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Ising Model with Frustration, Elasticity, and Competing Interactions

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The Ising model on a compressible triangular lattice with axial next-nearest-neighbor interactions is studied in the mean-field approximation. A representative phase diagram is generated, which exhibits first- and second-order phase transitions to commensurate modulated phases. The crossover point from first to second order transitions is calculated. The stability of the modulated phases is calculated analytically in a low-temperature approximation. These results are very different from the ANNNI model, which exhibits a second-order transition to a continuum of commensurate and incommensurate phases.

KEY WORDS: ANNNI models; modulated phases; frustration; elastic strain.

1. INTRODUCTION

Ising models with competing nearest- and next-nearest-neighbor interactions (ANNNI models) have been shown to exhibit modulated phases, both commensurate and incommensurate with the lattice.[1-4] These studies have been performed on a square or cubic lattice, where the nearest-neighbor ferromagnetic and antiferromagnetic cases are equivalent by a simple transformation of spins. Antiferromagnetic nearest-neighbor interactions on a triangular lattice with global lattice distortions coupled to the spin–spin interaction have also been studied,[5] and in this case a striped phase is found where the ordering is ferromagnetic in one axial direction and antiferromagnetic in the other two. We looked at a model similar to this triangular lattice model, with axial next-nearest-neighbor interactions along all three axes. In the mean-field approximation, we found that the striped phase was modulated with boundaries perpendicular to those of the stripes. While in the ANNNI model, an infinity of commensurate and

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incommensurate phases is found, our model had only a few commensurate modulated phases of relatively short modulation.

Our motivation for examining this model comes from studies of phase stability in CuAu alloys using the effective medium theory (EMT) of cohesion in metals.[7] These alloys exhibit commensurate modulated phases. Effective medium theory allows the approximate energy of an arbitrary configuration of atoms in a metal to be expressed in terms of the positions and species of the atoms. We make the further approximation that the atom positions are on the sites of a triangular lattice which is uniformly distorted such that the bond lengths in each of the three directions may be different.

2. THE MODEL

Our model consists of spins $S_i = \pm 1$ on the sites of a triangular lattice. There are three contributions to the energy. One is the elastic energy of the lattice, which has the form

$$\mathscr{E} = \frac{E}{2}(e_1^2 + e_2^2 + e_3^2) + \Lambda(e_1 e_2 + e_2 e_3 + e_3 e_1) \tag{1}$$

where the e_α are the relative changes in the bond lengths. Young's modulus and the Poisson ratio are functions of E and Λ. The magnetic coupling is assumed to depend linearly on the bond distortions, i.e., $J_{ij} = J(1 - e_\alpha)$. The third term is a size-effect or "packing" term, which yields an antiferromagnetic interaction between next-nearest neighbors along each of the three axes. The Hamiltonian obtained is the same as the one derived from effective-medium theory[7,2]:

$$H = \sum_\alpha J(1 - \varepsilon e_\alpha) \sum_{\langle ij \rangle_\alpha} S_i S_j + \sum_\alpha \frac{K}{2} \sum_i \left(\sum_{\langle ij \rangle_\alpha} S_j \right)^2 + N\mathscr{E} \tag{2}$$

The model analyzed in ref. 6 corresponded to fixed values of the e_α; thus there were different but fixed nearest-neighbor antiferromagnetic couplings. Under this approximation, the model was shown to behave very much like the ANNNI model.[7] The analysis of this paper shows that integrating out the displacements leads to a different type of behavior.

