic symmetry). The latter generalizes models [4–6] for manganites to $A \neq 0$ [17].

Additional motivations are as in [2]: Such terms are used to mimic the order-selecting effects of thermal, quantum, or dilution fluctuations (''order-by-disorder'' effects) [18,19], its ground-state phase diagram can be found analytically, and shows properties of interest in statistical mechanics and for manganites particularly.

The Luttinger-Tisza method and its generalizations (see the review [20]) appear to be not useful in connection

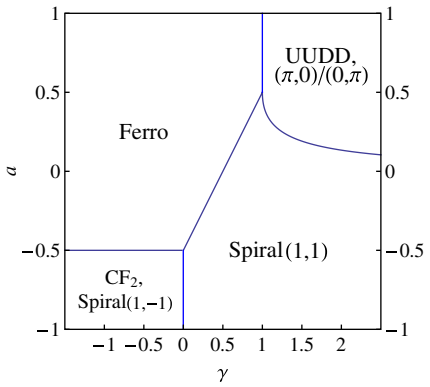

FIG. 1 (color online). Phase diagram, $\gamma' = 0$ (rhombic symmetry). In the upper-right and lower-left regions there is large degeneracy that is lifted by $\gamma' \neq 0$ in favor of the states given.

with (1) because of the nonlinearity in the equation for stationarity of H subject to the weak constraint, $\sum_j (J_{ij} - 2A_{ij}\mathbf{S}_i \cdot \mathbf{S}_j)\mathbf{S}_j = \lambda \mathbf{S}_i$.

Instead we turn to the rather unknown LK cluster method [7], which solves the problem exactly. Recall that method as applied here. Assume periodic boundary conditions, with the thermodynamic limit to be taken finally [7]. Then (1) can be written

$$H = \sum_{\mathbf{n}} H_c(\mathbf{S}_{\mathbf{n}}, \mathbf{S}_{\mathbf{n}+\hat{x}}, \mathbf{S}_{\mathbf{n}+\hat{x}+\hat{y}}, \mathbf{S}_{\mathbf{n}+\hat{y}}), \tag{2}$$

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

$$h_c(\mathbf{S}_1, \mathbf{S}_2, \mathbf{S}_3, \mathbf{S}_4) = -\frac{1}{2}\sum_{n=1}^{4}[\mathbf{S}_n \cdot \mathbf{S}_{n+1} + a(\mathbf{S}_n \cdot \mathbf{S}_{n+1})^2] + \gamma \mathbf{S}_1 \cdot \mathbf{S}_3 + \gamma' \mathbf{S}_2 \cdot \mathbf{S}_4, \tag{3}$$

where $\mathbf{S}_5 \equiv \mathbf{S}_1$, $a = A/|J_1|$, $\gamma = J_2/|J_1|$, $\gamma' = J_2'/|J_1|$, and we have taken $J_1 < 0$. ($\hat{x}$, $\hat{y}$ are primitive lattice vectors.) Clearly, $h \equiv H/|J_1|$ satisfies

$$h \geq \sum_{\mathbf{n}} \min h_c(\mathbf{S}_{\mathbf{n}}, \mathbf{S}_{\mathbf{n}+\hat{x}}, \mathbf{S}_{\mathbf{n}+\hat{x}+\hat{y}}, \mathbf{S}_{\mathbf{n}+\hat{y}}). \tag{4}$$

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 four 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 [21].

II. Results [21].—

Case 1. Infinitesimal γ' (rhombic symmetry): For clarity, we first consider coplanar spins (spin dimensionality $D = 2$, i.e., XY spins). Figure 1 is the phase diagram. The state, all spins parallel, occurs in the ferro region. In the upper-right region, the states UUDD or $(\pi, 0)/(0, \pi)$,

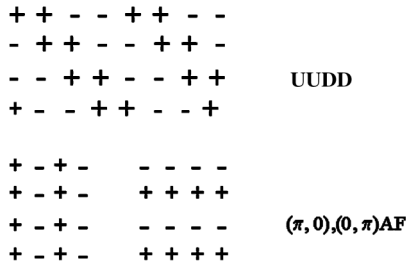

FIG. 2. The ground states in the UUDD, $(\pi, 0)/(0, \pi)$ region of Fig. 1.

shown in Fig. 2, are the ground states for $\gamma' <$ or > 0; $(\pi, 0)$ and $(0, \pi)$ refer to propagation vectors. The UUDD state is a wave with propagation vector $\mathbf{q}$ in the (1,1) direction. The notation is (q_x, q_y), x axis to the right, y up.

