Study of an Ising Model with Competing Long- and Short-Range Interactions

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A classical spin-one lattice gas model is used to study the competition between short-range ferromagnetic coupling and long-range antiferromagnetic Coulomb interactions. The model is a coarse-grained representation of frustrated phase seapration in high-temperature superconductors. The ground states are determined for the complete range of parameters by using a combination of numerical and analytical techniques. The crossover between ferromagnetic and antiferromagnetic states proceeds via a rich structure of highly symmetric striped and checkerboard phases. There is no devil's staircase behavior because mixtures of stripes with different period phase separate.

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Systems with competing interactions often show remarkably complex phase diagrams, even though they have quite simple Hamiltonians. In this Letter we study a model of this type, which was suggested by the physics of frustrated phase separation in high-temperature superconductors [1], although it is independently of considerable interest as a problem in statistical mechanics. The Hamiltonian is given by

$$H = K\sum_j S_j^2 - L\sum_{\langle ij\rangle} S_i S_j + \frac{Q}{2}\sum_{i\neq j}\frac{S_i S_j}{r_{ij}}, \qquad (1)$$

where $L, Q > 0$, and $S_j = \pm 1, 0$ specifies the charge at site j of a two-dimensional square lattice [2]. Evidently, this model is equivalent to a spin-one Ising model with a long-range antiferromagnetic Coulomb force of strength Q competing with a short-range ferromagnetic interaction of strength L, so we shall use spin terminology in discussing the phase diagram. The quadrupolar field K controls the concentration of sites at which $S_j^2 = 1$. For application to high-temperature superconductors, S_j is a coarse-grained variable, representing the local density of mobile holes [1]: each site j represents a small region of space for which $S_j = +1$ and $S_j = -1$ correspond to hole-rich and hole-poor phases, respectively, whereas $S_j = 0$ indicates that the local density is equal to the average value. In this realization, the fully phase separated state has $S_j^2 = 1$ and is ferromagnetically ordered, with $S_j = +1$ in one-half of the volume and $S_j = -1$ in the other, so as to maintain overall charge neutrality. This interpretation is different from that used in the more familiar spin-one Ising model [3] of tricritical phenomena, in which quadrupolar ordering $S_j^2 = 0, 1$ corresponds to phase separation. As we shall see, the long-range Coulomb interaction does not destroy all semblance of phase separation by forcing a state with $S_j = 0$ for all j. On the contrary, provided there is a short-distance clustering sufficient to justify our coarse-grained model, the ground state is typically inhomogeneous and shows a rather rich phase structure as the strength of the Coulomb interaction is varied [4]. In particular, striped phases of different period are the most prominent feature of the phase diagram.

The behavior of the model cannot be uncovered by purely analytical or purely numerical means. The problem is that the Coulomb interaction is difficult to handle analytically, whereas in a numerical study the energy differences between adjacent phases often are smaller than finite-size corrections. Accordingly, the zero-temperature properties of the model in the thermodynamic limit were determined by a combination of the two techniques. First of all, a general survey of the ordered phases was made by using a Monte Carlo simulation to heat up the system, followed by single spin flip dynamics to cool it to zero temperature. In this way it was found that the ground states of finite systems consist of simple periodic structures: The pure Coulomb interaction favors a Néel state, but as Q decreases the crossover to the ferromagnetic state proceeds via a series of striped phases of ever increasing width. Consequently, we were able to *assume* specific periodic ground state configurations for which the energy could be written

$$E = KE_{\text{self}} - LE_{\text{nn}} + (Q/2)E_{\text{Coul}}, \qquad (2)$$

where the self-energy E_{self}, the nearest neighbor energy E_{nn}, and the Coulomb energy E_{Coul} are constants, characteristic of the assumed state. It was then straightforward to obtain the phase boundaries as straight lines in the $(Q/K, L/K)$ plane by comparing the energies of the different configurations. As explained below, E_{Coul} may be reduced to a finite sum which may be calculated numerically to arbitrary accuracy. The other two constants may easily be evaluated analytically for each given configuration.