The lattice distortions can be eliminated and an effective spin Hamiltonian obtained by minimizing with respect to the three e_α. Solving

[2] Effective-medium theory actually yields the term $(\sum_j S_j)^2$, where the sum is over all nearest neighbors. This would introduce other interactions besides the axial next-nearest-neighbor interactions retained in the current model.

these three equations yields the values of the lattice distortions in terms of the nearest-neighbor pair correlations along the three axes. For example,

$$e_1 = \frac{\varepsilon J}{E^2 + E\Lambda - 2\Lambda^2}\left[(E+\Lambda)\sum_{\langle ij\rangle_1} S_iS_j + (-\Lambda)\sum_{\langle ij\rangle_{2,3}} S_iS_j\right] \tag{3}$$

By substituting the values found for the lattice distortions into (2), the elastic term and the spin–lattice coupling term both become four-spin terms where nearest-neighbor pairs are coupled by an infinite range interaction. The resulting Hamiltonian is

$$H = J\sum_{\alpha}\sum_{\langle ij\rangle_\alpha} S_iS_j + \sum_{\alpha}\frac{K}{2}\sum_i\left(\sum_{\langle ij\rangle_\alpha} S_j\right)^2 + \sum_{\alpha,\beta}\Gamma_{\alpha\beta}\sum_{\langle ij\rangle_\alpha} S_iS_j \sum_{\langle kl\rangle_\beta} S_kS_l \tag{4}$$

The $\Gamma_{\alpha\beta}$ are polynomial fractions of E and Λ which are found in the above substitution.

In ref. 5, an exact solution was obtained to the above model with $K=0$ and it was shown that there is always a strong first- order transition from the disordered to the striped phase. The striped phase is considered as the uniform phase in our work and we look for the appearance of modulated phases at finite K. In the alloy context, the striped phase is the two-dimensional analog of the $L1_0$, CuAu phase.

3. MEAN-FIELD CALCULATIONS

To find the spatially modulated phases in the mean-field approximation, we used the original Hamiltonian (2). We looked only for one-dimensional modulated phases, assuming that the average spin was the same in each vertical column (where the modulation is chosen to be in the horizontal direction). In addition, the striped phase, which is known to be the ground state for 0 K, is taken as the unmodulated phase, so the average spin in each column is multiplied by $(-1)^\mu$, where μ labels the column. This procedure yields a one-dimensional chain of average spins for which we can write the mean field equations. This procedure is similar to that used by Bak and von Boehm to find the mean-field phase diagram for the ANNNI model.[1] The effective field at site μ can be written as

$$h_\mu = \sum_\nu J_{\mu\nu}\langle S_\nu\rangle \tag{5}$$

The $J_{\mu\nu}$ are the projections of the $J(1-\varepsilon e_\alpha)$ onto the horizontal direction. If the fields are known, the spins can be calculated from the equation

$$\langle S_\mu \rangle = \tanh(h_\mu/T) \tag{6}$$

The two mean-field equations must be solved self-consistently. The $J_{\mu\nu}$ depend on the $\langle S_\mu \rangle$ through e_α and are recalculated at each step. For each value of K and T the equations were solved iteratively for initial conditions of varying periodicity. The longest initial periodicity was 20. There were several self-consistent solutions for most values of K and T, depending on their initial conditions chosen. When several self-consistent solutions were found for some K and T, the solution with the lowest free energy was taken as the solution.

4. PHASE DIAGRAM

Two representative phase diagrams are shown (Figs. 1 and 2). The parameters for each are the same except for the coupling strength ε, which is higher in Fig. 2. Each diagram exhibits three ordered regions. The low temperature, low-K state is a striped phase, consisting of alternating rows of up and down spins, the same as found by Chen and Kardar[5] in the exact 0 K solution. At higher values of K (and low temperature), this solution becomes modulated in a direction perpendicular to the stripes with the period of modulation equal to two lattice lengths. This phase is labeled the $\langle 2 \rangle$ phase. This phase is favored at large values of K since the second-nearest-neighbor pairs are antiferromagnetic along two of the three axes in the $\langle 2 \rangle$ phase and one of the three in the striped phase. This energy term competes with the elastic energy term, which favors the striped phase. In each phase diagram, there is a higher temperature modulated phase labeled the $\langle 3 \rangle$ phase, which retains the symmetry of the lattice. This phase is formed by dividing the lattice into three sublattices, one of up spins, one of down spins, and one which is disordered. The lattice distortions in this phase are the same in the three axial directions.