In the spiral (1, 1) region is a simple spiral [20] with propagation vector $\mathbf{q} = (q_0, q_0)$, $\cos q_0 = [2(\gamma - a)]^{-1}$. In the lower-left region a canted ferromagnet, CF_2, shown in Fig. 3, or a spiral are ground states for $\gamma' <$ or > 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.

Case 2. $\gamma = \gamma'$ (square symmetry): Figure 4 shows the phase diagrams for XY and for Heisenberg (HEIS) spins. **XY**: The ferro region is similar to that in Fig. 1. The $(\pi, 0)$, $(0, \pi)$ states no longer coexist with the UUDD states ($\gamma > 1/2$, $a > 0$). The ground state in the VL region, discussed previously by Henley [9] (who considered only $\gamma > 1/2$), can be described as an ordered array of vortices, which we call a vortex lattice. See Fig. 3 for an example, where the filled and unfilled circles indicate a pair of vortices with spin orientations as shown; the plaquette-spin patterns may be described as clockwise and counterclockwise, respectively. The vortices form a square lattice. In the region spiral-CF_4, a (q_0, q_0) spiral and a canted ferromagnet, CF_4 (see Fig. 3) are degenerate ground states. In the extreme lower left, the ground state CF_2 is no longer degenerate with a spiral. This canted ferromagnet was also found in [10]. HEIS: The main change from XY to HEIS is the replacement of the spiral-CF_4 phase by a noncoplanar state, discussed below.

*Noncoplanar states.—*The ground state is noncoplanar in the region conical VL. Figure 5 shows an example. The spins in each plaquette lie on the surface of a cone, of half-

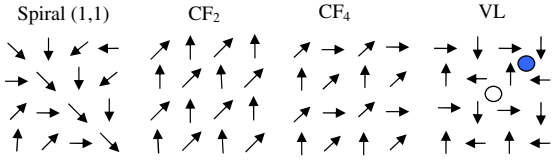

FIG. 3 (color online). Spiral and canted ferromagnets, CF_n (for illustrative value $\pi/4$ of the turn-angle q_0.). Vortex lattice: ground state in regions VL of Fig. 4.

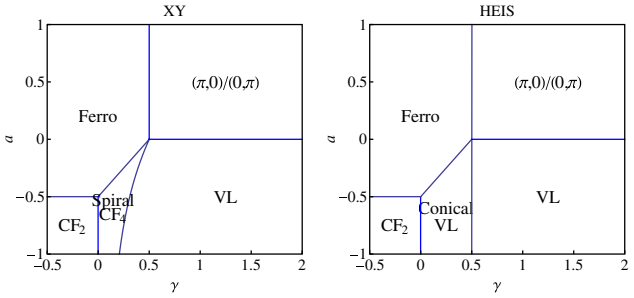

FIG. 4 (color online). Phase diagrams, $\gamma' = \gamma$ (square symmetry), for XY and Heisenberg (HEIS) models, respectively.

angle Ω, and the azimuthal angles are equally spaced. Thus the name "conical VL." Ω varies smoothly from 0 at the ferro boundary to $\pi/2$ at the VL boundary. 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 $\mathcal{S} = \ln$ (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 $\mathcal{S} \geq N^{1/2} \ln 2$, where N is the number of spins. Nonzero γ' removes this degeneracy. In the spiral-CF_4 region of Fig. 4 XY there is a similarly large degeneracy.

The propagation of the ferro and spiral states, Fig. 1, is unique. Similarly, all the regions in Fig. 4 other than spiral-CF_4 show unique propagation.

The line segments at $\gamma = 0$ in Figs. 1 and 4 are the $d = 2$ generalization of the TB disordered states [1]. In $d = 1$, $\mathcal{S} = N \ln 2$. Whether a similar conclusion holds in $d = 2$ is an interesting question that should be addressed. We find $\mathcal{S}$ is at least $O(N^{1/2})$ [21]. The line at $\gamma = 1$, Fig. 1, is the $d = 2$ isotropic generalization of the highly degenerate states of the ANNNI model [3] at the multiphase point.

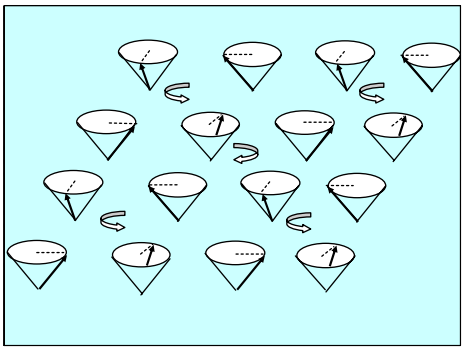

FIG. 5 (color online). Conical vortex lattice: ground state in the conical VL region of Fig. 4 HEIS. Open arrows indicate plaquette-spin patterns.

