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Stability conditions for the generalized Ising model: $S = 1$

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We consider the generalized Ising model for a spin-1 system, i.e., the most general static Hamiltonian with pair interactions in a spin-1 subspace. This Hamiltonian has previously been used to model lattice-gas systems, ternary alloys, and normal liquid mixtures, as well as other three-state systems. As special cases one obtains the Blume-Emery-Griffiths model for ^{3}He-^{4}He mixtures and the Blume-Capel model for singlet-ground-state systems. Here we use exact high-temperature expansions in field, to the linear term in $\beta = (k_B T)^{-1}$ for the pair-correlation functions, to construct the generalized static susceptibility. The singularity in the susceptibility as the system is lowered in temperature yields an expression for the temperature at which the system becomes unstable with respect to fluctuations in the order parameters. This stability temperature is a function of the order parameters and the interactions. Our result is equivalent to the mean-field approximation (with its incumbent limitations) and yields a surface in thermodynamic space which separates regions of stability (or metastability) and instability. We look specifically at the stability surfaces and stability temperatures for a number of special cases as well as the general result.

I. INTRODUCTION

The spin-$\frac{1}{2}$ Ising model in field has been used to model a multitude of two-state systems which exhibit cooperative phenomena.[1] This is in addition to its original purpose of describing the behavior of a highly anisotropic ferromagnet. The Hamiltonian for this system can be written

$$H = -\frac{1}{2} \sum_{gp} I(gp) S_g^z S_p^z - h \sum_g S_g^z \ , \tag{1.1}$$

where S_g^z is the z component of a spin-$\frac{1}{2}$ ($\hbar = 1$) vector at site g and $I(gp)$ is a translationally invariant pair interaction with

$$I(gp) = I(pg) = I(|\vec{g} - \vec{p}|) \ , \tag{1.2a}$$

$$I(gg) = 0 \ . \tag{1.2b}$$

The effective external field conjugate to S_g^z is given by h.

The study of Ising systems with spin greater than one-half as models for real systems is a relatively recent phenomenon. This has been prompted in part by (a) the discovery of magnetic systems which are indeed highly anisotropic and thus Eq. (1.1) is a reasonable description,[2] (b) the utility of describing multicomponent systems[3] with Hamiltonians similar to Eq. (1.1), and (c) the richness of the phase transitions exhibited by these multicomponent systems.[4] It therefore would appear worthwhile to generalize the concept of the usual Ising model to higher spin systems. Thus, in analogy to Eq. (1.1), we define the generalized Ising model (GIM)[5] for spin S as

$$\begin{aligned} H_S = &- \sum_{g}^{N} \sum_{\alpha}^{2S} h_\alpha (S_g^z)^\alpha - \frac{1}{2!} \sum_{g_1,g_2}^{N} \sum_{\alpha_1,\alpha_2}^{2S} I_{\alpha_1\alpha_2}(g_1 g_2)(S_{g_1}^z)^{\alpha_1}(S_{g_2}^z)^{\alpha_2} \\ &- \frac{1}{3!} \sum_{g_1,g_2,g_3}^{N} \sum_{\alpha_1,\alpha_2,\alpha_3}^{2S} I_{\alpha_1\alpha_2\alpha_3}(g_1 g_2 g_3)(S_{g_1}^z)^{\alpha_1}(S_{g_2}^z)^{\alpha_2}(S_{g_3}^z)^{\alpha_3} \\ &- \cdots - \frac{1}{n!} \sum_{g_1,g_2,\ldots,g_n}^{N} \sum_{\alpha_1,\alpha_2,\ldots,\alpha_n}^{2S} I_{\alpha_1\alpha_2\cdots\alpha_n}(g_1 g_2 \cdots g_n)(S_{g_1}^z)^{\alpha_1}(S_{g_2}^z)^{\alpha_2} \cdots (S_{g_n}^z)^{\alpha_n} \ , \end{aligned} \tag{1.3}$$

where h_α are effective external fields (possibly nonphysical) conjugate to the operators $(S_g^z)^\alpha$ and $I_{\alpha_1\alpha_2}(g_1g_2), I_{\alpha_1\alpha_2\alpha_3}(g_1g_2g_3), \ldots, I_{\alpha_1\alpha_2\cdots\alpha_n}(g_1g_2\cdots g_n)$ are two-body, three-body, and general n-body interactions, respectively. The limit on the spin summations in Eq. (1.3) is given by the kinematic constraint

$$\prod_{\alpha}^{2S+1} (S_g^z - S_\alpha) = 0 \ , \tag{1.4}$$

where $S_\alpha = -S, -S+1, \ldots, S-1, +S$. Since H_S

commutes with operators made up of $(S_g^z)^\alpha$, the GIM is particularly convenient for describing the thermodynamics of many classical multicomponent systems, or $(2S+1)$-state systems.

In the present paper we will be primarily interested in the GIM for spin one with only two-body interactions. We do note, however, that the use of both three-body[6] and four-body potentials[7] in Eq. (1.3) has occurred in the literature. For the spin-one system we write from Eq. (1.3)

$$\begin{aligned} H_1 = -\sum_g [h_1 S_g^z + h_2 (S_g^z)^2] \\ -\frac{1}{2}\sum_{g,p} [I_{11}(gp) S_g^z S_p^z + I_{22}(S_g^z)^2 (S_p^z)^2 \\ + I_{12}(gp) S_g^z (S_p^z)^2 \\ + I_{21}(gp)(S_g^z)^2 S_p^z] \ , \end{aligned} \tag{1.5}$$

which, for ease of notation, we rewrite

$$\begin{aligned} H \equiv H_1 = -\sum_g [h_1 S_g^z + h_2 (S_g^z)^2] \\ -\frac{1}{2}\sum_{g,p} [I_1(gp) S_g^z S_p^z \\ + I_2(gp)(S_g^z)^2 (S_p^z)^2 \\ + I_3(gp) S_g^z (S_p^z)^2] \ . \end{aligned} \tag{1.6}$$

The study of special cases of Eq. (1.6) began a little over a decade ago with the introduction of the Blume-Capel[8] model (BCM). The BCM is obtained from Eq. (1.6) by retaining only the bilinear interaction $I_1(gp)$ and the field h_2. At that time the model was used to describe a singlet-ground-state system in the presence of uniaxial single-ion anisotropy. The model is also of interest (among many other reasons) because it exhibits a tricritical point.