The transition to the striped phase involves two broken symmetries: one corresponding to the Ising spin and the other a three-state Potts symmetry associated with the change in bond lengths.[5] The transition to the $\langle 3 \rangle$ state does not break the three-state Potts symmetry.

Two representative mean-field phase diagrams for parameter values $E=5$, $\Lambda=2$, and $J=1$ are shown in Figs. 1 and 2. For $\varepsilon=1$, Fig. 1 shows a second-order transition from the disordered to the $\langle 3 \rangle$ phase for all values of K. There is a subsequent first-order transition from the $\langle 3 \rangle$ phase to the striped phase, or the $\langle 2 \rangle$ phase, at a lower temperatures. In their

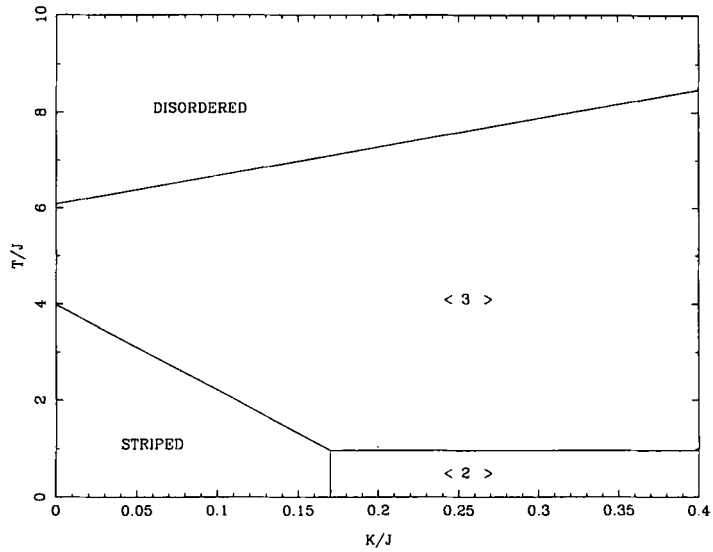

Fig. 1. The mean-field phase diagram for a small coupling to the lattice($E=5$, $A=5$, $J=1$, and $\varepsilon=1$). The transition to the $\langle 3 \rangle$ phase at $K=0$ is a mean-field artifact.[5]

exact solution of the $K=0$ case of this model, Chen and Kardar have shown that there is a first-order transition to the striped phase for all values of ε, E, and A. Thus at least at $K=0$, the $\langle 3 \rangle$ phase is a mean-field artifact. The long-range nature of the four-spin interaction in Eq. (4) could have led one to believe that a mean-field analysis of this model would be adequate; however, the numerical evidence is contrary to this expectation. It is also known from the analysis of nonfrustrated compressible Ising models that fluctuations beyond meanfield can change the nature of the transition.[6]

The phase digram for $\varepsilon=2$, shown in Fig. 2, is qualitatively different from the phase diagram for $\varepsilon=1$. For small values of K, there is a first-order transition from the disordered to the striped phase with no intervening $\langle 3 \rangle$ phase. At higher values of K, there is a sequence of two transitions: a second-order transition from the disordered to the $\langle 3 \rangle$ phase followed by a first-order transition either to the striped phase or the $\langle 3 \rangle$ phase. This is reminiscent of the sequence of transition seen in CuAu.

