Spatially frustrated lattice models

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A mean-field theory of a class of isotropic, spatially frustrated lattice-model Hamiltonians is constructed. Some general features of the mean-field equations are examined. Analytical techniques for determining the paramagnetic phase boundary and the low-temperature phases in the vicinity of this boundary are discussed. Finally, a numerical approach which has proved to be helpful in solving the mean-field equations is outlined.

I. DEFINTION OF MODEL HAMILTONIAN

We begin by defining a class of isotropic and periodic Ising spin Hamiltonians which have finite-ranged two-spin interactions on a simple-cubic lattice. By isotropy and periodicity we mean that the energy can be written

$$\mathcal{H}_l=\tfrac{1}{2}\sum_{\mathbf{n},\mathbf{n}'}\sigma_{\mathbf{n}}J_{\mathbf{n},\mathbf{n}'}\sigma_{\mathbf{n}}-H\sum_{\mathbf{n}}\sigma_{\mathbf{n}},\quad \mathbf{n}=(x,y,z)\ ,$$
$$\sigma_{\mathbf{n}}=\pm 1,\quad x,y,z=0,\pm 1,\pm 2,\ldots,\pm(l/2)\ , \tag{1.1}$$

where, in the limit $l\to\infty$, $\{J_{\mathbf{n},\mathbf{n}'}\}=\{J_{\mathbf{n}-\mathbf{n}'}\}$ is invariant under all the transformations of the lattice. The range of the interaction is defined to be that minimum number of bonds R from any central spin $\sigma_{\mathbf{n}}$, beyond which coupling constants are zero. The particular class of Hamiltonians which we shall study are those for which the linear operator $L_{\mathbf{n}}$, defined by

$$L_{\mathbf{n}}\sigma_{\mathbf{n}}=\sum_{\mathbf{n}'}^{(R)}J_{\mathbf{n},\mathbf{n}'}\sigma_{\mathbf{n}'}\ , \tag{1.2}$$

can be written as a polynomial in the lattice-difference operator $\Delta_{\mathbf{n}}^2$,

$$L_{\mathbf{n}}=\sum_{p=0}^{R/2}a_{2p}(\Delta_{\mathbf{n}}^2)^p\ . \tag{1.3}$$

Note that $\Delta_{\mathbf{n}}^2$ is the three-dimensional operator generalization of

$$\Delta_x^2 f_x=f_{x+1}-2f_x+f_{x-1},\quad x=0,\pm 1,\pm 2,\ldots\ . \tag{1.4}$$

That the linear operator $L_{\mathbf{n}}$ satisfies the conditions outlined above equation (1.2), follows from the observation that $\Delta_{\mathbf{n}}^2$ is invariant to all the lattice symmetries. To make contact between this notation and some familiar spin models we point out that a_0,a_2 finite, $a_{2p}=0(p>1)$ defines the nearest-neighbor (NN) Ising model, while a_0,a_2,a_4 finite, $a_{2p}=0(p>2)$ corresponds to a recently introduced[1] lattice model of microemulsions with NN and next-nearest-neighbor (NNN), both linear (LNNN) and diagonal (DNNN), interactions. This model has a rich variety of spatially modulated phases and, within mean-field theory and for certain regions of parameter space, it reduces to the Bak–von Bohm axial NNN Ising (ANNNI) model.

Thus, Hamiltonians of general form

$$\mathcal{H}_l=\tfrac{1}{2}\sum_{\mathbf{n}}\sigma_{\mathbf{n}}L_{\mathbf{n}}\sigma_{\mathbf{n}}-H\sum_{\mathbf{n}}\sigma_{\mathbf{n}} \tag{1.5}$$

can produce very rich phase diagrams due to the possibility of spatial frustration or competing interactions,[2,3] and one would like to understand some of their qualitative properties. To achieve this we will construct a local-density-mean-field theory.

For some systems of interest the mean-field theory is not a good approximation.[4] For this reason it may be necessary to justify its application to spatially frustrated lattice models.

There are many interesting problems in statistical physics where, at the beginning of the study, it is not obvious what general types of thermodynamic phases will appear on the phase diagram. In these cases local-density mean-field theory is the simplest statistical-mechanical treatment which preserves all the symmetries of the Hamiltonian. This is an important advantage since one does not want to exclude some complicated spatially ordered phase which may result from a spontaneous symmetry breaking of the Hamiltonian. Where the interactions in the Hamiltonian cause a high degree of spatial frustration Monte Carlo simulations may not be reliable because of problems of metastability or anomalous finite-size effects and, in any case, are best undertaken with prior knowledge of the mean-field phase diagram. Mean-field theory becomes formally exact in the limits of zero and infinite temperature, infinite range of interaction, or infinite lattice coordination number. However, the experience of many researchers with, for example, the ANNNI model seems to indicate that a properly constructed mean-field theory in three dimensions gives a qualitatively correct and sometimes quantitatively meaningful description.

Now, if one accepts that local-density mean-field theory is a useful framework for initial studies, it may still be difficult to solve the equations for the order parameters. Most of this paper is devoted to the derivation and discussion of useful analytical and numerical methods for studying lattice models with Hamiltonians of form (1.1).

II. THE LOCAL-MEAN-FIELD THEORY

The mean-field free-energy functional is most easily derived using the Gibbs-Bogoliubov inequality. Thus,

one can show that the exact free energy $\tilde{G}$, corresponding to a Hamiltonian $\mathcal{H}$ satisfies

$$\tilde{G} < G = G_s + \langle (\mathcal{H} - \mathcal{H}_0) \rangle_0 , \tag{2.1}$$

where G_s is the free energy of the Hamiltonian $\mathcal{H}_0$ and $\langle X \rangle_0$ means the thermal average of X with $e^{-\mathcal{H}_0/kT}$ as weight factor. If one chooses

$$\mathcal{H}_0 = -\sum_{\mathbf{n}} h_{\mathbf{n}} \sigma_{\mathbf{n}} ,$$

then $\{h_{\mathbf{n}}\}$ can be treated as a set of parameters which can be chosen to minimize G. It then follows that the free-energy density can be written as a functional only of $S_{\mathbf{n}} = \langle \sigma_{\mathbf{n}} \rangle_0$,

$$\begin{aligned} \mathcal{G}(S_n, a) = & -H \langle S_{\mathbf{n}} \rangle + \tfrac{1}{2} \langle S_{\mathbf{n}} L_{\mathbf{n}} S_{\mathbf{n}} \rangle \\ & + \tfrac{1}{2} kT \langle \{ (1+S_{\mathbf{n}}) \ln[\tfrac{1}{2}(1+S_{\mathbf{n}})] \\ & + (1-S_{\mathbf{n}}) \ln[\tfrac{1}{2}(1-S_{\mathbf{n}})] \} \rangle , \end{aligned} \tag{2.2}$$

where $\mathbf{a}$ stands for the set of coupling parameters in Eq. (1.3) and

$$\langle x_{\mathbf{n}} \rangle = \lim_{l \to \infty} \left[[1/(l+1)^3 \sum_{\mathbf{n}} X_{\mathbf{n}} \right] . \tag{2.3}$$