If, in addition to $I_1(gp)$, we also retain the quadrupolar interaction $I_2(gp)$ we obtain the Blume-Emery-Griffiths (BEG)[9] model for ^{3}He-^{4}He mixtures. Also, in this form, the GIM is applicable to magnetic systems having both dipolar and quadrupolar interactions.[10]

The full GIM Hamiltonian of Eq. (1.5) has been used by Mukamel and Blume[11] in the mean-field approximation (MFA) to study tricritical points in ternary mixtures. Also Sivardière and Lajzerowicz[12] have used Eq. (1.5) as the basis for a spin-one lattice-gas model, in a MFA, to study the properties of simple fluids and binary and ternary fluid mixtures. The GIM has also been used to study the properties of ternary alloys in both the ordered[13] and disordered[14] regimes. The global phase diagram for this system in the MFA has recently been considered by Furman, Dattagupta, and Griffiths.[15]

While most of the above cited works have used the MFA in determining the system thermodynamics, the methods of the renormalization-group approach have also been applied to special cases of Eq. (1.5), i.e., the BCM[16] and the BEG[17] model. This interest has been prompted by the fact that these systems exhibit tricritical behavior, as well as other interesting critical phenomena.

For a thermodynamic system knowledge of the regions on the phase diagram where the system becomes unstable with respect to fluctuations in the order parameters is vital to understanding the properties of the phase transition. For a binary phase-separating solid solution,[18] for example, the phase diagram is divided by the miscibility gap and the spinodal into regions of stability, metastability, and instability. Likewise in magnetic systems the phase diagram is divided up by the magnetization curve and the stability curve.[8] The behavior of these curves determines whether a phase transition will be first or second order. In either case it is important to locate the stability curve in order to better understand the phase transition and also since it plays an important role in the transformation kinetics.[19]

In the present paper we present a straightforward and physically appealing way of calculating stability surfaces for classical multicomponent lattice systems, i.e., systems described by the GIM. The method is based on classical fluctuation theory and uses high-temperature series expansions for the pair-correlation functions in field. The pair-correlation functions are used to construct the generalized static susceptibility, and an appropriate series inversion is performed. Singularities in the generalized susceptibility then yield expressions for the stability surfaces.

As an example of the technique we calculate the stability surfaces for the spin-one GIM within the MFA only. Consequently our results are not different from many of those mentioned above, and also suffer from problems intrinsic to the MFA.[20] However, the technique used here is directly amenable to extensions beyond the MFA, and we have recently presented these results for the ternary alloy.[21] After presenting the MFA results for the spin-one GIM we will look specifically at a number of the more common special cases.

The outline of the paper is as follows: in Sec. II we present the results for the pair correlation functions based on the spin-one GIM and in Sec. III discuss the correlation-function approach (CFA) and construct the static generalized susceptibility. From this generalized susceptibility we calculate the stability temperature as a function of the order parameters and the interactions. In Sec. IV we discuss stability surfaces and investigate the surfaces for a number of special cases. We conclude in Sec. V with a discussion of the results and the technique.

II. HIGH-TEMPERATURE EXPANSIONS

The usual approach to determining the equilibrium thermodynamics of systems governed by Hamiltonians of the form (1.3) is to calculate the free energy by using some appropriate approximation: usually a cluster approximation or a variational technique. The free energy can then be used to determine the behavior of the order parameters and consequently the properties of the phase transition. This procedure and the various approximations are well documented in the literature.[1,22]

Although the calculation of the exact free energy is the ultimate goal of any thermodynamic problem, in most cases it is not feasible and sometimes not necessary. Indeed the approach taken here works directly with the correlation functions rather than the free energy. While we do not have access to all the information available from the free energy, we do have a straightforward and physically appealing procedure for working in the disordered regime using correlation functions calculated by means of high-temperature series expansions. In this section we will discuss the calculation of the correlation functions, while we will determine the stability surface in Sec. III.

A number of papers have considered the correlation functions, and hence the susceptibility, for special cases of the GIM for spin one by using high-temperature series expansions.[23–26] However, all of these studies, besides only looking at special cases of the GIM, have either considered only zero field or have not calculated all of the correlation functions relevant to the GIM. Also most of these studies have been concerned only with the critical point for second-order phase transitions.

The exception is the work of Tahir-Kheli[14] in which all correlation functions relevant to the spin-one GIM are calculated for arbitrary fields. This expansion itself is based on exact formal relationships for the correlation functions, which are unique to the GIM.[27] Besides forming the basis for high-temperature expansions,[28–30] the correlation identities can serve as a starting point for calculations in the ordered regime[13] and have recently been used to investigate the critical region.[31]

We refer the interested reader to Ref. 14 for the details of both the derivation of the correlation function identities and the series expansion techniques for the three-state system. We do point out that in order to keep the fields arbitrary the expansion coefficients are temperature dependent, but due to grand canonical averaging our final expression contains only the interactions and the order parameters.