III. Discussion.—

Case 1. $\gamma' = 0$, extreme rhombic symmetry: The speculation [2] that the $d = 2$ version of the rhombic model would be qualitatively similar to the $d = 1$ case, is borne out: the phase diagram Fig. 1 is topologically the same as that for $d = 1$ [2]. There are, however, three major differences. The ferro-UUDD boundary occurs at $\gamma = 1$ for $d = 2$, vs $\gamma = 1/2$ for $d = 1$. While the UUDD state is the only state in its region for $d = 1$, in $d = 2$ there are other degenerate states, e.g., $(\pi, 0)$, $(0, \pi)$. Similarly, in $d = 1$ the CF_2 state appears alone in its region, while in $d = 2$ it is degenerate with other states.

Experimentally it is UUDD, not $(\pi, 0)$, $(0, \pi)$, that is observed [4,8]. As seen from Fig. 2, a small γ' will remove that degeneracy, a *ferromagnetic* γ' will favor the UUDD state. Interestingly, the calculations of Kimura *et al.* [4] find a small ferromagnetic γ'.

The fact that the value of γ needed to get into this UUDD region is now >1 might be discouraging. Also, a needs to be $\approx 1/2$, which also might not bode well for the present mechanism. However, the unoccupied Mn orbital (e_g) in the manganites gives rise to a ferromagnetic contribution to the Heisenberg exchange in addition to the usual antiferromagnetic contribution [22]. The resulting cancellation can be large if the unoccupied orbital lies close in energy to the occupied orbitals, with the biquadratic exchange not suffering such cancellation [23]. And the Mn ion in the manganites apparently satisfies this requirement. This close cancellation has been invoked for the NN exchange in a different mechanism for the origin of UUDD [8]. It has also been invoked to justify very large anisotropies compared to $|J_1|$ [6,24]. But the latter, particularly the Dzyaloshinskii-Moriya interaction, is expected to be $\ll$ the antiferromagnetic term, being $\approx (g - 2)/g$ times that term [25] (e.g., in $LaMnO_3$, this is 1% [26], compared to the tens of % for the biquadratic terms). In this light, a mechanism along the present lines (i.e., involving isotropic corrections to Heisenberg interactions) is clearly a strong candidate for the origin of the UUDD state in manganites. The ferro (which leads to the A-type ordering [6]), and spiral regions also essentially account for the other ground-state orderings observed. The existence of spirals appears to rule out the nonfrustrated model [8] as a general theory.

Case 2. $\gamma = \gamma'$, square symmetry: Under the NN interaction $J_1 \rightarrow -J_1$, the net spin in the CF_2 and CF_4 remains nonzero, 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. 3 changes it to one of the $(\pi, 0)$, $(0, \pi)$ states of Fig. 2. At $a = 0$, such a uniform rotation through an arbitrary angle ϕ has energy independent of ϕ for any γ [9,27], explaining why the boundary between VL and $(\pi, 0)$, $(0, \pi)$ is the line $a = 0$.

The question of what removes the degeneracy was considered: Randomness due to dilution was found to give preference to $\phi = 0$ [9,18,28] while quantum fluctuations stabilize $\phi = \pm\pi/2$, i.e., the collinear states $(\pi, 0)$, $(0, \pi)$ [9,27]. Furthermore, as we have seen, the same effect is caused by the biquadratic terms, illustrating the use of the latter to mimic the fluctuations [18,19]. In view of the appreciable size of the biquadratic terms, shown by experiment [11,12], true biquadratic interactions might be at least as important as the fluctuations.

The purely electronic mechanism for the (two-body) biquadratic terms also gives, in the same order in the hopping amplitude, three-body, e.g., $\mathbf{S}_1 \cdot \mathbf{S}_2\mathbf{S}_2 \cdot \mathbf{S}_3$, and four-body terms, like $\mathbf{S}_1 \cdot \mathbf{S}_2\mathbf{S}_3 \cdot \mathbf{S}_4$. To be complete one needs information about the coefficients of these various terms. The only unambiguous experiments, in that they can contain only two-body terms, are studies of magnetic dimers. Two examples: Mn impurities in MgO [11], where Mn-Mn pairs were studied, and an example involving Ni^{2+} dimers [29]. 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.* [30]. Unfortunately, such a conclusive result is not available for the three- and four-body terms, as far as we are aware. There is a calculation of the three-body terms for a rather special case [30], and the four-body terms have been calculated only for $S = 1/2$ spins [10,31]. The lattice-induced mechanism is similar in that it also gives four-body terms [16], and sufficiently general explicit calculations of these terms are not available. Fortunately, the experiments on MnO, NiO [12], where these extra terms will appear, show the same physics as represented by the NN 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 (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 [7] 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.

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