We will be interested in the global minima of the functional (2.2) for every choice of $\mathbf{a}$ and T. The functional Taylor series about a minimum with free-energy density $\mathcal{G}_0$ may be written

$$\begin{aligned} & \mathcal{G} = \mathcal{G}_0 + \sum_{r=1}^{\infty} K_{\mathbf{n}_1, \mathbf{n}_2, \ldots, \mathbf{n}_r} \delta S_{\mathbf{n}_1} \delta S_{\mathbf{n}_2} \cdots \delta S_{\mathbf{n}_r} , \\ & K^{(r)}_{\mathbf{n}_1, \mathbf{n}_2, \ldots, \mathbf{n}_r} = \frac{1}{r!} \frac{\delta^r \mathcal{G}}{\delta S_{\mathbf{n}_1} \delta S_{\mathbf{n}_2} \cdots \delta S_{\mathbf{n}_r}} \bigg|_{S_{\mathbf{n}}} . \end{aligned} \tag{2.4}$$

The kernels $K^{(r)}$ contain much information about the phase transitions of the system. For our purposes the most important derivatives are

$$K^{(1)} = \frac{\delta \mathcal{G}}{\delta S_{\mathbf{n}}} = L_{\mathbf{n}} S_{\mathbf{n}} + kT \tanh^{-1} S_{\mathbf{n}} - H = 0 , \tag{2.5}$$

$$K^{(2)} = \frac{\delta^2 \mathcal{G}}{\delta S_{\mathbf{n}} \delta S_{\mathbf{n}'}} = [L_{\mathbf{n}} + kT/(1 - S_{\mathbf{n}}^2)] \delta_{\mathbf{n},\mathbf{n}'} . \tag{2.6}$$

For any set of local spin densities to correspond to a local minimum of $\mathcal{G}$, it is necessary that

$$\langle K^{(2)}_{\mathbf{n},\mathbf{n}'} \delta S_{\mathbf{n}} \delta S_{\mathbf{n}'} \rangle > 0 \quad \text{for all } \delta S_{\mathbf{n}} , \tag{2.7}$$

or, if there is some choice $\{\delta \mathbf{S}_{\mathbf{n}}\}$ for which this is zero then $K^{(3)}$, $K^{(4)}$, or some higher derivative must be such that

$$\langle K^{(r)}_{\mathbf{n}_1, \mathbf{n}_2, \ldots \mathbf{n}_r} \delta S_{\mathbf{n}_1} \delta S_{\mathbf{n}_2} \cdots \delta S_{\mathbf{n}_r} \rangle > 0 . \tag{2.8}$$

Thus, $K^{(2)}$ is positive semidefinite and can be written

$$K^{(2)}_{\mathbf{n},\mathbf{n}'} = \sum_i K_i \phi_i(\mathbf{n}) \phi_i^*(\mathbf{n}'), \quad K_i \geq 0 \quad \text{all } i$$

where the K_i's are the eigenvalues of

$$\sum_{\mathbf{n}'} K_{\mathbf{n},\mathbf{n}'} \phi_i(\mathbf{n}') = K_i \phi_i(\mathbf{n}) . \tag{2.9}$$

A second-order phase transition occurs when at least one of these eigenvalues (K_0) is zero. This means that there is some choice, $\delta \bar{S}_{\mathbf{n}} = \phi_0(\mathbf{n})$, for which the left-hand side of Eq. (2.7) is zero. One then checks that the next nonzero term in the Taylor series is positive, in order to ensure that the $\{S_{\mathbf{n}}\}$ was indeed a stable solution of Eqs. (2.5). If the critical phase is spatially homogeneous then the eigenfunctions ϕ_i are plane waves. In this case it is possible to Fourier transform Eqs. (2.6) and obtain a closed formula for the eigenvalues $K_{\mathbf{q}}$. In general, the equations which determine the critical hypersurface in the parameters $\mathbf{a}$ are

$$K_j = 0 \quad \text{with } j = 0, 1, \ldots, t, \quad K_j > 0 \quad \text{with } j > t \tag{2.10}$$

where $j = 0, 1, \ldots, t$ labels those eigenvalues which satisfy

$$\min_i \{K_i\} = 0 . \tag{2.11}$$

When the critical phase is uniform ($S_{\mathbf{n}} = M_0$ for all $\mathbf{n}$) we have,

$$K^{M_0}_{\mathbf{q}_c} = 0 , \tag{2.12}$$

$$K^{M_0}_{\mathbf{q}_c} = \min_{\mathbf{q}} \{K^{M_0}_{\mathbf{q}}\} \Longrightarrow \nabla_q K^{M_0}_q \,|_{\mathbf{q}=\mathbf{q}_c} = 0 . \tag{2.13}$$

Equation (2.12) expresses the fact that there is no change in free energy [to $O(A^4)$] if the uniform solution $S_n = M_0$ is replaced by $S_n = M_0 + A\phi_{\mathbf{q}_c}$

$$\phi_q = \cos(q_1 x + \alpha_1) \cos(q_2 y + \alpha_2) \cos(q_3 z + \alpha_3) . \tag{2.14}$$