For the spin-one system there are two order parameters,

$$m \equiv \langle S_g^z \rangle \ , \tag{2.1a}$$

and

$$Q \equiv \langle (S_g^z)^2 \rangle \ , \tag{2.1b}$$

where $\langle \cdots \rangle$ is the usual thermal average. The results for the pair-correlation functions from Tahir-Kheli's paper[14] for an expansion to the first order in $\beta = (k_B T)^{-1}$ are

$$\begin{aligned}\langle S_g^z S_p^z \rangle - m^2 = {} & \delta_{gp}(Q-m^2) + \beta I_1(gp)(Q-m^2)^2 \\ & + \beta I_2(gp) m^2 (1-Q)^2 \\ & + \beta I_3(gp) m (1-Q)(Q-m^2) \\ & + O(\beta^2) \ ,\end{aligned} \tag{2.2}$$

$$\begin{aligned}\langle (S_g^z)^2 (S_p^z)^2 \rangle - Q^2 = {} & \delta_{gp} Q(1-Q) \\ & + \beta(1-Q)^2 [I_1(gp) m^2 + I_2(gp) Q^2 \\ & + I_3(gp) mQ] + O(\beta^2) \ ,\end{aligned} \tag{2.3}$$

and

$$\begin{aligned}\langle S_g^z (S_p^z)^2 \rangle - mQ = {} & \delta_{gp} m(1-Q) \\ & + \beta m(1-Q)[I_1(gp)(Q-m^2) \\ & + I_2(gp) Q(1-Q)] \\ & + \tfrac{1}{2}\beta I_3(gp)(1-Q)(m^2+Q^2-2m^2Q) \\ & + O(\beta^2) \ .\end{aligned} \tag{2.4}$$

As it turns out, these results to the first order in β are equivalent to the MFA.[14]

An equivalent problem to that described by Eq. (1.6) is the ternary alloy of arbitrary concentration.[14] The ternary alloy formalism affords us a particularly convenient way of describing the kinematic constraints on m and Q. Solving for the free energy would enable us to calculate m and Q explicitly. However, since we have not done this here we consider the limits for these parameters by way of kinematic constraints.

Now in the ternary alloy the concentrations of components A, B, and C are given by m_A, m_B, and m_C, respectively. The concentrations are positive and obey the sum rule

$$m_A + m_B + m_C = 1 \ . \tag{2.5}$$

We can relate the concentrations of the ternary alloy to the order parameters of the GIM by[14]

$$m_A = \tfrac{1}{2}(Q+m) \ , \tag{2.6a}$$

$$m_B = 1 - Q \ , \tag{2.6b}$$

$$m_C = \tfrac{1}{2}(Q-m) \ . \tag{2.6c}$$

We immediately see (Fig. 1) that we have the

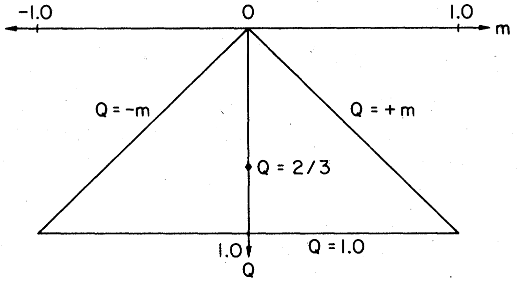

FIG. 1. Kinematically allowed region defined by Eq. (2.7) in the $\tau = 0$ plane. The Potts point $m = 0$, $Q = \frac{2}{3}$ is indicated.

kinematical constraints on m and Q,

$$0 \leq m \leq Q \ , \tag{2.7a}$$

$$0 \leq Q \leq 1 \ , \tag{2.7b}$$

$$0 \leq -m \leq Q \ . \tag{2.7c}$$

Basically, this is the usual Gibbs triangle for the concentrations in the ternary alloy. We also note that in the zero-field limit ($h_1 = h_2 = 0$),

$$m = 0 \tag{2.8a}$$

and

$$Q = \tfrac{2}{3} \ , \tag{2.8b}$$

which are the usual paramagnetic results for spin one and in the ternary alloy problem corresponds to the equicomposition limit. We have indicated this point, Eq. (2.8), in Fig. 1. The limit of $m = Q = 0$ corresponds to a pure B system, while $Q = +1$, $m = -1$ and $Q = +1$, $m = +1$ correspond to pure C and A, respectively.

We are now ready to calculate the stability temperature using the correlation functions.

III. CORRELATION-FUNCTION APPROACH

The correlation functions are the natural variables for describing a system if we approach the transition temperature from the high, or disordered side. Thus we see from Eqs. (2.2)–(2.4) that if $T \rightarrow \infty$ ($\beta \rightarrow 0$) and $g \neq p$, there are no correlations in the system, and in some sense it is behaving as an ideal gas (no interactions). However, as the temperature is lowered the interactions come into play, and the system becomes more like a real gas. Local regions of clustering or ordering appear and many-site correlations come into play. This behavior is built into the high-temperature expansions of Sec. II in a self-consistent way.[14,28]

Finally, as the temperature is lowered further the correlation function will possibly diverge[4] at some temperature, T_s. We can represent this divergence by recognizing that the Fourier transform of the correlation function is proportional to the static susceptibility, or scattering intensity,[32] i.e.,

$$\chi(\vec{k}) = \sum_{g,p} \chi(gp) e^{-i\vec{k}\cdot(\vec{g}-\vec{p})} \ , \tag{3.1a}$$

$$\chi(gp) = \frac{1}{N} \sum_{\vec{k}} \chi(k) e^{i\vec{k}\cdot(\vec{g}-\vec{p})} \ , \tag{3.1b}$$

where

$$\chi(gp) = \langle S_g^z S_p^z \rangle - m^2 \ . \tag{3.2}$$

Thus the divergence in the correlation function can be written

$$\chi(\vec{k}) \rightarrow \infty \tag{3.3}$$

as $\vec{k} \rightarrow \vec{k}_s$ and $T \rightarrow T_s$. Here $\vec{k}_s$ is a lattice vector associated with the ground state, i.e., $\vec{k}_s = (0,0,0)$ for a ferromagnetic ground state (phase separation) and $\vec{k}_s$ is finite for an antiferromagnetic ground state (ordered system).

For the spin-one GIM we have in general three pair-correlation functions, or susceptibilities, given by Eqs. (2.2)–(2.4). Consequently, instead of dealing only with the usual susceptibility above, i.e., Eq. (3.2), we must consider all three. Tahir-Kheli[33] has recently proposed, in the context of the ternary alloy problem, that we should consider the divergences in the correlation matrix, i.e.,

$$|\underline{\chi}(\vec{k})| \rightarrow \infty \ . \tag{3.4}$$

In other words, we should look at the behavior of the correlation matrix,

$$\chi_{\alpha\beta}(gp) = \langle (S_g^z)^\alpha (S_p^z)^\beta \rangle - \langle (S_g^z)^\alpha \rangle \langle (S_p^z)^\beta \rangle \ , \tag{3.5}$$

where $\alpha, \beta = 1, 2, \ldots, 2S$, instead of the individual susceptibilities.