In order to analyze the mean-field phase diagrams, we transform Eq. (2) using the Hubbard–Stratanovich transformation[3] and study the Lagrangian density in q-space. If the transition is second order, the quadratic

[3] For a review of the Hubbard–Stratanovich transformation, see Amit.[8]

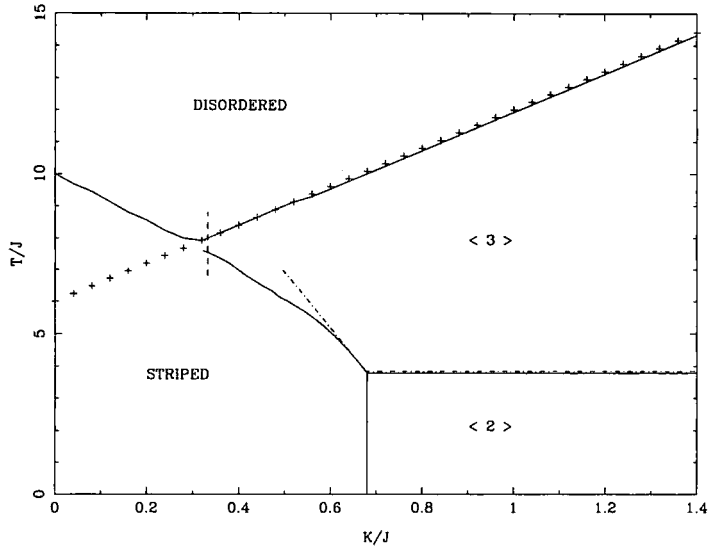

Fig. 2. The mean-field phase diagram for a larger coupling to the lattice ($\varepsilon = 2$) but the same values as Fig. 1 of all the other parameters. The dashed-dotted lines are results of low-temperature calculations (see discussion in text). The line of crosses denotes the line of anticipated second-order transitions to the $\langle 3 \rangle$ phase (see discussion in text), and the vertical line marks the value of K where the transition to the $\langle 3 \rangle$ phase becomes first order.

term determines the transition temperature and the transition is to the phase where $J(q)$ is a minimum. In our model, the transformed interaction term $J(q)$ can be split into a part independent of the lattice deformations, $J_0(q)$, and a part linearly dependent on the deformations, $J_1(q)$. The latter term contributes in higher order after the deformations are eliminated by performing the Gaussian integral over the e_α and therefore the quadratic term involves $J_0(q)$ only:

$$\mathscr{L}^{\text{quadratic}} = \beta \sum_q J_0(q)[1 - 2\beta J_0(q)]\, \psi_q \psi_{-q} \tag{7}$$

In our model, $J_0(q)$ is a minimum for $q = 2\pi/3$ for all K, corresponding to the $\langle 3 \rangle$ phase. This is in contrast to the ANNNI model, where q varies with the second-neighbor coupling. The second-order transition temperature to the $\langle 3 \rangle$ phase is $T_c = 2J_0(q)$ at $q = 2\pi/3$. Mean-field theory would therefore predict a second-order transition from the disordered to the $\langle 3 \rangle$ phase for all values of K and ε unless this is preempted by a first-order transition. Thy transition from the disordered to the striped phase is

therefore always first order and stabilized by the quartic term in the Lagrangian density. The transition from the disordered to the $\langle 3 \rangle$ phase can also become first order in mean-field theory if the coefficient of the quartic term becomes negative. In that case, the sequence of transitions is determined by the numerical values of the first-order transition temperatures from the disordered to the striped phas and the disordered to the $\langle 3 \rangle$ phase.

To locate the point where the transition to the $\langle 3 \rangle$ phase changes from second order to first order, we look at the quartic part of $\mathscr{L}$. If the sign of the coefficient of the ψ^4 term is negative at the second-order transition temperature, the second-order transition is preempted by a first-order transition. Keeping terms up to second order in e_α, the displacement integrals are Gaussian and the e_α can be eliminated to obtain an effective spin Lagrangian. We first solve for the e_α in much the same way as we did to get Eq. (4), but now specifically in the $\langle 3 \rangle$ phase, where the e_α are all equal, as the $\langle 3 \rangle$ phase retains the symmetry of the lattice. By minimizing $\mathscr{L}(e_\alpha)$ we find that

$$e_\alpha = \frac{\varepsilon J(1-12\beta(J+K))}{N(E+2\Lambda)-36\varepsilon^2 J^2 \beta \psi_q \psi_{-q}} \psi_q \psi_{-q} \tag{8}$$