The second of these two equations ensures that the zero eigenvalues lie at the minimum of the spectrum. The matrix $K^{(2)}$ is positive semidefinite and if one then shows that $K^{(3)}$ or $K^{(4)}$ are positive, then the solution for the critical phase is at least a local minimum of the free energy at the phase transition. However, there remains the possibility that if the uniform solution is not a global minimum of the free energy, then solutions to Eqs. (2.12) and (2.13) do not determine a true phase transition. In fact, they would define a spinodal hypersurface which divides parameter space into regions where the solution M_0 is a stable or unstable solution of the Euler equations. Such a circumstance arises when the second-order phase transition we seek to study is preempted by a first order transition. Well-known examples are found in certain mean-field treatments of the Potts model,[5] and also similar behavior has been observed in a model of microemulsions.[6] Indeed, for lattice models with Hamiltonians of form (2.5) ($a_{2p} = 0$, $p > 1$) it is known that even mean-field descriptions can result in first-order transitions from disordered to ordered phases. However, the following proof shows that this can happen only if H in Eqs. (2.5) is nonzero. Local mean-field descriptions of all two-body Hamiltonians with only finite-ranged interactions and $H = 0$ have only second-order order-disorder transitions.[7]

We define a norm of the bounded linear operator $L_{\mathbf{n}}$ by

$$\|L_{\mathbf{n}}\| = \max_{-1 \leq S_{\mathbf{n}} \leq 1} \left\{ \frac{\|L_{\mathbf{n}} S_{\mathbf{n}}\|}{\|S_{\mathbf{n}}\|} \right\} . \tag{2.15}$$

The operator is Fourier transformed and its minimum eigenvalue determined from

$$L_{\mathbf{n}} f_{\mathbf{q}} = \lambda_{\mathbf{q}}^{\min} f_{\mathbf{q}}, \quad \mathbf{q} \text{ solution of } \nabla_{\mathbf{q}} L_{\mathbf{q}} = 0 \ . \tag{2.16}$$

Now turn to the equations (2.5) and define $f_{\mathbf{n}} = (1kT) L_{\mathbf{n}} S_{\mathbf{n}}$, where $-\infty < f_{\mathbf{n}} < \infty$. Taking the hyperbolic tangent of both sides of Eqs. (2.5) and then applying the operator $L_{\mathbf{n}}$ to both sides, we find

$$L_{\mathbf{n}} \tanh^{-1} f_{\mathbf{n}} = -kT f_{\mathbf{n}} \ , \tag{2.17}$$

which has a solution $f_{\mathbf{n}} = 0$ for all $\mathbf{a}$ and T.

Now take the norm of Eqs. (2.16) and, since

$$|kT| \, \|f_{\mathbf{n}}\| \leq \|L_{\mathbf{n}}\| \, |\tanh f_{\mathbf{n}}| \leq \|L_{\mathbf{n}}\| \, \|f_{\mathbf{n}}\| \tag{2.18}$$

can be satisfied only for $|kT| \leq |\lambda_{\mathbf{q}}^{\min}|$ unless $f_{\mathbf{n}} = 0$ (all $\mathbf{n}$), we see that they must have only the zero solutions for $|kT| \geq |\lambda_{\mathbf{q}}^{\min}| \equiv kT_c$. Thus, above kT_c one has only the paramagnetic phase where $S_{\mathbf{n}} = 0$. Note that a wave vector q_c, which satisfies (2.16) is also a solution of Eq. (2.13) and $K_{\mathbf{q}_c}^0 = L_{\mathbf{q}_c} - kT_c = 0$. In addition, one can show that $K^{(3)}$ is zero and Eq. (2.8) for $r=4$ is satisfied. Thus, the surface in parameter space which is defined by $(T_c, \mathbf{a})$ is actually a surface of second-order transitions. Above and upon this surface ($T \geq T_c$) the paramagnetic phase is the global minimum of the free-energy density. Beneath it ($T < T_c$) the paramagnetic phase is an unstable solution since some of the eigenvalues $K_{\mathbf{q}}$ are negative.

There are some features of the critical hypersurfaces of models defined by the operator in (1.3) which are quite general. Thus, using the Fourier-transformed representation of the lattice-difference operator,

$$\Delta_{\mathbf{n}}^2 \rightarrow 2(e_{\mathbf{q}} - 3), \quad e_{\mathbf{q}} = \sum_{i=1}^{3} \cos q_i \ , \tag{2.19}$$

one can calculate $K_{\mathbf{q}}(S_{\mathbf{n}} = 0)$ from Eq. (2.6). The result is a polynomial of order p in the function $e_{\mathbf{q}}$,

$$K_{\mathbf{q}}^0 = \sum_{n=0}^{p} b_n e_{\mathbf{q}}^n, \quad -3 \leq e_{\mathbf{q}} \leq 3, \quad b_0 = kT + \text{const} \ . \tag{2.20}$$

We determine the critical surface from Eqs. (2.12) and (2.13), the latter now expressed as

$$\left[-\sum_{n=0}^{p} n b_n e_{\mathbf{q}}^{n-1} \right] \sin q_i = P_{\mathbf{q}} \sin q_i = 0, \quad i = 1,2,3 \ ; \qquad -\pi < q_i \leq \pi, \quad -3 \leq e_{\mathbf{q}} \leq 3 \ , \tag{2.21}$$

$$P_{\mathbf{q}} = -\sum_{n=0}^{b} n b_n e_{\mathbf{q}}^{n-1} \ .$$

There are various ways in which the left-hand side of Eq. (2.13) can become zero; the bracketed polynomial, $P(e_{\mathbf{q}})$, of order $p-1$ may be zero, all three wave numbers may be zero or π, or some combination of these possibilities. However, in the case where $(q_1, q_2, q_3) = (0,0,\pi)$ or $(0,\pi,\pi)$ one can show that (2.13) gives a local minimum only if $P(e_{\mathbf{q}}) = 0$. Thus, the elements of the Hessian matrix of $K_{\mathbf{q}}^0$ are given by

$$\frac{\partial K_q^0}{\partial q_i \, \partial q_j} = \frac{\partial P}{\partial e_{\mathbf{q}}} \sin q_j \sin q_i - P_{\mathbf{q}} \cos q_i \, \delta_{ij} \tag{2.22}$$

and one can verify that the wave vectors $(0,0,\pi)$ and $(0,\pi,\pi)$ give local maxima unless $P=0$ and that $(0,0,0)$ and (π,π,π) are local minima if $P > 0$. The solution to Eqs. (2.20) and (2.21) can thus be divided into three regions. The equation

$$\sum_{n=0}^{b} b_n e_{\mathbf{q}}^n = 0, \quad -3 \leq e_{\mathbf{q}} \leq 3 \ , \tag{2.23}$$