The Fourier-transform of Eq. (3.5) can then be expanded in a high-temperature series as in Eqs. (2.2)–(2.4),

$$\underline{\chi}(\vec{k}) = \underline{\chi}_0 + \beta \underline{\chi}_1(\vec{k}) + \cdots \ . \tag{3.6}$$

We note that Eq. (3.6) does not exhibit singularities as $T \rightarrow T_s$ and $\vec{k} \rightarrow \vec{k}_s$. In order to introduce singularities we therefore perform a self-consistent series inversion on Eq. (3.6) so that

$$\underline{\chi}(\vec{k}) = \underline{\Delta}[\underline{1} - \beta \underline{\gamma}_1(\vec{k}) - \cdots]^{-1} \ . \tag{3.7}$$

[For details of the series inversion, as well as justifications, for binary systems see Ref. 28.] $\underline{\Delta}$ is a wave-vector independent normalization constant determined by the conservation of the scattering intensity, i.e.,

$$\frac{1}{N} \sum_{\vec{k}} \underline{\chi}(\vec{k}) = \underline{\chi}_0 \ . \tag{3.8}$$

We see from Eq. (3.7) that the divergence in the magnitude of the scattering intensity, i.e., Eq. (3.4), can thus be written

$$|\underline{1}-\beta\underline{\gamma}_1(\vec{k})-\cdots|\to 0 \ , \tag{3.9}$$

as $T\to T_s$ and $\vec{k}\to\vec{k}_s$. The resultant equation is a polynomial for β_s^{-1} as a function of the order parameters and the interactions.[34]

This technique has recently been used to look at the ternary alloy in the MFA[33] and for the first-order corrections to the MFA.[21] Also the influence of three-body potentials on Eq. (1.6) has been considered in the MFA for the tenary alloy.[6] The technique, which we have called the correlation-function approach, has also been applied to quaternary ($S=\frac{3}{2}$) alloys.[34]

Applying this technique to the spin-one GIM we formally have for the divergence in the static generalized susceptibility,

$$\begin{vmatrix}\chi_{11}(\vec{k}_s) & \chi_{12}(\vec{k}_s)\\ \chi_{21}(\vec{k}_s) & \chi_{22}(\vec{k}_s)\end{vmatrix}=\infty \ . \tag{3.10}$$

Substituting from the series-expansion results for $\chi_{\alpha\beta}(\vec{k}_s)$, i.e., Eqs. (2.2)–(2.4), we can solve Eq. (3.10), or (3.9), to yield

$$\begin{aligned}\beta_s^{-2}-\beta_s^{-1}[&J_1(\vec{k}_s)(Q-m^2)+J_2(\vec{k}_s)Q(1-Q)+J_3(\vec{k}_s)m(1-Q)]\\ &-(1-Q)(m^2-Q^2)[J_1(\vec{k}_s)J_2(\vec{k}_s)-\tfrac{1}{4}J_3(\vec{k}_s)^2]=0 \ ,\end{aligned} \tag{3.11}$$

where $J_j(\vec{k}_s)$ are the Fourier transforms of the interactions $I_j(gp)$. Generally, in the MFA, the solution to Eq. (3.4) is a polynomial of order $2S$ for a spin-S GIM.

The solution to Eq. (3.11) can immediately be written

$$\begin{aligned}\beta_s^{-1}=\tfrac{1}{2}[&J_1(\vec{k}_s)(Q-m^2)+J_2(\vec{k}_s)Q(1-Q)+J_3(\vec{k}_s)m(1-Q)]\\ &\pm\tfrac{1}{2}\{[J_1(\vec{k}_s)(Q-m^2)+J_2(\vec{k}_s)Q(1-Q)+J_3(\vec{k}_s)m(1-Q)]^2\\ &\qquad+4(1-Q)(m^2-Q^2)[J_1(\vec{k}_s)J_2(\vec{k}_s)-\tfrac{1}{4}J_3(\vec{k}_s)^2]\}^{1/2} \ .\end{aligned} \tag{3.12}$$

It is perhaps worthwhile re-emphasizing that this solution is the most general one for the spin-one GIM with two-body interactions within the MFA.

A few words may be in order here concerning the approximations used in the series expansion (3.6) and the series inversion (3.7). [A more detailed discussion is given in Refs. 14 and 28.] First, as mentioned in Sec. II, in order to accommodate arbitrary external fields we have allowed the expansion coefficients to be temperature dependent.[24] Thus Eq. (3.6) should not be looked at as a controlled term-by-term expansion in β, although it is a self-consistent expansion. Second, the series inversion (3.7) is undertaken so that, as with the expansion (3.6), the sum rule (3.8) is obeyed. This causes the normalization factor, $\underline{\Delta}$, to be temperature dependent. Even though Eqs. (3.6) and (3.7) are related in a self-consistent way, there is consequently an additional uncertainty in the order of β in Eq. (3.7). The self-consistency is maintained between Eqs. (3.6) and (3.7) as long as the wave-vector dependence is given as in Eq. (3.7).[28] Finally, the technique and the solutions, Eq. (3.12), given here for an expansion to first order in β can be justified *a posteriori* by noting that the results for two,[28] three,[6,21,33] and four[34] component systems agree with their corresponding regular solution (MFA) results.

Thus the technique used here employs an uncontrolled, but self-consistent, series expansion in β. Consequently the stability condition (3.9) should be viewed as a necessary condition for the existence of solutions, and not in terms of a controlled series expansion in β, i.e., expansions of $\underline{\chi}(\vec{k})$ correct to order β yield a polynomial of order $2S$ in β_s.