Substituting this back into $\mathscr{L}$, there are several contributions to the quartic term of the effective spin Lagrangian. The first is from the expansion of the $\log[\cosh(2\beta J)]$ yielded by the Hubbard–Stratanovich transformation. The second is from the substitution of e_α into the quadratic term, and the third is the quartic part of the elastic term. We have

$$\mathscr{L}^{\text{quartic}} = \left[108\beta^4(J+K)^4 - \frac{3\varepsilon^2 J^2 \beta[1-12\beta(J+K)]^2}{E+2\Lambda} + \frac{\frac{3}{2}\varepsilon^2 J^2 \beta[1-12\beta(J+K)]^2}{E+2\Lambda} \right] \psi^4 \tag{9}$$

Note that the elastic term is opposite in sign and exactly one-half the magnitude of the coupling term. Near the second-order transition, the β can be replaced by $1/[6(J+K)]$. The condition for the quartic term to be negative, and thus for the phase transition to be first order, is

$$K < \frac{3\varepsilon^2 J^2}{E+2\Lambda} - J \tag{10}$$

This condition is shown as a vertical line in Fig. 1. This line would move to higher values of K with increasing ε and one could envisage a

situation where there is a first-order transition to the $\langle 3 \rangle$ phase followed by a first-order transition to the striped phase as the temperature is lowered. Our numerical results, for the range of ε studied, show a first-order transition only to the striped phase, suggesting that the quartic term favors the striped phase over the full range of K.

The transition from the disordered to the $\langle 3 \rangle$ phase is exactly analogous to the ferromagnetic transition in a *compressible* Ising model.[6] This transition is known to be driven first order, for all values of coupling to the lattice, by fluctuations beyond mean field and we expect the same scenario for the disordered $\Rightarrow \langle 3 \rangle$ transition.

The line of crosses in Fig. 2 show the anticipated second-order transition into the $\langle 3 \rangle$ phase obtained from $T_c = 2J_0(q)$ at $q = 2\pi/3$, As seen from the phase diagram, there is a range of K for which this second-order transition is preempted by the first-order transition to the striped phase. The calculations suggest that there is a point in this phase diagram where the disordered, striped, and $\langle 3 \rangle$ phases meet. Numerically, this point is close to the critical value of K where the transition to the $\langle 3 \rangle$ phase becomes first order. However, the accuracy of our numerical calculations was not sufficient to conclusively demonstrate the existence of this point. This triple point is reminiscent of the Lifshitz point found in ANNNI models, but is not a multicritical point, since we have two first-order and one second-order transition lines meeting at this point. Moreover, the ordering wavevector has a finite jump from zero to $2\pi/3$ at this point, in contrast to the ANNNI model, where this wavevector changes continuously.

At low temperatures, the transitions between the phases can be calculated analytically, if we make the approximation that the average spins, the $\langle S_i \rangle$, are either 1, -1, or 0. In this case the energy for each phase can be calculated, and the entropy per spin is ln(2) times the fraction of zeros. In the striped phase

$$\langle S_i S_j \rangle_{\langle ij \rangle_1} = +1 \qquad \text{and} \qquad \langle S_i S_j \rangle_{\langle ij \rangle_{2,3}} = -1$$

Substituting these values into (4) and calculating the K term in a similar manner, one obtains

$$H^{\text{striped}} = -J + 6K - \varepsilon^2 J^2 \frac{3E + 5\Lambda}{E^2 + E\Lambda - 2\Lambda^2} \tag{11}$$

In the $\langle 3 \rangle$ phase, $\langle S_i S_j \rangle_{\langle ij \rangle_\alpha}$ for all α. In this phase, the energy is

$$H^{\langle 3 \rangle} = -J + 2K - \varepsilon^2 J^2 \frac{E - \Lambda}{3(E^2 + E\Lambda - 2\Lambda^2)} \tag{12}$$