where $e_{\mathbf{q}}$ is given by $P(e_{\mathbf{q}}) = 0$ is valid for the region defined by $-3 \leq e_q \leq 3$. The second-order surfaces dividing paramagnetic from ferromagnetic and antiferromagnetic phases are given, respectively, by

$$\sum_{n=0}^{b} 3^n b_n = 0 \ , \tag{2.24}$$

$$\sum_{n=0}^{b} (-3)^n b_n = 0 \ . \tag{2.25}$$

Above ($T > T_c$ or $K_{\mathbf{q}_c}^0 > 0$) each of these surfaces the paramagnetic phase is the only stable solution, while just beneath ($T < T_c$ or $K_{\mathbf{q}_c}^0 < 0$) the only stable solutions are, respectively, the ferromagnetic and antiferromagnetic solutions. However, the second-order surface described by (2.23) is more complicated in a number of respects. To begin with, the equation $P(e_{\mathbf{q}}) = 0$ may have more than one solution for which the Hessian (2.22) is positive semidefinite. In this case one should choose the solution $e_{\mathbf{q}}$ which gives global minima of $K_{\mathbf{q}}^0$ [recall Eq. (2.13)]. Even when this choice is made there will, in general, only be one relation to determine the three wave numbers $\mathbf{q}_c = (q_1^c, q_2^c, q_3^c)$ in the region $-3 \leq e_{\mathbf{q}} \leq 3$. This means that the precise nature of the solutions to the Euler equations just beneath the paramagnetic-modulated critical surface cannot be determined by Eqs. (2.12) and (2.13) alone.

For this reason we develop a perturbation expansion for the solutions in terms of the small (positive) parameter $\varepsilon = 1 - T/T_c$. This method has been outlined briefly in an earlier paper.[10] It has two merits. The first is that the analysis described by Eqs. (2.10) and (2.11) can be placed in context, being merely the conditions arising from the zeroth order in a perturbation expansion. The second advantage is that the remaining symmetry in the first-order solutions (2.14) is removed and the solutions which correspond to the global minima of the free-energy density are constructed.

The Euler-Lagrange equations (2.5) are first divided by kT_c and then written ($H = 0$)

$$\begin{aligned} &L_{\mathbf{n}} S_{\mathbf{n}} + (1-\varepsilon) \tanh^{-1} S_{\mathbf{n}} = 0 \ , \\ &L_{\mathbf{n}} = \sum_{p=0}^{k/2} \alpha_{2p} (\Delta_{\mathbf{n}}^2)^p, \quad \alpha_{2p} = a_{2p}/kT_c \ . \end{aligned} \tag{2.26}$$

It can be shown that solutions to these equations have at most a regular singularity at $\varepsilon = 0$. In fact, by expanding $S_{\mathbf{n}}$ in the series,

$$S_{\mathbf{n}}=\sum_{p=0}^{\infty}\varepsilon^{\beta+p}S^{(p)} \tag{2.27}$$

(where $S^{(p)}\equiv S_{\mathbf{n}}^{(p)}$ is the pth-order contribution from perturbation theory), one sees that $\beta=\frac{1}{2}$, the expected mean-field order parameter exponent at a critical point. Then, by expanding the nonlinearity in Eqs. (2.26) and equating equal powers in ε, one obtains an infinite set of coupled equations which define the perturbation theory. To $O(\varepsilon^{5/2})$ we obtain

$$(L_{\mathbf{n}}+1)S^{(0)}=0\ ; \tag{2.28}$$

$$(L_{\mathbf{n}}+1)S^{(1)}=f^{(1)},\quad f^{(1)}=S^{(0)}-\tfrac{1}{3}(S^{(0)})^3\ ; \tag{2.29}$$

$$\begin{aligned}&(L_{\mathbf{n}}+1)S^{(2)}=f^{(2)}\ ,\\ &f^{(2)}=S^{(1)}+\tfrac{1}{3}(S^{(0)})^3-(S^{(0)})^2S^{(1)}-\tfrac{1}{5}(S^{(0)})^5\ .\end{aligned} \tag{2.30}$$

Equation (2.28) is solved by Fourier transformation. Thus, either $S^{(0)}=0$ or $K_{\mathbf{q}}=0$ and

$$S^{(0)}=\sum_{\mathbf{q}}{}'\,A_{\mathbf{q}}\phi_{\mathbf{q}}\delta(K_{\mathbf{q}})\ \text{ with } A_{\mathbf{q}}\text{ arbitrary}\ , \tag{2.31}$$

where elements of $\{\phi_q\}$ have the form given in (2.14) and K_q is the Fourier transform of the operator $(L_{\mathbf{n}}+1)$. Note that it is related to $K_{\mathbf{q}}^0$ by division of the latter by kT_c. By the prime on the sum in Eq. (2.31) we mean the following. In addition to ensuring $K_{\mathbf{q}}=0$ one must, by virtue of the requirement that $K_{\mathbf{q}}\geq 0$, only includes $\mathbf{q}$'s which satisfy conditions (2.13) or $P(\{\alpha_{2p}\},e_{\mathbf{q}})=0$. The other situations, $\mathbf{q}=(0,0,0)$ or (π,π,π), are much simpler (there is no degeneracy) and can be considered separately. We now make the following ansatz.[8] Let $S^{(0)}$ be represented by only one mode

$$S^{(0)}=A_{\mathbf{q}}\prod_{i=1}^{3}\cos(q_ix_i+\alpha_i),\quad x_1=x,\ x_2=y,\ x_3=z$$

all integer values . (2.32)

We can now make use of the Fredholm alternative to solve for the constants $A_{\mathbf{q}}$ and α_i in Eq. (2.29). Thus, one knowns that a nonzero solution to Eqs. (2.29) exists only if $f^{(1)}$ is orthogonal to every solution of the homogeneous equations defined by setting $f^{(n)}=0$ in any of the coupled equations (2.28) and (2.30). Orthogonality of two functions $p_{\mathbf{n}}$ and $g_{\mathbf{n}}$ on a lattice means [see Eq. (2.31)] $\langle p_{\mathbf{n}}g_{\mathbf{n}}\rangle=0$. Thus, the requirement

$$\langle S^{(0)}f^{(1)}\rangle=0$$

in Eq. (2.29) may be written

$$\langle (S^{(0)})^2\rangle=\tfrac{1}{3}\langle (S^{(0)})^4\rangle\ . \tag{2.33}$$