IV. STABILITY SURFACES

The correlation-function approach of Sec. III can be related to the system thermodynamics by using the thermodynamic sum rule[35]

$$\chi_{\alpha\beta}(\vec{k})=Nk_BT\left(\frac{\partial^2 f(\vec{k})}{\partial\sigma_\alpha\partial\sigma_\beta}\right)^{-1} \ , \tag{4.1}$$

as $T\to T_s$ and $\vec{k}\to\vec{k}_s$. Here $f(\vec{k})$ is the free-energy density and the general order parameters are

$$\sigma_\alpha\equiv\langle(S_g^z)^\alpha\rangle \ . \tag{4.2}$$

Thus we see that the condition for the divergence in $|\underline{\chi}(\vec{k})|$ is equivalent to

$$\left|\frac{\partial^2 f(\vec{k})}{\partial\sigma_\alpha\partial\sigma_\beta}\right|\to 0 \ , \tag{4.3}$$

which is the equation for the stability surface in the classical Gibbs sense. Solution of Eq. (4.3) yields a $(2S+1)$-dimensional surface in the space of $[T,\sigma_\alpha; \alpha=1,2,\ldots,2S]$ which separates stable, or metastable, regions from unstable regions. The spirit of the CFA is to calculate $\chi(\vec{k})$. As mentioned above, the usual approach is to calculate $f(\vec{k})$, and then $\chi(\vec{k})$.

The properties of a phase transition for a system governed by a free energy with a single-order parameter,[1,8] σ, are determined from

$$\frac{\partial f}{\partial \sigma}=0 \qquad (4.4)$$

and

$$\frac{\partial^2 f}{\partial \sigma^2} \geq 0 \ . \qquad (4.5)$$

The first condition determines the extremum condition and yields the magnetization curve, while the second condition insures that the extremum is a minimum. Using the equality in Eq. (4.5) gives the stability curve. When the two equalities are simultaneously satisfied we have a continuous phase transition. If we are interested in first-order transitions or in tricritical points we need additional information: particularly the free energy. However, regardless of the type of transition knowledge of Eq. (4.5) is a necessary, but not a sufficient, condition for a phase transition since it separates regions of stability from regions of instability.

For the spin-one GIM this stability surface is given by

$$\begin{vmatrix} \dfrac{\partial^2 f(\vec{k}_s)}{\partial m^2} & \dfrac{\partial^2 f(\vec{k}_s)}{\partial m\,\partial Q} \\ \dfrac{\partial^2 f(\vec{k}_s)}{\partial Q\,\partial m} & \dfrac{\partial^2 f(\vec{k}_s)}{\partial Q^2} \end{vmatrix} = 0 \ , \qquad (4.6)$$

which is a surface in the three-dimensional space of (T,m,Q). By Eq. (4.1) we see that this surface is given by Eq. (3.12).

In order to demonstrate the properties of the solutions to Eq. (3.12) we will restrict ourselves to nearest-neighbor "ferromagnetic" interactions, and a "ferromagnetic" (phase separating) ground state, i.e.,

$$\vec{k}_s=(0,0,0) \ , \qquad (4.7a)$$

$$J_j(\vec{k}_s)=zJ_j>0 \ , \qquad (4.7b)$$

where z is the number of nearest neighbors. It would be straightforward to include further neighbor effects in Eq. (3.12). There has been a fair amount of work on these types of systems within the context of the ternary alloy.[36–38]

We again note that the phase diagram is in the thermodynamic space of (T,m,Q). A good deal of previous work on critical surfaces has been in the space of fields, (T,h_1,h_2),[15,39] rather than densities. However most work in ternary alloys has used the space of densities,[36] or order parameters, (T,m_A,m_B). In actual practice measured variables are usually a mixture of the densities and the fields.

In order to make contact with previous work on three-state systems we will consider a number of specific limits to the general result before considering the full solution.

A. Spin-one Ising model

In this case we set $J_2=J_3=0$ and set the field conjugate to $(S_g^z)^2$ to zero, i.e., $h_2=0$. The GIM then assumes the form of the traditional $S=\frac{1}{2}$ Ising Hamiltonian, (1.1). We immediately obtain from Eq. (3.12)

$$\tau(+)=Q-m^2 \ , \qquad (4.8a)$$

$$\tau(-)=0 \ , \qquad (4.8b)$$

where we have defined,

$$\tau(\pm)\equiv[\beta_s(\pm)zJ_1]^{-1} \ . \qquad (4.9)$$

The solution $\tau(+)$ is restricted by the fact that we have set $h_2=0$. This specifies a particular kinematic relationship between m and Q. From the series expansion for the correlation functions[14] we obtain

$$Q=\tfrac{4}{3}-\tfrac{1}{3}(4-3m^2)^{1/2} \ , \qquad (4.10)$$

which, when placed in Eq. (4.8), yields

$$\tau(m)=\tfrac{1}{3}(4-3m^2)-\tfrac{1}{3}(4-3m^2)^{1/2} \ . \qquad (4.11)$$

We have plotted the solutions to Eq. (4.11) in Fig.

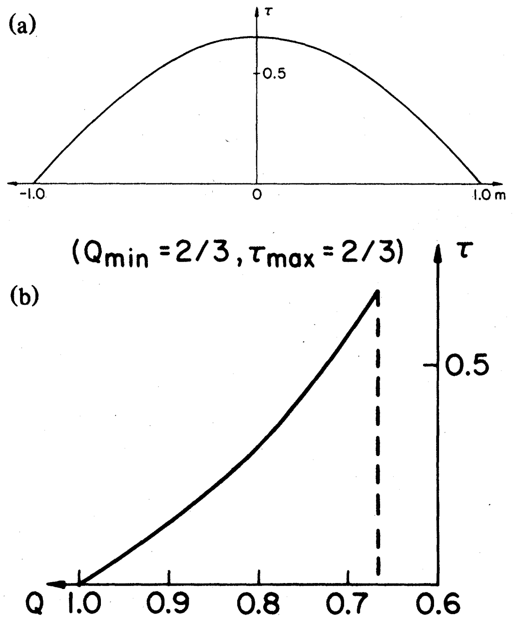

FIG. 2. Stability curve for the spin-one Ising model, Eq. (4.1). (a) The projection on the (τ,m) plane; (b) the projection of the (τ,Q) plane.