In the $\langle 3 \rangle$ phase, $\langle S_i S_j \rangle_{\langle ij \rangle_1}$ and $\langle S_i S_j \rangle_{\langle ij \rangle_{2,3}}$ Here, the energy is

$$H^{\langle 2 \rangle} = -J + 2K - \varepsilon^2 J^2 \frac{E + \Lambda}{E^2 + E\Lambda - 2\Lambda^2} \tag{13}$$

The calculated transition line between two states is where the free energies of the two states are equal. The entropy is zero in both the striped phase and the $\langle 2 \rangle$ phase, and is $[N \log(2)]/3$ in the $\langle 3 \rangle$ phase. The calculated transition lines are shown as broken lines in Fig. 2. The calculated line between the striped and the $\langle 2 \rangle$ phases is too close to the numerical results to appear separate.

5. DISCUSSION

The mean-field phase diagram of our model exhibits both first-and second-order phase transitions to ordered unmodulated and modulated phases. The transitions differ in nature from the ANNNI model, with the striped phase (corresponding to the ferromagnetic phase of the ANNNI model) being stabilized by the elastic interactions. The $K=0$ model has only a single, first-order, transition from the disordered to the striped phase.[5] In the absence of elastic interactions, there is no ordering in this model. Our mean-field calculations show that the the stability of the $\langle 3 \rangle$ phase is enhanced at finite K. The numerical mean-field results did not exhibit any first-order transition to the modulated $\langle 3 \rangle$ phase. However, we have found no arguments to rule out this possibility. Moreover, all the transitions become first-order beyond mean field because of the existence of the long-range four-spin interaction [cf. Eq. (4)].

This model was inspired by microscopic calculations of alloy phase diagrams where there is a sequence of *disordered* $\Rightarrow$ *modulated* $\Rightarrow$ *striped* transitions as the temperature is lowered. The model considered in this paper shows such a sequence for a range of K values. The modulated phase found in this model is, however, not a true long-period structure. The modulated phases found in the alloys are closer to $\langle 10 \rangle$ than to $\langle 3 \rangle$. Finding such long-period structures may require the minimum of $J(q)$ to vary as a function of the magnitude of the longer range interactions, as measured by K.

The original EMT interactions, which were approximated in this model to axial second-neighbor interactions (see Footnote 2), can be expressed as

$$\cdots + K \sum_i \left[\sum_{\langle ij \rangle} S_j \right]^2 + \cdots \tag{14}$$

where all the nearest-neighbor spins are summed and squared, does have this property. The minimum of $J(q)$ for this model is not a constant and changes continuously from $2\pi/3$ at small K to $4\pi/3$ at large K. This model might be the right candidate for finding the type of modulated structures that we are looking for. Future work may involve examining similar models on a face-centered-cubic lattice; such models may shed further light on modulated ordering in binary alloys.

In conclusion, we have shown that there exists a class of models which can be derived from microscopic descriptions of interactions in metallic alloys which (i) have short-range competing Ising interactions and (ii) exhibit modulated phases separated by first-order transitions from the disordered and the unmodulated structures. This combination of properties is commonly observed in metallic alloys. The model is interesting in its own right because it combines short-range competing interactions with long-range four-spin interactions arising from the strain fields.

Our analysis has certain implications for Fermi-surface driven instabilities in alloys. The scenario, in the absence of elastic interactions, is that the disordered state has an instability at a certain $\mathbf{q}$ vector which is determined by Fermi surface nesting, and orders into this particular ordered structure (see ref. 9 for a review). At lower temperatures, locking into commensurate phases can occur, just as in the ANNNI model. We would like to suggest an alternate scenario where the lower temperature phase is favored by the coupling to elastic strain just as the striped phase is in our model. This would explain the occurrence of a ground state which is different from that prejdicted from Fermi-surface nesting, and it would explain why the transitions are first order.

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