Using the result,

$$I_l=\frac{1}{(l+1)}\sum_{x=-l/2}^{l/2}\cos(qx+\alpha)\cos(q'x+\beta)$$

$$\sim\tfrac{1}{2}[\delta(q+q'+2n\pi)\cos(\alpha+\beta)+\delta(q-q'+2n\pi)\cos(\alpha+\beta)]+O(1/l),\quad \text{as } l\to\infty \text{ with } n=0,+1,\ldots, \tag{2.34}$$

along with (2.33), one finds

$$A_{\mathbf{q}}^2=3\times 4^3\prod_{i=1}^{3}\left[\frac{1+\delta(q_i+2n\pi)\cos(2\alpha_i)}{3+4\delta(2q_i+2n\pi)\cos(2\alpha_i)+\delta(4q_i+2n\pi)\cos(4\alpha_i)}\right]\ . \tag{2.35}$$

These results give the coefficients in first-order perturbation theory in terms of phase angles appearing in the function (2.32). In turn these can be determined by minimizing the free-energy density with respect to variational parameters α_i. To do this we must develop an expansion for the free-energy density.

Consider the functional (2.2) for $H=0$. Using Eq. (2.26) the interaction term can be eliminated and then expanding $S_{\mathbf{n}}$ in the series (2.27) we find

$$\mathcal{G}/kT_c\sim-\ln 2-\tfrac{1}{12}\varepsilon^2\langle (S^{(0)})^4\rangle -\varepsilon^3[\tfrac{1}{3}\langle (S^{(0)})^3S^{(1)}\rangle+\tfrac{1}{15}\langle (S^{(0)})^6\rangle]+O(\varepsilon^4)\ . \tag{2.36}$$

However, the $O(\varepsilon^2)$ term can be further simplified with the help of Eq. (2.33),

$$\mathcal{G}/kT_c\sim-\ln 2-\tfrac{1}{4}\varepsilon^2\langle (S^{(0)})^2\rangle+O(\varepsilon^3)\ . \tag{2.37}$$

Equation (2.32) along with (2.35) becomes

$$\begin{aligned}&\langle (S^{(0)})^2\rangle=\prod_{i=1}^{2}r_i\ ,\\ &r_i=(3)^{1/2}2\left[\frac{1+\delta(2q_i+2n\pi)\cos(2\alpha_i)}{3+4\delta(2q_i+2n\pi)\cos(2\alpha_i)+\delta(4q_i+2n\pi)\cos(4\alpha_i)}\right]\ .\end{aligned} \tag{2.38}$$

The free-energy density is minimized by finding the maxima of (2.38). There are clearly special values of q_i for which the phase angles contribute different amounts to r_i. The various possibilities are, after maximizing with respect to α_i,

(i) $q_i \neq 0, \pi/2, \pi$; $r_i = \frac{1}{3}$,

(ii) $q_i = 0, \pi$; $r_i = \frac{1}{2}$ (independent of α_i) ,

(iii) $q_i = \pi/2$; $r_i = \frac{1}{2}$ ($\alpha_i = \pi/4 + n\pi/2$, n integer) .

Thus, one seeks solutions (2.32) where as many of the q_i as possible are chosen from the set $0, \pi/2$, and π consistent with the constraint

$$\frac{P}{kT_c} = -\sum_n n\beta_n e_q^{n-1}, \quad \beta_n = b_n/kT_c , \tag{2.39}$$

where P is the polynomial defined in Eq. (2.21). Now imagine fixing the values b_n and kT_c [or α_n in Eq. (2.26)]. When there is a solution to Eq. (2.39), e_q lies between -3 and 3. However, one can always find $e_q = \pm 3, \pm 2, \pm 1, 0$ using only the values $q_i = 0, \pi/2, \pi$. Thus, in constructing first-order solutions (2.32) it is always possible to choose at least two of the q_i to have these special values which have an extra negative free-energy contribution. The third value will then be fixed by Eq. (2.39). One draws the conclusion that lattice models of the class we are studying will have modulations of maximum period four in two directions. In the third direction, and depending on the point in coupling parameter space, the modulation will have some period between one and infinity.

However, to this order in perturbation theory one cannot decide which combination of special values will be selected at any chosen value of the coupling parameters. To break this remaining degeneracy one must solve Eq. (2.30). Note that the various solutions to Eq. (2.29) which are degenerate at first order, each of which has q_1 and $q_2 = 0, \pi/2, \pi$, are not unique since one can always add a function BS^0 to the particular solution. However, one can apply the expression of the Fredholm alternative described beneath Eq. (2.32) to Eq. (2.30). In this way B can be determined. The three types of solutions which have to be considered are (i) $q_1 = q_2 = 0, \pi$; $q_3 \neq 0, \pi/2, \pi$; (ii) $q_1 = \pi/2$; $q_2 = 0, \pi$; $q_3 \neq 0, \pi/2, \pi$; and (iii) $q_1 = q_2 = \pi/2$; $q_3 \neq 0, \pi/2, \pi$. When the calculation outlined above is performed for each, one finds

$$B = \frac{1}{2}\left[\frac{1}{3K_{113}} - 1\right] , \tag{2.40}$$

where K_{113} means K_q evaluated at $q_1, q_2, 3q_3$ and q_1 and q_2 are 0, $\pi/2$, or π. The different solutions are now fully determined and are listed below:

(i) $$S_n \sim 2\varepsilon^{1/2}\phi_q + 2\varepsilon^{3/2}B\phi_q - \frac{2\varepsilon^{3/2}}{3K_{113}}\phi_{\bar{q}_1\bar{q}_2 3q_3} + O(\varepsilon^{5/2}) \quad \text{as } \varepsilon \to 0 ,$$

$$\phi_q = \cos(\bar{q}_1 x)\cos(\bar{q}_2 y)\cos(q_3 z); \quad \bar{q}_1, \bar{q}_2 = 0, \pi , \tag{2.41}$$