2. We note that Q goes from 2/3 at $m=0$ to 1 at $m=\pm 1$. $\tau(m)$ is a maximum when $m=0$, or $\tau(0)=2/3$. Regions below the curve are unstable with respect to fluctuations in the order parameters.

B. Diagonal limit

A related problem to the Ising model is the diagonal limit. Here we want to choose parameters such that the generalized susceptibility assumes a diagonal structure, i.e., $\chi_{12}(\vec{k}_s)=\chi_{21}(\vec{k}_s)=0$. Investigation of Eq. (2.4) reveals that this will occur when $Q=1$, i.e., we have complete quadrupolar ordering, or when very special relationships exist among the interactions. Assuming that the former is the case we set $Q=1$. This causes $\chi_{22}(\vec{k}_s)$ to vanish also and the interactions J_2 and J_3 drop out. Thus we are left with what will turn out to be a special case of the BCM. The stability temperatures become

$$\tau(+)=1-m^2 \ , \tag{4.12a}$$

$$\tau(-)=0 \ . \tag{4.12b}$$

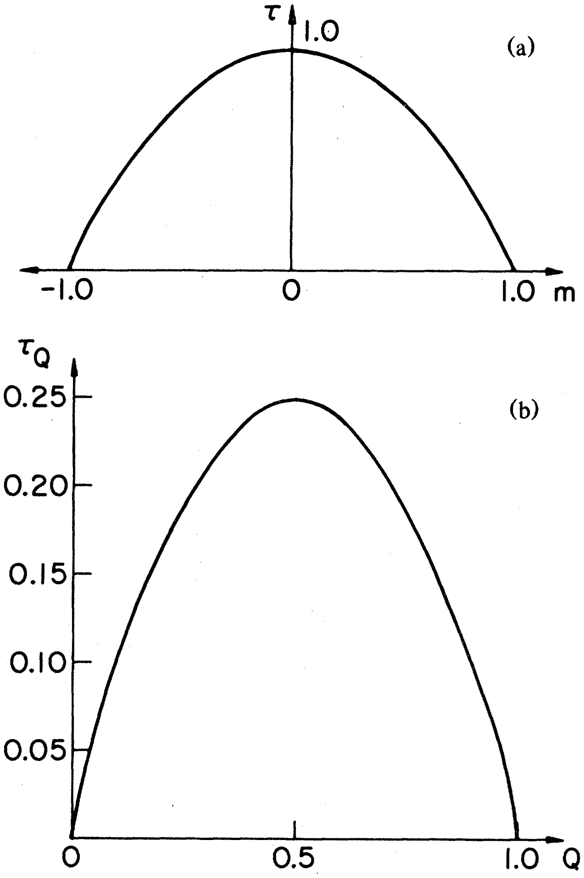

FIG. 3. (a) Stability curve for the diagonal limit, Eq. (4.12); (b) the stability curve for pure quadrupolar interactions, Eq. (4.13).

We have plotted this solution in Fig. 3(a). We note that the condition $Q=1$ corresponds to $m_B=0$, and consequently we have an AC mixture, or the stability curve for a spin-$\frac{1}{2}$ Ising model. If the disordered mixture is quenched across the stability curve the system will phase separate to a point on the stability curve determined by initial conditions.

C. Pure quadrupolar model

Setting $J_1=J_3=0$ we find that

$$\tau_Q(+)\equiv[\beta_s(+)zJ_2]^{-1}=Q(1-Q) \ , \tag{4.13a}$$

$$\tau_Q(-)=0 \ . \tag{4.13b}$$

These solutions should correspond to a magnetic system which has pure quadrupolar ordering. Since m does not appear in the solution it is effectively zero. Thus we effectively have $h_1=0$, or $m_A=m_C$ and $Q=1-m_B=2m_A$. We have plotted the parabola $\tau_Q(+)$ in Fig. 3(b). The peak occurs at $Q=\frac{1}{2}$, $\tau_Q(+)=\frac{1}{4}$.

D. BCM

The BCM is arrived at by setting $J_2=J_3=0$ in Eq. (1.6) and requiring that $h_2\neq 0$. If $Q=1$ we obtain the diagonal limit. We can have either $m=0$ or $m\neq 0$ depending on whether the field h_1 is zero or nonzero. For $m\neq 0$ $(m_A\neq m_C)$ we have

$$\tau(+)=Q-m^2 \tag{4.14a}$$

and

$$\tau(-)=0 \ . \tag{4.14b}$$

The stability surface in this case is a bisected paraboloid of revolution having its apex at the origin $(Q=0)$ and its base in the plane $Q=1$. We have plotted projections of this surface in Fig. 4. In the limit that $m=0$ $(m_A=m_C)$, $\tau(+)$ degenerates to a straight line in the (τ,Q) plane, i.e.,

$$\tau(+)=Q \ , \tag{4.15a}$$

$$\tau(-)=0 \ . \tag{4.15b}$$

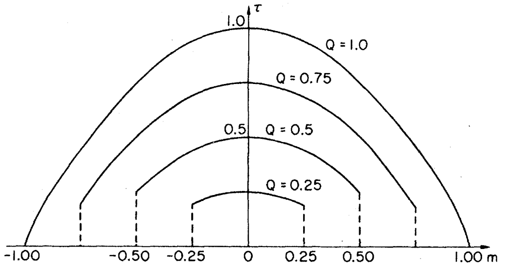

FIG. 4. Stability curves for the BCM. We are looking along the Q axis at the projections in the τ-m plane. The dashed lines indicate the kinematic constraints.

We see from Fig. 4 that for $Q < 1$ the stability surface is cutoff by the kinematic conditions (2.7). What this means is that for a given $Q = Q_0$, if the system is quenched through a temperature difference, below $\tau_{\max} = Q_0$, greater than $\Delta\tau = Q_0^2$, the system will have to move to a new value of $Q < Q_0$ in order to achieve stability.