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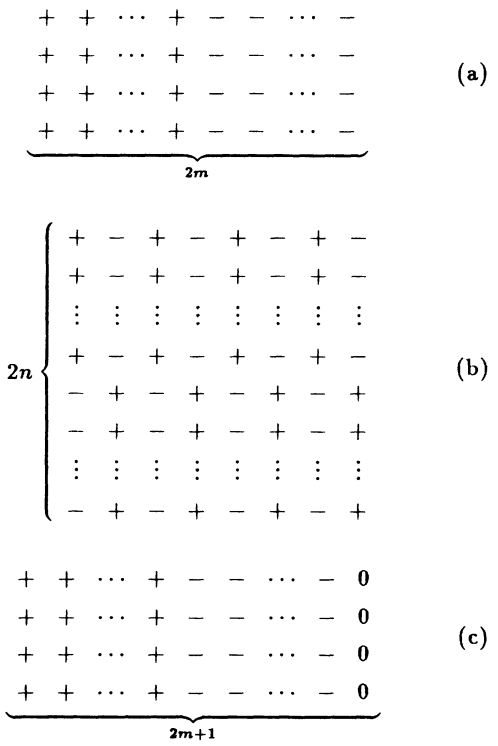

FIG. 1. Types of ground states. (a) Phases consisting of stripes of m up and m down spins. (b) Checkerboard phases of size $(2\times 2n)$. (c) Stripes with one row of zeros.

In order to describe the phase diagram obtained by this procedure, the periodic states will be characterized as $(M_x \times M_y)$ phases, where M_x and M_y are the sizes of the unit cell in the x and y directions, respectively. Thus the Néel state is a (2×2) phase, whereas a phase consisting of positive and negative vertical stripes each of width m, as shown in Fig. 1(a), corresponds to $(2m\times 1)$ for $m = 1,2,\ldots$. The checkerboard phase shown in Fig. 1(b) is specified by $(2\times 2n)$.

First consider $K=0$, for which the model has only one free parameter Q/L. There are no stable phases containing macroscopic numbers of sites with average spin equal to zero. When $\sqrt{L/Q}$ is decreased from infinity to 0.355, the system passes through a succession of striped phases, from $m=\infty$ to $m=1$, as shown in Fig. 2. If $\sqrt{L/Q}$ is decreased further, the system passes from (2×1) to the Néel state (2×2) via a series of $(2\times 2n)$ checkerboard phases, illustrated in Fig. 1(b). This second staircase is much narrower, being confined to a region $0.33 < \sqrt{L/Q} < 0.35$. A thorough study did not reveal any intervening phases between the regular stripes and the $(2\times 2n)$ checkerboards. This will be discussed in greater detail later.

If $K<0$, states containing zeros are even less favorable, and the phase diagram is the same as for $K=0$. This is no longer true when K is positive, and then it is necessary to use two parameters Q/K and L/K to specify

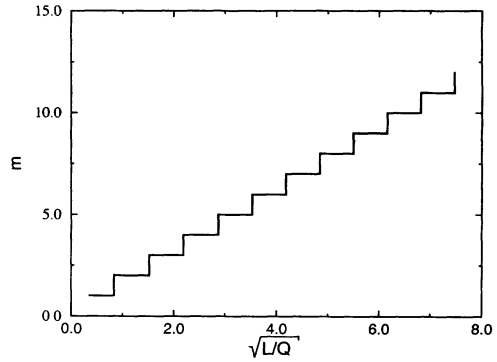

FIG. 2. The half-width m of a striped phase plotted versus $\sqrt{L/Q}$ for $K=0$.

the phase diagram. In this case, new phases consisting of stripes of width $2m$ separated by stripes of zeros, as illustrated in Fig. 1(c), appear for Q/K and L/K small. When Q/K and L/K are large, the phase diagram is similar to the case of $K=0$. The complete phase diagram for $K>0$ is shown in Fig. 3.

Since the relevant states are doubly periodic with periods M_x and M_y along the x and y directions, respectively, it is possible to obtain a deeper understanding of the phase diagram on the infinite lattice by reducing the problem to an effective model within the $(M_x \times M_y)$ unit cell. The argument will be presented for the striped phases, which are of greatest interest here. They correspond to $M_x = 2m$, $M_y = 1$, and, in-cell, the model is a one-dimensional spin-half Ising model on a periodic lattice of $2m$ sites. The effective Hamiltonian may be written