(ii) $$S_n \sim 2\sqrt{2}\varepsilon^{1/2}\phi_q + \frac{2\sqrt{2}\varepsilon^{3/2}}{3K_{113}}B\phi_q - \frac{2\sqrt{2}\varepsilon^{3/2}}{3K_{113}}\phi_{\bar{q}_1\bar{q}_2 3q_3} + O(\varepsilon^{5/2}) \quad \text{as } \varepsilon \to 0 ,$$

$$\phi_q = \cos(\bar{q}_1 x + \alpha_1)\cos(\bar{q}_2 y)\cos(\bar{q}_3 z); \quad \bar{q}_1 = \pi/2, \quad \bar{q}_2 = 0, \pi, \quad \alpha_1 = \pi/4 + n\pi/2 , \tag{2.42}$$

(iii) $$S_n \sim 4\varepsilon^{1/2}\phi_q + 4\varepsilon^{3/2}B\phi_q - \frac{4\varepsilon^{3/2}}{3K_{113}}\phi_{\bar{q}_1\bar{q}_2 3q_3} + O(\varepsilon^{5/2}) \quad \text{as } \varepsilon \to 0 ,$$

$$\phi_q = \cos(\bar{q}_1 x + \alpha_1)\cos(\bar{q}_2 y + \alpha_2)\cos(q_3 z), \quad \bar{q}_1 = \bar{q}_2 = \frac{\pi}{2}, \quad \alpha_i = \frac{\pi}{4} + \frac{n\pi}{2} . \tag{2.43}$$

Note that, since the original model was isotropic, one can generate different solutions simply by interchanging the x, y, and z axes in Eqs. (2.41)–(2.43). These formulas along with Eqs. (2.33) and

$$\langle S^{(0)} f^{(2)} \rangle = \langle S^{(0)} S^{(1)} \rangle + \tfrac{1}{3}\langle (S^{(0)})^4 \rangle - \langle (S^{(0)})^3 S^{(1)} \rangle - \tfrac{1}{5}\langle (S^{(0)})^6 \rangle$$

in Eq. (2.36), give

$$\mathcal{G}/kT_c \sim -\ln 2 - \frac{\varepsilon^2}{2} - \frac{\varepsilon^2}{3}\left[\frac{1}{3K_{113}} + 1\right] + O(\varepsilon^4) , \tag{2.44}$$

which is valid for all of the states [(i)–(iii)]. These states are degenerate at the level of first-order perturbation theory. Now further degeneracies will be broken by the term containing K_{113}. Recall that K_{113} stands for $K_{q_1 q_2 3q_3}$, where $q_1, q_2 = 0, \pi/2, \pi$. To decide which choice of these first two wave numbers yields the lowest free-energy density, one miniizes K_{113} over $q_1, q_2 = 0, \pi/2, \pi$ with q_3 fixed by the constraint (2.39). An example is given in Sec. III.

The formulas (2.12), (2.13), and (2.44) contain most of the important conclusions of this analysis. From them one concludes that it is possible to classify the local-density mean-field solutions of any Hamiltonian of form (1.5) for regions of parameters space near to the second-order transition. This is done simply by locating the transition using Eqs. (2.12) and (2.13) and then minimizing K_{113} subject to the constraint (2.13). It is possible to

show that, providing the single-mode ansatz is valid,[8] the remaining degeneracies can be broken by minimizing $K_{q_1 q_2 (2l+1) q_3}$, unless they are global degeneracies of the mean-field theory.

Before turning to a specific example it may be worth contemplating the physical significance of the series (2.53). The eigenvalues $K_{\mathbf{q}}$ may be interpreted as the cost in free energy of imposing a distortion (excitation or density wave) of wave vector $\mathbf{q}$ on the critical phase at the critical point. One is interested in determining which phase develops just beneath $T=T_c$. It is now natural to suppose that those perturbations with wave vector $\mathbf{q}_c$ which minimize the appropriate eigenvalues at the critical point indicate that the phase which develops beneath T_c will have wave vector $\mathbf{q}_c$. This is the essential content of the formulas (2.12), (2.13), and (2.44).

We now turn to some examples where the above analysis has been a useful tool in constructing the phase diagram.

III. APPLICATIONS OF THE PERTURBATION METHOD AND COMMENTS ON NUMERICAL METHODS FOR STUDYING LOCAL MEAN-FIELD THEORY

The simplest nontrivial example is defined by the linear operator

$$L_{\mathbf{n}} = a_4 \Delta_{\mathbf{n}}^4 + a_2 \Delta_{\mathbf{n}}^2 + a_0 \ , \qquad \mathcal{H} = \tfrac{1}{2} \sum_{\mathbf{n}} \sigma_{\mathbf{n}} L_{\mathbf{n}} \sigma_{\mathbf{n}} \ . \tag{3.1}$$

Recall that if $a_4=0$ then (3.1) defines the nearest-neighbor Ising model. The term in Δ_n^4 represents a sort of bending energy in the same way that terms in $(\nabla^2)^2$ have been used to approximate curvature energy in continuum mechanics. Models having energy contributions like those in Eq. (3.1) can be used to study microemulsions, biological membranes, and liquid crystals.[1,9] The choice of parameters,

$$a_4 = -M, \quad a_2 = -(J+12M), \quad a_0 = -6(J+5M) \ , \tag{3.2}$$

corresponds to a lattice model of microemulsions introduced by Widom[1] to which we have referred earlier. In this case, J and M are related to the activities and interaction energies of three types of molecules, oil, water, and amphiphile. The interested reader should consult the references for further details.[1,10–13]

From the arguments given beneath (2.15) we know that the Hamiltonian defined by Eq. (3.1) must have a second-order transition from the paramagnetic to all other phases. Thus, after Fourier transformation of the second-order derivatives defined by Eq. (2.6) ($S_{\mathbf{n}}=0$), Eqs. (2.12) and (2.13) become, respectively, (3.3) and (3.4),

$$-4me_{\mathbf{q}}^2 - 2je_{\mathbf{q}} + 6m + 1 = 0, \quad -3 \le e_{\mathbf{q}} \le 3 \ , \tag{3.3}$$

$$e_{\mathbf{q}} = -j_c/4m_c \ , \tag{3.4a}$$

or

$$\mathbf{q} = (0,0,0), (\pi,\pi,\pi) \ . \tag{3.4b}$$

Equations (3.3) and (3.4a) apply only between $-3 \le -j_c/4m_c \le 3$ and in this interval they describe a segment of the ellipse