E. BEG model

In this case we set $J_3 = 0$ and Eq. (3.12) readily becomes

$$\tau(\pm) = \tfrac{1}{2}[Q - m^2 + \alpha_{21} Q(1-Q)] \pm \tfrac{1}{2}\{[Q - m^2 + \alpha_{21} Q(1-Q)]^2 + 4(1-Q)(m^2 - Q^2)\alpha_{21}\}^{1/2} , \quad (4.16)$$

where we have set $\alpha_{21} = J_2/J_1$. In the limit that $\alpha_{21} \to 0$ we regain the BCM, and in the limit that $Q \to 1$ we regain the diagonal limit.

For the symmetric case ($m = 0$, or $m_A = m_C$), Eq. (4.16) becomes

$$\tau(+) = \alpha_{21} Q(1-Q) , \quad (4.17a)$$

$$\tau(-) = Q . \quad (4.17b)$$

We have plotted Eq. (4.17) in Fig. 5. We denote the point where we have simultaneous solutions of Eq. (4.17) as τ_D, the degeneracy temperature. This point may, or may not, be a tricritical point. For this case we have

$$\tau_D = 1 - \alpha_{21}^{-1} = Q = \frac{J_2 - J_1}{J_2} , \quad (4.18)$$

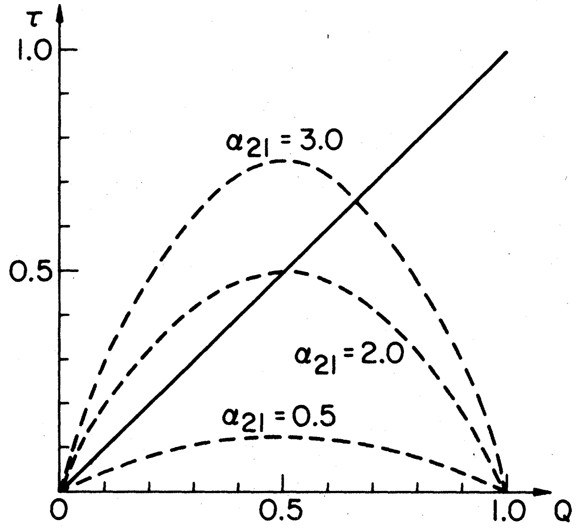

FIG. 5. Stability curves for the BEG model for various values of $\alpha_{21} = J_2/J_1$ and $m = 0$. The solid line is $\tau(-) = Q$; the dashed line $\tau(+) = \alpha_{21} Q(1-Q)$.

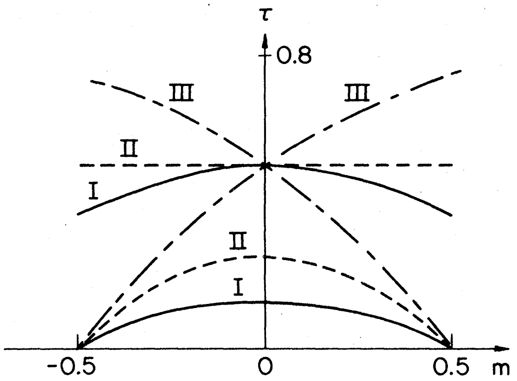

FIG. 6. Stability curves for the BEG model for $Q = 0.5$ and $m \neq 0$. I (solid line), II (dashed line), and III (dot-dash) correspond to $\alpha_{21} = \frac{1}{2}$, 1, and 2, respectively.

which, since $Q > 0$, requires that $J_2 > J_1$.

We have plotted the general solution, Eq. (4.16), for the BEG model ($m \neq 0$) in Fig. 6 for the particular case $Q = 0.5$ and for various values of α_{21}. Setting $\tau(+) = \tau(-)$ from Eq. (4.16) shows that there exist no degeneracy temperatures for $m \neq 0$.

F. Potts symmetry

The Potts symmetry[11,40] requires symmetry between interchange of particles in a three component system, i.e., $m_A = m_B = m_C$. We see from Eq. (2.6) that this requires $h_1 = h_2 = 0$, or

$$m = m_A - m_C = 0 \quad (4.19a)$$

and

$$Q = m_A + m_C = \tfrac{2}{3} , \quad (4.19b)$$

or the point in Fig. 1. We also require that the odd interaction vanish ($J_3 = 0$).

We now have a special case of the BEG model, and immediately have for the stability, and degeneracy temperature,

$$\tau_D = \tfrac{2}{3} , \quad (4.20)$$

which from Eq. (4.18) requires that

$$J_2 = 3J_1 . \quad (4.21)$$

G. Mean interaction

We see from Eq. (3.12) that the radical goes to zero if we choose the interactions to be related by their geometric mean, i.e.,

$$J_3 = 2(J_1 J_2)^{1/2} . \quad (4.22)$$

We call this the mean interaction and note that it is basically the same as the Shiba condition[41] in the theory of disordered binary alloys. Consequently Eq. (3.12) becomes

$$\tau(+) = Q - m^2 + \alpha_{21} Q(1-Q) + 2\sqrt{\alpha_{21}}\, m(1-Q) \ , \tag{4.23a}$$

$$\tau(-) = 0 \ . \tag{4.23b}$$

If $\alpha_{21} = 0$, or $Q = 1$, we obtain the results for the BCM and the diagonal limit, respectively. If $m = 0$ ($m_A = m_C$; $h_1 = 0$) we obtain

$$\tau(+) = Q + \alpha_{21} Q(1-Q) \ , \tag{4.24a}$$

$$\tau(-) = 0 \ . \tag{4.24b}$$

We have plotted Eq. (4.24) in Fig. 7(a). For $\alpha_{21} > 1$ we get a peak given by

$$Q_{\max} = (1+\alpha_{21})(2\alpha_{21})^{-1} \ , \tag{4.25a}$$

$$\tau_{\max} = (1+\alpha_{21})^2(4\alpha_{21})^{-1} \ . \tag{4.25b}$$

For $\alpha_{21} > 1$ we have a region,

$$\Delta\tau = \tau_{\max} - 1 = (\alpha_{21}-1)^2(4\alpha_{21})^{-1} \ , \tag{4.26}$$

for which the quenched system can go to either increasing or decreasing Q.