$$H = \sum_{i,j=1}^{2m} \sigma_i \sigma_i V(i-j)\,, \tag{3}$$

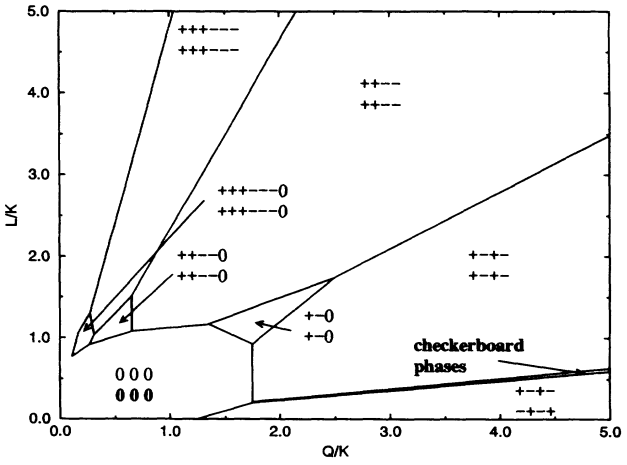

FIG. 3. Phase diagram for $K>0$. The structures of the ground states are marked symbolically. The approach to the L/K axis proceeds via an infinite sequence of striped phases (not shown).

where the spin variables take the values $\sigma_i = \pm 1$ and $V(i-j)$ is the effective interaction within the unit cell. The contribution $V_C(i-j)$ from the Coulomb interaction [the term proportional to Q in Eq. (1)] may be obtained by using the Poisson sum formula to evaluate the potential due to the rows of charges along the y direction, and then carrying out the sum over unit cells in the x direction:

$$V_C(j) = -2\ln\left|\sin\left(\frac{\pi j}{2m}\right)\right| + 4\sum_{l_x=-\infty}^{+\infty}\sum_{n=1}^{\infty} K_0(2\pi n(2ml_x+j)) \quad (4)$$

for $j \neq 0$. Here K_0 is the modified Bessel function of order 0, and infinite terms have canceled in virtue of the assumption of charge-neutral states. The first term in this equation comes from the uniform component of the charge distribution in a row, and it is the usual potential between columns of charge or line vortices: When $j=0$, it should be replaced by $2[\ln(m/\pi)+C]$, where C is Euler's constant. The second term in Eq. (4) comes from the finite wave vector components of the charge distribution of a stripe, and it gives the short-distance transient contributions to the potential. For many purposes, this term is numerically unimportant and may be omitted.

This formulation of the problem enables us to understand the simple structure of Fig. 2, and to address the question of whether there are additional more complex ground states, possibly giving rise to a devil's staircase [5], i.e., regions of the phase diagram in which any two steps are separated by an infinity of extra steps. Bak and Bruinsma [5] showed that for a one-dimensional Ising model with an infinite range convex interaction, the average magnetization displayed a complete devil's staircase as the applied magnetic field was varied. For the problem defined in Eq. (1), charge neutrality forces the average magnetization to be zero. But a devil's staircase might arise in the concentration of spin *flips* (i.e., adjacent spins pointing in opposite directions) as its conjugate variable, the ferromagnetic coupling L, is changed. Consequently, a systematic study of the Coulomb energies within subspaces $S_{m,l}$ with $2l$ spin slips was undertaken. For such a subspace the near-neighbor energy per site is fixed and equal to $-LE_{nn} = (2l/m-2)L$, and the Coulomb energies may be obtained from Eqs. (3) and (4) by using a computer algorithm to generate states in the subspaces $S_{m,l}$. In practice, we considered all states with $m \leq 13$ (several hundred thousand states), and found that, when l is a divisor of m, the ground state is given by a periodic pattern of m/l up spins followed by m/l down spins (stripes of width m/l). In other words, the spin flips are equally spaced. When l was not a divisor of m, the ground state in the subspace $S_{m,l}$ separated into two phases: one an extended striped phase of period $2p$, the other an extended striped phase of period $2(p+1)$, where $p < l/m < p+1$. For finite m, the energy of such a state is higher than the energy of each of the two coexisting phases because of a finite boundary energy.

According to this discussion it is clear that the ground states are simply periodic, except possibly at the values of L/Q for which the period jumps (as shown in Fig. 2) where two phases may coexist. For the simply periodic phases, m may always be chosen so that the ground state for any L/Q has $l=1$, i.e., within the unit cell it is "ferromagnetic," with total spin zero. From Fig. 2 it is also clear that, in considering states with period up to 26, the ground state has been identified for L/Q less than ~ 50. However, the same structure will persist for larger values of L/Q, since the ferromagnetic state is favored by L. It should be emphasized that these conclusions did not involve a finite-size approximation: Equations (3) and (4) are *exact* representations of the Hamiltonian for $(2m \times 1)$ periodic states, and the computer algorithm was simply used to carry out a systematic examination of the states.