$$6j_c^2 + (12m_c+1)^2 = 1 \ . \tag{3.5}$$

For $-j_c/4m_c > 3$ and $-j_c/4m_c < -3$ the respective solutions are $\mathbf{q}=(0,0,0)$ and (π,π,π). In these cases the second-order lines separating the paramagnetic and ferromagnetic or antiferromagnetic phases are respectively given by

$$j_c + 5m_c = \tfrac{1}{6} \ , \tag{3.6}$$

$$j_c - 5m_c = -\tfrac{1}{6} \ . \tag{3.7}$$

We can now follow the method described beneath Eq. (2.43) to determine the types of phase which correspond to global minima of the free-energy density beneath the ellipse (3.5). This is necessary because, as was pointed out in Sec. II, Eq. (3.4a) gives only one equation to determine three wavenumbers (q_1,q_2,q_3). Our task, then, is to minimize K_{113} on the ellipse (3.5), subject to the constraint (3.4a). Since

$$K_{113} = -m_c Y^2_{\bar{q}_1 \bar{q}_2 3 q_3} - j_c Y_{\bar{q}_1 \bar{q}_2 3 q_3} + 6m_c + 1; \quad Y_{\mathbf{q}} = 2e_{\mathbf{q}} \tag{3.8}$$

and

$$m_c = -1/(6+Y^2), \quad j_c = 2Y/(6+Y^2) \ , \tag{3.9}$$

$$Y = 2e_{\mathbf{q}_c}; \quad -6 \le Y \le 6 \ , \tag{3.10}$$

one finds that

$$K_{113} = \frac{64}{(6+Y^2)} e_3^2 (e_3^2 - 1)^2; \quad e_i = \cos q_i, \quad i = 1,2,3 \tag{3.11}$$

is to be minimized subject to the constraint $Y = -j_c/2m_c$. The results are most easily expressed in the following way. For every fixed value of Y (or $j_c/2m_c$) we can now give those sets of values $[e_1,e_2,e_3]$ which label the wavelengths of the modulations in the phases which minimize the free-energy density just beneath T_c. This allows us to construct a phase diagram which accurately describes all the paramagnetic region and some neighborhood of the second-order transition to the other phases.[11] In Fig. 1 we have exhibited these results and have also drawn the important first-order lines which separate the generic types of phase. Thus, for example, $[1,1,e_3]$ labels states which are translationally invariant in two directions, but which have modulations of wave number

$$q_3 = \cos^{-1} e_3; \quad e_3 = \tfrac{1}{2}Y - 2: \ \text{Region 1} \tag{3.12}$$

in the third direction. The symbol $[1,0,e_3]$ labels states which are translationally invariant in one direction, have period of oscillation four in the second, and are modulated with wave number determined by

$$e_3 = \tfrac{1}{2}Y - 1 \tag{3.13}$$

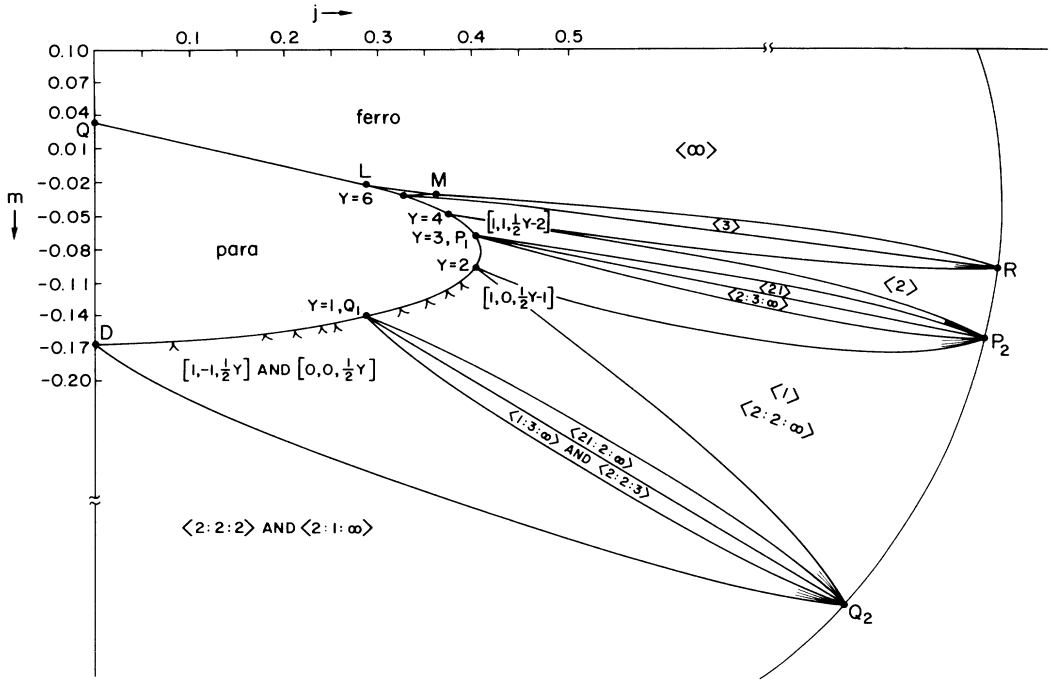

FIG. 1. Phase diagram (m vs j plot) for the lattice model of microemulsions (after Ref. 10). The straight line QL and the segment of the ellipse LD are the loci of second-order transition points between the paramagnetic and the other phases. All remaining phase transitions in the j,m plane are first order. Just outside the paramagnetic region and close to the curve LP_1 there are only planar phases described by the symbol $[1,1,\frac{1}{2}y-2]$ (Sec. III). Near the curve P_1Q_1 one finds states with modulations in two directions, labeled $[1,0,\frac{1}{2}y-1]$. In the region near the curve Q_1D there are degenerate phases described by $[1,-1,\frac{1}{2}y]$ and $[0,0,\frac{1}{2}y]$. The cusps marked on the curve LD are meant to indicate that commensurate-phase regions, of which there are an infinite number, grow from points to finite areas upon departure from the critical point. In the region beneath the line P_1P_2 only the periodic phases have been marked on the diagrams. Note that the multiphase points R, P_2, and Q_2 occur at zero temperature, and this corresponds to $j,m\to\infty$. The high-temperature analysis is consistent with the known zero-tempeature states (Refs. 10 and 13). The notation $\langle lm:kp:jn\rangle$ used to denote the various phases is discussed in detail in Ref. 10. This symbol denotes the following state. In the x direction of the lattice there are l up (or down) spins followed by m down (or up) spins. This configuration is repeated and the state thus has period $l+m$ in this direction. When l and m are equal we need only one integer to designate the arrangement. The spin configurations in the x, y, and z directions are thus designated by the three sets of integers separated by colons.