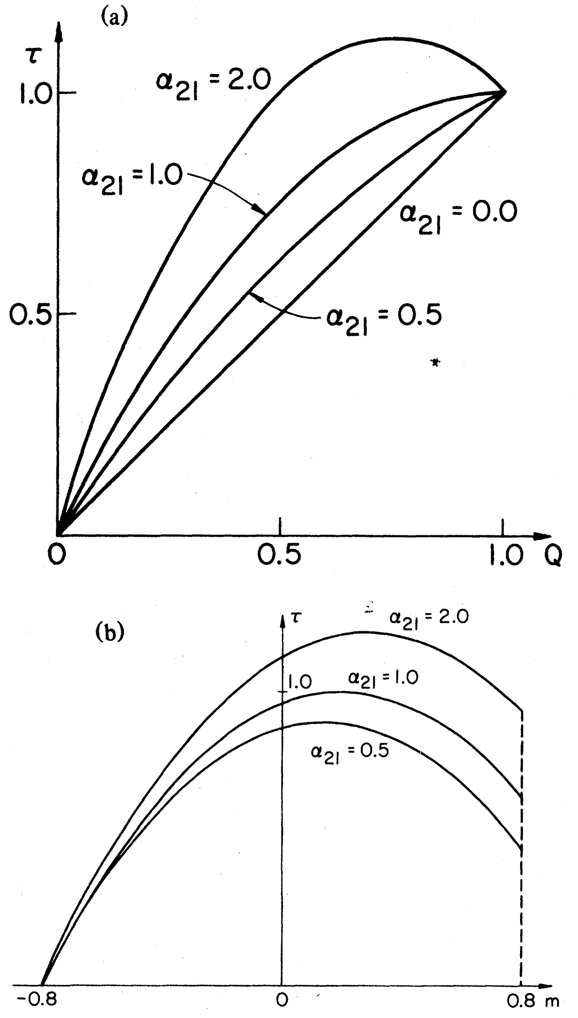

FIG. 7. (a) Stability curves for the mean interaction model for various values of α_{21} and $m = 0$; (b) the stability curves for the mean interaction model for $Q = 0.8$ and various values of α_{21}. The dashed lines indicate kinematic constraints.

The solutions for the case $m \neq 0$, i.e., Eq. (4.23), are plotted in Fig. 7(b) for a particular value of $Q = Q_0$ and for various α_{21}. The maximum value of τ is given by

$$m_{\max} = \sqrt{\alpha_{21}}(1-Q_0) \tag{4.27a}$$

and

$$\tau_{\max} = Q_0 + \alpha_{21}(1-Q_0) \ . \tag{4.27b}$$

Thus if the system is quenched below $\tau_{\max}$ by

$$\Delta\tau = \alpha_{21}(1-Q_0) \ , \tag{4.28}$$

the system must move to negative m, or smaller Q.

H. Uniform interaction

Setting $J_1 = J_2 = J_3$ in Eq. (3.12) the stability temperature becomes

$$\tau(\pm) = \tfrac{1}{2}[(Q-m^2) + Q(1-Q) + m(1-Q)] \pm \tfrac{1}{2}\{[(Q-m^2) + Q(1-Q) + m(1-Q)]^2 + 3(1-Q)(m^2-Q^2)\}^{1/2} \ . \tag{4.29}$$

We note in Fig. 8 the asymmetry of the stability surface with the peak shifting from $m = 0$ to outside of the kinematic region as Q decreases.

I. General solution

We have plotted the general solution (3.12) in Fig. 9 for a given value of α_{31} and Q. The remaining

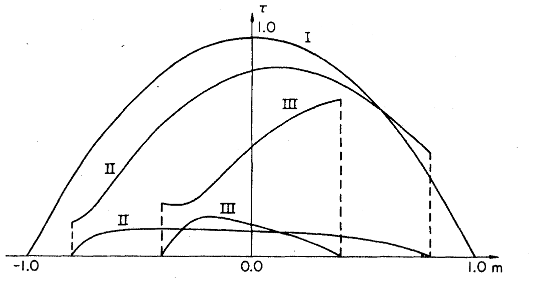

FIG. 8. Stability curves for the uniform interaction model for various values of Q. I, II, and III correspond to $Q = 1$, 0.8, and 0.4, respectively. The dashed lines indicate kinematic constraints.

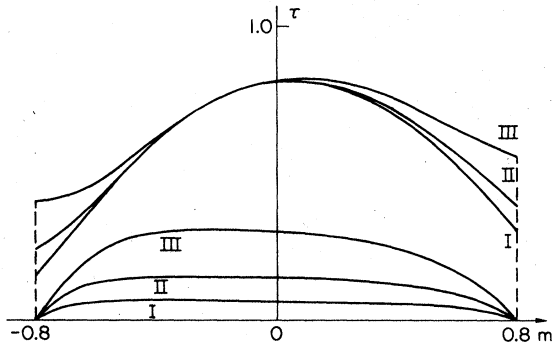

FIG. 9. Typical stability curves for the general solution with $Q=0.8$, $\alpha_{31}=\frac{1}{2}$ and I, II, and III corresponding to $\alpha_{21}=\frac{1}{2}$, 1, and 2, respectively. The dashed lines indicate kinematic constraints.

solutions for α_{31} do not appear to exhibit appreciably different behavior.

V. CONCLUSIONS

Having introduced the concept of the GIM we have seen that we can take advantage of correlation identities which are unique to the GIM in order to generate high-temperature series expansions. These expansions are valid for arbitrary field, and we have seen that the linear expansion yields the MFA. Obviously we should be able to improve on the MFA by going to higher orders in β.[21,42] By using a series inversion and the thermodynamic sum rule we can then construct the stability surface for $(2S+1)$-state systems. We have demonstrated this explicitly within the MFA for the most general three-state GIM with two-body potentials. It would be straightforward to include further neighbor effects and many-body potentials. The stability surfaces so generated are necessary in order to understand the properties of the phase transition and the transformation kinetics.

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