As we have seen, the possibility of a devil's staircase, in which the states would consist of a mixture of the adjacent phases of period $2n$ and $2(n+1)$ [5], is pre-empted by phase separation, and the transition from one periodicity to another is first order. A physical feeling for the origin of the phase separation may be obtained by the following argument. In a phase of period $2n$, each unit cell may be regarded as a column of dipoles. It is straightforward to show from Eqs. (3) and (4) that the interaction between dipoles pointing in the same direction is attractive. Moreover we find that, starting from a phase-separated state, (1) it costs energy to transfer a $2n$ dipole into the $2(n+1)$ phase and vice versa and (2) there is an attractive interaction between two $2n$ dipoles in the $2(n+1)$ phase and vice versa. Consequently, the phase separated state is stable, and, conversely, the mixed state tends to phase separate.

Once it is recognized that the ground states are ferromagnetic within the unit cell, it is possible to proceed analytically by evaluating the energy for each $(2m \times 1)$ unit cell and minimizing with respect to m for fixed Q/L. Using Eqs. (3) and (4), it is straightforward to show that the Coulomb contribution to the energy per site for the ferromagnetic unit cell may be rewritten:

$$\frac{Q}{2}E_{\mathrm{Coul}}(m) = Q\sum_{n=1}^{m-1}\left(1-\frac{2n}{m}\right)V_C(n) + \frac{Q}{2}V_c(0). \quad (5)$$

For large m, the leading contribution to E_{Coul} may be obtained by changing the sum to an integral and using Eq. (4):

$$\frac{Q}{2}E_{\mathrm{Coul}}(m) \sim -4Qm\int_0^{1/2} dx(1-4x)\ln\sin(\pi x). \quad (6)$$

Here the K_0 terms have been omitted because they turn out to be numerically unimportant. On evaluating the integral and adding in the near-neighbor contribution, the total energy per site becomes

$$E(m) \sim 0.85Qm + 2L/m + c_1, \quad (7)$$

where c_1 is a function of the parameters of the Hamiltonian but is independent of m. If we fit c_1, the error in this expression is less than 1.5% for $m > 2$, and the Coulomb energies are linear in m for essentially the whole range of interest. The period of the ground state for any L/Q may now be obtained by minimizing $E(m)$ with respect to integral values of m. Then the transition from m to $m+1$ occurs when $E(m) = E(m+1)$ or

$$m(m+1) = 2.35L/Q. \quad (8)$$

This expression is in excellent agreement with Fig. 2 over the whole range of L/Q. A good approximation to the solution of Eq. (8) is $m = -\frac{1}{2} + 1.53\sqrt{L/Q}$, for the relevant values of $\sqrt{L/Q}$. This expression shows why the points at which the value of m jumps lie on a straight line and the steps are of length 0.63, independent of m.

Stability of the ferromagnetic unit cells for all m requires that the "compressibility" (or, equivalently, the second derivative of the total energy with respect to the volume) is positive. Since m is an integer, this condition is equivalent to the requirement that the second *difference* of the total energy $2mE(m)$ with respect to m is positive. It is evident that this condition is satisfied by the approximation (7). We have verified numerically that the condition is also satisfied for small m, where there are small deviations from (7).

It also is possible to derive an effective one-dimensional potential for the phases containing zeros, as shown in Fig. 1(c). Once again the Coulomb energies $E_{\mathrm{Coul}}(m)$ of these phases are linear in the stripe width for large m:

$$\frac{E(m)}{K} \sim 0.85m\frac{Q}{K} - \frac{L}{K}\frac{4m-3}{2m+1} + c_2, \quad (9)$$

where c_2 is a constant. Following the same argument as before, it is easy to show that there is an infinite set of stripes. The regions occupied by the new phases are entirely determined by the constants in Eqs. (7) and (9).

In conclusion, we have shown that a simple Ising model with short-range exchange and a competing long-range Coulomb interaction has an interesting and remarkably complex phase diagram, which illustrates the consequences of frustrated ferromagnetic order (or phase separation in the "parent" model). It would be of some interest to extend this study to finite temperature. In variants of the model considered here, the conclusion that the ground states are inhomogeneous should persist, but the ground state may not be simply periodic. In particular, introducing quantum fluctuations or allowing the charge to be different for $S_i = +1$ and $S_i = -1$ should lead to disordered states.

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