in the third direction. There is a first-order line (P_1P_2) between these two types of states which extends from zero temperature to the critical point on the ellipse. It should be noted, however, that at lower temperatures there are many first-order lines dividing the planar structures of differing commensurate periods from each other. The same is true at some low enough temperature for any commensurate structure. Actually, the phase transition defined by the line (P_1P_2) is due to a curvature instability and is reminiscent of the liquid crystalline transition from lamellar to rippled phases. This is not purely fortuitous since one would expect terms like those appearing in Eq. (3.1) to be present in some model of liquid crystals. At present we believe that this lattice model is capable of describing oil, water, liquid crystalline, and microemulsion phase equilibria.

We end this section by pointing out that analogous results can be obtained for the lattice model (3.1) in two dimensions. In both cases the conclusions one draws from the expansion around the critical line are consistent with our knowledge of the zero-temperature states and of extensive numerical calculations.[12] The nature of these calculations, which will be described in detail in a later publication, is worth mentioning because conventional techniques were found to fail for the models we have studied. Thus, one would naturally try to solve Eqs. (2.5) either directly or by a variational treatment based on the functional (2.2). In practice, for models which have competing interactions, this is found to be an extremely difficult process because there are so many metastable minima. For a three-dimensional model such as that defined by Eq. (3.1) we have not found it possible to apply either method with any success. Even when one attempts to use the method of simulated annealing there is little improvement in matters. However, if one carries out all calculations in Fourier space, global minima are always located with acceptable annealing times. Thus, Eqs. (2.5) can be replaced by a set of coupled Langevin equations,

$$\frac{\partial S_k}{\partial t}=-\frac{\partial F}{\partial S_k}+\lambda\eta_k S_k\ . \tag{3.14}$$

The functional F is the free-energy density as a functional of the Fourier transforms of the local order parame-

ters S_n. The parameters λ and η are, respectively, a coupling parameter (which couples the "heat bath" to the objective functional F) and a random number. Of course the functional F is not known explicitly, but can be calculated numerically by fast-Fourier transformation. In essence, Eqs. (3.14) are merely being used to sample the mean-field free-energy density in an efficient way. They are solved iteratively by reducing the coupling parameter in some logarithmic way. To understand the relative success of this procedure one must recall that the spin densities of the phases are usually represented by rather few terms in a Fourier series. In a sense, therefore, one is transforming to coordinates S_k in which the global minima are represented by rather few parameters which are relatively large. Finally, we note that the functional F in Eq. (3.14) is not confined to those which are from Hamiltonians of the type defined in Eq. (1.5). We have been able to use it successfully in investigations of lattice models with three-body interactions and a magnetic field.

IV. CONCLUSIONS

The success of local-density mean-field theory when applied to isotropic Hamiltonians is not assured. Even in that region of the phase diagram, Fig. 1, which corresponds to the ANNNI model, one must bear in mind that, unlike the case of the ANNNI model, the planar phases arose as a result of spontaneous symmetry breaking. Thus, the well-attested success of the mean-field theory of the ANNNI model does not necessarily ensure that isotropic models are well described by the method. The fluctuations in a model which is intrinsically one dimensional are expected to be quite different from those where the spatial frustration is three dimensional. For example, the validity of mean-field theory for one example of an isotropic frustrated lattice model has been discussed in Refs. 13. Preliminary Monte Carlo studies[14] indicate that the qualitative nature of the phase diagram of the model described in Sec. III is well described by mean-field theory. There is, however, a marked decrease in the transition temperature for a given layered phase to the paramagnetic phase. Also, when the mean-field solutions are infinitely degenerate the periodic phases do not appear to be stable when used as initial configurations in a Monte Carlo simulation. For example, this would be true of the $\langle 1 \rangle$ phase in Fig. 1 which, within mean-field theory, is degenerate at all temperatures with an infinite number of nonperiodic structures. However, it seems that the version of mean-field theory described in this article is valid (in three dimensions) whenever one obtains locally stable solutions to Eqs. (2.5). If, as we believe, this is a general principle, then the method has the important advantage of being self-consistent. When, for certain values of the parameters, the mean-field solutions are not locally stable we cannot make any firm prediction about the nature of the thermodynamic phase. However, it is possible that, in the example discussed in Sec. III, the paramagnetic phase replaces the unstable periodic phases predicted by mean-field theory.

In conclusion, then, we believe that since the local-density mean-field theory is usually a worthwhile first approximation it is important to be able to solve it properly. We believe that Eqs. (2.12), (2.13), and (2.44) already give much useful information about the theory. In addition, they are useful in providing independent checks on numerical procedures such as that outlined beneath Eq. (3.14).

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[7]For example, the mean-field treatment of the three-state Potts model in three dimensions predicts a first-order order-disorder transition. However, the mean-field entropy term is different from that of the Ising model and one cannot write the inequality (2.18). In fact, upon expanding the free-energy density in powers of the magnetization, one finds a cubic term with a negative coefficient. It is this term which is responsible for the first-order transition.

[8]In this paper, the use of only one mode in the sum (2.31) is viewed as an ansatz. However, there are reasons for believing that this choice is, in general, quite appropriate. At present we merely note that the following rules, deduced on the basis of a two-model analysis, seem to hold. If the state predicted by Eq. (2.43) is not degenerate then the inclusion of more terms at zeroth order is unnecessary since the state is the unique global minimum of the free-energy density. If the choice of a single mode [Eq. (2.32)] at zeroth-order perturbation theory results in degenerate solutions, then these are still global minima of the free-energy density. However, there may exist yet other distinct solutions to Eqs. (2.26) with which they are degenerate. This situation arises, for ex-

ample, in the phase diagram, Fig. 1. In the region bounded by the lines Q_1Q_2, the segment of the ellipse from Q_1 to D and the m axis, there are two degenerate states produced by this perturbation theory. However, one can show that these are degenerate with other states which cannot be represented by a single mode at the zeroth order, Eq. (2.31).

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